

We thank the reviewer for the careful reading and the useful comments that helped improving our manuscript. All comments have been addressed in the revised version. In particular to address the model resolution comment, we have performed all simulations again in the higher horizontal resolution of our model. The figures have been appropriately redrawn. The major conclusion of the paper remains unchanged since the discussion paper. Discussion has been improved to highlight also the importance of primary dissolved Fe emissions in the computed atmospheric deposition changes.

Specific comments:

1) *Page 3969 lines 2-5: “Sensitivity simulations show that increases in anthropogenic emissions since 1850 resulted in more acidic environment and thus an increase (50 %) in DFe deposition ($0.230 \text{ Tg-Fe yr}^{-1}$ in the past against $0.489 \text{ Tg-Fe yr}^{-1}$ nowadays)”.* I can think of at least three changes that can explain the difference in DFe deposition : 1) The biomass burning is increased by a factor of 10 in the model between preindustrial and present-day, 2) Iron is produced from combustion that appeared during the industrial revolution, 3) Atmospheric processing has changed since the atmospheric composition changed radically from preindustrial to present. Hence the reasons for this increase DFe are multiple and the results of the paper give us the combined result from these processes. The authors need to rewrite the conclusions to make sure there is no misinterpretation from the reader. It would have been even a stronger paper if the different causes for the preindustrial to present and the present to future causes for changes had been disentangled.

- Indeed, the changes in the primary emissions of DFe from biomass burning and anthropogenic combustion are also important for the calculated changes in the deposition of DFe. We have rephrased this part in the conclusions to explicitly discuss the role of primary DFe emission changes to the calculated DFe deposition changes. *“Sensitivity simulations show that increases in anthropogenic and biomass burning emissions since 1850 resulted in both enhanced Fe combustion emissions and a more acidic environment and thus more than double DFe deposition ($\sim 0.213 \text{ Tg-Fe yr}^{-1}$ in the year 1850 against $\sim 0.496 \text{ Tg-Fe yr}^{-1}$ nowadays). Air-quality regulations are projected to decrease anthropogenic emissions and thus atmospheric acidity in 2100. Our model results show a 5-fold decrease in Fe emissions from anthropogenic combustion sources ($\sim 0.013 \text{ Tg-Fe yr}^{-1}$ in the year 2100 against $\sim 0.070 \text{ Tg-Fe yr}^{-1}$ nowadays), and about 45% reduction in mineral Fe dissolution ($\sim 0.078 \text{ Tg-Fe yr}^{-1}$) compared to the present day ($\sim 0.175 \text{ Tg-Fe yr}^{-1}$), while DFe biomass burning emissions are enhanced by 20% ($\sim 0.155 \text{ Tg-Fe yr}^{-1}$ in the year 2100 against $\sim 0.127 \text{ Tg-Fe yr}^{-1}$ nowadays) Overall, the model calculates for 2100 a global DFe deposition of $\sim 0.369 \text{ Tg-Fe yr}^{-1}$ that is lower than the present day deposition.”*
- We have also appropriately changed the discussion in section 4.3 and added the following text in the abstract: *“The calculated changes also show that the atmospheric deposition of DFe supply to the globe has more than doubled since the preindustrial period due to 8-fold increases in the primary non-dust emissions and about 3-fold increase in the dust-Fe dissolution flux. However, the DFe*

deposition flux is expected to decrease (by about 25%) due to reductions in the primary non-dust emissions (about 15%) and in the dust-Fe dissolution flux (about 56%). Over the global ocean in present atmospheric deposition of DFe is calculated to be about 3 times higher than for 1850 emissions and about 30% decrease is projected for 2100 emissions. These changes are expected to impact most on the High Nutrient Low Chlorophyll oceanic regions”

2) *For the aspect of presentation, the Figures that 2D-maps (Fig. 2, 3, 4, 6, 8 & 9) are much too small to be captured by the naked eye. The reader has to use magnifying glasses to see them. An effort should be put on these Figures, together with the presentation of Fig. 5 that permits to evaluate the model against observations.*

- *We have tried to improve the readability of the Figures. In particular we have increased the fonts in Figure 5, since they were indeed very small. The other figures are of good quality and they can be magnified as needed to fit the full page available when they will be inserted in the main text for the final edited manuscript. In addition the journal is electronic which allows the magnification of any figure before printing.*

3) *The authors’ state that the comparison with the Atlantic Ocean TFe deposition is satisfying (Figures 7a through d). They should comment on the very large overestimate seen in Regions 2 and 3 in the periods April-May-June and Sept-Oct.-Nov. Such difference with the observations of Baker et al. (2013) needs to be resolved or at least noticed to try to advance our understanding of dissolved iron.*

- *Both regions 2 and 3 are strongly affected by Sahara dust outflow. Thus the model overestimate of TFe observations by Baker et al. (2013), while DFe observations are much better captured by the model, could be due to a longer lifetime of TFe in the model than in the atmosphere resulting from smaller size distributions of TFe in the model than in reality. Note also that these estimates are associated with large uncertainty. This is now discussed in section 3.6.*

4) *The regions in Figure 4c where the ratio of %Fe(II)/DFe is greater than 10% are regions for which dust concentrations are very small and this should be explicitly noted in the text.*

