Response to Anonymous Referee #2

We thank Referee #2 for the time and effort devoted to the review of our manuscript. Below we reproduce his/her comments and address them point by point. The reviewer's comments are in regular font black with our responses in green. Text from the manuscript is shown in italics and changes are shown with deletions in strikethrough and additions in bold.

The present study of distribution of iron and its isotopes along the 600 km long Lena River freshwater plume in the Laptev Sea is aimed to understand the iron pathways to the Arctic Ocean. For this purpose authors have studied the Fe concentrations and its isotope distribution in three phases mainly, particulate, colloidal and truly dissolved. The study is important to understand how the riverine Fe is exported to open ocean. Authors have a very limited particulate Fe data (6 stations out of 10) and also colloidal Fe data (4 stations out of 10) presented in this study. Based on this small data set, I believe it is difficult to explain the Fe pathways. Apart from that, the truly dissolved iron isotope composition of any station is not given and explained. But in the objectives and in methodology they have mentioned about the truly dissolved iron isotope composition. It is difficult to understand the iron isotope fractionation during the estuarine mixing without truly dissolved iron isotope composition. There are many other short comings in the MS for example, very crucial analytical methodology for iron isotopes was not written clearly, units in the text and figures are different, citing the actual concentrations in the text are not correct etc. So I can't recommend the manuscript in its present form to publish in BGD journal. Please find the comments below as page wise.

Comments: Please find the comments and question in the font in bold.

Page 1

Line 8: Number 3 for author affiliation is missing.

Thank you for pointing out this mistake. It has been corrected.

"Department of Geosciences, Swedish Museum of Natural History, Stockholm, Sweden"

Page 3

Line4-5: third reference is not cited properly

The reference has been removed, as it just provides measurements from other studies.

Line 18: This study presents Fe concentrations and Fe isotope compositions in the particulate, colloidal, and truly dissolved phase along the Lena River freshwater plume in the Laptev Sea.

I couldn't find the Fe isotope data for the truly dissolved phase.

That is correct. We do not present Fe isotope data for the truly dissolved phase. We rephrased the sentence. The Fe concentration of the truly dissolved phase is too small to achieve reliable Fe isotope data.

This study presents Fe concentrations and Fe isotope compositions in the particulate **and** colloidal phase along the Lena River freshwater plume in the Laptev Sea, **as well as Fe concentrations in the truly dissolved phase**.

Line 21: The main objectives were to study the distribution of Feinthe Lena River–Laptev Sea transect and the variations in the partitioning of Fe between the different size fractions, as well as to identify the impact of processes such as mixing, transformation, and removal by settling on the export of Fe to the deeper ocean.

To explain the above mentioned processes and partitioning of Fe between different size fractions truly dissolved phase studies are very important (both concentration and isotope composition). But the concentrations are reported only for 4 samples out of 10 samples and the isotope compositions have not reported for truly dissolved phase.

This concern was also raised by reviewer #1.

We agree that it would be good to have a more detailed profile along the Lena River freshwater plume with more samples separated by ultrafiltration. The Lena River freshwater plume was sampled during the 50-days ISSS-08 field campaign. The number of samples, which can be obtained during sampling such a remote and inaccessible area is limited by logistical constraints, e.g. sampling permissions. Furthermore, the process of cross-flow ultrafiltration is quite time consuming and could not be carried out at all stations. In total, water from 15 stations were filtered with this methodology.

The samples from the Lena River freshwater plume might be limited, but they give insights into the different size fraction for isotopes and the riverine transport of Fe to the open ocean. The distribution of Fe between the three size fractions (particulate, colloidal, and truly dissolved) and their concentration range along the Lena River freshwater plume can be used to identify different estuarine processes, e.g. mixing, transformation, and removal by settling. The Fe isotope composition sheds light on the composition of Fe particles and colloids, which are transported into the Arctic Ocean. We think this information are valuable and potential future sampling regimes, with a more comprehensive sampling along the Lena River freshwater plume, will deepen the knowledge further and confirm our conclusions.

The reason that there are no Fe isotope values reported is that they have not been measured during this project. The samples have been measured at ALS Scandinavia AB, where we have a cooperation with I. Rodushkin and E. Engström. The minimum amount of Fe concentration in a sample should be 0.1 mg/L to measure reliable Fe isotope concentrations.

Page 5

Table 1: unit for salinity?

