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 review is amazingly thorough and much needed by the community as a summary of Fe isotope values as they relate to aerosols. The supplementary tables are invaluable as comprehensive summaries of published Fe isotope values. I am very excited to see this paper published.

Reply: We would like to thank ref#2 for recommending our manuscript for publication and his/her comments which have help us further improve our manuscript. We have addressed these comments and updated the manuscript accordingly; when we do not quite agree with ref #2, we have provided proper explanation. Please find more details below.

There is some work, however, that needs to be done to make the manuscript a more cohesive unit. Terms need to be summarized the first time they are used (this is especially true for operational definitions such as “soluble”), and word choice needs to be used more consistently (for example, desert dust and UCC should not be used interchangeable/have different definitions in different sections).

Reply: It is a good suggestion. We have made the following changes in the revised manuscript: 1) we have tried to use replace desert dust with UCC whenever possible; 2) for terms which need definition, we have defined them where they appear for the first time or in the appendix (page 36-37).

Further, I suggest re-organizing the manuscript as right now it reads as separately written sections that are sometimes redundant and do not lead the reader to the comprehensive takeaways for different processes.

Right now, I feel like this paper is a very detailed review of all applicable literature, but I think it could be made stronger by streamlining the details and adding an introductory/summary as things relate to aerosols for all sections. This will allow the relevance and summary of the literature to be clear to readers.

Possible new, simplified outline (would require reorganizing current paragraphs into the correct section:

Reply: We fully understand that these comments raised ref #2 kindly are intended to help us further improve our manuscript. The structure which ref #2 suggested may also be a good option. However, we prefer to not to change the structure of our manuscript, because in our opinion the current structure delivers very well the information we want to deliver.

We feel that overall our manuscript has been streamlined quite well. For example, we have used Section 1 as the introduction and Section 4 as the summary; in addition, we have also provide necessary introductions or summaries for some key subsections (for example, Sections 3.2 and 3.3).

In the revised manuscript (page 29), we have added a few sentence to inform the readers why we discuss Fe isotopic fractionation in Section 3.5: "Chemical processes which change the speciation of Fe in the environment may also lead to isotopic fractionation (Table S1), and thereby may attenuate Fe isotopic signatures observed in ambient aerosols. However, when using Fe isotopic composition to trace sources of total and soluble aerosol Fe, only a few studies have discussed possible Fe isotopic fractionation induced by atmospheric chemical processing (Labatut et al., 2014; Camin et al., 2024); most previous studies assumed that total and soluble Fe from a given source have the same endmember value, implicitly assuming no Fe isotopic fractionation. On the other hand, Conway et al. (2019) showed that total, water-soluble and seawater-soluble Fe in Saharan dust aerosol over the North Atlantic all had the equivalent δ^{56}Fe to the UCC, suggesting no Fe isotopic fractionation and supporting the use of endmembers without fractionation."

In addition, in the revised manuscript we have also tried to reduce redundancies or to provide further discussion when possible. Please refer to our reply to the following comments and our revised manuscript for more details.

Specific line comments/questions:

L23-40: Abstract would need to be reorganized if the suggestion of changed manuscript outline is selected.

Reply: As we prefer not to change the overall structure of our manuscript (please see our reply to previous comments), we decide not to re-organize the abstract either.

L45: Define dissolved, also consider using the abbreviation dFe throughout the paper.

Reply: As suggested by both referees, we have added an appendix in the revised manuscript (page 36-37) to define several terms used in our manuscript, including total Fe and dissolved Fe. Since dissolved Fe is not much longer than dFe, we prefer to use dissolved Fe for better readability.

L48-49: Define HNLC

Reply: In fact we have already defined HNLC in our original manuscript.

L50: Define the multiple ways you use the word soluble in this paper, i.e. size fraction and chemical leaching method

Reply: As suggested by both referees, we have added an appendix in the revised manuscript (page 36-3) to define several terms used in our manuscript, including total Fe and dissolved Fe (soluble Fe). When we define dissolved Fe (soluble Fe), we mention that multiple ways are used in different studies.

L73: The , after troposphere makes it seem like it's part of the list that follows.

