Response to Anonymous Referee #1 comments (RC1)

Major comments

1) RC1 The in-depth characterization of the Arctic's Laptev and East Siberian Seas provided here is an essential contribution to understanding the biogeochemical and physical processes controlling trace metals budgets, especially given the recent accelerating changes in the region. The authors present a compelling mechanistic synthesis, but the manuscript requires refinement in its presentation. Overall, the manuscript is generally grammatically clean. However, it needs improvements in structure and readability.

We are grateful for evaluating our manuscript and encouraging us to improve the manuscript. We tried improving the manuscript's readability after receiving the reviewer's comments.

First, the manuscript reads more like a report or book chapter, overly descriptive and lacking a clear hypothesis, or answering specific questions. Though a valuable contribution, it is unclear why readers should read it. For instance, focusing on contrasting the physical and biogeochemical controls of iron (Fe) and manganese (Mn) in the Arctic's Laptev and East Siberian Seas could provide a better framework for the study.

In response to the reviewer's comment, we revised the abstract to give a clearer hypothesis for this study and explain how we tackled our research questions. The revised abstract is as follows:

> "**Abstract.** The Arctic's Laptev and East Siberian Seas (LESS) are areas with high biogeochemical activity. Nutrient inputs associated with river runoff and shelf sediment-water exchange processes are vital for supporting primary production in the LESS. Relative to macronutrients, data on dissolved iron (dFe) and manganese (dMn), essential micronutrients for primary producers, have been historically sparse in the LESS. Some dFe and dMn are reportedly carried in the Central Arctic by the Transpolar Drift, a major current that directly transports Eurasian shelf water, river waters, and sea ice from the LESS continental margins. However, dFe and dMn supply to the surface waters of the LESS and subsequent biogeochemical processes are not well constrained. In the summer of 2021, we investigated the questions: *what are the sources of dFe and dMn to the surface layer* and *what factors control their concentrations and distributions on the LESS continental margins*? We demonstrated strong regional controls in dFe and dMn distribution based on distinct hydrographic regimes between the eastern side of the LESS (the East Siberian Sea and the Chukchi Abyssal Plain) and the western side (the Makarov and Amundsen Basins). Specifically, the East Siberian Sea and Chukchi Abyssal plain were governed by Pacific-sourced water and the Makarov and Amundsen Basins were influenced by Atlantic-sourced water. Pacific-sourced waters contained higher levels of dMn released from the continental shelf sediments than Atlantic-sourced water. In contrast, elevated dFe signals were not observed; which is likely because sedimentary dFe was more rapidly removed from the water column through oxidation or scavenging than dMn. The impact of river water discharge on the dFe distributions of Pacific- and Atlantic-sourced water

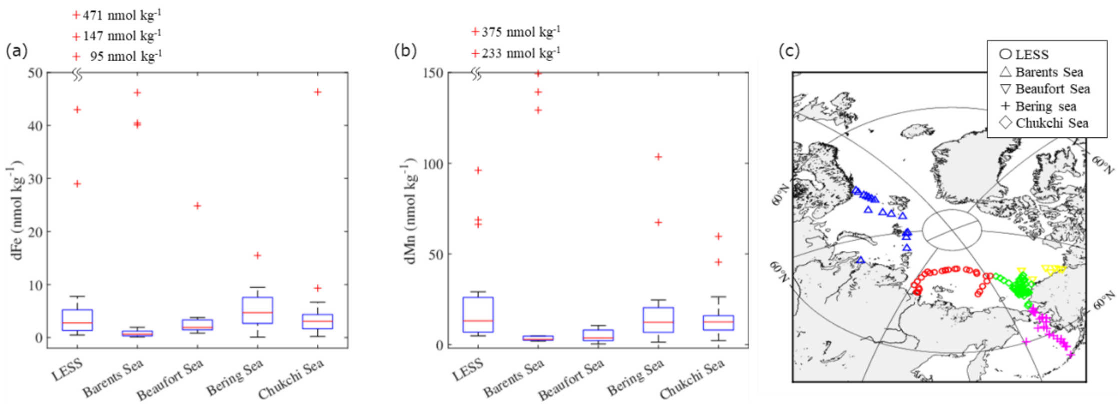
is significant. A positive correlation between the fraction of meteoric water (river water and precipitation), dFe, and humic-like colored dissolved organic matter (CDOM) in these waters confirmed a common freshwater source for dFe and CDOM. Terrigenous organic ligands likely stabilized Fe in the dissolved phase, which was not the case for Mn. Sea ice melting/formation was not a significant source during our observation period. We conclude that the major sources controlling dFe and dMn distributions in the LESS continental margins are river discharge and the input of shelf sediment. "

We had already focused on contrasting the physical and biogeochemical controls of Fe and Mn in the LESS throughout the manuscript. For example, we discussed preferential removal of Fe relative to Mn with *N**, an indicator of water passing through the reductive Chukchi Shelf. We further added a summary figure and some discussion in section 4.4. to explain how Fe and Mn data from this study compares with the previous study. We are sure that we responded well with this discussion for the reviewer's comment. The added discussions and the figure are as follows:

Section 4.4.

"To investigate the distribution patterns of dFe and dMn in surface waters, we combined our dataset with available data reported for the Arctic Ocean (Cid et al., 2011, 2012; Gerringa et al., 2021; Hölemann et al., 2005; Jensen et al., 2020; Klunder et al., 2012; Kondo et al., 2016; Middag et al., 2011; Rijkenberg et al., 2018; Savenko and Pokrovsky, 2019; GEOTRACES Intermediate Data Product Group, 2023). The dFe concentrations are relatively low (up to 2 nmol kg^{-1}) in the Atlantic sectors of the Arctic Ocean (Fig. 12a). In the surface of the Nansen Basin and Barents Sea, Fe is expected to be the first nutrient to be depleted by primary producers (Rijkenberg et al., 2018), and phytoplankton consumption could be an important sink for Fe. In the LESS continental margins, however, surface dFe concentrations are significant (>5 nmol kg^{-1}) and even persist in the late summer of 2021 (Fig. 12a). Figure 13a shows a boxplot of the surface dFe concentration combined our dataset with available data reported for each region in the Arctic Ocean (Fig. 13 a and c). All sampled water in Figure 13 has salinity larger than 25. The result shows that the median value is the highest in Bering Sea (4.7 nmol kg^{-1}) followed by Chukchi Sea (3.1 nmol kg^{-1}) and by LESS (2.8 nmol kg^{-1}). We deduce that sedimentary Fe originating from reductive Bering Shelf is gradually removed in Chukchi Sea after entering it from the Bering Strait (Jansen et al., 2020), and then penetrate East Siberian Sea. The dFe concentrations also increased toward the estuaries of the Lena, Yenisei, and Mackenzie Rivers (Fig. 12a). Other studies determined that dFe concentration in the estuary water (salinity < 25) of the Lena River was as high as 9,000 nmol kg^{-1}, as well as in the estuary waters of the Yenisei and Mackenzie Rivers. Fe-binding organic ligands in the form of humic substances originating from Lena River strongly affect the dFe concentration here, as discussed in the previous section. The natural humic substances Fe ligands of the surface Arctic Ocean have known to belong to the group of strong ligands ubiquitous in surface ocean waters (Laglera et al., 2019). The strongly complexed Fe may be less biologically available to the phytoplankton community than the weakly complexed Fe released from grazing and bacterial remineralization of organic matter (Gledhill and Buck, 2012). The river-influenced water from the LESS continental margins is the source water of the Trans Polar Drift, which enriches in dFe in the central Arctic Ocean (Fig. 12a) (Charette et al., 2020; Gerringa et al., 2021; Klunder et al., 2012).