The salinity is displayed in the practical salinity scale. The International Association for the Physical Science of the Ocean (IAPSO) recommends that the practical salinity is expressed by dimensionless numbers only. We added the information that the salinity is expressed on the Practical Salinity Scale to the table caption.

Salinity is based on the Practical Salinity Scale PSS-78.

Line 10: Which acid and what concentration was used for acidification?

We added the missing information. The water samples were acidified with ultrapure HNO₃.

All water samples were stored in acid-cleaned polyethylene (PE) bottles and acidified with ultrapure HNO₃ to a pH <2, and all nitrocellulose filters (0.22 μ m, Millipore[®]) were stored at -18°C until further analysis (Ödman et al., 1999).

Page 6

Line 19: For element analysis, the water samples were diluted (2-200 fold) with 10 % HNO3.

What grade acid was used and what are the blank levels?

We used high purity Suprapure ® acids during the whole sample treatment and the analysis. The information can be found in the second sentence under 2.3 Analytical Methods.

Rodushkin, Engström, and Baxter (2010) summarized the routines at ALS Scandinavia AB and reported concentrations of Fe in Milli_Q water with 1% HNO₃ after several purification steps (3.6 ng/L (SD: 0.7 ng/L).

High purity Suprapure[®] acids were used throughout sample treatment and analysis.

Line 20: For Fe analysis, the samples were diluted by a factor of 50.

Fe concentrations at such a low level (nano-molar level) require a proper sample handling and preprocessing of samples in clean rooms etc. Authors have not mentioned anything about that and also what standard was used?

We added the following information to the text.

All sample manipulations were performed in a clean laboratory (Class 10 000) by personnel wearing clean room gear and following all general precautions to reduce contamination (Rodushkin, Engström, and Baxter, 2010).

The analytical procedure was validated with different reference materials (SLRS-4 River Water CRM for Trace Metals, SLEW-2 Estuarine Water CRM for Trace Metals and NASS-4 open ocean water (all supplied from National Research Council, Ottawa, Canada) (Rodushkin et al., 2005; Rodushkin et al. 2016).

Page 7

Line 6-7: For the Fe isotope ratio measurements, water samples and digested filters were evaporated to dryness, and the residue was redissolved in 1 mL 9M HCI.

There is no data shown for the water samples in the whole manuscript, I really do not know how they wrote about water samples. They might have analyzed the water samples for Fe isotope composition but not included in the MS?

When we wrote water samples we were referring to the colloidal fraction (1kDa-0.22µM). We added additional information (under 2.3 Analytical methods) to the elemental analysis, where the colloidal and truly dissolved phase are the water samples and to the Fe isotope analysis, where just the colloidal phase was measured. Please see text below.

For element analysis, the water samples (colloidal: 1 kDa to 0.22µm; truly dissolved: <1 kDa) were diluted (2-200 fold) with 10 % HNO₃.

For the Fe isotope ratio measurements, water **samples (colloidal: 1 kDa to 0.22µm)** and digested filters were evaporated to dryness, and the residue was re-dissolved in 1 mL 9M HCl.

Line 7: Iron was separated from the matrix by ion exchange, with a recovery rate above 95%.

Details about the column chemistry is necessary. I wonder how come they haven't cited any paper for their methodology.

Thank you for pointing out this flaw. Apparently, we missed to cite the column chemistry. We added some additional text about the column chemistry to the method section. Please see below.

Iron was separated from the matrix elements by using an AG-MP-1M ion-exchange resin (Ingri et al., 2006; Rodushkin and Ruth, 1997). After the sample was loaded, the matrix was washed with 9.6 M HCl, and Cu was eluted with 8 ml 5M Cl. Afterwards, Fe was eluted with 6 ml 2 M HCl and can be used for further steps (Rodushkin et al. 2015). After evaporating to dryness, 50 μL of concentrated HNO₃ was pipetted directly to the residue followed by the addition of 5 mL MQ-water. Samples with high Fe content were diluted with 0.2M HNO₃ to a concentration of 2 mgL⁻¹ in the measurement solutions. Low Fe concentration water samples were further diluted to 40-50 μgL⁻¹ and measured using high-efficiency desolvation nebulizer (Aridus) in a separate analytical sequence.

Line 14-15: We only discuss the δ 56Fe in this study, although all Fe isotope data are reported in Table 3 including 2σ (n=4;).