Reply: To avoid this misunderstanding, in the revised manuscript (page 4) we have changed this sentence to "Natural, anthropogenic and wildfire aerosols may undergo various chemical and physical processes in the atmosphere, which can "solubilize" insoluble Fe minerals to soluble Fe"

L82: At this point, it is difficult to understand what you mean by 'ambient aerosols'. I would explicitly define it. This is an example also of where using desert dust and UCC consistently and making the definitions clear is important.

Reply: Here ambient aerosols mean aerosol particles in the troposphere. To explicitly define ambient aerosol, in the revised manuscript (page 4) we have made the following change: "…Fe solubility can be much higher for ambient aerosols (aerosol particles in the troposphere) collected over the oceans…"

We have checked the entire manuscript, and replaced desert dust with UCC when possible.

L95: Define enrichment factors and add a sentence for how they are used before you mention that there isn't a consensus.

Reply: As suggested, in the revised manuscript (page 36-37) we have added an appendix where we have defined several terms, including enrichment factors.

L104: Single particle analysis through what methods?

Reply: In the revised manuscript (page 6) we have made the following change to mention the typical methods used for single particle analysis: "Single particle analysis, typically using electron microscopy, X-ray micro-spectroscopy or single particle mass spectrometry, can also be very useful for source identification of aerosol Fe in individual particles…" In addition, references cited here have also been updated.

L116-121: An example of a section that can be moved to the modeling section. Also, it was unclear, how they determined coal combustion as a major anthropogenic source. Seems like a key takeaway.

Reply: The two papers (Wang et al., 2022; Wei et al., 2024) are review papers which review studies related to aerosol Fe isotopes, and they both use a statistical model (MIXSIAR) to re-analyze literature data. As a result, we mention them in Section 1 (Introduction) instead of in Section 3.4 (modeling studies). In the revised manuscript (page 28), we have also changed the title of Section 3.4 to reflect its content more properly: "3.4 Atmospheric modeling studies of isotopic compositions of ambient aerosol Fe".

It is beyond the scope of our review paper to explain why Wei et al. (2024) identified coal combustion as the major anthropogenic source of aerosol Fe. This is perhaps linked to the endmember values they used for coal combustion.

L137-139: Add information about how IRMM-014 is running out and what standard the field is moving to.

Reply: In the revised manuscript (page 7) we have added the following sentence to provide more information related to IRMM-014 and its substitutes: "Production of IRMM-014 has ceased and current stocks will likely run out in the future; as a result, new reference materials, such as IRMM-524a, are beginning to be used in place of IRMM-014 (González De Vega et al., 2020; Xu et al., 2022)."

L148: I would argue that 1-3permille is a more realistic range across transects.

Reply: We agree with the referee, and have changed it to "…typically 1-3‰…" in the revised manuscript (page 8).

L150: short -> shorter

Reply: In the revised manuscript (page 8) we have implemented the suggested change.

L172: Choose either SE, 1sigma, and 2sigma error for consistent notation throughout the paper.

Reply: In our manuscript we usually use 2σ to represent errors. In response to this specific comment, in the revised manuscript (page 8) we have made the following change: "…to achieve acceptable uncertainties (<0.15‰, 2σ) in solutions…"

As suggested, in the revised manuscript we have made changes so that we always used.

L174-187: Talking in ng of Fe is not common for many of the target readers of this article. I would consider explaining how you get these numbers from concentration to make sure readers are on the same page.

Reply: The minimum amount of Fe required by Fe isotope analysis, is typically expressed in mass. Interested readers can easily obtain the minimum concentration of aerosol Fe (in the troposphere) required if they know the air volume samples (and vice versa).

L174: Add "of Fe" after a minimum mass

Reply: We have made the suggested change in the revised manuscript (page 8): "…a minimum mass of Fe depends on…"

L186: Define "such precision"

Reply: As suggested, in the revised manuscript (page 8) we have made the following change to define the precision: "Such precision (<0.1‰) is often essential for resolving variability between aerosol samples."

L191: Add sources

Reply: We have carefully checked L191. We feel that we cannot add "sources" to this line and that the sentence to which L191 belongs looks fine.

L198: This is an example of a section that is out of place in my opinion. It comes right after you finish explaining that you won't discuss in-depth marine studies. It also references fractionation processes in the surface that you have yet to discuss in the manuscript. Also, it is unclear how you call things "dust" when you later explicitly state that the surface has to be ignored through fractionation.