High dMn found in the central Arctic Ocean (Fig. 12b) is also related to the presence of Trans Polar Drift (Charette et al., 2020; Gerringa et al., 2021). In addition to the riverine inputs, sediment-water column exchange over the shelves leads to relatively dMn-rich water in the Pacific sectors of the Arctic Ocean. The dMn concentrations increased toward the broad shelves of the East Siberian Sea (~68 nmol kg^{-1}), Chukchi Sea (~45 nmol kg^{-1}), and Bering Sea (~103 nmol kg^{-1}), whereas the concentration was relatively low (~8 nmol kg^{-1}) over a narrow shelf of Beaufort Sea (Fig. 12b). Boxplot of the surface dMn concentration in Figure 13b shows that the difference of median value is relatively small among LESS (13.0 nmol kg^{-1}), Chukchi Sea (12.4 nmol kg^{-1}), and Bering Sea (12.2 nmol kg^{-1}). As previously discussed, the shelf sediment-water exchange processes over the Chukchi Shelf largely influence the Fe and Mn distributions in the East Siberian Sea. Vieira et al. (2019) provided the first estimate of the benthic flux of the radium isotope (Ra^{228}) in the Chukchi Sea as tracers of benthic trace metal inputs, which was among the highest rates reported globally. The low-N* water spreads over regions where nitrate is already depleted relative to phosphate, mainly due to the oxidation of organic matter by bacteria in the reductive shelf sediment (Fig. 12c). The East Siberian Sea, as well as Chukchi Sea, are likely hotspots of sediment-sourced dMn via the reductive dissolution of Mn oxide in the sediment. A multi-step removal process of dMn has been suggested in the Arctic (Jansen et al., 2020): dMn is rapidly removed to the particulate phase within 150 km of the shelf break, but some dMn remains conserved through the next 1000 km away from the shelf. The dMn originating from the LESS continental margins appears to be exported by the Trans Polar Drift to the central Arctic Ocean effectively (Fig. 12b), even though stabilization by organic complexation is unlikely for Mn during offshore transport. “



“**Figure 13** Boxplots of (a) dFe and (b) dMn concentrations in surface water (< 25 m depth) in each region of the Arctic Ocean. (c) Location of stations used in this boxplot. The bottom and top of the box in (a)–(b) indicate the 25th and 75th percentiles, respectively, and the line inside the box indicates the median. The bottom and top error bar in (a)–(b) show minimum and maximum values, respectively, and the outliers are plotted individually using the '+' marker symbol. Some outliers are plotted the outside of figures to better show differences between regions. “

Second, the labeling of the figures is inadequate; nearly all figures lack titles, necessitating readers to refer to figure descriptions for context. Moreover, the figure descriptions do not adequately explain the terms used, and there is a notable presence of technical jargon throughout the study.

We have carefully and thoroughly revised the figures and their captions. All figures were provided titles for conciseness, and their descriptions were strived for clarity. Please check the revised all figures and their captions at the end of this file.

Third, the manuscript overuses acronyms, which can hinder readability.

Following the reviewer's indications, we avoided using acronyms as much as we can throughout the text. For example, we decided not to use acronyms of Amundsen Basin, Makarov Basin, Chukchi Abyssal Plain, and East Siberian Sea in the text, which strongly hinder readability.

Finally, some sentences contain incomplete reasoning, which detracts from the overall coherence of the manuscript. Below, I highlight few examples.

We revised the text following the reviewer's comments below. Detailed responses to the reviewer's comments are as follows.

<u>Minor comments</u>

2) RC1 Line 165: Too many acronyms already, you don't need S and T for temperature and salinity.

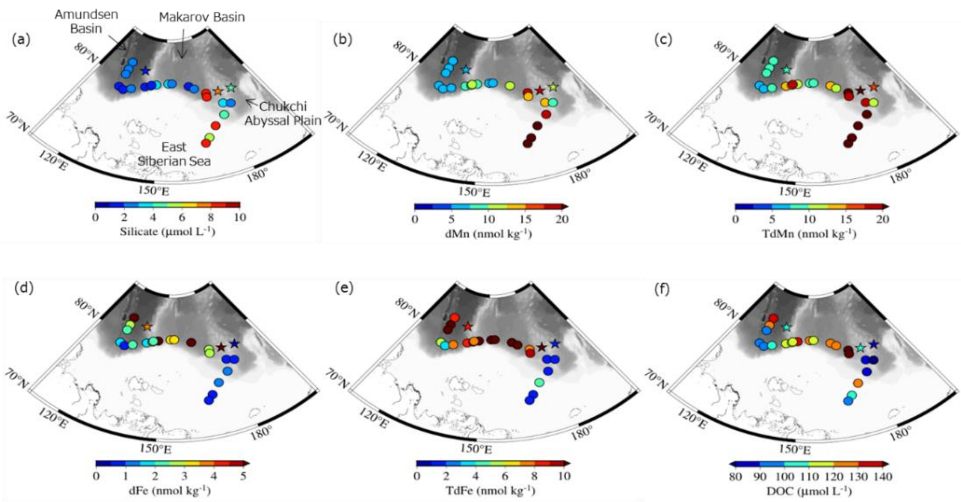
We deleted them as the reviewer suggested.

3) RC1 Line 165: "The T of water above 25 m exceeded 0°C at 125−145 °E (Figs. S1 and S2), owing to the atmospheric radiative forcing" It is not clear what you mean in this sentence. What does radiative forcing have to do with surface ocean temperature? These two concepts are obviously related, but it is unclear how you are using them

We realized that this sentence had been over-interpreted. We did not measure atmospheric radiative forcing. Therefore, we removed this sentence from the text.

4) RC1 Line 190: Define the terms in your figures.

We defined the terms in the figure caption as the reviewer suggested. The revised figure and its caption are as follows:



"**Figure 3** Spatial distributions of (a) silicate, (b) dMn, (c) TdMn, (d) dFe, (e) TdFe, and (f) DOC in the surface of the Arctic's Laptev and East Siberian Seas. "

5) RC1 Line 215: "In contrast, a negative value of fsim along the continental slope suggests that sea ice formation is dominant in the region," which region?

