

4 September 2026

Dear Dr Janssen,

Thank you for handling our manuscript, submitted for inclusion in the “RUSTED: Reducing Uncertainty in Soluble aerosol Trace Element Deposition” special issue of Biogeosciences (MS#: egusphere-2026-1916, Title: “Variations in Arctic aerosol iron solubility in relation to leaching methodology, air mass characteristics, and seasonality”).

After previously addressing the comments provided by three referees, we have also now addressed your additional comments. Below we include our response to editor and reviewer comments, with the latter edited to reflect the further changes made to the manuscript before resubmission.

We have also revised the manuscript based on these comments, including the removal of the iron stable isotope data. Two authors whose contribution centred around the isotope data are no longer involved in the manuscript and have been removed from the author list. In addition, we have made a handful of minor changes to correct typographic errors in the manuscript. Both clean and tracked changes versions of the revised manuscript are submitted here.

We are grateful for the constructive comments of the reviewers, which we believe have led to an improved manuscript.

Dr Chris Marsay

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Associate Editor Comment:

Reviewer comments are in black.

Our responses are in red. Text in the manuscript appears in quotation marks, with additions/changes in italics. **Line numbers of the revised manuscript are highlighted yellow.**

Dear Dr. Marsay and co-authors,

I appreciate the efforts made to comprehensively address the comments of the reviewers, and I would be happy to receive a revised submission in line with the responses to reviewer comments.

I make the following additional comments, to guide the revision.

I agree with reviewer 3 that the treatment of the blank is problematic, and while I value the efforts made to address this in the reply, this is not a great solution. I also understand that there is not an ideal solution possible here, given the data availability, since the blanks contribute a significant portion of some samples and no blank $\delta^{56}\text{Fe}$ values were determined in this study. The other blanks from another cruise may be from different batches of filters, cleaned at different times, and handled at different times or settings both at sea and in the lab. Given that, I am not convinced that a 1:1 transfer of those blanks to this study is guaranteed to be accurate.

As the authors note in their response to reviewer 3, the isotope data are a small part of this manuscript. There are plenty of other great data here, and I think given the caveats needed, the addition of the isotope data is at best minimal, and may distract from the other points of the manuscript. I therefore think the isotope data are best removed from this manuscript.

While we still think that the relatively limited use of the iron stable isotope data made in the manuscript would hold up even if direct blanks had been measured and applied accordingly, we have followed the recommendation and removed the isotope data from the manuscript. This results in the removal of one subsection of the methods (formerly section 2.5), a couple of paragraphs of discussion in section 3.3, the original Figure 3, and a couple of rows of Table 3. The remaining figures have been renumbered as needed.

A couple small comments:

- Some of the data in table S3 are within uncertainty of 0. Rather than giving these data, I suggest replacing these with some designation like 'indistinguishable from 0' in this table, and for the interpretations throughout.

Rather than replacing the data in question, the values have been italicised in Table S3 and marked with a "*", with a note in the caption that this indicates "*blank-corrected data indistinguishable from zero within propagated uncertainty*". In addition, we have excluded these values from any average values in the text and in Tables 2, 3, and 4.

We have added the following text to section 3.1:

Concerning UPW leach concentrations: "*One sample (Aer08) was measured at less than the filter blank, while two others (Aer02, Aer09) were indistinguishable from zero within propagated uncertainty once blank-corrected.*" **(line 216)**

Concentrating SW leach concentrations: "*Higher filter blanks for the SW leach, which included an Fe contribution dependent upon the SW batch used (Table S1, S2), resulted in two samples (Aer08, Aer13) being measured at less than the filter blank and three others (Aer02, Aer04, Aer12) indistinguishable from the blank within measurement uncertainty.*" **(line 219)**

- Please include a supplemental table where the relative contribution of the blank to each sample is presented. Otherwise, given the different units of these two (pmol and pmol/m3), it is difficult for the readers to compare them.

This was also suggested by Reviewer 3 and has been done in the revised manuscript as Table S4.

Referee Comment 1:

Reviewer comments are in black.

Our responses are in red. Text in the manuscript appears in quotation marks, with additions/changes in italics. Line numbers of the revised manuscript are highlighted yellow.

This article combines together Fe solubility measurements in the Arctic Ocean and, for the first time, explores the seasonal variability of both bulk deposition and relative solubility of Fe transported and deposited via aerosol deposition. The Arctic provides a unique template for aerosol exploration given the year-round sea ice coverage that prevents complete deposition into the seawater where Fe is likely to have the largest impact on biology. The authors do a very complete comparison with other aerosol studies in the Arctic from the same cruises they focus on (GEOTRACES GN01 and the MOSAiC expedition). Ultimately, they conclude that deposition of soluble Fe is more substantial in winter due in part to increased anthropogenic emissions despite lower atmospheric depositional rates. The authors also make some guesses of how this pattern may evolve or shift with projected reduced sea ice cover in the Arctic.

I enjoyed reading this manuscript and have no major comments or revisions for the authors, only a few questions and small technical corrections.