Which standards were analyzed prior to the sample analysis? It is very important to report the standard values along with their precision and accuracy to make sure the presented data is of good quality.

The Fe isotope method was validated using two reference materials from the US Geological Survey (A1 and P1) by Malinovsky et al. 2003. Furthermore, to assure the quality of the procedure and measurement the Fe standard IRMM-14 undergoes the entire separation process as a known sample.

Page 8

Table 3: Fe isotope data for the sediment and the particulate and colloidal phase.

The description in the legend is not followed in the table. The 2 σ is high for the samples with highest concentrations.

We changed the caption of the figure to make it easier for the reader to follow the manuscript. Furthermore, we would like to make you aware of the fact, that the former table 3 is now table 4. The second reviewer suggested to include a table with the Fe concentration to the manuscript, which can be found in table 3.

Table 4: Fe isotope data for the sediment and the particulate and the colloidal phase, as well as Fe isotope data for the surface sediments.

We cannot see a correlation between high Fe concentrations and high 2σ , which we would expect if there is any systematic error in the measurements. Furthermore, we checked if we could find a relationship between DOC and 2 σ , as it can occur that Fe isotope measurements can be problematic. In samples with high organic carbon contents. We could not find a correlation between the two parameters, why we would exclude problems caused by DOC at the Fe isotope measurements.

Page 9

Line 1: I really do not see the necessity for the supplementary material S2 in which only organic carbon data is given and rest is duplication of Table 1.

We agree and included the organic carbon concentrations into Table 1 of the manuscript. Furthermore, we added pH and oxygen saturation values. The new supplement consists of Fig. S1, which contains the three-isotope plot and Tab. S2, which includes all Fe isotope values.

Line 4: Please cite the S2 after numbers. And change the concentration 300 to 320.

Changed accordingly. We refer to Table 1, as we moved the DOC data to that table.

The DOC concentrations show a small variation of between 320 and 440 μ M in the surface waters of the inner and outer plume (**Tab. 1**; Fig. 3).

Line 6-8: It has been shown that DOC is behaving conservatively during mixing between Lena River water and Arctic Ocean water along the sampling profile (Alling et al., 2010; Opsahl et al., 1999; Pugachetal., 2018).

Does sampling profile means the same transect of the present study?

The references Alling et al. (2010) and Pugach et al. (2018) use samples from the same cruise (ISSS-08) in their studies. So yes, they are from the same transect as data from this study. Opsahl et al. published data from the Arctic Ocean, e.g. in the Laptev Sea. These samples were not taken at the same place as samples from the ISSS-08 cruise.

 $Line 16: The pFe concentration decreased from 56 to 0.1 \mu M \ along the Lena River freshwater plume (Fig. 4).$

Figure 4 do not have the same units of μ M. It is good practice to use the same units in both table and figure for easy understanding.

Thank you for pointing this out. We changed it accordingly throughout the text, tables and figures towards µM. We decided to keep the truly dissolved Fe concentration in nM.

Line 16-17: Between the inner and the outer plumes (i.e. between YS-10 and YS-9), the pFe concentration dropped to 0.9 μ M, a loss of 98% of pFe.

I do not find any data either in table or in figure.

We understand the confusion. Indeed we do not have measurements at station YS-9 or YS-10. In an earlier version of Fig. 4, we included all stations of the profile for a better visualization where each station is located. Then it occurred that the Fe drops between these two stations. We changed the station numbers in the text towards station YS-11 and YS8.

Between the inner and the outer plumes (i.e. between **YS-11** and **YS-8**), the PFe concentration dropped to 0.9 μ M, a loss of 98% of PFe. The CFe concentration decreased from 0.6 to 0.1 μ M along the freshwater plume, a loss of about 85% CFe (Fig 4).

Page 10

Line 4-5: In the Laptev Sea close to the river mouth about 18% of the total OC was present as POC and this was apparently rapidly lost during mixing (Fig. 5)

Please put full stop after the sentence.

We added the full stop. Furthermore, we referred to the wrong figure, which we changed.

In the Laptev Sea close to the river mouth about 18% of the total OC was present as POC and this was apparently rapidly lost during mixing (Fig. **3**).

Line 11-12: The pFe concentrations found in the Laptev Sea close to the shore are higher than the average pFe concentration in the Lena River, but similar to the highest river values up to 32 μ M(Hirst et al., 2017).