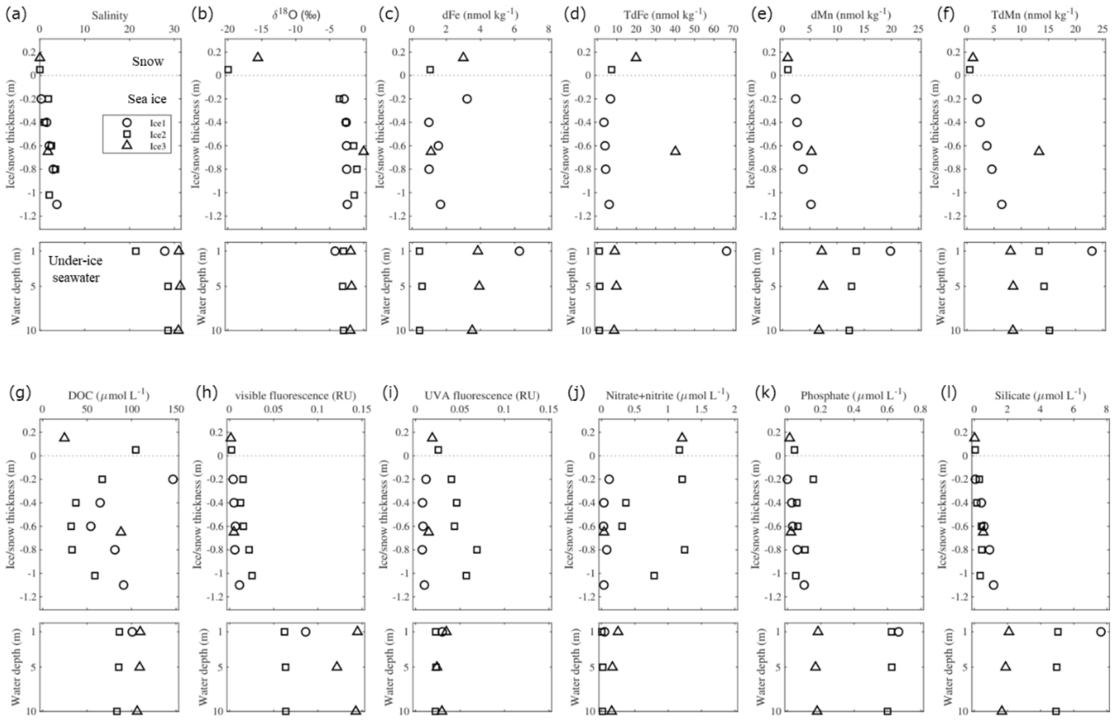
We revised the sentence as follows:

Line 215:

"In contrast, a negative value of f_{sim} in the Makarov Basin suggests that sea ice formation is dominant, in agreement with..."

6) RC1 Figure 4: Same issue with Figure 3, but you sometimes use full names. Be consistent.

We revised the terms in the figure following the reviewer's comment. We decided not to use 'Si', 'N+N', and 'P' because these terms were not used frequently throughout the manuscript. We also do not use the acronym of salinity (S). The revised figure and its caption are as follows:



"**Figure 5** Vertical profiles of (a) salinity, (b) δ^{18}O, (c) dFe, (d) TdFe, (e) dMn, (f) TdMn, (g) DOC, (h) visible fluorescence, (i) UVA fluorescence, (j) nitrate+nitrite, (k) phosphate, and (l) silicate in snow, sea ice, and under-ice water, respectively. "

7) RC1 Line 255: {The dMn concentration in the surface waters gradually increased with decreasing salinity, whereas the dFe concentration did not show a similar increasing trend (Fig. 5)} This statement is not a complete interpretation of figure 5, can you explain the exceptions

In response to the comments from the reviewer, we revised the sentence as follows:

Line 255:

"The dMn concentration in the surface waters gradually increased with decreasing salinity (Fig. 6b). The dFe concentration also shows a similar increasing trend among the salinity ranges from 31 to 34 (Fig. 6a), however, the dFe concentration in less saline waters (salinity < 31) is irrespective of salinity variation. "

Figure 1

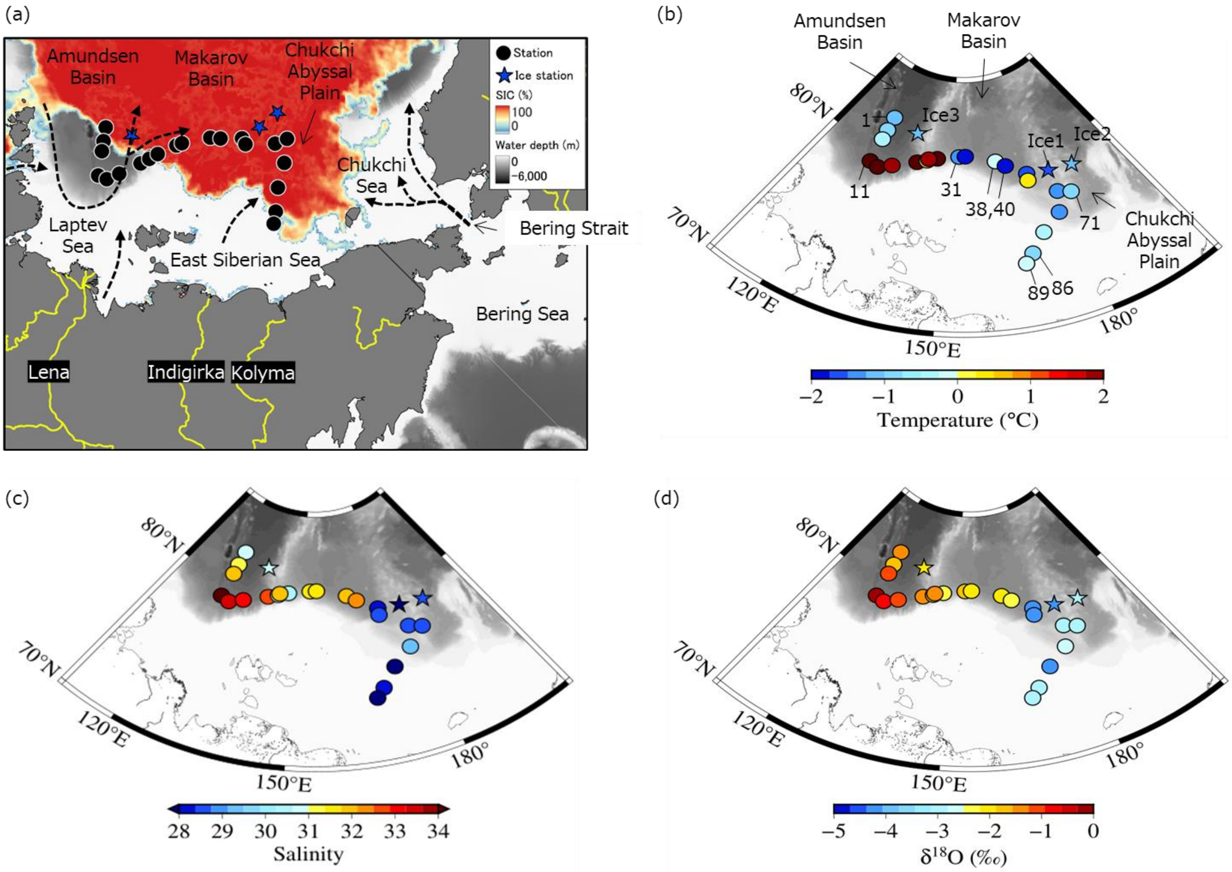


Figure 1 (a) Location of stations sampled in the Arctic's Laptev and East Siberian Seas (LESS) in the late summer of 2021. Spatial distributions of (b) water temperature, (c) salinity, and (d) $\delta^{18}O$ in the surface. Sea ice concentration (SIC, %) on 1 October 2021 (GCOM-W/AMSR2, Japan Aerospace Exploration Agency) and general water flow on the surface of LESS (Anderson et al., 2015; Bauch et al., 2018; Clement Kinney et al., 2022; Doglioni et al., 2022; Rudels et al., 2004; Stabeno et al., 2018) were depicted in (a).

