

Response to Referee #1

We appreciate Dr. Mónica Zamora Zapatas's helpful and constructive comments and accordingly improve our manuscript. The Referee's comments are marked in blue.

This study shows the effects of varying the iron emission size distribution on global-scale simulations to diagnose iron concentration in the atmosphere and ocean deposition. Four different size distributions are studied, properly justified, and their results are compared to aircraft observations, allowing them to conclude on the closest simulated distributions. The methods lack some details about the iron emission and deposition processes. Finally, some comments on the possible biases of using a global model could be useful, reflecting on the possibility of higher resolution models being helpful in further elucidating this phenomenon. Therefore, I recommend a minor revision. Some minor comments follow:

Response: Accepted. We add more description about the iron emission and deposition processes (Please see Line 104-107 and Line 92-93). The higher resolution models can be used at regions scales to explore the deposition and chemical aging processes over different polluted environments (Please see Line 345-346). Detailed revisions are shown in the following.

Revisions:

Line 92-93: "Both dry deposition (Zhang et al., 2001) and wet deposition (Liu et al., 2012) of aerosols are treated in our model. The improvement of aerosol in-cloud wet scavenging process was included to improve the modelling of aerosol long-range transport efficiency (Liu and Matsui, 2021)."

Line 104-107: "For anthropogenic iron, we consider five different minerals, namely magnetite, hematite, illite, kaolinite, and sulfate iron, following the global emission inventory by Rathod et al. (2020), which was developed by a bottom-up approach at 1° spatial resolution and 1-month temporal resolution. The reference year of the inventory was 2010. We account for a wide range of anthropogenic sources including iron smelting and fossil fuel combustion sources."

Line 345-346: "Higher resolution models with finer grids and detailed microphysics are useful to explore iron aerosol deposition and chemical aging processes at regional scales."

In the abstract, it would be great to highlight which findings are new.

Response: Accepted. This study, by performing a series of global aerosol simulations, demonstrates the importance of representing iron aerosol size distributions at emission in understanding global iron deposition to the ocean. This property has been overlooked in previous modelling analysis and should be well treated to narrow model uncertainties. Please see the revised statement at Line 21-26.

Revisions:

Line 21-26: “Shifting the iron aerosol size distribution toward finer particle sizes (<1 μm) enables longer atmospheric lifetime (a doubling), promoting atmospheric processing that enhances the soluble iron deposition to ocean basins by up to 50% on an annual basis. The monthly enhancements reach 110% and 80% over the Southern Ocean and North Pacific Ocean, respectively. Uniquely, our results highlight that compared with emission flux variability, iron emission size distribution plays an equally important role in regulating soluble iron deposition, especially to the remote oceans. Our new findings can help to interpret inter-model differences in iron deposition estimation and to better quantify the effects of atmospheric nutrients input on marine biogeochemistry, including but not limited to iron, phosphorus, and others.”

L55 are all of these papers working with global models? Is there a variety of local to global approaches and if so, how does grid resolution and microphysics models differ?

Response: Accepted. All of those papers applied global models. Only global chemistry transport and general circulation models can be used to reflect the long-range transport effect on iron aerosol distribution. Therefore, we present them to explore why the size distribution of iron is a critical factor. Grid resolution and microphysics may be also important, but not the scope of this study, and we mention them in the discussion part (Line 345-346). Please see the revised sentence at Line 55-56 and Line 59-61.

Revisions:

Line 55-56: “To elucidate atmospheric flux of iron-containing aerosols to the ocean, global-scale aerosol models have been developed to include a range of iron emission sources that currently show a large intermodal difference in flux estimates (Myriokefalitakis et al., 2018).”

Line 59-61: “Among those global aerosol simulations, the size distribution of iron, which is an important consideration when determining aerosol lifetimes and thus its long-range transport potential, is key in shaping atmospheric iron distributions (Hamilton et al., 2020a; Myriokefalitakis et al., 2018).”

Line 345-346: “Higher resolution models with finer grids and detailed microphysics are useful to explore iron aerosol deposition and chemical aging processes at regional scales.”

L118 Is it also possible that iron emissions have a different size distribution in other parts of the world? How could this simplification be considered for future work?

Response: Accepted. We tested four different size distributions for anthropogenic iron, all of which were applied on a global scale. This method is commonly used in previous modeling works. Applying region-specific size distributions may be more realistic but challenging due to limited knowledge about that. We add more discussions on this uncertainty. Please see Line 152-154 and Line 281-282.

Revisions:

Line 152-154: “To enable the intercomparison among these cases, we used the global-

scale anthropogenic iron emission mass inventory from Rathod et al. (2020) but with different allocations between fine and coarse sizes in each case. The size distribution of anthropogenic iron emission in each case was treated uniformly on a global scale.”

Line 281-282: “Because the size distributions of anthropogenic iron minerals may depend on different combustion processes, source- and region-specific size distribution representation is desirable in the future work.”

L95 I don't completely follow how the emissions are prescribed or calculated. Is it homogeneous over all surfaces in the world? Same with its absorption. Do they change over time?

Response: We would like to state that in this paper, the emission means the amount of iron particles released into the atmosphere during the fossil fuel combustion and industrial activities. The iron emissions are provided by a grid-resolved emission inventory (Rathod et al., 2020). The emission fluxes are different in time and space around the world, with a temporal resolution of one month. The reference year of the inventory is 2010. Absorption is beyond the scope of this study. Please see Line 104-107.

Revisions:

Line 104-107: “For anthropogenic iron, we consider five different minerals, namely magnetite, hematite, illite, kaolinite, and sulfate iron, following the global emission inventory by Rathod et al. (2020), which was developed by a bottom-up approach at 1° spatial resolution and 1-month temporal resolution. The reference year of the inventory was 2010. We account for a wide range of anthropogenic sources including iron smelting and fossil fuel combustion sources.”

Fig. 4,5,6: Are these plots derived from yearly averaged values?

Response: Accepted. Yes, we describe that these plots are made using either yearly averaged or yearly accumulated model results. Please see the revised figure captions for Figs. 4, 5, 6.

Reference:

Rathod, S. D., Hamilton, D. S., Mahowald, N. M., Klimont, Z., Corbett, J. J., and Bond, T. C.: A Mineralogy-Based Anthropogenic Combustion-Iron Emission Inventory, *J. Geophys. Res.-Atmos*, 125, e2019JD032114, 10.1029/2019jd032114, 2020.