Reply: We feel that Section 3.1 is in the right place. Before we review application of Fe isotopic analysis in aerosol research, we have a short section (Section 3.1) we present a few examples of relevant ocean observational and modeling studies to illustrate how δ^{56}Fe may help constrain sources of dissolved Fe to the ocean, focusing on aerosol deposition case studies; in other words, Section 3.1 sets the context for the following contents in Section 2. This is mentioned at the beginning of Section 3 and further elaborated in the first paragraph of Section 3.1.

We mention fractionation when we discuss the three examples used in Section 3.1, because the measured Fe isotopic composition of a sample is not only affected by its sources, but may also be impacted by isotopic fractionation.

L203: Different precision range than earlier in the manuscript.

Reply: To keep the precision ranges consistent, in the revised manuscript (page 10) we have made the following change: "…with a precision of better than ±0.10‰."

L220: Confusing that you call it 2 component mixing model when there are more than two sources.

Reply: At each station, Fe is assumed to originate from two sources, and one source is always dust, and the other source varies with stations. In the revised manuscript (page 11), we have made the following change for better clarification: "…two component mixing was used to quantitatively constrain sources at each station across the basin…"

Because 1) we only use this study (Conway and John, 2014) as one of the three examples to demonstrate the usefulness of Fe isotopes to understand the sources and processes of marine Fe and 2) the paragraph is already quite long, we decide not to provide further details but refer the interested readers to the work by Conway and John (2014).

L226: I recommend adding more sources that discuss the fractionation process at the surface ocean.

Reply: Similar to our reply to last comment, we only use this study (Conway and John, 2014) as one of the three examples to demonstrate the usefulness of Fe isotopes to understand the sources and processes of marine Fe, and it is beyond the scope of our manuscript to discuss in depth Fe isotope fractionation at the surface ocean; in addition, the paragraph is already quite long. As a result, we prefer not to discuss the fractionation process at the surface ocean in more details.

L247: This would probably make more sense being moved to the modeling section.

Reply: We respectively disagree with the referee. The modeling study by Konig et al. (2021) is focused on biogeochemical processes of Fe in the ocean, and we use this study as one of the three examples to demonstrate the usefulness of Fe isotopes to understand the sources and processes of marine Fe; on the other hand, Section 3.4 only discusses modeling studies of aerosol Fe in the atmosphere. As a result, we prefer to discuss the work by Konig et al. (2021) in Section 3.1.

To better reflect the content of Section 3.4, in the revised manuscript (page 34), we have changed its title: "3.4 Atmospheric modeling studies of isotopic compositions of ambient aerosol Fe".

L256: More background information on how aerosols are shown to be a mixture of natural and anthropogenic sources and choose the words that fit into each category that you will consistently use throughout.

Reply: We have a paragraph in our original manuscript (line 59-72, Section 1) to discuss natural and anthropogenic sources of aerosol Fe in the atmosphere; as a result, we prefer not to repeat it in Section 3.2.

We agree with the suggestion on using consistent terms for aerosol sources. In the revised manuscript, we have made the following changes (page 12): "…and aerosol particles emitted by various anthropogenic sources...", and updated the title of Section 3.2.3 (page 15): "3.2.3 Aerosol particles emitted by anthropogenic sources".

L275-279: Missing + before heavier Fe isotope values.

Reply: We have checked the entire manuscript (and supplement), and added "+" for all the positive δ^{56}Fe values.

L276: Define ATD.

Reply: As suggested, in the revised manuscript (page 13) we have defined ATD: "…namely Arizona Test Dust (ATD)…"

L288: Calculate an actual average or state something like, "for natural dust contributions, we can assume an endmember equal to that of UCC (+0.09 permille). Also, I think it is important to mention in the natural aerosol section that not all natural aerosols are from deserts.

Reply: As suggested, in the revised manuscript (page 14) we have modified the first sentence in this paragraph: "Most of the δ^{56}Fe values for desert dust Fe, as reported in previous studies, fall into a small range (-0.1 to +0.19‰); as a result, one may conclude that the Fe isotopic composition of desert dust is very similar to the UCC, characterized by an endmember value of around +0.09‰."