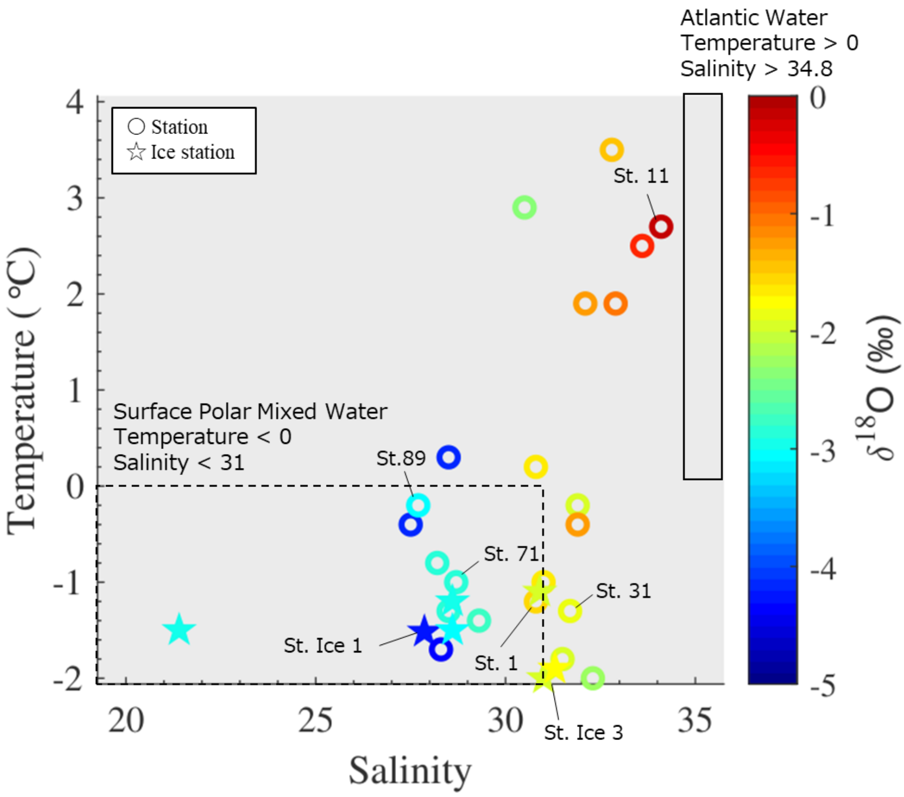


Figure 2 Temperature versus salinity diagram on the surface with stations sampled in the Arctic's Laptev and East Siberian Seas. The color scale shows the $\delta^{18}O$ values in each water sample. The temperature and salinity ranges of Surface Polar Mixed Water and Atlantic Water are indicated by the area surrounded by dashed and solid lines, respectively.

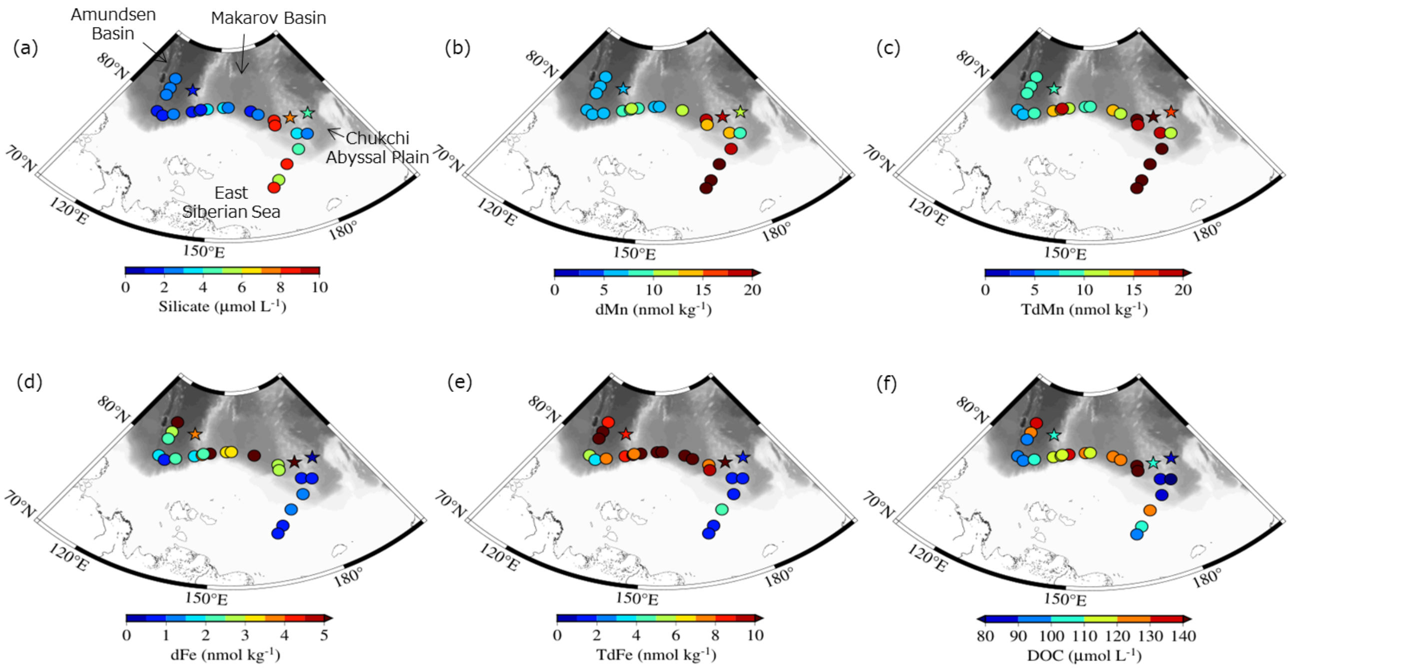


Figure 3 Spatial distributions of (a) silicate, (b) dMn, (c) TdMn, (d) dFe, (e) TdFe, and (f) DOC in the surface of the Arctic's Laptev and East Siberian Seas.

