Review of manuscript egusphere-2022-254,

In this manuscript, Debyser and co-workers present combined isotopic data of dissolved nitrate (δ^{15}N-NO$_3$ and δ^{18}O-NO$_3$) and silicon (δ^{30}Si(OH)$_4$) in seawaters from the Fram Strait. By comparing the dataset between the inflowing Atlantic waters and the outflowing Pacific surface waters, the authors evaluate the modification of Si and N within the Arctic ocean and highlight the importance of denitrification on Arctic shelves as well as its impact on DSi utilization. The authors also try quantify the contribution of terrestrial input to the total DSi exported through Fram Strait.

This manuscript is well-constructed and well-written. This work improves the understanding of the modification of major nutrients (Si&N) within the Arctic Ocean, and has key implications for the future change of nutrient supply from the Arctic ocean to the Atlantic Ocean. As such, this work is a useful addition to the field that would be suitable for publication in BG. But there are a few points where I think the authors need to be more careful in their interpretation, as I outlined below. I recommend this manuscript for moderate to major revisions before the final publication.

Major comments:

1. The justification of measuring δ^{29}Si instead of δ^{30}Si

Method part, L145-L148: I understand that it is difficult to measure δ^{30}Si of samples with low DSi concentrations. However, it is not common nowadays to report δ^{30}Si values based on the measurements of δ^{29}Si. This practice will be more justified if the authors could provide more details (can be in the supplementary) on why it was not possible to measure δ^{30}Si directly. For example, what kind of efforts the authors have already put into trying to resolve the matrix effects and interferences? Normally anion doping (sulfate, nitrate, etc…) or pre-removal of organic matter could help to diminish the matrix effect (Closset et al., 2016; Hughes et al., 2011). Isobaric interferences of e.g., nitrogen (^{14}N$_2$) and nitric oxide (^{14}N^{16}O) can be avoided by medium resolution mode focusing on the left side of the peak shoulder (Liguori et al., 2020). At least it would be nice to see that the authors have already tried all these approaches before giving up on directly measuring δ^{30}Si.

2. The interpretation of the isotopic fractionation model in section 4.1.1

   1) The initial level of the nutrients before utilization should be the subsurface water from the time period with strong mixing prior to the sampling, i.e. winter season. Initial condition for both spring and summer seasons should be the same (i.e. upwelled winter subsurface water). The authors did not give too much details of their choice on the initial condition, but it seems like they simply chose the subsurface waters in each sampling event as the initial condition. On the other hand, considering the horizontal transport of the water mass (i.e. PSW and AW), the initial condition might be found horizontally.

Therefore, I wonder whether the authors' choice of the initial condition of the model is correct and I recommend the authors to provide more information on this aspect.

2) It makes more sense to sperate PSW and AW dataset into different models because their initial conditions are different. Spring and summer dataset from the same water mass should be combined into the same model, because they belong to the same fractionation system, i.e. the nutrient kept being utilized in summer after the utilization in spring.

3) In figure 6, δ^{30}Si data do not fit any of the models during any of the sampling events, so the discussion between L257 to L260 is not valid. This might point to the incorrect choice of initial condition that the authors applied to calculate the nutrient utilization.

3. The estimation of the terrestrial Si sources at Fram Strait in section 4.2.6

It is not justified to estimate the PSW Si:N (that free of terrestrial influence) based on the assumption of "further modification of marine δ^{30}Si(OH)$_4$ and DSi:N through the Bering Strait is linear with North Pacific trends". Especially after the authors already concluded that nitrate was largely removed via denitrification in the Arctic, which will clearly modify the Si:N within the Arctic. It is thus not convincing to conclude DSi:N>0.78 in the PSW must originate from terrestrial riverine sources. Therefore, the estimation in the paragraph L434-441 is not valid. Additionally, as the authors illustrated the mixing scheme in Figure 10, it is quite obvious that Si:N of the PSW is located within the error of the mixing line between the AW and E.S. Shelf. This indicate that the increase of Si:N in the PSW is more a result of the shelf CPND. The only prominent outliers of PSW dataset beyond the mixing lines are the three data points with lower Si:N/heavier δ^{15}N.