These values are similar to the pFe values of Hirst et al., 2017? Please state which phase it is? Yes, we were referring to the PFe values in the Lena River, published by Hirst et al. (2017). We added this information.

The PFe concentrations found in the Laptev Sea close to the shore are higher than the average PFe concentration in the Lena River, but similar to the highest **PFe** river values up to $32 \mu M$ (Hirst et al., 2017).

Line 12-13: The cFe and dFe in the Lena River (Hirst et al., 2017) showed higher average concentrations (cFe: 1.5 μ M; dFe: 54 nM) that are similar to concentrations found in the Lena River – Laptev Sea transect.

I do not see any values close to 1.5 µM in the present study. They are only half of the reported values.

That is correct, we changed the sentence. We furthermore added some explanation, why the concentration of CFe and DFe is higher in the Lena River than in the Lena River freshwater plume.

The CFe and DFe in the Lena River (Hirst et al., 2017) showed higher average concentrations (CFe: 1.5 μ M; DFe: 54 nM) thant are similar to concentrations found in the Lena River – Laptev Sea transect. Most likely some of the CFe and DFe from Lena River already flocculated at salinities below 1, where the first sample of our sampling profile was taken (YS-14).

Line 24: We observed non-conservative mixing of pFe at salinities lower than 5 and conservative mixing at salinities higher than 5.

Please do cite the figure.

We added the reference to Figure 5 here.

We observed non-conservative mixing of PFe at salinities lower than 5 and conservative mixing at salinities higher than 5 (Fig. 5).

Line 26-28: Organic C hinders the coagulation of the particles during riverine transport, but in the estuarine mixing zone the negatively charged particles will react with seawater cations and form largeraggregates (Boyle et al., 1977).

What are these negatively charged particles?

We added the information.

Organic C hinders the coagulation of the particles during riverine transport, but in the estuarine mixing zone the negatively charged **iron-bearing** particles will react with seawater cations and form larger aggregates (Boyle et al., 1977).

Page 11

Line 1: freshwater plume is likely due removal of

Freshwater plume is likely due to removal of ...

Added to the text.

The large amount of pFe (98%) lost in the inner Lena River freshwater plume is likely due **to** removal of chemically reactive ferrihydrite, which is the main form of particulate Fe in the Lena River.

Line 8: The truly dissolved Fe (<1kDa) concentrations along the freshwater plume are almost constant around 8nM

But the concentration in the river mouth is about 1 nM (S2). Is the value not correct?

Yes, the DFe concentration at station YS-14, closest to the coastline, was 1 nM, according to our measurements. This low concentration is surprising, when looking at the DFe in Lena River (about 54 nM, Hirst et al. 2017) and at the stations further out in the Lena River freshwater plume (about 8 nM, this study). Furthermore, earlier studies

showed DFe concentrations in a similar range (3 to 10 nM in the upper 20 m in the Laptev Sea (Klunder et al. 2012) and 36 to 44 nM (Ob River) and 9 nM (Yenisey River) with almost constant concentrations along their estuaries (Dai and martin, 1995)). We cannot explain this low concentration at station YS-14, but the overall trend of this study (and earlier studies) is that the truly dissolved Fe concentration behaves almost constant along the freshwater plume.

Line 8-10: These observations are in accordance with previous studies in the Laptev Sea where dissolved Fe concentrations of >10 nM has been 10 reported (Klunder et al., 2012)

These Fe concentrations are from the surface? Because the concentrations may vary from surface to the bottom.

Thank you for pointing this out. You are correct that concentrations > 10 nM were measured in the bottom water samples in the Laptev Sea by Klunder et al (2012). In the upper 20 m they measured values of 3 to 10 nM. We changed this accordingly in the manuscript.

These observations are in accordance with previous studies in the Laptev Sea where dissolved Fe concentrations of **3 to**10 nM **in the upper 20 m** has been reported (Klunder et al., 2012).

Line 27-29: In these areas, within the fully oxidized water column, the pFe phase show negative δ56Fe values, while the dissolved phase generally shows higher values (Escoube et al., 2015, 2009; Ingri et al., 2006; Staubwasser et al., 2013; Zhang et al., 2015).

What does it mean by higher values? Author means positive $\delta 56Fe$?

