Comments by referees are in blue.
Our replies are in black.
Changes to the manuscript are highlighted in red both here and in the revised manuscript.

## Reply to referee #2

This manuscript presents new measurements of iron (Fe) content and solubility across a range of anthropogenic fuel sources, with a particular focus on residential combustion (coal and biofuel). These observations are subsequently integrated into the MIMI–CESM2 model framework to assess impacts on soluble Fe deposition to the ocean. The study addresses a critical knowledge gap in quantifying anthropogenic aerosol Fe emissions and their influence on ocean biogeochemistry, especially in high-nutrient, low-chlorophyll (HNLC) regions. The experimental dataset is extensive and covers key combustion sources in China, while the modeling analysis provides valuable constraints on the climatological and future patterns of soluble Fe deposition. The finding that residential combustion disproportionately contributes to soluble Fe fluxes to the ocean is novel and has significant implications for anthropogenic Fe cycling and marine productivity. The manuscript is generally well-written, well-structured, and scientifically solid. However, several points require clarification and enhanced discussion before the manuscript can be recommended for publication.

**Reply:** We would like to thank ref #2 for reviewing our manuscript and recommending it for publication after minor revision. We have carefully addressed all the comments, as detailed below.

Major comments:

1. Line 136–137: Please provide the detection limit for soluble Fe. In addition, briefly describe the QA/QC procedures for the measurement method to ensure data reliability.

**Reply:** The detection limit of Fe was 0.5 μg/L. In the revised manuscript (page 9) we have added one section (Section 2.1.3) to provide information for the detection limit and the major QA/QC procedures we used: "The detection limit of Fe in solutions was determined to be 0.5 μg/L in this work. A reference solution (NIST 1643f) was used to check the accuracy of ICP-MS analysis, and the difference between actual and measured concentrations was found to be <1%. Furthermore, three blanks (with no fly ash or filters not loaded with any particles) were used in each batch when we measured total or soluble Fe. The background levels of soluble Fe were always below the detection limit; the background levels of total Fe, ranging from 4.3-5.7 μg/L, were much lower than total Fe concentrations for most of our samples and subtracted when we reported our results."

2. Line 448–490: While the manuscript highlights the importance of acid processing in promoting Fe dissolution, the evidence supporting atmospheric processing-induced release of soluble Fe from power-plant coal fly ash is not clearly demonstrated. Please provide more explicit evidence or discussion.

**Reply:** We think here ref #2 refers to Line 488-490, instead of Line 488-490. As we stated in the original manuscript (Line 484-490), previous laboratory studies (Chen et al., 2012; Fu et al., 2012; Baldo et al., 2022) found that acid processing could significantly increase promote Fe dissolution of coal fly ash. We may not understand very well what ref #2 means; in this case, we are happy to make further revision if ref #2 can explain this comment more specifically.

3. Line 556–559: The statement "This is because Fe in emitted particles is mainly highly soluble Fe sulfates for low combustion" requires clarification. What is meant by "low

combustion"? Could the authors elaborate on the formation mechanism and emission source of highly soluble Fe sulfates in this context?

**Reply:** In our original manuscript we made a typo, and "..low combustion.." should have been "low temperature combustion". In the revised manuscript (page 30) we have corrected it.

In our original manuscript (Section 3.2.2, page 498-502) we have provided a few sentences to explain why combustion at low and high temperatures will lead to aerosol Fe with different solubility: "Pyrite ($FeS_2$) is the major Fe-containing mineral in coal (Deng et al., 2015; Oliveira et al., 2016; Rathod et al., 2020). In low-temperature combustion, pyrite is mainly transformed to Fe sulfate (Bhargava et al., 2009) which has very high Fe solubility; as the temperature increases to >1000 K, Fe sulfate is further transformed to hematite and magnetite which exhibit very low solubility (Hu et al., 2006; Ram et al., 1995; Rathod et al., 2020)." As a result, we feel that it is not necessary to repeat this explanation in Section 3.2.6.

4. Line 590–594: Figure 3b shows that the largest relative increases in soluble Fe deposition occur primarily over equatorial/tropical regions such as the Congo Basin and Amazon rainforest. Does this imply that enhanced soluble Fe deposition over the South Atlantic originates largely from long-range transport of soluble Fe associated with South American residential biofuel combustion? Please clarify this interpretation.

**Reply:** Yes, this is our interpretation of Figure 3 as well. In the revised manuscript (page XX) we have made the following change to enhance our discussion: "Changes to soluble Fe fluxes from biofuel burning were most concentrated across the South Atlantic (Figure 3), likely due to the long range transport of emissions from the Amazon rainforest and across the Congo River basin where biofuel-burning in cook stoves is a common residential practice (García-López et al., 2025; Stoner et al., 2021)."