Minor comments:

L130-139: The two-step co-precipitation has been widely used previously, so there is no need to elaborate its necessity here. It can be directly cited from previous work, for example (Reynolds et al., 2006), (Grasse et al., 2013), (Liguori et al., 2020) etc…

L137: What does "regrouped" mean?

L152: Please note whether the uncertainties are 1SD or 2SD.

L181: Please add "in the upper 400m"

L185: Please add (Figure 3c)

L189: Please add (Figure 3b)

L193-195: Please tone down the argument here, as (5.42 ± 0.70 µM) and (6.65 ± 1.67 µM) are within error identical.

L196: Please add (Figure 3d)

Section 3.3: The description in this section is bouncing back and forth between figure 3 and figure 4&5, i.e. between whole depth profile and surface data. It will be clearer if the authors can give clearer information on which figure/panel the sentence refers to, and describe the distribution from the surface to the deep for both Si and N/O isotopes.

L234-235: "Nutrient utilization" is normally defined as the fraction of nutrient that has been utilized. The way that the authors define it here is against the common cognition.

L291-293: The whole sentence "settling particulate nitrogen… sediment interface." reads a bit repetitive, please rephrase.

Section 4.1.2 The authors try to discuss the modification of nitrate and DSi in the Arctic ocean in this section, so maybe the authors should exclude the dataset within the mixed layer, which are largely impacted by the local biological uptake. From Figure 7, only panel (c) excludes samples from within the mid-layer depth.

L355: It is not easy to understand "merging towards signatures resembling riverine endmembers" here, please give the values of the riverine endmembers.

L367: TDP → TPD

L372: The larger variability in Si isotope signatures of PSW (R^2>0.3) at Fram strait might reflect the combination of mixing and local biological uptake.

L376: valuated → evaluated

L397: ~~while~~

L409: ply?

Line 429-430: Please show the linear relationship between δ^{30}Si and DSi:N in North Pacific waters.

L489: I would not describe a 0.11‰ increase of δ^{30}Si(OH)$_4$ as "significantly" enriched, as the long-term reproducibility of the ALOHA$_{1000m}$ measurement is 0.08‰, the two values with a difference of 0.11‰ even overlap within error.

Figures:

Figure 2: the scale of temperature (left panel) is missing

Figure 6: I wonder whether it is necessary to add the fractionation lines of the products, as there is no data from the biogenic phase and there's no discussion of the fractionation of the products. Removing these unnecessary lines can make the plots cleaner.

Figure 10: Please correct the sentence in the caption: "~~Dotted lines~~ <u>Solid line</u> shows the regression (conservative mixing line) between AW and shelf ~~endembers~~ <u>endmembers</u>, dotted

lines are for one standard deviation." Also, if the line is conservative mixing line, then it is not regression line. They are not the same.

References:

Closset, I., Cardinal, D., Rembauville, M., Thil, F., & Blain, S. (2016). Unveiling the Si cycle using isotopes in an iron-fertilized zone of the Southern Ocean: From mixed-layer supply to export. *Biogeosciences*, *13*(21), 6049–6066. https://doi.org/10.5194/bg-13-6049-2016

Grasse, P., Ehlert, C., & Frank, M. (2013). The influence of water mass mixing on the dissolved Si isotope composition in the Eastern Equatorial Pacific. *Earth and Planetary Science Letters*, *380*, 60–71. https://doi.org/10.1016/j.epsl.2013.07.033

Hughes, H. J., Delvigne, C., Korntheuer, M., De Jong, J., André, L., & Cardinal, D. (2011). Controlling the mass bias introduced by anionic and organic matrices in silicon isotopic measurements by MC-ICP-MS. *Journal of Analytical Atomic Spectrometry*, *26*(9), 1892–1896. https://doi.org/10.1039/c1ja10110b

Liguori, B. T. P., Ehlert, C., & Pahnke, K. (2020). The Influence of Water Mass Mixing and Particle Dissolution on the Silicon Cycle in the Central Arctic Ocean. *Frontiers in Marine Science*, *7*(April), 1–16. https://doi.org/10.3389/fmars.2020.00202

Reynolds, B. C., Frank, M., & Halliday, A. N. (2006). Silicon isotope fractionation during nutrient utilization in the North Pacific. *Earth and Planetary Science Letters*, *244*(1–2), 431–443. https://doi.org/10.1016/j.epsl.2006.02.002