In addition, we have also added one sentence in the revised manuscript (page 14) to mention that not all the natural aerosol Fe originates from desert dust: "In addition to desert dust, there are other natural sources for aerosol Fe which may have different isotopic composition, such as soil particles entrained into the atmosphere during wildfires (Hamilton et al., 2022)."

L353-261: This is a paragraph that I think would be a better introduction to the anthropogenic aerosol section.

Reply: This paragraph serves to summarize the first three paragraphs in Section 3.2.3, and it also sets the background for next paragraph (also the last paragraph in Section 3.2.3). As a result, we believe that it is suitable to keep it in the current place.

L368: If you are using leach data, talk about how the leaches are proven to fractionate or not fractionate Fe and the sources that support this.

Reply: Most of studies reviewed in Sections 3.2 and 3.3 does not consider Fe isotopic fractionation, and this is why we discuss Fe isotopic fractionation in Section 3.5. This has been discussed in our original manuscript (page 38, line 795-797).

To make this more explicit, we have made the following change in the revised manuscript (page 29): "Chemical processes which change the speciation of Fe in the environment may also lead to isotopic fractionation (Table S1), and thereby may attenuate Fe isotopic signatures observed in ambient aerosols. However, when using Fe isotopic composition to trace sources of total and soluble aerosol Fe, only a few studies have discussed possible Fe isotopic fractionation induced by atmospheric chemical processing (Labatut et al., 2014; Camin et al., 2024); most previous studies assumed that total and soluble Fe from a given source have the same endmember value, implicitly assuming no Fe isotopic fractionation. On the other hand, Conway et al. (2019) showed that total, water-soluble and seawater-soluble Fe in Saharan dust aerosol over the North Atlantic all had the equivalent δ^{56}Fe to the UCC, suggesting no Fe isotopic fractionation and supporting the use of endmembers without fractionation."

L381-391: I think this paragraph would be a better introduction to an endmembers section.

Reply: This paragraph and the other paragraph in Section 3.2.4, are used to summarize previous studies on endmember values. As a result, instead of putting it in the front of Section 3.2 (as an introduction), we prefer to have it as the end of Section 3.2 (as a summary).

L399: Define "fine" and "coarse" and let the reader know if these definitions change from study to study.

Reply: The cutoff size between fine and coarse particles varies in different studies. That is why we provide the size ranges for fine and coarse particles in our manuscript whenever possible.

L409: If you keep this outline, why is there a 2016 cutoff?

Reply: In the revised manuscript (page 18) we have added one sentence to explain why we choose 2016 as the cutoff year: "We choose the year of 2016 as the cutoff, because Kurisu and co-workers who published several papers in this field published the first paper in 2016."

L411: Define TSP

Reply: We have already defined TSP previously (line 367 in our original manuscript).

L458: These are the lightest isotope values included in the paper. Might be worth positing why it makes sense they are so light (or why it is surprising).

Reply: In fact the lightest Fe so far was reported by Hsieh and Ho (2024). It is a good idea to highlight the lightest Fe. In the revised manuscript (page 25) we have added one sentence to highlight it: "This study reported lowest δ^{56}Fe values so far for total and soluble Fe in the troposphere (down to -3.35 and -4.46‰, see Table S2).” The possible reason for the highest Fe is implied in the sentence immediately after the sentence we have added.

L480-501: This is an example location where I think this information would flow better in the applicable endmember section.

Reply: The work by Kurisu and Takahashi (2019) investigated ambient aerosol impacted by biomass burning, and can also be placed in Section 3.2.3 where we discuss aerosol particles emitted by anthropogenic sources. However, because the ambient aerosols examined by Kurisu and Takahashi (2019) also originated from other sources (in addition to biomass burning), we prefer to discuss this work in Section 3.3 where we discuss Fe isotopic composition of ambient aerosols.

L519-579: These sections are examples of where it feels like there was a different author. They are a lot more detailed/lend a full paragraph to one study, compared to elsewhere in the manuscript. I think it would help the reader to streamline this and draw connections between the studies as applicable.

Reply: The two papers (Kurisu et al., 2021, Bunnell et al., 2025) are key studies and present very rich and insightful information; therefore, the two paragraphs which analyze and discuss these two studies are quite long. Understandably, different people have different opinions on the extent to which details should be provided; for example, ref #1 asked us to provide endmember values Kurisu et al. used in their two component mixing model.