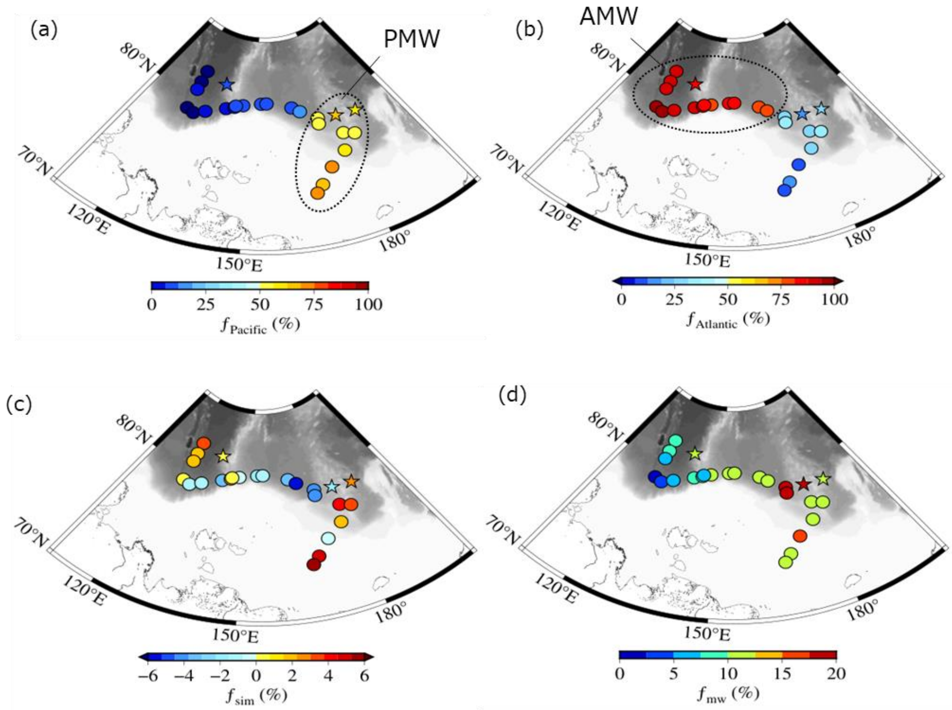


Figure 4 Spatial distributions of fractional (a) Pacific Water (f_{Pacific}), (b) Atlantic Water (f_{Atlantic}), (c) sea ice meltwater (f_{sim}), and (d) meteoric water (f_{mw}) in the surface of the Arctic's Laptev and East Siberian Seas. Abbreviations in (a) and (b): Surface Polar Mixed Water (PMW) and Surface Atlantic Mixed Water (AMW).

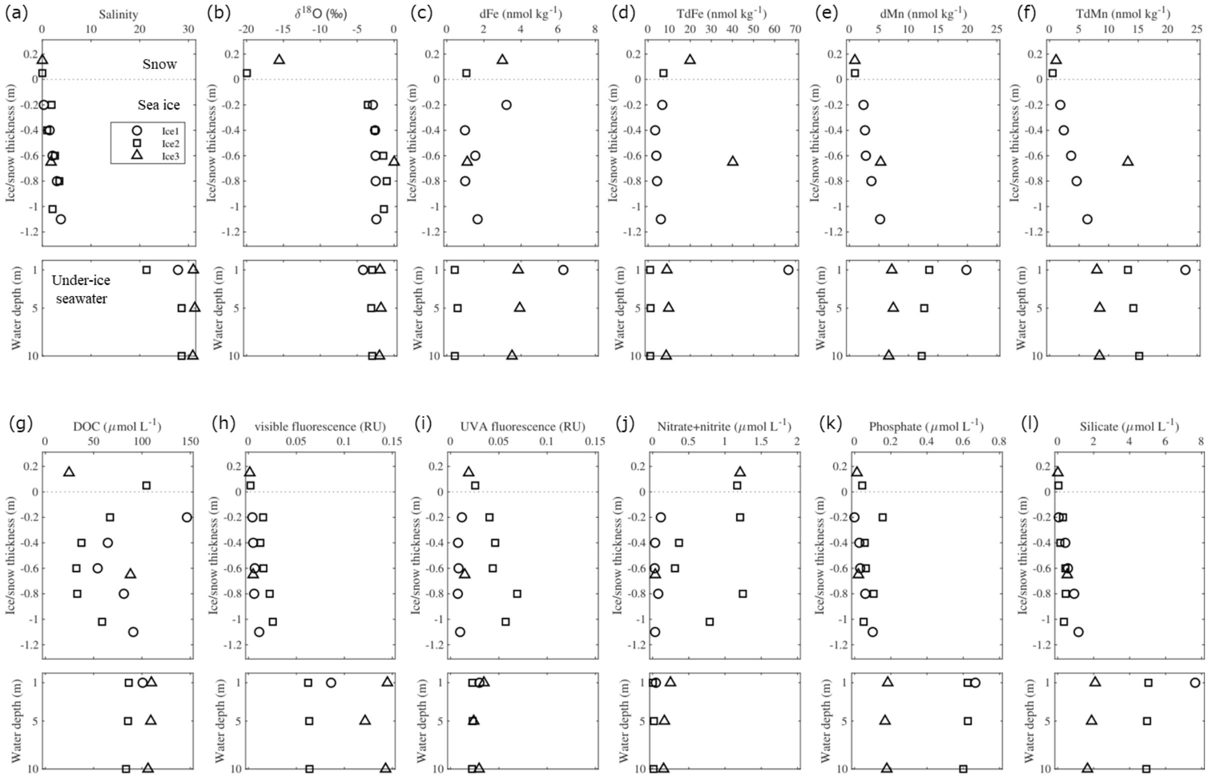


Figure 5 Vertical profiles of (a) salinity, (b) $\delta^{18}O$, (c) dFe, (d) TdFe, (e) dMn, (f) TdMn, (g) DOC, (h) visible fluorescence, (i) UVA fluorescence, (j) nitrate+nitrite, (k) phosphate, and (l) silicate in snow, sea ice, and under-ice water, respectively.