Minor comments:

Figure 5/Lines 461-462: I think the data shown in the Figure 5 (now Figure 4) inset does not support the statement that no inverse pattern is noticeable within the Arctic data. I think there is a slight inverse pattern with the MOSAiC data. The Arctic data falls squarely within the range of previous data, particularly in the Atlantic datasets. I think you are trying to emphasize the very limited range in concentration compared to Atlantic/Pacific aerosols but I would rephrase this sentence.

To investigate this statistically, we took logs of the total Fe concentration and percent solubility, which is often done to display the relationship between the two as approximating a straight line (e.g., Shelley et al, 2018; Jickells et al, 2016 – both cited in the manuscript) and then conducted correlation analysis (Spearman Rank Order) of the datasets. The combined dataset of Pacific and Atlantic samples had a statistically significant correlation ($\rho = -0.437$, $p < 0.001$). Neither the combined Arctic dataset (GN01 and MOSAiC) or either of the two Arctic datasets considered separately results in a linear correlation.

We have adjusted the sentence slightly so that it reads “Whereas the *overall* dataset reflects the *previously described* relationship between total Fe concentration and Fe solubility (Sholkovitz et al., 2012), there is no clear relationship for the Arctic aerosol data. (line 450)

Lines 519-520: repeated use of “resulting”, sounds a bit messy.

We have changed the sentence to “Potentially, *the longer wintertime residence times* for particles would allow for greater internal mixing of crustal aerosols and anthropogenic emissions, resulting in greater solubilization of crustal Fe by acidic species.” (line 514)

Lines 609-612: Same thing, both sentences here start with “As a result”. I would do a careful read through of the manuscript to make sure phrases like “as a result” and “suggests/suggesting” aren’t over-used in quick succession.

Start of the second sentence has been changed to “*This means that...*” (line 615).

Conclusions: I thought this discussion was pretty brief. I wonder if the authors could comment on whether Fe making it to open water may relieve potential emerging Fe limitation in the Eurasian sector (Rijkenberg et al., 2018) or whether larger scale shifts in atmospheric weather patterns (like the Arctic Oscillation Index) may exert a control on summer vs winter deposition.

The conclusions section has been used primarily to summarize the discussion aspects earlier in the manuscript. To address this comment, we have instead split the discussion of deposition flux seasonality into two sections, with the first focusing on quantifying the flux and a new section (3.4.4) that discusses how changing conditions in the Arctic may influence atmospheric Fe supply and its effects. We include a couple of sentences about Fe limitation in the Arctic:

“While the Arctic Ocean is typically not considered to be Fe limited due to the influence of shelf processes, fluvial inputs, and the transpolar drift upon Fe supply, there is some evidence that low dissolved Fe concentration can become important for limiting productivity in the summer (Rijkenberg et al., 2018). Thus, the magnitude and relative solubility of atmospheric Fe deposition (either direct or from seasonally melting sea ice) could become increasingly important in the future.” (line 581)

This discussion already contains a brief discussion about expected changes in atmospheric transport and precipitation patterns, and the potentially conflicting influences on atmospheric Fe supply. We have now added a sentence to this to emphasize the importance of further studies to try and pick apart how this will pan out:

“As such, further studies investigating the relative importance of atmospheric processing versus emission type (e.g., soluble combustion emissions, refractory mineral aerosol) are needed to predict how changing atmospheric weather patterns will influence supply of bioavailable iron to the Arctic Ocean.” (line 600)

Referee Comment 2:

Reviewer comments are in black.

Our responses are in red. Text in the manuscript appears in quotation marks, with additions/changes in italics. **Line numbers of the revised manuscript are highlighted yellow.**

The manuscript entitled 'Variations in Arctic aerosol iron solubility in relation to leaching methodology, air mass characteristics, and seasonality' by Marsay et al., presents a comprehensive dataset on aerosol deposition as a significant source of iron to the Arctic Ocean. Given the ongoing changes in sea ice cover, a detailed analysis of aerosol iron solubility is essential for understanding the implications for marine productivity in the Arctic Ocean and, consequently, for regional climate change. Overall, this is a well-conceived study, and the authors appropriately highlight the importance of seasonal variations in Arctic aerosol iron solubility.

However, prior to further processing of this manuscript, I have a general comment concerning the interpretation of variations in Arctic aerosol iron solubility in relation to leaching methodology..., or actually the pH instead. It is well established that measurements of trace metal/element solubility are conducted using extraction in pure water, seawater, or a buffered acidic solution. I assume that the chemical properties of these solvents, particularly the final pH of each solution, exert a substantial influence on metal solubility (Mahowald et al., 2018; Wang et al., 2022).

We agree that pH likely plays a role in metal solubility, along with other aspects of the leaching approach used (e.g., exposure time, heat). Unfortunately, in this study, the final pH of leachates was not measured. We know that the UPW and SW leaches had notably different *starting* pHs (5.6 and 8.1, respectively), and we discuss in the manuscript how, in some previous studies, this has resulted in higher Fe solubility measured using the UPW leach than the SW leach – in addition to having a higher initial pH, SW presumably has more buffering capacity to maintain pH when it contacts any acidic components of the aerosol. However, in this study, we did not find this (higher solubility in UPW compared to SW) to be the case, though we discuss how this is probably at least partly due to the low aerosol Fe concentrations, which make it difficult to distinguish between the two methods. The HAc leach has a much lower starting pH again (pH 2), and results in much higher Fe solubility estimates (heat and presence of a reducing agent could also contribute to this).

