Response to Referee #2

This manuscript reports on a numerical modelling study that examines the impact of the size distribution of iron-bearing anthropogenic emissions on the transport and deposition of soluble iron to the oceans. The paper is extremely well written and the work is presented clearly and concisely. I have no hesitation in recommending that it is suitable for publication in ACP with only minor revisions.

Response: We appreciate the reviewer's positive comments and make minor revisions as follows.

Specific Comments

Line 1. The work reported focusses exclusively on the size distribution at emission of anthropogenic iron. The title of the paper does not reflect this. I suggest that "Representation of iron aerosol size distributions of anthropogenic emissions is critical in evaluating atmospheric soluble iron input to the ocean" would give a better representation of the subject of the paper.

Response: Accepted. We revise the title as: Representation of iron aerosol size distributions of anthropogenic emissions is critical in evaluating atmospheric soluble iron input to the ocean.

L 84. Given the dominance of dust as a source of Fe, it might be helpful for the reader to be given a little more detail on the representation (and validation) of the size distribution of dust Fe in the model. Similarly, the model described identifies various Fe-bearing minerals in anthropogenic emissions (L 100 – 101). Presumably at least some of these minerals also occur in dust. How does the model treat these minerals in dust, and how are the anthropogenic and dust fractions handled in the comparison to the aircraft observations (e.g. Fig. 3)?

Response: Accepted. Following these points, we revise the manuscript below.

1) We add more details on the treatment of dust Fe size distribution in our model. As a part of dust aerosols, dust Fe features the same size distribution as dust, derived by Kok (2011). Please see Line 108-110 in the revised manuscript.

2) As commented by the reviewer, this study focuses on anthropogenic iron emission, and natural dust mineralogy was not considered in our simulations. Though some minerals, e.g., magnetite, can come from both anthropogenic and dust emissions, the dust fractions have been excluded from the aircraft observations (by Lamb et al., 2021). So that we can use them directly for comparison with our anthropogenic iron simulations results. We add such sentences at Line 113-116.

Revisions:

Line 108-110: "Dust iron emission was calculated by assuming a constant iron content of 3.5% in dust aerosol emission (Shi et al., 2012). The model estimated total dust emission fluxes using the scheme of Zender et al. (2003), with modifications by Albani et al. (2014) and the size distribution from Kok (2011)."

Line 113-116: “We validated our modeled anthropogenic iron oxide concentrations against a global-scale aircraft measurement in the troposphere consisting of eight campaigns for the periods of 2009–2011 and 2016–2018 (Lamb et al., 2021). These observations provide mass concentrations of anthropogenic iron oxide, i.e., magnetite, with volume equivalent diameters between 180-1290 nm.”

L 96-97. Please comment on the implications of the assumption of internal mixing of Fe with all other aerosol components for your results.

Response: Accepted. This assumption is reasonable for anthropogenic iron aerosols, which are often mixed together with other aerosol compounds in the polluted environments, e.g., East Asia (Li et al., 2017). The internal mixing enables rapid growth of small aerosols into larger ones via condensation and coagulation and makes them deposited more efficiently than aerosols in external mixing state. Please see our added description at Line 100-102.

Revisions:

Line 100-102: “The internal mixing assumption is reasonable for anthropogenic iron aerosols, which are often mixed together with other aerosol compounds in the polluted environments, e.g., East Asia, and enable the growth of iron aerosols via condensation and coagulation.”

L 189 – 190. Please explain this statement further.

Response: Accepted. We would like to state that the size range of aerosols for comparison should be consistent between observation and simulation. The measurement data is available for sub-micron mode of aerosols (i.e., 180-1290 nm in diameter), and correspondingly the model results with the similar size range were extracted. Please see the revised description at Line 113-116 and Line 197-198.

Revisions:

Line 113-116: “We validated our modeled anthropogenic iron oxide concentrations against a global-scale aircraft measurement in the troposphere consisting of eight campaigns for the periods of 2009–2011 and 2016–2018 (Lamb et al., 2021). These observations provide mass concentrations of anthropogenic iron oxide, i.e., magnetite, with volume equivalent diameters between 180-1290 nm.”

Line 197-198: “We extracted the modeled mass concentrations of iron aerosols with the size range similar to that of the measurements.”

L 223 – 224. Perhaps some clarification is needed here? The differences referred to do appear to be more pronounced over much of the global ocean, but not over the Southern Ocean.

Response: Accepted. We reword this sentence and clarify that the differences appear to be more pronounced over much of the global ocean, except the Southern Ocean. Please see Line 230-231.

Revisions:
Line 230-231: "By contrast, the differences for soluble iron are more pronounced than for total iron over much of the global Ocean (Fig 4c), because of the higher solubility of anthropogenic iron than dust iron."

L 225 – 227. Is the solubility enhancement referred to here relevant to dust Fe, anthropogenic Fe, or Fe in general?
Response: Accepted. This refers to anthropogenic Fe. Over East Asia, anthropogenic iron emissions are pronounced and subject to efficient aging processes during transport. Please see Line 231-233.

Revisions:
Line 231-233: "Over East Asia and its outflow areas, the rapid aging process in the polluted environments are capable of enhancing iron solubility, particularly those of anthropogenic origin"

Technical Corrections

L 102. Delete "taken into account".
Response: Deleted. Please see Line 106-107.

L 117. Also add " of anthropogenic emissions" to this section heading?
Response: Corrected. Please see Line 124.

L 139. "A similar…"
Response: Corrected. Please see Line 146. -thanks for correcting typos.

L 190. "… used as an …"
Response: Corrected. Please see Line 199.

L 250. "iron source", rather than "iron emission"?
Response: Corrected. We reword it as "iron source". Please see Line 257.

Fig. 8. Please add a description of the two panels. (b) shows a percentage difference. What is this difference relative to?
Response: Accepted. We add a description of the two panels as: The panels display (a) absolute differences and (b) percentage differences in net primary production of the fine-sized group relative to the coarse-sized group.
Please see the revised caption below Fig. 8.

References:
Kok, J. F.: A scaling theory for the size distribution of emitted dust aerosols suggests climate models underestimate the size of the global dust cycle, Proceedings of the

National Academy of Sciences of the United States of America, 108, 1016, 10.1073/pnas.1014798108, 2011.

Li, W., Xu, L., Liu, X., Zhang, J., Lin, Y., Yao, X., Gao, H., Zhang, D., Chen, J., Wang, W., Harrison, R. M., Zhang, X., Shao, L., Fu, P., Nenes, A., and Shi, Z.: Air pollution–aerosol interactions produce more bioavailable iron for ocean ecosystems, Sci. Adv., 3, e1601749, 10.1126/sciadv.1601749, 2017.

Lamb, K. D., Matsui, H., Katich, J. M., Perring, A. E., Spackman, J. R., Weinzierl, B., Dollner, M., and Schwarz, J. P.: Global-scale constraints on light-absorbing anthropogenic iron oxide aerosols, npj Clim. Atmos. Sci., 4, 10.1038/s41612-021-00171-0, 2021.