By higher values we were referring to δ^{56} Fe values in the DFe phase, which are more enriched in the heavy Fe(III) compared to the δ^{56} Fe of the PFe phase. This does not have to be a positive δ^{56} Fe, e.g. Staubwasser et al. (2013), report negative values for the PFe and less negative (and positive) δ^{56} Fe values for the DFe. Other studies, e.g. Escoube et al. (2009), report negative δ^{56} Fe for the PFe phase and positive d56Fe for the DFe phase. We rephrased the sentence for the better understanding.

In these areas, within the fully oxidized water column, the PFe phase show negative δ^{56} Fe values, while the dissolved phase generally shows values, enriched in *Fe(III) compared to the PFe phase* (Escoube et al., 2015, 2009; Ingri et al., 2006; Staubwasser et al., 2013; Zhang et al., 2015).

Line 27-29: Hirst et al. (in prep.) show a seasonal dependence of the δ 56Fe composition of the dissolved fraction (colloidal and truly dissolved) in the Lena River, with summer flow δ 56 30 Fe values higher than those of the continental crust, and spring flood δ 56Fe values of the dissolved phase lower than those of the continental crust.

Why the δ 56Fe composition varies with the summer and spring flood? The reason for such variations are not explained.

Hirst et al. (2017, conference contribution) show that the sources of Fe vary throughout the season. This change in sources results in varying Fe isotope compositions. They observed a shift from negative δ^{56} Fe to crustal values from winter to spring. During winter the system is ice-dominated, while during spring the system is fluvial dominated causing a change in δ^{56} Fe. Furthermore, Ingri et al. (accepted at GCA in July) showed that temporal variations of the

 δ^{56} Fe can be explained by changing sources of Fe within the soil throughout the year. We modified the text to clarify the discussion.

Hirst et al. (in prep.) show a seasonal dependence of the δ^{56} Fe composition of the dissolved fraction (colloidal and truly dissolved) in the Lena River, with summer flow δ^{56} Fe values higher than those of the continental crust, and spring flood δ^{56} Fe values of the dissolved phase lower than those of the continental crust. Therefore, the Lena River can be a source of CFe with both negative and positive isotope compositions. It has been shown that the Fe isotope composition is affected by seasonal variations of water flow pathes to the river (Hirst et al., 2017). Ingri et al. (accepted at GCA) showed that the Fe isotope composition is an indiactor for different Fe aggregates and for changing primary Fe sources throughout the season. Page 12

Line 5: flood discharge, which has much higher DOC concentrations (1170 µM), their samples would plot on a different mixingline (Allingetal., 2010).

Font is smaller than the previous lines. Please make uniform font.

Thank you, we changed the font size to fit the other text.

Line 7-9: The variations in the distributions of Fe between the different species in the iron-organic complexes are controlled by pH and OC concentrations (Neubauer et al., 2013; Sundman et al., 2013).

Authors have cited the importance of pH but have not given the pH data of the present study.

As mentioned above we added the pH and oxygen to table 1. The pH ranges from 7.5 to 7.9. In the studies of Neubauer et al (2013) and Sundman et al. (2013) the chemistry of Fe in much lower pH are discussed. We intended to use these references to describe the origin of the different Fe isotope compositions in the Lena River freshwater plume. We do not think that the change of 0.4 ph units in the freshwater plume has an influence of the Fe isotope composition. The change of pH might in fact increase the possibility to form solid-phase ferric iron oxides or hydroxides within the estuaries (e.g. Daneshvar 2015).

Line 12-13: Laboratory experiments of the oxidative precipitation of Fe(II) to Fe(III), which can occur in natural streams, show an overall fractionation factor of 0.9.

Authors have not given the reference for this fractionation factor. It is very important to see under which conditions the laboratory experiments were conducted?

We added the references to the sentence and rewrote it slightly. We did not want to give a fractionation factor for laboratory studies, but for measured natural values.

The fractionation factor of 0.9‰ was measured by Bullen et al. (2001) in spring water of the lower western flank of Mount Ruapehu, a composite volcano in Tongariro National Park, New Zealand. Fe isotope compositions of ferrihydrite and coexisting aqueous Fe samples along the first 600 m of down- stream reached a δ^{56} Fe of ferrihydrite, which is >0.9‰ greater than that of coexisting aqueous Fe.

Laboratory experiments **showed the existence** of oxidative precipitation of Fe(II) to Fe(III) **(e.g. Welche et al., 2003)**, which can occur in natural streams. **Bullen et al. (2001) measured an** overall fractionation factor of **about** 0.9 **in natural streams**.