The two references mentioned by the reviewer both consider factors that influence the pH of aerosols themselves (Wang et al., 2022) and the effect that this has on metal solubility (Mahowald et al., 2018). We suspect that, of the three leaching agents, UPW is most prone to a change in pH during the leaching process due to its contact with acidic aerosol species. This is based on the potential buffering capacity of seawater due to its higher ionic strength compared to UPW, and the already very low pH of the HAc leach. However, certainly in the GN01 dataset, this does not appear to be a significant effect, given that SW solubility is similar to UPW solubility despite its expected greater buffering capacity.

Specific comments:

Line 126 (2.2 Aerosol sample processing): It would be useful to report the final pH measured in each leaching experiment, if available.

Unfortunately, pH was not routinely measured after leaching each filter in this study.

Line 220 (Figure 2 caption): Please include essential information regarding the fractional percentages, even if mentioned in line 229. I misunderstood when only reading the figure.

The start of the figure caption (**line 225**) has been edited to read:

“Figure 2: Aerosol Fe concentrations and Fe fractional solubility in GN01 aerosols, where fractional solubility is the ratio of soluble to total Fe expressed as a percentage...”

Line 239: While the statement may be true, it should be noted that greater variability in solubility seems to be observed between aerosols from different sources than between different leaching solutions (Mackey et al., 2015).

We have edited the two paragraphs around this line to include this point. At [line 248](#) we have added the following:

In addition, direct comparison of the two leaches (i.e., using the same flow-through method but with UPW or SW) has previously demonstrated that, although Fe solubility is typically slightly higher in the UPW leach than in SW, aerosol source typically has a greater influence on variability in Fe solubility than the choice of UPW or SW as leaching agent (Aguilar-Islas et al., 2010; Buck et al., 2006, 2010; Shelley et al., 2018).

As comparison of the two leaching mediums continues in the following paragraph, we have edited the first sentence of it to read ([line 252](#)):

“Reports in previous studies of generally higher Fe solubility in UPW, relative to SW, have been attributed to the lower pH of UPW versus seawater (Buck et al., 2006, 2010, 2013; Shelley et al., 2018).”

The point about aerosols from different sources providing greater variability than different leaching solutions was previously alluded to in the introduction, and we have now given more emphasis to this ([line 50](#)):

*“Although comparison of data across different studies is complicated by the range of leaching methodologies used, there is a consensus that source and mineralogy are *more* important controls on Fe solubility than nuances of leaching approach (Aguilar-Islas et al., 2010; Mackey et al., 2015).”*

Lines 242-244: Please provide the specific numbers used for such comparisons.

The sentence has been edited to include approximate pHs of the liquids mentioned ([line 246](#)):

“It’s (UPW’s) lower pH (~5.6), relative to seawater (pH ~8.1) is also more representative of the lower pH of rainfall (typically in the range of pH 4 to pH 6), which can play a major role in atmospheric deposition in some regions (Duce et al., 1991; Spokes et al., 1994).”

As a result of adding pH of UPW and SW in this sentence, we have removed those values from the following sentence ([line 248](#)) to avoid needless repetition.

Line 286 and lines 304-309: I suspect that pH is the key factor here. Was pH measured in each experiment?

Neither the de Vera et al paper or the Laing et al paper provide a pH of the leaching solution at the start or end of their filter extractions. The authors of each paper state that UPW/Milli-Q was used, from which we assume that the starting pH was close to the 5.6 measured for UPW used in our study and so unlikely to account for the differences between studies.

The longer extraction time of the Laing et al study may have led to a decrease in pH of the solution over the course of the extraction, but this is speculation. As stated in the manuscript, the Laing et al study did involve higher loading of anthropogenic aerosols than those measured in this study, including sulfate concentrations of at least an order of magnitude higher, while Ca^{2+} concentrations were of a similar average concentration to this study. This suggests a more acidic aerosol sampled in the Laing et al study. In addition, as stated in this manuscript, the authors of that study acknowledged that aluminosilicates were not completely digested, which would influence their Fe percent solubility calculations to some degree.

For the de Vera study, higher aerosol V concentrations suggest that there may be more anthropogenic aerosol influence in those samples, which could mean higher concentrations of acidic species that would influence Fe solubility. However, we also note in the manuscript that there were slight differences in the UPW leaching approach. In particular, there is no mention of the use of a backing filter when leaching the W41 filters with UPW. Previous studies suggest that this can have a

significant influence on measured Fe solubility, due to nanoparticles being washed through into the leachate solution and being measured as “soluble” Fe (Meskhidze et al., 2016; Tang et al., 2025).

Line 461: The statement is difficult to follow, even only considering the larger dataset. Are there any statistical data available to support that?

To investigate this statistically, we took logs of the total Fe concentration and percent solubility, which is often done to display the relationship between the two as approximating a straight line (e.g., Shelley et al, 2018; Jickells et al, 2016 – both cited in the manuscript) and then conducted correlation analysis (Spearman Rank Order) of the datasets. The combined dataset of Pacific and Atlantic samples had a statistically significant correlation ($\rho = -0.437$, $p < 0.001$). Neither the combined Arctic dataset (GN01 and MOSAiC) or either of the two Arctic datasets considered separately results in a linear correlation.