- *We rephrased as: “This ratio also exceeds ~10% at several other locations around the globe, in particular over the tropical Pacific and the Southern Ocean; implying that chemical aging of dust due to atmospheric processing and long-range transport enhances significantly the production of Fe(II). As also discussed in Sec. 2.6, in relatively basic pH environments (e.g the Southern Ocean due to the buffering capacity of sea-salt particles; see Fig. S2) and due to high OXL concentrations (e.g. tropical Pacific ocean) the production of Fe(II) is favoured (Fig. S4e and Fig. S4h, respectively). Thus, our model calculations indicate that the enhanced fraction of Fe(II) over the remote oceans (Fig. 4c), characterized by low concentrations of dust and non-negligible OXL concentrations (see Fig. S3) due to the aqueous-phase oxidation of organic compounds of marine origin NMVOCs (e.g. isoprene) could be attributed to the production of ferrous-oxalato complexes.”*

Minor comments:

1) Page 3496, line 15: change, ‘At the surface waters, the phytoplankton photosynthetic activity uses CO₂ and nutrients. . .’ to ‘In surface waters, the phytoplankton photosynthetic activity uses CO₂ and nutrients. . .’

- Corrected

2) Page 3947 line 20 : ‘3% of Hematite in dust’ please indicate if this content refers to mass or to volume.

- Clarification has been added and typo corrected (see also Reply to rev. #1). It now reads: ‘5% mass fraction of Hematite in dust’

3) Page 3948, lines 3 to 6 : ‘ However, the large acid buffering ability of the carbonate from minerals like CaCO₃ and MgCO₃ in coarse dust particles can regulate mineral-Fe proton promoted dissolution, creating an inverse relationship between SFe and particle size (Ito and Feng, 2010). ’ There is much debate about the cause for this inverse relationship, several factors are at play and mineral-Fe proton promoted dissolution is just one of them. Be more conservative when putting this cause forward.

- This sentence has been rephrased as follows: ‘However, the buffering capacity of minerals like CaCO₃ and MgCO₃ which reside in coarse dust particles may regulate mineral-Fe proton-promoted dissolution, contributing, among others together with combustion emissions of DFe on fine particles and atmospheric transport, to the observed an inverse relationship between SFe and particle size (Ito and Feng, 2010)’

4) You use a model resolution of 6° in longitude by 4° in latitude (line 27 page 3950). You should state that this coarse resolution will smooth the gradient in dust and Fe concentrations in or near-source regions.

- We agree with the reviewer that the highest is the spatial resolution of the model, the more accurate are the results. Therefore, for the revised version of this paper, we have performed the simulations in the fine resolution of the model (3x2 with 34 hybrid levels in the vertical). About 50% of uncertainty in the computed atmospheric deposition is associated with the model horizontal resolution. A comment has been added in section 3.5: ‘In addition, at least a 50% of uncertainty in the calculated deposition fluxes was found to be associated with the applied horizontal resolution of the model, with higher fluxes calculated with the higher model resolution.’

5) Page 3952, line 6 The iron-content for hematite and goethite differ by 7% (62.8% for goethite and 69.94% for hematite (see <http://webmineral.com>), this is worth taking into account rather than using the same content of 66% in your computations.

- The Nickovic et al. (2012) mineralogy database that we are using provides the distribution of the sum of Hematite and Goethite (as iron oxides) with a mean iron-content of 66%. Therefore we do not have the information needed to split the

database in hematite and goethite. However, a relevant comment has been added in section 2.2: “Despite differences in the chemical reactivity and iron content of goethite and hematite (e.g. see <http://webmineral.com>), these minerals are here considered as one surrogate species, the hematite, used as proxy for Fe oxides as suggested by Nickovic et al. (2012).”

6) In Table 1, you have to clearly point out that it is a Fe content that you refer to and not the relative abundance of the mineral in dust. I got confused by it the first time I read through this Table.

- To avoid confusion, the title of this Table has been modified and it is now clearly stated that the emissions of Iron contained in dust minerals are shown.

7) Page 3957 lines 14-15: Please indicate the percentage of content of Fe in dust that is calculated here (as a global mean).

- In section 2.2, first paragraph, we mention that:
“Given this, the annual global mean Fe content of emitted dust particles in TM4-ECPL is calculated to be ~ 3.2 %”

8) In paragraph 3.5 the maximum values of deposition you give are strongly linked to the model resolution. With a higher model resolution, your values would be increased. It is worth mentioning it here.

- see reply to comment 4.

9) Page 3967, bottom of page: What method did you use to delimit the HNLC regions?

- For the characterisation of the HNLC oceanic regions for the present study, the annual mean global NO_3^- surface water concentrations from the LEVITUS94 World Ocean Atlas (<http://iridl.ldeo.columbia.edu/SOURCES/.LEVITUS94/>) and the monthly chlorophyll a (Chl a) concentrations MODIS retrievals taken into account in the model (Myriokefalitakis et al., 2010) for the year 2008 have been used. As we explain in p. 3967 line 16 of the discussion paper (1st paragraph section 4.3) the model grid boxes corresponding to HNLC waters are here defined based on the co-occurrence of surface seawater NO_3^- concentrations of $> 4 \mu\text{M}$ (Duce et al., 2008) and Chl-a concentrations of $< 0.1 \text{ mg m}^{-3}$ (Boyd et al., 2007). This is an off-line calculation and the figure of HNLC regions as delimited for the present study is now added in the supplement – New figure S7e.