Line 17-18: The Fe isotope variation along the plume and the composition of the surface sediment suggest that the chemically reactive ferrihydrite represent colloids and particles, with a negative $\delta 56$ Fe value, sedimenting close to the shoreline.

Which phase of the Fe isotope variation along the plume?

We were referring to the negative δ^{56} Fe values of the colloidal and particulate phase. We added this information to the sentence.

The Fe isotope variation values of CFe and PFe along the plume and the composition of the surface sediment suggest that the chemically reactive ferrihydrite represent colloids and particles, with a negative δ^{56} Fe value, sedimenting close to the shoreline.

Line 17-18: The surface sediments in the shelf areas along the Laptev Sea have δ56Fe values of - 0.2‰ (Figure 6). This value results from the removal of particulate and colloidal Fe(II, III)oxyhydroxides from the water column and burial in the sediment.

Does the sedimentary resuspension and diagenesis do not affect the iron isotope composition of the sediment?

Yes, we think that both processes are possible. The wave base can reach the sediment, but we do not have data on resuspension. We believe that the uppermost sediment is newly sedimented in the form of colloidal and particulate ferrihydrites, with negative δ^{56} Fe values. The data across the ESAS show that the Fe isotope composition of the uppermost sediment is the same.

The resuspension of sediment would favor a non-reductive dissolution of sediment to the seawater (Radic et al., 2011). The word dissolution expresses a flux from the particulate to the dissolved phase (<0.4µm).

The slightly negative δ^{56} Fe of the colloidal and particulate fraction (0.0 to -0.4‰) indicates that iron is not produced by dissimilatory iron reduction and redox cycling (characterized by a very negative δ^{56} Fe; -3.3 to -1.7‰), (Homoky et al. 2009; Severmann et al. 2006; 2010). Furthermore, non-reductive dissolution of sediment particles to the seawater would result in positive δ^{56} Fe of the DFe in the water column (Radic et al., 2011).

We added some lines to the discussion to rule out resuspension and non-reductive dissolution responsible for the Fe isotope composition of the uppermost sediment.

Other processes, as resuspension of sediment and non-reductive dissolution of sediment to the seawater (Radic et al., 2011) would lead to a much more negative (-3.3‰ to -1.7‰) Fe isotope composition of the sediment (Homoky et al., 2009; Severmann et al., 2006; 2010). Therefore, the δ^{56} Fe of the uppermost sediment reflecting the δ^{56} Fe of the sedimenting colloids and particles from the water column seems reasonable.

Page 13

Line 10-12: Climate warming is increasing discharge and accompanying OC and Fe from land to the ocean. Increasing the amount of colloidal and truly dissolved Fe, which is passing the estuarine mixing zone will lead to a higher Fe flux towards the Arctic Ocean.

It is very difficult to say that higher Fe flux towards the Arctic Ocean with this small data set.

We deleted this sentence, as it is no conclusion we can justify with our limited data set.

Page 22

Figure 1 Legend: Please give the full abbreviation of ESAS.

Changed accordingly.

Figure 1: Sampling stations in the Arctic Ocean. Black dots mark the stations in the detailed **East Siberian Arctic Shelf** <u>ESAS</u> map. Along the Lena River-Laptev Sea transect membrane filtration and/or ultrafiltration was carried out. The sampling stations of this study follow the Lena River freshwater plume. The green numbers display δ 56Fe values, measured in the uppermost sediment.

Page 23

Figure 2 Legend: Lena River transect is same as in the Figure 1? If yes please use uniform name. Changed accordingly.

Figure 2: The salinity gradient along the Lena River-Laptev Sea transect. Salinity is based on the Practical Salinity Scale PSS-78. The freshwater builds an almost 10 m thick surface layer in the Laptev Sea, and the plume itself extends over an area of about 50 times 600 km. The plume is divided into an inner and outer plume between station YS-8 and YS-11 by a sharp increase of salinity.

Page 24

Figure 3 Legend: Please use the same terminology for the Lena River Laptev Sea transect. I do not see the outermost station YS-128 in the graph.

Unfortunately, there are no DOC and POC data available for station YS-128.

Changed accordingly. Furthermore, we corrected 20 % towards 18% to be more concise.