The sentence has been revised to read (450):

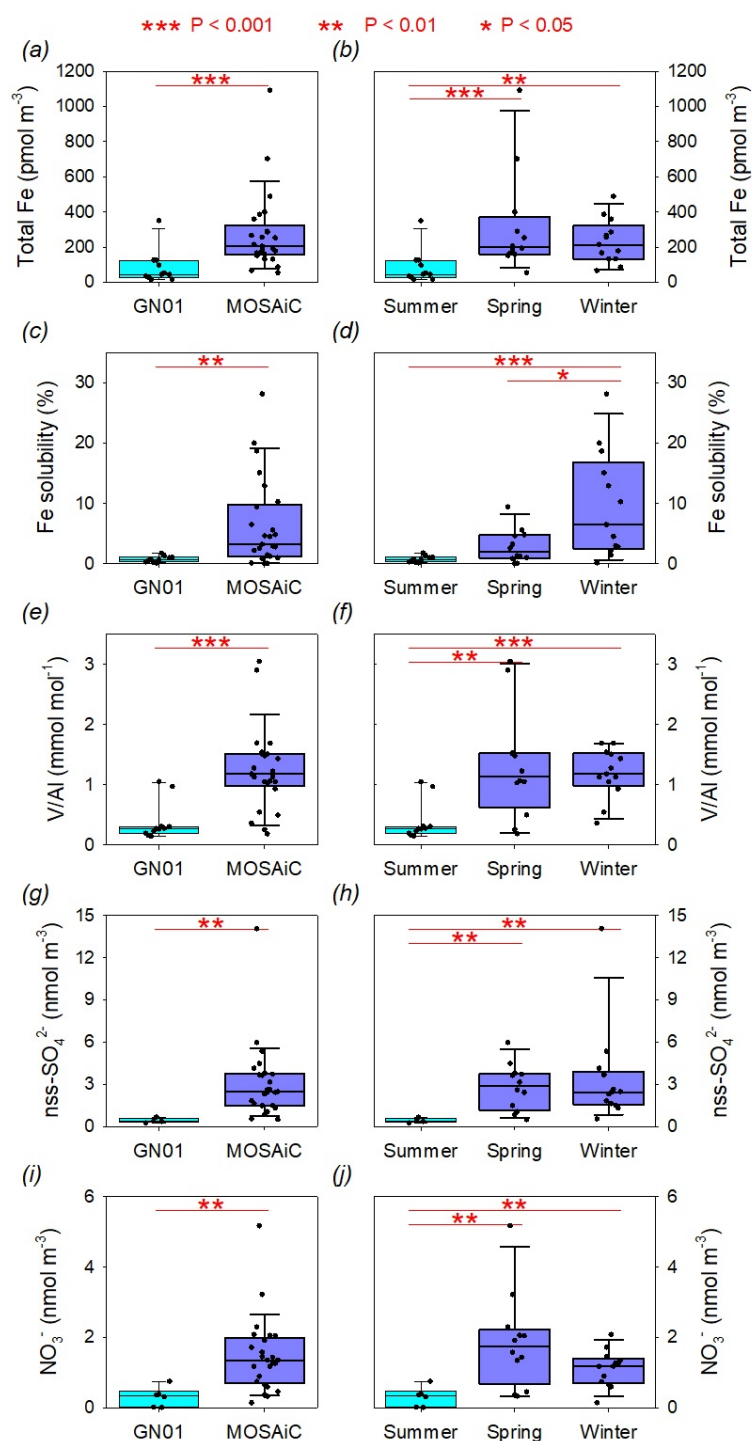
“Whereas the *overall* dataset reflects the *previously described* inverse relationship between total Fe concentration and Fe solubility (Sholkovitz et al., 2012), there is no clear relationship for the Arctic aerosol data.”

See also the response to Reviewer 1.

Line 510 (Figure 6 caption): It would be helpful to indicate the sample size within each boxplot (each season), e.g. jitter plot.

The previous Figure 6 (now Figure 5) has been adapted into a jitter plot (see below) to show the variability of individual sample values within each dataset. In addition, the caption has been edited to include details about sample sizes for each grouping and about the plot itself (line 501):

“Figure 5: Comparison of aerosol characteristics between GN01 (*cyan*) and MOSAiC (*purple*). (a, b) Total aerosol Fe concentration, (c, d) Fe UPW-solubility, (e, f) V/Al molar ratio, (g, h) nss-sulfate concentration, and (i, j) nitrate concentration. Comparisons are made between the two projects (left column) and between seasonal datasets (right column), with MOSAiC data ($n = 26$) split between winter (deployments 1-13, $n = 13$) and spring (deployments 14-26, $n = 13$). GN01 data excludes samples collected south of 70°S (Aer01, Aer14; $n = 11$, or less where some measurements are below detection). Each box spans 25th to 75th percentiles of data and is bisected by the median, with whiskers extending to 10th and 90th percentiles. Individual datapoints are superimposed as dots. Red lines indicate statistically significant differences between datasets (Mann-Whitney Rank Sum Test), with level of significance indicated by asterisks.”



