

Reviewer 2:

Here my reply (in blue) to your questions. Changes are added in the manuscript

**1.)** Page 13743: Line 12- 17: Please explain in more detail the fjord hydrography. How low was the Salinity in the surface layer and below! I presume that the pycnocline was right between the brackish and deep waters at ~5 m. Would be nice to have a plot that shows T,P,S, O<sub>2</sub> (very important for the redox sensitive trace metal Fe) in the fjord. What was the average depth of the fjord? This all should be mentioned when planning to extrapolate the results to fjord waters.

**ANS:** Data for salinity in the LSL were added together with some other descriptive features of the fjord. References to extended literature on the fjord hydrography have been added.

**2.)** Page 13743: Line 19-25: It is really hard to understand what was exactly done, please rewrite. "Seawater was pumped to a 33 L container via a peristaltic pump deployed both on a peer. The collected water was then distributed equally to 1m<sup>3</sup> mesocosms."

**ANS:** Seawater was pumped to a mixing container by a peristaltic pump and then distributed simultaneously to all 1m<sup>3</sup> mesocosms tanks by gravity-fed pipe flow.

I have several questions here:

**3.)** When you work on trace metals, such as Fe, was the hose and the 33L container acid washed.

**ANS:** Yes. Changes added to the manuscript.

**4.)** Were the 1m<sup>3</sup> tanks cleaned too?

**ANS:** No. Tanks were washed the ambient seawater thoroughly and preconditioned by same seawater. Changes added to the manuscript.

Were the mesocosms closed after filling, or were they left open.

**ANS:** Containers were closed all time except during sampling. Changes added to the manuscript.

**5.)** When open how much additional fresh water was introduced by rain?

**ANS:** Containers remain closed all time except during sampling, no additional freshwater was introduced.

**6.)** Why was the water not prescreened?

**ANS:** Since this study was part of a major experiment which made focused on different oceanographic aspects (ecology, biology and chemistry), it was important to minimize perturbations and keep community composition as natural as possible.

**7.)** how much Fe was in the fjord (without pumping, ect.), that could at least used to quantify the amount of added by the sampling procedure!

**ANS:** Initial concentrations for all treatments corresponded to water collected by the peristaltic pump directly at 2 and 10m for the brackish and marine system respectively and before being pumped into the containers. Changes added to the manuscript.

The concentrations measured we believe represent accurate values for the fjord (compared to previous measurements carried out in the fjord), given that either by Go-Flo bottle or peristaltic pump there is similar chance of contamination. Since the focus of our study was the enclosed environment, our main objective was to track the changes undergoing in the different fractions of Fe, rather to compare or extrapolate directly to the biogeochemistry of the fjord.

**8.)** Page 13744: Line 1-15: Nutrient solutions were cleaned, and received from where, also the acids and the ammonia/acetic acid?

**ANS:** Yes, all macronutrients solutions were prepared from reagent grade and treated by adding Chelex-100 resin to remove Fe. Changes added to the manuscript.

**9.)** Page 13744: Line 14: “nutrient concentration” there is no concentration in Table 1; just some strange rates! Please correct the table, see comments for Table 1.

**ANS:** The nutrient table display the rate applied per day for the 22 days, to reach final target concentration. The table now is modified and final concentrations attained in the mesocosms are listed. Changes added to the manuscript.

**10.)** Page 13744: Line 17: How the water samples were collected on the raft? Any standard seawater was run?

**ANS:** Collection of samples from the tanks was performed every third day, after proper mixing of tanks and before any other kind of samples (Table 1). A hose connected to peristaltic pump was introduced in each tank pumping 2-3 Lt into 5 L plastic containers, which were taken to laboratory for processing. All material used, except from tanks were acid washed in ultra-pure HNO<sub>3</sub>. (Double quartz distilled from reagent grade).

**11.)** What was the pH of the buffer solution, and the final seawater solution?

**ANS:** This section has been expanded. Changes added to the manuscript.

**12.)** Was the filtration performed before the addition of Chelex or afterwards. It is not very clear from the text. The seawater chelex matrix was transferred into a column with a frit?