Figure 3: Dissolved (< 0.70 μm) and particulate (> 0.70 μm) organic carbon concentrations along the Lena River-Laptev Sea transect freshwater plume in the Laptev Sea. Close to the Lena River mouth POC constitutes about **18%** of the TOC input, while at the outermost station it is only 2% of the TOC.

Page 25

Figure 4 Legend: Particulate, colloidal and truly dissolved Fe concentrations along the Lena River freshwater plume. Concentrations of pFe 5 and cFe decreased along the salinity gradient, while the concentrations of truly dissolved Fe is almost constant. Note the logarithmic scale and the sharp decrease of pFe between the inner and the outer plume.

I do not see the Particulate Fe in the graph. The labels say that tFe. I'm not sure whether author mean this as particulate Fe. I did not find the stations YS-5, 7, 9 & 10 in the graph. The manuscript is based on this graph and there are only 6 stations particulate and colloidal Fe data and only four truly dissolved Fe data out of 10 stations. The units used in this figure are in nano-moles. But in the text, authors have discussed that data in micro-moles. I also find the difference in citing the exact values.

This is right we do not show PFe in the graph, indeed we show TFe. Particle Fe is indicated as the difference between TFe and CFe. We changed the figure caption accordingly. We changed the unit in the figure towards µM and we added data for the truly dissolved Fe in the Lena River (Hirst et al., 2017) as reference for the river endmember.

Unfortunately, we do not have PFe, CFe, and DFe concentration for the stations YS-5, YS7, YS-9, and YS-10.

Figure 4: **Total**, colloidal and truly dissolved Fe concentrations along the Lena River freshwater plume. Concentrations of **P**Fe and **C**Fe decreased along the salinity gradient, while the concentrations of **D**Fe is almost constant. Note the logarithmic scale and the sharp decrease of **P**Fe between the inner and the outer plume. **The reference for the Lena River is an average of all analyzed samples (PFe n=3; CFe and DFe n=5) by Hirst et al. 2017.**

Page 26

Figure 5 Legend: Please give unit for Salinity

We changed figure 5 slightly, by adding a legend (please see below). We added information for the salinity to the figure caption.



Figure 5: The colloidal and particulate Fe concentrations plotted versus salinity. **Salinity is based on the Practical Salinity Scale PSS-78.** Note the y-axis break due to the high range of PFe in the inner plume. The linear correlation between PFe and salinity is based on the data points below $1\mu M$ PFe. In the low salinity environment, the PFe is much higher compared to the CFe, whereas at salinities above 5 the differences are smaller.

Page 27

Figure 6 Legend: What is ESAS? I do not see truly dissolved Fe isotope data in this figure. But in the methodology it was mentioned.

We added the information to the figure caption. There are no Fe isotope data for the truly dissolved phase. As mentioned earlier, the DFe Fe isotope values were not measured due to the low Fe concentrations in that phase. We changed the description of what we did in the introduction. Please see below.

Figure 6: Iron isotope values along the Lena River freshwater plume and the uppermost sediment of the **East Siberian Arctic Shelf** (ESAS). The error bars represent $\pm 2 \sigma$, in some cases the symbol is larger than the error. The δ^{56} Fe values of PFe are negative at all stations, values close to zero close to the coast and more negative towards the open sea. The δ^{56} Fe values of the CFe are negative in the inner plume and positive in the outer plume. The δ^{56} Fe of the sediment samples were around -0.2 ‰, displaying the overall composition of the entire ESAS area.

From the introduction:

This study presents Fe concentrations and Fe isotope compositions in the particulate **and** colloidal phase along the Lena River freshwater plume in the Laptev Sea, **as well as Fe concentrations in the truly dissolved phase.**

References added to the manuscript and the response to the reviewer:

Andersson, Leif; Jutterstrøm, Sara (2008): Seawater carbonate chemistry and nutrients measured on water bottle samples during the International Siberian Shelf Study 2008 (ISSS-08) in the Laptev, East Siberian and Chukchi Seas. Department of Chemistry, University of Gothenburg, PANGAEA, https://doi.org/10.1594/PANGAEA.715045

Asmala, E., Bowers, D., Autio, R., Kaartokallio, H., Thomas, D.N., 2014. Qualitative changes of riverine dissolved organic matter at low salinities due to flocculation. J. Geophy. Res.: Biogeosci. 119,1919–1933, doi:10.1002/2014JG002722.

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