Refs:

Mahowald, N. M., D. S. Hamilton, K. R. M. Mackey, J. K. Moore, A. R. Baker, R. A. Scanza, and Y. Zhang (2018), Aerosol trace metal leaching and impacts on marine microorganisms, *Nat Commun*, 9(1), 2614.

Mackey, K. R. M., Chien, C.-T. C.-T., Post, A. F., Saito, M. A. & Paytan, A. Rapid and gradual modes of aerosol trace metal dissolution in seawater. *Front. Microbiol.* 5,1–11 (2015).

Wang, G., et al. (2022), Quantitative Decomposition of Influencing Factors to Aerosol pH Variation over the Coasts of the South China Sea, East China Sea, and Bohai Sea, Environmental Science & Technology Letters, 9(10), 815-821.

References used in response to reviewer (if not cited in manuscript):

Meskhidze et al (2016) "Influence of measurement uncertainties on fractional solubility of iron in mineral aerosols over the oceans", Aeolian Research, 22, 85-92,
<https://doi.org/10.1016/j.aeolia.2016.07.002>

Referee Comment 3:

Reviewer comments are in black.

Our responses are in red. Text in the manuscript appears in quotation marks, with additions/changes in italics. **Line numbers of the revised manuscript are highlighted yellow.**

Review of egusphere-2026-1916 “Variations in Arctic aerosol iron solubility in relation to leaching methodology, air mass characteristics, and seasonality” by Marsay et al.

Summary:

This article presents iron concentrations and isotopic compositions in aerosols, either as bulk values or as an estimate of the soluble or labile fraction of aerosol iron. It also reports trace element concentrations in wet deposition samples.

The approach taken in this article is interesting and original. It benefits greatly from the first author's participation in several sampling campaigns in the Arctic Ocean across different seasons. Temporal comparisons within a single region are uncommon and particularly relevant in an area undergoing rapid change.

However, a major problem affects this article. The isotopic data obtained after UPW and SW leaching are presented with error bars that are largely underestimated. The reported measurement uncertainty corresponds only to the internal error of the mass spectrometer, and does not account for the fact that the blank represents a very significant fraction of the samples (blank/sample ratio of 58% on average for UPW and 60% for SW). I think that the uncertainties are at least 1‰, meaning the values are indistinguishable from one another (Fig. 6).

In my view, as they stand, the isotopic compositions of the UPW and SW leaches are not publishable. These data, and the interpretations derived from them, must be either removed or substantially revised to reflect the true measurement uncertainty. A detailed explanation is provided below.

We initially addressed the issues raised by the reviewer regarding blank corrections for Fe isotope data by using filter blank data from a different study that used the same procedures. However, in our revised submission of the paper, following further comments from the Associate Editor, we have removed the Fe isotope data and discussion relating to it. Responses to the specific comments below have since been edited reflect this.

Major scientific comment: These samples show a blank/sample ratio of 58% (UPW) and 60% (SW) on average (line 217). This is not surprising given the small amounts of iron that can be leached by UPW or SW. The iron concentrations of these samples were corrected accordingly, as mentioned on line 193. However, there is no mention of any blank correction applied to the isotopic compositions. The uncertainty associated with the isotopic compositions reported by the authors is that of the internal error (lines 205–206), which is not representative.

The isotope data has now been removed.

If the authors still wish to present these data by correcting the isotopic compositions of the UPW and SW leaches, they should:

- Report the blank/sample ratios for all samples, at least in a supplementary table;

This has now been done as supplementary table S4.

- Provide an isotopic composition of the blank with a realistic confidence interval, justifying their choice;

The isotope data has now been removed.

- Correct the isotopic compositions of the samples for the isotopic composition of the blank, and account for the error propagation arising from the isotopic composition and concentration of the blank;

The isotope data has now been removed.

- Revisit all interpretations based on these data, taking their uncertainty into account (likely larger than 1‰).

The isotope data has now been removed.

Regarding the HAC leaching, the blank/sample ratio is probably lower (this still needs to be reported), but it may remain significant which is the case whenever the blank accounts for more than roughly 5 percent of the sample. Indeed, there is generally an uncertainty associated with the isotopic composition of the blank, and in the absence of proper characterization, the hypothesis that this uncertainty is on the order of 1‰ cannot be excluded. If the blank is significant, the same approach as for the UPW and SW leaching should be applied.

No Fe stable isotope measurements were made on SW leach or HAC leach samples and so no isotope data for these leaches were included in the manuscript. Thus, no action is taken here.

I suggest that the authors also discuss the potential isotopic fractionation induced by these leaching steps.

The isotope data has now been removed.

Other scientific comments:

Line 28. It is unclear whether the UPW-soluble Fe flux refers only to the summer or to the annual average.