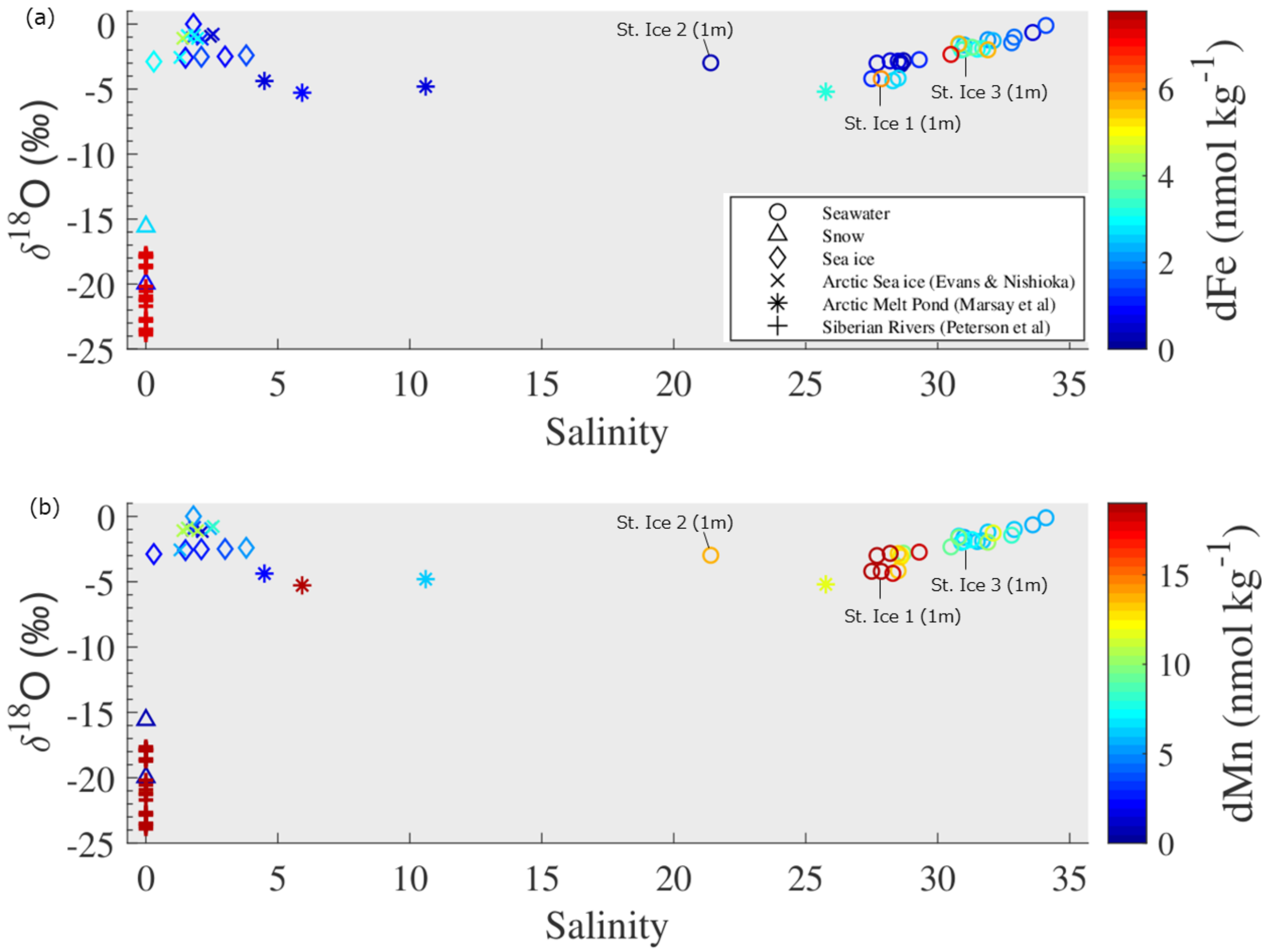


Figure 6 Salinity versus δ^{18}O diagram in seawater, snow, and sea ice. The color scale shows concentrations of (a) dFe and (b) dMn in each water sample. The values of other freshwater sources are cited from Evans and Nishioka (2019), Marsay et al. (2018), and Peterson et al. (2016).

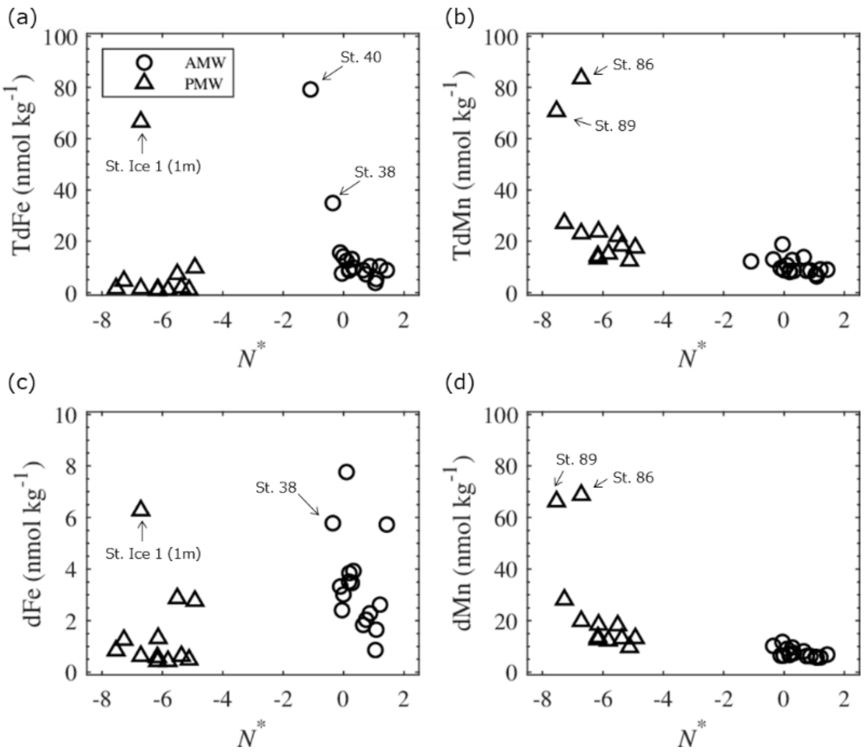


Figure 7 Plots of (a) TdFe, (b) TdMn, (c) dFe, and (d) dMn in Surface Atlantic Mixed Water (AMW) and Surface Polar Mixed Water (PMW) against *N** values.

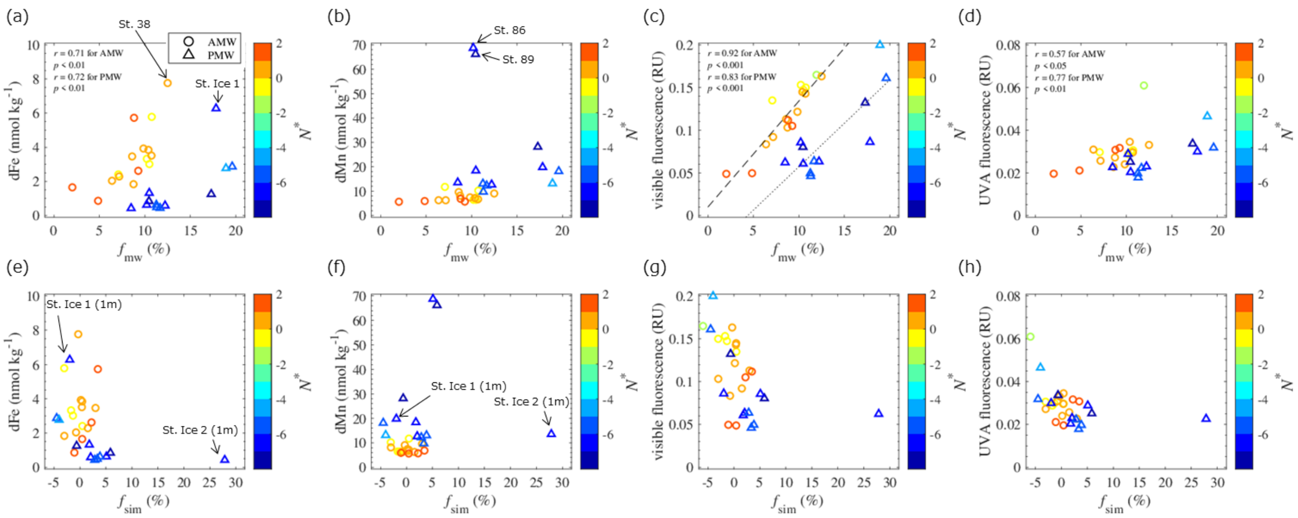


Figure 8 Plots of dFe, dMn, visible fluorescence, and UVA fluorescence in Surface Atlantic Mixed Water (AMW) and Surface Polar Mixed Water (PMW) against fractional meteoric water (f_{mw}) in (a)–(d) and sea ice meltwater (f_{sim}) in (e)–(h). The color scale shows the N* values of each water sample. Linear relationships were evaluated based on Pearson correlation coefficients (r). The liner fits of the visible fluorescence-f_{mw} relationships in the AMW and PMW are shown by dashed and dotted lines in (c), respectively.