**ANS:** This section has been expanded. Changes added to the manuscript.

**13.)** Replace “Milli-Q” by “deionized 18.2MΩ cm water”!

**ANS:** Changes added to the manuscript

**14.)** Am I correct that when you collect the TFeCh fraction also other particles (e.g., inorganic sediments introduced by filling the tanks) were collected during column separation?

**ANS:** This is a deep fjord and the sampling spot was ca 30 – 45 m, so we argue for a low re-suspension possibility. Even if there is some inorganic PFe component, the difference in TPF<sub>e</sub> from initial conditions and when we have highest productivity is very significant.

**15.)** Grade of used acid?

**ANS:** double quartz distilled, Changes added to the manuscript

**16.)** Page 13745: Line 20ff: UP: Ultra-pure?

**ANS:** Yes, Changes added to the manuscript

**17.)** How was the gel/resin and the HNO<sub>3</sub> separated from each other?

**ANS:** using plastic tweezers. Changes added to the manuscript

**18.)** Did you transfer all added HNO<sub>3</sub>? When 4mL 3M HNO and 4mL MQ are added together you end up with 8mL 1.5M HnO<sub>3</sub>!

**ANS:** 1 mL 3M UP HNO<sub>3</sub> was added (typing error), then 4 ml deionized water was added to make a final ml 0.6 M HNO<sub>3</sub>. Changes added to the manuscript

**19.)** Page 13746: Line 12: Washed with deionized water!

**ANS:** Changes added to the manuscript

**20.)** Page 13746: Line 22ff: Any recovery test for the Chelex performed? Results

**ANS:** Additional information on procedural blanks together with results on recovery tests of certified reference materials are now included.

**21.)** Page 13747: Line 3ff: I don't see any errors for TFe and DFe. Please include in Fig. 2. I was reading earlier that the experiment was conducted for 22 days, but just 8 days are shown, why?

**ANS:** There was only sample measurement when it comes to Chelex fractions. We privileged the measure over a larger number of points on time (gradient fashion) rather than replicates. The samples were collected every 3<sup>rd</sup> day, hence Sday corresponds to the sampling day and not the consecutive day. Table with the type of samples and day of collection is now added.

**22.)** However, I'm not sure what the labile TFeCh is telling you! It is just a leach of the particulate fraction, or when you collect the Chelex with the particles suspended then you do a leach with the HNO<sub>3</sub> later in the lab, right. So, I'm really concerned what kind of total fraction we are talking about here!

**ANS:** Here we are using 2 or 3 days equilibrium process, while we mix Chelex-100 (100-300  $\mu$ m mesh size) with non-filtrated seawater by gently shaking this mixture. After these process, easily releasable Fe complexed with Chelex-100 when mixture reach an equilibrium and easily leachable Fe from mostly biological materials may give us idea about the amount of iron has been removed by biological activities, mainly by phytoplankton, in the studied system (in our case in the mesocosm containers).

**23.)** Similar problems I have with the labile DFe fraction. Some organically complexed Fe won't be scavenged by the Chelex at pH8. Especially the strong ligands will compete. Any numbers of how much was in the real dissolved Fe fraction.

**ANS:** That's probably, but chelex and DGT have been used to estimate the capture the most easily available fractions of Fe.

**24.)** Page 13747: Line 21ff: With respect to the wide scattering of FeDGT I don't see any trend! With respect to the difficulty of the procedure DGT, I personally would also take contaminations into account? Or why should be the DGT Fe concentration higher than the labile dissolved?

**ANS:** DGT data, expected to resemble the truly soluble fraction since pore size of diffusion gel is 2 – 20 nm, was intended to be correlated with the dissolved chelex labile fraction, as values of the former keep within the range of the latter. Unfortunately to describe the temporal variability we could only describe a gross pattern, perhaps point the differences between initial and final concentrations. We rather would be more able to compare between treatments at a given sampling time.