The fluxes here refer only to the summer period. This has been clarified in the abstract text (line 21):

“Summertime bulk deposition fluxes, calculated by applying the beryllium-7 method to GN01 data, were estimated at $0.9 \pm 1.3 \text{ nmol m}^{-2} \text{ d}^{-1}$ UPW-soluble Fe, $2.4 \pm 1.9 \text{ nmol m}^{-2} \text{ d}^{-1}$ SW-soluble Fe, and $46 \pm 48 \text{ nmol m}^{-2} \text{ d}^{-1}$ HAC-soluble Fe. Thus, the UPW-soluble Fe flux during GN01 was around an order of magnitude lower than that measured during the winter months despite bulk deposition velocity being almost twice as fast during the summer”.

Lines 45-47. I would suggest placing more emphasis on the importance of iron (Fe) for marine life (a nutrient, sometimes a limiting factor in marine primary production, etc.).

We have edited the sentence slightly for clarity. However, the point about the importance of iron for marine life is previously made in the first sentence of the introduction. We have given more emphasis to the importance of atmospheric Fe supply by adding to the second sentence of the introduction (line 29):

“Deposition of continentally derived material from the atmosphere (either through gravitational settling or carried by precipitation) is an important supply mechanism of Fe to the surface ocean (Jickells et al., 2005; Jickells and Moore, 2015), and observational and experimental evidence indicates that this input can promote increased productivity by relieving iron limitation (Han et al., 2011; Achterberg et al., 2013; Tang et al., 2021).”

Lines 76-78. This sentence is hard to follow. Starting with a definition of the Arctic haze and its seasonality would probably help the reader.

The paragraph has been restructured (line 71) as:

“Composition and concentration of aerosols in the Arctic atmosphere are characterized by strong seasonality. In the winter and spring, long-range transport to the Arctic from lower latitudes, combined with low precipitation rates, leads to a build-up of anthropogenic aerosols known as Arctic haze, characterized by higher concentrations of organic and acidic aerosols (Barrie, 1986; Schmale et al., 2022; Shaw, 1995). This transport includes mineral aerosol carried from distant sources to the south, which contribute more to dust loading during the spring, whereas locally sourced mineral aerosol dominates in the summer and autumn months (Groot Zwaagftink et al., 2016).”

Lines 99-101. Figure 1, b). It is difficult to understand what Ppt05, Ppt06, Ppt07 and Ppt08 are, even though Table 1 helps. I was looking, for instance, for an explanation of the black square. I would suggest describing this in the caption: Ppt05 and Ppt07 are cutoff bottle samples, while Ppt06 and Ppt08 are N-con samples, and they are geographically very close, which explains why they are hard to distinguish on the figure.

The caption has been edited to include the following explanation (italics) (line 97):

“Figure 1: Locations of (a) bulk aerosol and (b) precipitation samples collected during GN01. Cruise track is shown by thin blue line with aerosol or N-CON precipitation sample collections overlain in black and light blue. Red triangles in (b) indicate locations of blowing snow samples collected with cutoff bottles, while the black square indicates two N-CON samples (Ppt06, Ppt08) collected close to each other and to cutoff bottle samples Ppt05 and Ppt07. Figure produced using Ocean Data View (Schlitzer, 2025).”

Line 104. How was this uncertainty on the filtered air taken into account for aer01 (Table 1, Marsay et al., 2018a)? Could this uncertainty be propagated through all the subsequent concentration and flux calculations, in order to avoid any partial conclusions? Please check that the conclusions drawn for aer01 are consistent across all possible cases, i.e., a filtered volume of 444.3, 559.7 or 675.1 m⁻³.

The uncertainty associated with filtered air volume for Aer01 was already incorporated into reported uncertainties for digest Fe and HAC-leachable Fe. However, this additional uncertainty had previously been missed for the UPW and SW leaches. It has now been incorporated and is included in both concentration and solubility data in Figure 2. We also now include the following in the caption for Figure 2 (line 229):

“Error bars are propagated as described in the text, and include air volume uncertainty for Aer01.”

Line 131. It might help the reader to specify that the collected aerosols are "bulk", meaning that they include both the wet and dry fractions of an aerosol, and that different protocols were then applied to assess the total, soluble or labile fraction of a bulk aerosol. Alternatively, the definition of a bulk aerosol could simply be given around line 90.

We have added *“(i.e., with no size fractionation)”* at line 88.

Line 131. Are the filters acid-cleaned too?

Yes. At line 91 it states that aerosols were collected on “...acid-washed cellulose Whatman 41 filters.” In addition, we have now amended the text in section 2.2.2 to clarify that backing filters were also acid-washed using established protocols (line 140):

“A 0.4 µm polycarbonate membrane, previously acid-washed following procedures describe in Planquette and Sherrell (2012), was loaded on the filter rig beneath the aerosol filter...”

Line 135. At which temperature?

Sentence has been edited to include *“...at 140 °C”* (line 135)

Line 155. Given the large UPW rinsing volumes, did you measure the iron blanks of the UPW? If so, could you report the Fe concentration in the UPW, or at least provide the values in a supplementary table?

Yes, UPW blanks were analysed. We have now added the median and maximum rinse UPW contribution to rinse sample measurements (in %) to Table S5. In the main text, we have slightly edited the text in Section 2.3 to include the % contribution to Fe measurements from rinses (line 175):

“In such cases, a UPW blank was also kept to *correct* for any potential rinse contribution to measured TE concentrations (*generally <1 % for Fe measurements*).”