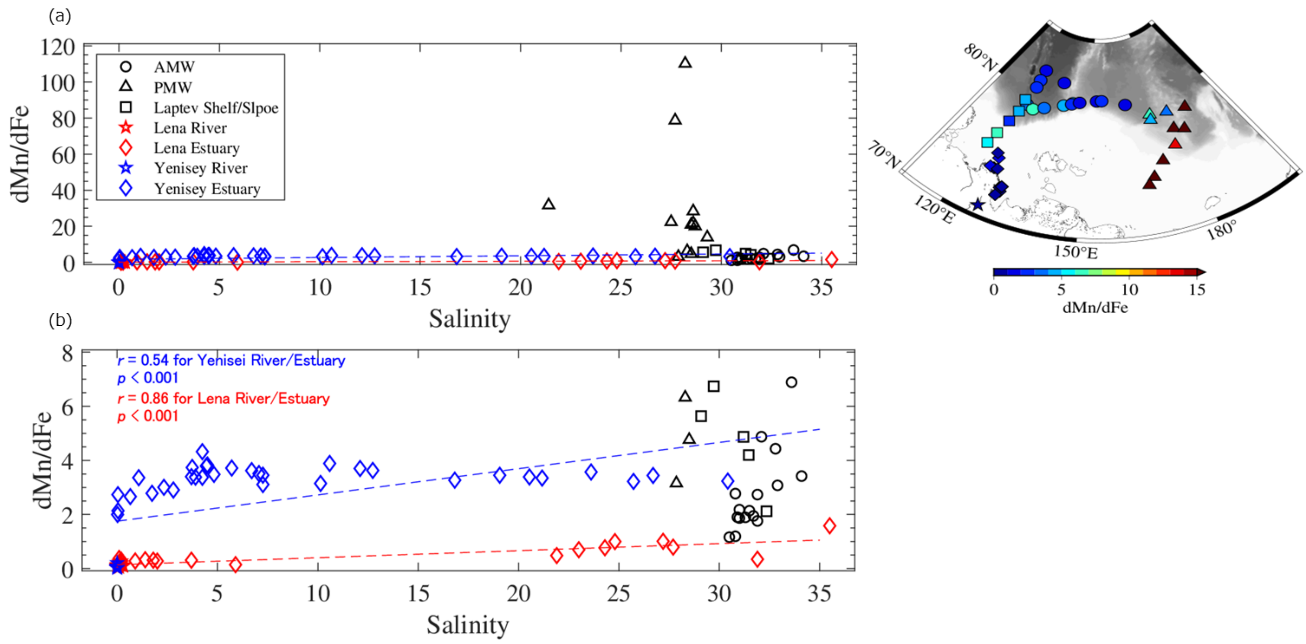


Figure 9 (a) Relationships between salinity and dFe/dMn ratio in specific water mass. A smaller scale of the Y-axis is shown in (b). The geographical distribution of each sample is indicated by different symbols as shown in the map. Mixing lines of freshwater (Lena and Yenisei Rivers) and seawater (Lena and Yenisei estuaries) are derived from Hölemann et al. (2005), Peterson et al. (2016), and Savenko and Pokrovsky (2019). Linear relationships were evaluated against the freshwater-seawater line based on Pearson correlation coefficients (*r*). The value for surface water on the Laptev Shelf/Slope (Klunder et al., 2012; Middag et al., 2011) is plotted for comparison.

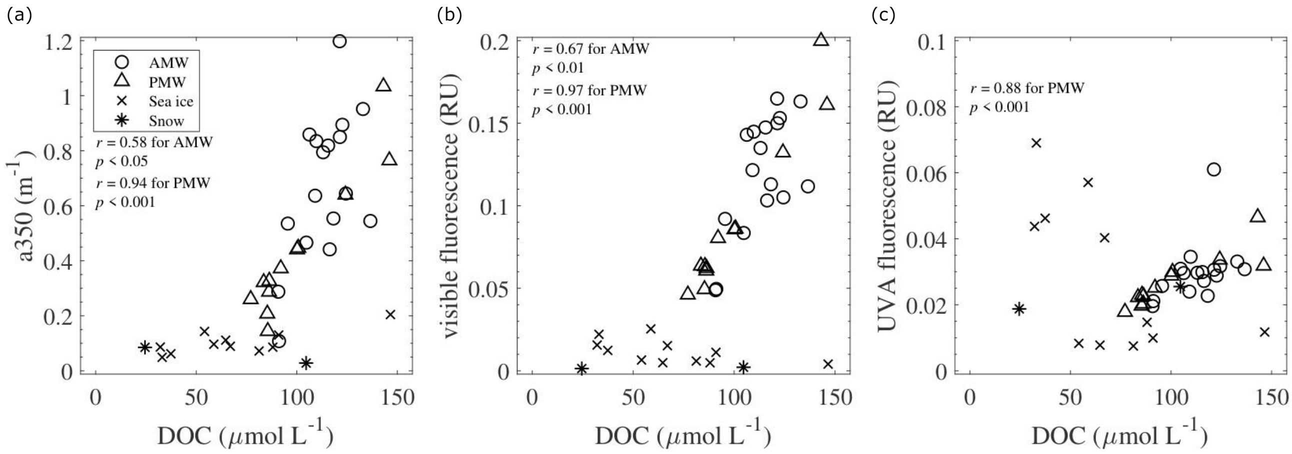


Figure 10 Relationships of DOC with (a) a_{350}, (b) visible fluorescence, and (c) UVA fluorescence in Surface Atlantic Mixed Water (AMW), Surface Polar Mixed Water (PMW), sea ice, and snow. Linear relationships were evaluated based on Pearson correlation coefficients (r).

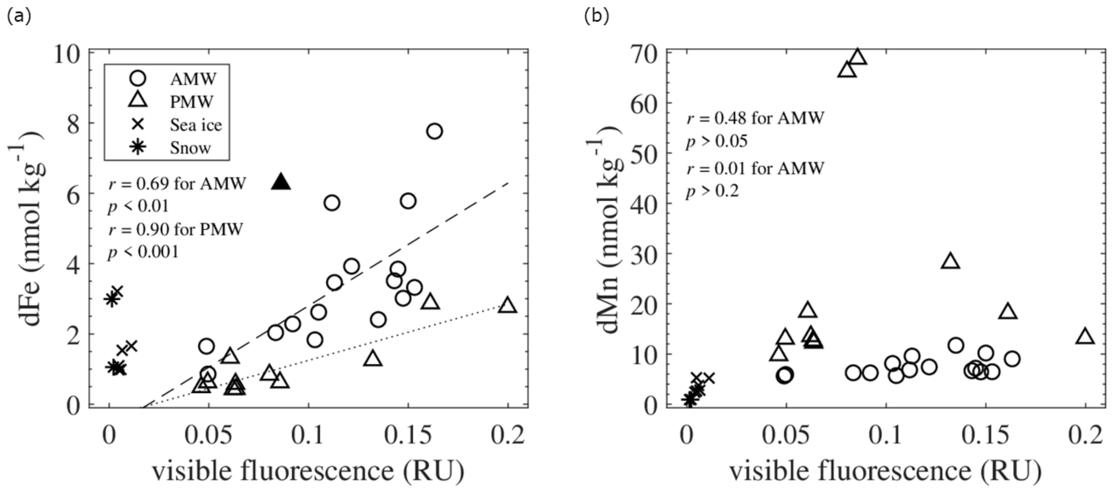


Figure 11 Relationships of visible fluorescence with (a) dFe and (b) dMn in Surface Atlantic Mixed Water (AMW), Surface Polar Mixed Water (PMW), sea ice, and snow. Linear relationships were evaluated based on Pearson correlation coefficients (*r*), except for an outlier (▲) in (a). The linear fits of the relationships in the AMW and PMW are shown by dashed and dotted lines in (a), respectively.