Nevertheless, we agree with ref #2 in principle, and have tried our best to make these two paragraphs shorter. The paragraph which discusses the work by Kurisu et al. (2021) has been shortened from 15 to 13 lines (page 23-24 in the revised manuscript), and the one discusses the work by Bunnell et al. (2025) has been reduced from 17 to 16 lines (page 25 in the revised manuscript).

L644: Discuss why different endmembers were used in this study.

Reply: We carefully checked this paper (Kurisu et al., 2021) which states “The representative δ^{56}Fe value of combustion Fe was estimated to be -3.9‰ to -4.7‰ based on the δ^{56}Fe values of the aerosol samples collected at various sites, which included suburban areas and sites near sources of anthropogenic emissions.” We know how the authors assigned endmember values to combustion Fe, but do not know why they used different values; as a result, we are not able to discuss why different endmember values were used by Kurisu et al. (2021).

L644: I understand why you have included this section, as aerosols are greatly chemically altered before deposition, however this has not been explained to the readers. If you leave these sections, make sure to include examples of how this is applicable to aerosol studies and the main takeaways aerosol people should know from this information.

Reply: This is a good suggestion. In the revised manuscript (page 29) we have made the following change to inform the readers why we discuss Fe isotopic fractionation in Section 3.5: "Chemical processes which change the speciation of Fe in the environment may also lead to isotopic fractionation (Table S1), and thereby may attenuate Fe isotopic signatures observed in ambient aerosols. However, when using Fe isotopic composition to trace sources of total and soluble aerosol Fe, only a few studies have discussed possible Fe isotopic fractionation induced by atmospheric chemical processing (Labatut et al., 2014; Camin et al., 2024); most previous studies assumed that total and soluble Fe from a given source have the same endmember value, implicitly assuming no Fe isotopic fractionation. On the other hand, Conway et al. (2019) showed that total, water-soluble and seawater-soluble Fe in Saharan dust aerosol over the North Atlantic all had the equivalent δ^{56}Fe to the UCC, suggesting no Fe isotopic fractionation and supporting the use of endmembers without fractionation."

L764: I really liked the perspectives suggestion, I would just suggest that you make it even more strongly stated that these are the recommended steps that Fe isotope aerosol research needs to take. You say it, but stronger language would make it stand out. Also make sure to have some sort of conclusion at the end and that the manuscript doesn't just end at the end of the last suggestion.

Reply: We would like to thank referee #2 for his/her highly positive comment on Section 4. As suggested, we have made the following two changes to make our recommendation stronger. The first change is (page 33): "After reviewing and discussing previous studies in a critical manner, we recommend several future research directions below to further enhance the usefulness of Fe isotopes in atmospheric aerosol research", and the second change is (page 34): "Therefore, measurements of size-dependent δ^{56}Fe of total and soluble Fe in aerosol particles emitted by various anthropogenic and combustion sources are highly recommended."

We think we have already have a number of conclusions in Section 4 which has eight paragraphs. In each of the last seven paragraphs, we first provide a summary/conclusion in one aspect, and discuss future work we recommend in this direction.

Fig 1: Not everything in this figure is discussed in the paper (i.e. SO2, NOx, VOC, DMS, CO2). Either include them in the fractionation process section or remove them from the figure.

Reply: We use Figure 1 to provide a schematic overview of emission, transport, processing and deposition of aerosol Fe, and thus I put in in Section 1. We include trace gases (such as SO_2 and NO_x) in this figure to illustrate that reactions of these trace gases lead to aging of aerosol particles (including aerosol Fe).

Fig 4: I think this figure would be even stronger if there was some indication on the figure itself of the concentration differences between the two seasons.

Reply: This figure comes from a previous paper by Kurisu et al., and it is difficult for us to add another information to this figure. Therefore, we prefer to use this figure as it is. Please note that we have underscored and discussed the differences in Fe solubility in our original manuscript (line 463-466).

Supp Table S1&S2: Make sure to define your size fractions

Reply: In our original manuscript, we have provided size information in Tables S1 and S2 (ambient aerosols) whenever possible. Please note that the results summarized in Table S2 usually do not contain size information (in many cases the samples examined are not aerosol particles).