Line 157. Why this specific protocol of 100 mL in a single step?

Although there are several variations of the flow-through UPW leach, using different volumes and number of steps, the single-step, 100 mL method is well established and has been used in many studies since first published by two of the PIs involved in this project.

Line 250. It could be useful to also draw the lower error bar in Figure 2. Currently only the positive error is displayed, which makes it difficult to see whether Total/HAc and UPW/SW differ at the 95% level. Switching from a bar to a point value might help. Moreover, I cannot find the information on the error bar for concentration - is it 2sd or rsd (line 195)? Please make this clearer and consistent in the methods section.

We have kept the bar chart format for Figure 2 but have added the lower error bar in each panel as suggested and have adjusted the colour scheme so that these can be seen within the bar.

In the text, we have updated section 2.4 to indicate that errors are propagated from one standard deviation of measurements in samples and blanks (lines 192 and 199):

“For replicate treatments, the average and *one* standard deviation (1σ) of replicates were then calculated, and the average filter blank subtracted from the sample value.”

“In all cases, reported uncertainties of blank-corrected aerosol concentrations represent propagation of the precision (1σ) from samples and filter blanks.”

Line 254. Is the volume uncertainty of aer01 taken into account in the solubility calculation?

It does now – see the response to Line 104 comment.

Line 267. Yes, even though the lithogenic fraction is conventionally considered refractory, it can be dissolved in seawater (Rousseau et al., 2015) and during atmospheric transport (references should be available).

The main point being made here was that the amount of Fe solubilised by the HAc leach was surprisingly high if it is assumed that mineral aerosol was the main contributor to aerosol Fe load. We have tried to emphasize this more by rewording from “some fraction” to “*a substantial* fraction” (line 270).

Line 272. The intercomparison has been well described.

Thank you.

Line 297. Consequently, for future cruises it could be interesting to compare the efficiency of the 1×100 mL and 3×50 mL protocols.

I agree that more testing of this method variability would be helpful in terms of comparing studies with slightly different approaches.

Line 331. If Ppt13 is removed, does this still hold?

If Ppt13 is excluded, the average (mean) Fe solubility in precipitation samples is still higher than that based on the UPW-leach, but this drops to being twice as high. This is now clarified in the text (line 330):

“From these four precipitation samples, average Fe solubility was roughly 4 times higher than for aerosol Fe UPW-solubility (Table 2), though this *drops to 2 times higher if* Ppt13, which had a notably higher dissolved Fe percentage than the other three filtered precipitation samples, *is excluded* (Figure 3b).”

Line 351. This is an open question (no need to answer it in the article): do you think there is a way to estimate this "overestimate" due to the unknown Fe content of refractory particles?

I don't. I think the best way to address this would be a dedicated study collecting paired rain samples – one that is filtered for analysis of total particulate (and dissolved) Fe and one for analysis of total dissolvable (and dissolved) Fe – that cover different transport pathways/Fe sources and assessing the variability of dFe/TDFe versus dFe/(dFe + pFe).

Line 390. The -1.6‰ to -4.7‰ range is partial. Wang et al. (2022) published a review of iron isotope signatures in aerosols. Signatures heavier than -1.6‰ can also be attributed to anthropogenic aerosols, including positive ones (Flament et al., 2008). Kurisu et al. (2016) and Kurisu & Takahashi (2019) reported values close to the one observed for aer01. This is mentioned later in the article, but you could also briefly note that a mixing between anthropogenic and crustal aerosols can lead to such a slightly negative value.

The isotope data has now been removed.

Lines 398 and 400. Camin et al. (2025) published complementary positive signatures in the equatorial Pacific and discussed isotopic fractionation during transport.

The citation was added in relation to the discussion about positive Fe isotope signatures. With the removal of the isotope data, we have kept this citation as the isotope fractionation is discussed in relation to possible loss of soluble Fe from aerosols during transport, which. The revised text now reads (line 400):

“Studies in the remote Pacific Ocean have encountered similarly low Fe/Al ratios along with a distinct heavy stable Fe isotope signature, which has led to speculation that this may be due to loss of a soluble fraction of Fe during transport, along with isotope fractionation (Kurisu et al., 2021; Camin et al., 2025). This would explain the low Fe solubility in Group 2 samples. However, more work is needed to verify how the soluble fraction could be separated, and whether Al and Ti would undergo similar processing. Notably, the similarly low Fe solubility in Group 3 samples was associated in some cases with more direct transport from terrestrial sources (e.g., Aer11; Marsay et al., 2018a), rather than material that had been airborne for a long period.”

Line 408. The Fe/Al and Fe/Ti elemental ratios presented in Marsay et al. (2018) are compared to the global crustal value from Taylor and McLennan (1995). Crustal values can show considerable heterogeneity, so comparing them to a more regional ratio could be more accurate, or at least, it would be worth checking that the regional ratio does not differ significantly from the one given in Taylor and McLennan (1995).