Regarding contamination, given that DGT is based on the diffusion coefficient, it always possible that larger Fe organic complexes formed may have slower diffusion coefficient, hence leading to overestimation of concentration.

**25.)** I also do not see the need for that technique since fluxes are important for sediments for instance, but in the water column. Probably of interest at the pycnocline, but other than this?

**ANS:** DGT technique is not only to determine the flux, this technique can also be used to estimate the bulk concentration of iron and other trace metals by using the diffusion coefficient. Although the technique is not settled yet, but it has some advantage to determined labile Fe (all Fe species smaller than pore size of diffusion gel (in the range 2–20 nm ).

**26.)** Page 13747: Line 2ff: TFe and DFe values in Figure 3b and 3c represent the final concentration or the average over all days?

**ANS:** Values represent the average for all days.

**27.)** Page 13748: Line 12ff: The particulate samples were collected when (after 22 days?)? First you talk about average concentration which is fine deciding between brackish and seawater, but from my point it is also of interest how the particulate Fe fraction evolved?

**ANS:** The particulate samples were collected as can be seen in figures 4 and 6. Both legend say "along time ". Table 1 added display which and when parameters were sampled.

**28.)** By combining PFe and Chl/POC you show that the intracellular ratio changes from the beginning towards the end of the experiment. However, I have no idea what the PFe:Chl to Chl diagram is telling us and which idea it supports!

**ANS:** In plotting PFe standardized by either Chl or POC (fig. 4) we can observe the trend in time for each treatment as well as the difference against the control and natural system. In plotting already standardized PFe against Chl or POC (fig. 6), we can see specifically the exponential decrease relation with each parameter at increasing NH<sub>4</sub> concentration.

**29.)** Have you thought about an terrestrial particulate Fe fraction (sediment resuspension etc. that were sucked into the mesocosms when filling)

**ANS:** Same as 14. We argue for a low re-suspension possibility.

**30.)** Page 13748: Line 25ff: I do not understand that sentence. When Fe concentrations of all size fractions are added together then we end up with PFe, or?

**ANS:** PFeSF renders information of the Fe distribution within the different fractions of the natural plankton community. The objective was to see the evolution in time and variation in the distribution of PFe. We could observe that change in the 20-140 and 2-20µm for the different systems. However the information on the other fractions is not discussed in depth within the manuscript. Paragraph has been modified and changes added in the manuscript

**31.)** I do not also understand what we learn from the mean total PFeSF what we have not learned already from PFe!

**ANS:** Same as 28

**32.)** Page 13749: Line 7: Seawater is decreasing!

**ANS:** Paragraph has been modified and changes added in the manuscript

**33.)** Page 13749: Line 9-18: It is really hard to understand that paragraph, since the following section is dealing especially with the size fraction. Better to put all the size fraction results in one paragraph! Page 13749:

**ANS:** Paragraph has been modified and changes added in the manuscript

**34.)** Line 23: What happened with Fig. 7. Page 13750: Line 2ff: Last sentence should come earlier in the paragraph. It would increase the chance to understand the ratio better.

**ANS:** Paragraph has been modified and changes added in the manuscript.

**35.)** Page 13750: Line 12ff: Rewrite "A long time, : : ." It sounds like a fairy tail.

**ANS:** Typing error, changes added in the manuscript

## Tables

**36.)** Table 1: As I understood correctly you added every third day macro nutrients to the mesocosms. Here you show a rate per day. Be more consistent and state that you added nutrients every third day. For that reason numbers should be stated in target concentrations. Column mesocosm, should read "1 and 6", "2 and 7", etc. Otherwise, people might think from mesocosm 1 to 6.

**ANS:** The rate displayed was to show the equivalent incremental concentration of nutrient along time. Now final concentrations are displayed. Changes added in the manuscript

**37.)** Table 2: Fe Concentrations?

**ANS:** Additional information on procedural blanks together with results on recovery tests of certified reference materials are now included