5. Line 638–645: Figure 4 indicates a systematic model underestimation of total Fe (A1) and a systematic overestimation of soluble Fe (B1–B4), leading to an overestimate of aerosol Fe solubility. Please explain the potential causes of these biases and implications for the modeled Fe dissolution scheme.

**Reply:** There are several considerations when examining Figure 4 that need to be taken into account. First, the model-observation comparison uses observations that represent a single snapshot of the atmospheric state from highly transient and spatially sparse shipborne campagins. Often this can mean we have a single day observation representing 1000's of kilometers. As a full spatiotemporal data set is not available, the goal here is not to attain a perfect model-observational match but to gain a level of constraint at the regional level that is acceptable given the large uncertainty. The only region that the model – observation range for total (and thus soluble) does not represent well is the Southern Ocean. However, Southern Ocean solubility is not well captured – an issue other Fe models struggle with (see Myriokefalitakis et al. 2018).

Second, the figure itself is a function of the chosen region definition over which to calculate medians. Here we have chosen "classic" physical ocean definitions. However, if the regions were modified to instead represent aerosol source provinces the results will look different (e.g., the recent evaluation by Bergas-Masso et al. (2025)). This is once again due to the sparse data being used.

Third, the ultimate goal of the MIMI model is to capture soluble iron, as that is the parameter used for biogeochemical modelling. In Figure 4 it can be seen that for all regions, apart from the Southern Ocean, the model-observation relaionship fall slightly low, but are relatively aligned with each other. This suggests that the spatial relationship between regions is robust, but the magnitude itself may be low. This, coupled with the low bias in total iron, suggests that there may be a missing source of Fe in the model rather than any dissolution chemistry bias.

One conclusion of this paper is that residental iron does not fill that knowledge gap. But moreover, we have a dataset of observations that do not well capture downwind residental iron plumes and suggest that this is considered in future campaigns.

6. The comparison with ship-based observations is useful; however, as noted by the authors, some ship measurements are outside biofuel-influenced regions. Such data should not be included for model validation in this specific context, as they do not reflect the emission regime of interest.

**Reply:** In lieu of source-apportioned observations, we replotted the model-observation comparison data only including observations collected in ocean regions downwind of strong residential iron influences, which we defined as areas where soluble Fe fluxes increased by a factor of 2 or more in the PD-BIOF simulations. However, this resulted in dropping our n value from n = 990 to n = 25 (media aggregated data within 25 grid cell locations), so this approach was ultimately not desirable for model evaluation purposes.

Those plots using the "biofuel-influenced' regions are now provided in Figure S4 of the revised supplement (page 15); in addition, in the revised manuscript (page 38) we have added one paragraph to discuss this further: "Withstanding source-apportioned measurements of residential coal or biofuel aerosol in our observational dataset, we performed additional model-observation comparisons only using measurements collected in ocean regions downwind of strong residential burning influences. These regions were defined as model-resolution grid cells wherein soluble Fe fluxes increased by 100% or more in the PD-BIOF simulations (Figure 3). However, this reduced median-aggregated observational data points from n=990 to n=25, limiting statistical capacity to constrain model fluxes. When using the smaller observational dataset, model-observational comparisons for Total Fe, soluble Fe, and solubility mirrored agreement trends using the larger dataset (Figure 4); those regression analyses are provided in the Supplemental Information with results from the PD-IND simulation (Figures S4-S5)."

Minor comments

1. l.116: Please report the effective number of valid filter samples for each biofuel type in the main text.

**Reply:** We feel that such information may not be critical, and therefore we include it in Tables S2 and S4. In the revised manuscript (page 8) we have made the following change to specifically refer reader to the supplement: "…the number of effective filter samples (for which Fe content and solubility were reported) were usually <8 for each fuel type (see Tables S2 and S4 for further information)."

2. Line 74–75: grammatical issue with "to and from?" — please revise.

**Reply:** In the revised manuscript (page 4) we have change "…to and from the atmosphere and surface ocean..." to "…from the atmosphere and to the surface ocean…" to make it clearer.

3. Line 97: clarify whether "aerosol solubility" refers specifically to Fe solubility.

**Reply:** Yes, it refers to Fe solubility. In the revised manuscript (page 5) we have changed "…Fe content and solubility of aerosol emitted by…" to "…Fe content and solubility for aerosol emitted by…", in order to be clearer.

4. Line 114: revise "in order….." to "in order to……".

**Reply:** In the revised manuscript (page 9) we have changed "…in order…" to "…in order to …".

5. Several minor grammatical issues exist; a careful proofreading is recommended.

**Reply:** We would like to thank ref #2 for the careful review. As suggested, during our revision we have carefully checked the entire manuscript in order to minimize typos and defer any remaining oversights to the editorial team.