The difficulty with the suggestion of using a regional Fe/Al or Fe/Ti ratio is determining which region is appropriate to use. Air mass back trajectories calculated for the period of this study highlight that the Arctic Ocean is characterized both by transport from multiple regions (Siberia, North America, the North Pacific and landmasses further south) and by winds circulating around the polar region for several days, which increases uncertainty about the initial transport pathway from continental regions. As such, we feel that using an average crustal value is appropriate in this case.

Lines 428-430. This is an interesting result.

It is and is something that could probably be addressed in laboratory-based experiments.

Line 458. Maters et al. (2022) observed this process as well (Fig. 1 of their article).

Thank you. We have incorporated this citation into the manuscript as follows (line 444):

“Alternatively, the relationship has been hypothesized to represent the combined effects of aerosol removal by deposition during atmospheric transport and physicochemical processing of the remaining material. *In this scenario*, Fe solubility increases as mineral aerosol undergoes increasing interaction with other aerosol components, including acidic species and organic ligands (Mahowald et al., 2018), *as observed during laboratory experiments (Maters et al., 2022).*”

Line 509. The statistical approach is interesting and strengthens the conclusions of the article.

Thank you.

Technical comments

Lines 42-44. I recommend choosing between 'photic' and 'euphotic' and using the same word throughout to avoid losing the reader. You could also define it quickly.

Good point. The word at line 41 has been changed to “euphotic” for consistency.

Lines 45-47. This sentence is hard to follow, maybe separate it in two parts.

The sentence has been adjusted slightly to read (line 42):

“Understanding the degree to which atmospherically supplied Fe is bioavailable *once in the ocean* is crucial *for* modelling the impact of atmospheric deposition *upon* the marine ecosystem and the carbon cycle.”

Line 130. For clarity purpose, to precise that ‘digests’ means total, e.g. ‘digests (total) ...’.

We have added “total” before the word “digests” (line 130).

Line 163. Please define the acronym HAc.

The HAc acronym was previously defined at line 129. To avoid confusion, we have now moved the first use of HAc to section 2.2.4 (Acetic acid leach) and first use of SW to section 2.2.3 (Filtered seawater leach). All three leach abbreviations (UPW, SW, HAc) are also now defined in the titles for subsections 2.2.2, 2.2.3, and 2.2.4.

Line 354. A space is missing between ‘(2021)’ and ‘discussed’.

Corrected. (line 356)

Line 575. Marsay et al. is repeated twice.

Corrected. (line 570)

References

Camin, C., Lacan, F., Pradoux, C., Labatut, M., Johansen, A., and Murray, J. W.: Iron isotopes suggest significant aerosol dissolution over the Pacific Ocean, *Atmos Chem Phys*, 25, 8213–8228, <https://doi.org/10.5194/acp-25-8213-2025>, 2025.

Flament, P., Mattielli, N., Aimoz, L., Choël, M., Deboudt, K., Jong, J. de, Rimetz-Planchon, J., and Weis, D.: Iron isotopic fractionation in industrial emissions and urban aerosols, *Chemosphere*, 73, 1793–1798, <https://doi.org/10.1016/j.chemosphere.2008.08.042>, 2008.

Kurisu, M. and Takahashi, Y.: Testing Iron Stable Isotope Ratios as a Signature of Biomass Burning, *Atmosphere-Basel*, 10, 76, <https://doi.org/10.3390/atmos10020076>, 2019.

Kurisu, M., Sakata, K., Miyamoto, C., Takaku, Y., Iizuka, T., and Takahashi, Y.: Variation of Iron Isotope Ratios in Anthropogenic Materials Emitted through Combustion Processes, *Chem Lett*, 45, 970–972, <https://doi.org/10.1246/cl.160451>, 2016.

Marsay, C. M., Lam, P. J., Heller, M. I., Lee, J.-M., and John, S. G.: Distribution and isotopic signature of ligand-leachable particulate iron along the GEOTRACES GP16 East Pacific Zonal Transect, *Mar Chem*, 201, 198–211, <https://doi.org/10.1016/j.marchem.2017.07.003>, 2018.

Maters, E. C., Mulholland, D. S., Flament, P., de Jong, J., Mattielli, N., Deboudt, K., Dhont, G., and Bychkov, E.: Laboratory study of iron isotope fractionation during dissolution of mineral dust and industrial ash in simulated cloud water, *Chemosphere*, 299, 134472, <https://doi.org/10.1016/j.chemosphere.2022.134472>, 2022.

Rousseau, T. C. C., Sonke, J. E., Chmeleff, J., van Beek, P., Souhaut, M., Boaventura, G., Seyler, P., and Jeandel, C.: Rapid neodymium release to marine waters from lithogenic sediments in the Amazon estuary, *Nat Commun*, 6, 7592, <https://doi.org/10.1038/ncomms8592>, 2015.

Taylor, S. R. and McLennan, S. M.: The geochemical evolution of the continental crust, *Reviews of Geophysics*, 33, 241–265, <https://doi.org/10.1029/95RG00262>, 1995.

Wang, Y., Wu, L., Hu, W., Li, W., Shi, Z., Harrison, R. M., and Fu, P.: Stable iron isotopic composition of atmospheric aerosols: An overview, *npj Clim Atmos Sci*, 5, 1–13, <https://doi.org/10.1038/s41612-022-00299-7>, 2022.