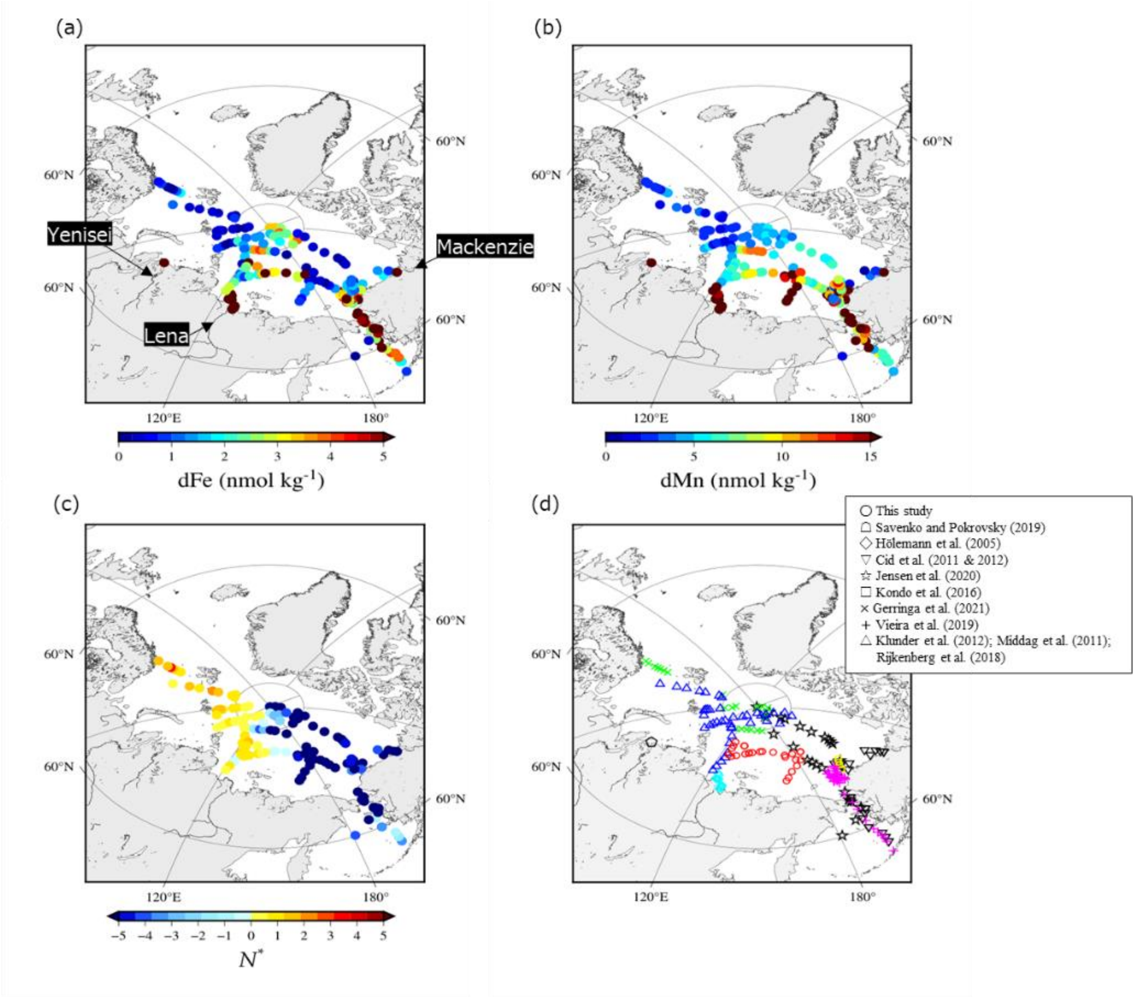


Figure 12 Spatial distributions in (a) dFe, (b) dMn, and (c) N^* in surface water (< 25 m depth) in the Arctic Ocean. (d) Location of stations on dFe, dMn, and N^* reported by this study and the previous studies.

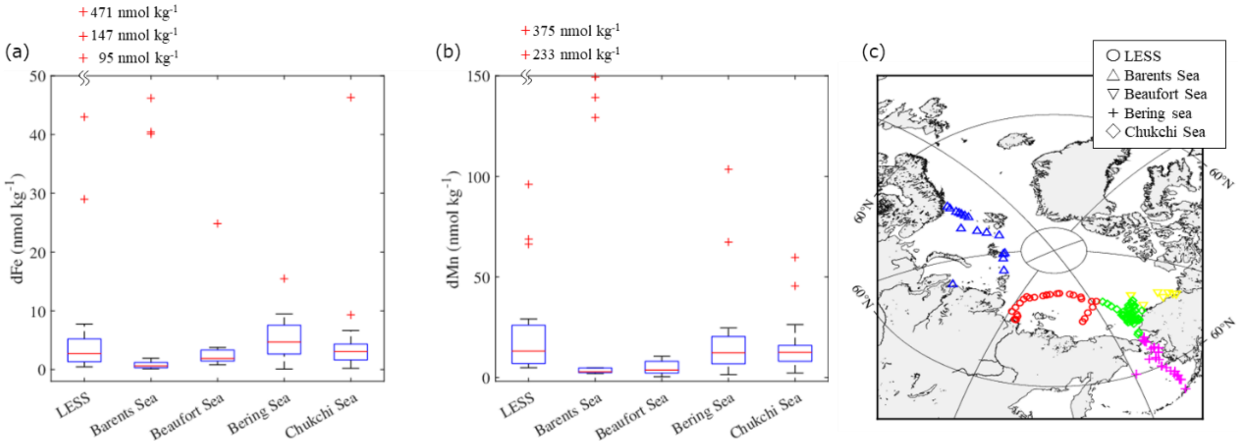


Figure 13 Boxplots of (a) dFe and (b) dMn concentrations in surface water (< 25 m depth) in each region of the Arctic Ocean. (c) Location of stations used in this boxplot. The bottom and top of the box in (a)–(b) indicate the 25th and 75th percentiles, respectively, and the line inside the box indicates the median. The bottom and top error bar in (a)–(b) show minimum and maximum values, respectively, and the outliers are plotted individually using the '+' marker symbol. Some outliers are plotted the outside of figures to better show differences between regions.