

This manuscript presents new nutrient and nutrient isotope data from across the Fram Strait, with the aim of deconvolving the controls on nitrogen and silicon cycling, and nitrate and dissolved silicon (DSi) biological uptake, in the Arctic. Given the rapid environmental (and biogeochemical) change in the Arctic, and the importance of this gateway for supplying nutrients to the North Atlantic, this paper is timely, the findings have key implications for our understanding of future change in the Arctic, and are of importance to the community. The manuscript is well-written and clearly presented, and an enjoyable read. As such, the manuscript is suitable for publication in an EGU journal. I just have a few suggestions for moderate revisions and broader discussions. I would also suggest the inclusion of some additional recent papers. These papers are only recently published (some only as pre-prints) so it's understandable that these could have been missed before submission, but I think it would be beneficial to include them now.

Most of my comments relate to the interpretation of the silicon isotope data, which have implications for the conclusions of the manuscript.

1) One of the key challenges in this paper is in the interpretation of Si isotope systematics at very low DSi concentrations ([DSi]). Specifically, I am concerned that without the very depleted DSi waters included in the isotopic analysis, it's challenging to determine the isotopic behaviour of DSi in the mixed layer (relevant for the discussion on line 269 onwards regarding the decoupling of N and Si[†]). From Figure 4, it's clear that there are several profiles missing between -5 and 5 degrees longitude. Furthermore, several very low [DSi] surface samples do not have corresponding $\delta^{30}\text{Si}(\text{OH})_4$ measurements (which is entirely understandable as these are very challenging measurements to make!). It appears from Figure 4 that there is only one datapoint from within the mixed layer in the transect (although it's not clear how this corresponds to the data shown in Figure 6 – what depths are these data from?). So, interpreting the relationship between uptake/utilisation and isotopic composition (i.e., closed vs. open systematics) in the mixed layer is going to be challenging. As such, I would suggest perhaps toning down this aspect of the interpretation, relating to closed vs. open nature of DSi uptake. I was hoping to test out some of these plots myself, but I found that the data link was for the incorrect year of sampling, and the $\delta^{30}\text{Si}$ data were not available.

[[†]Note that if the discussion of N and Si decoupling is included, please note that this could be due to other related processes in addition to shifting limitation e.g., algal population structure changes throughout the season. If there are any data available on algal population structure, they would be useful to include or refer to here.]

Related to this question, I'm also intrigued as to why the observational data consistently fall 'below' the $\delta^{30}\text{Si}(\text{OH})_4$ model curve in Figure 6 (c,d). It seems that, unlike for the N isotope model (Fig. 6 a,b), the model predicts heavier isotopic compositions in the seawater for DSi, regardless of assumptions about whether the system is open or closed. Why could this be? Is this related to an incorrect assumption about the endmember compositions, or related to recycling/dissolution?

2) I don't think it's necessarily clear from the evidence presented that "DSi uptake is being regulated by nitrate availability" across the Arctic (line 278, and very relevant for discussion on line 319 onwards). This could be the case in some regions of the Arctic, but I

think DSi uptake and biogenic silica formation will ultimately, on a Pan Arctic scale, be regulated by multiple factors. I think a lot of the nuance here lies in the different interpretations of limitation at different spatial and temporal scales, and different biological/ecological contexts. For example, the evidence noted by the authors for DSi limitation in summer seasons (especially in AW dominated surface waters) based on silicon-32 uptake experiments (e.g. Krause et al. papers) in addition to the relationship between [DSi] and nitrate concentrations. Giesbrecht et al., 2021, used similar experiments to show that Si uptake by diatoms was limited by DSi even high [DSi] availability in Pacific Arctic Region, likely because the diatoms were acclimated to high 'background' [DSi] conditions, and that there was also strong interaction between DSi and light limitation (also relevant for the discussion on line 466). In other words, Si uptake and biogenic silica formation can become limited before exhaustion of DSi in the water column, and can be promoted through addition of DSi even at relatively high background levels. Again, I would suggest that the authors rephrase these aspects of the paper to account for these complexities.

NB: Giesbrecht et al., 2022, also present $\delta^{30}\text{Si}(\text{OH})_4$ data from the Pacific Arctic Region that needs to be included in this manuscript (and there is some very relevant content for the discussion on sea-ice on line 388 onwards).

3) There could be more discussion surrounding the delivery of Si from glacial meltwater sources. As mentioned on line 57, although there is a lot of both DSi and amorphous silica (ASi) in glacial meltwaters, we don't yet have a good handle on fjord and estuary processes (perhaps the authors could cite Meire et al., 2016, and Hopwood et al., 2020 here for discussion on this subject). I agree that the majority of studies show that surface waters within glacial fjords can be depleted to very low [DSi], and it's highly likely that much of the ASi will be trapped in fjord sediments (line 381 onwards). However, given the very high [ASi] in glacial meltwaters (an order of magnitude higher than [DSi], Hawkings et al., 2017) even a small percentage of ASi escaping the fjord system will contribute a significant amount towards the total silicon supply from glaciers. For example, a study of benthic Si cycling off SW Greenland revealed evidence for slow remineralization of isotopically light Si (a good proportion of which is likely to be glacial ASi) in coastal shelf sediments, driving a significant diffusive flux of DSi into overlying shelf waters (Ng et al., 2020). This ASi contribution would shift the glacial contribution in Figure 8 towards high $\ln(\text{Si})$, with only a minor shift in $\delta^{30}\text{Si}$, closer towards the riverine endmember (but likely with an isotopically lighter composition). As such, this would make the glacial endmember more challenging to exclude. The questions of how much amorphous silica is present in Arctic river particulates, and how it impacts the overall isotopic composition of runoff, sadly remain unanswered.

4) There is further information that could be included regarding the benthic supply of DSi to Arctic (shelf) waters (relevant to discussion on e.g., line 307, 312, 320, 330). Ward et al., 2022, (GCA and Biogeosciences discussion) present new porewater/solid phase silicon isotope data from the Arctic. Given that these data were from samples that were collected in the Barents Sea, and so more relevant geographically than the study cited on line 211, I would suggest that this study is referenced here in addition to Ehlert et al., 2016. It might be possible (c. line 214), with the additional data and model results from Ward et al., to attempt to calculate possible fluxes of benthic DSi and their contribution to the overall water column signature (see their BGC discussion paper, Figure 6).

Further, these authors show that lithogenic silica (LSi) plays an important role in supplying DSi to Arctic shelf waters. Given that this LSi is isotopically light relative to river waters, this additional source exacerbates the Arctic isotope balance 'problem' (from Brzezinski et al., 2021). However, Ward et al. also discuss the possibility of abiotic *uptake* of Si into sediments (adsorption and – critically – authigenic phase formation) during early diagenesis that could form a sink of isotopically light Si. The longer-term BSi burial found by these authors is actually quite low due to strong benthic-pelagic coupling on a seasonal timescale.

Minor points:

Please add limits of detection and precision/accuracy data for the dissolved inorganic nutrient measurements section in the Methods (line 120-121). This is important when discussing such low nutrient concentrations, and using these low nutrient concentrations as an indication of ecosystem-scale nutrient limitation.

It's a shame that the $\delta^{30}\text{Si}$ values were not able to be measured, as using the three isotopes is an extra check on data quality (through the determination of mass dependency). However, the $\delta^{29}\text{Si}$ values of reference standards do seem to reproduce well and agree with published data. Please rephrase the sentence on line 146: whilst I agree it's a useful exercise for comparison with other datasets, I don't think that the conversion of $\delta^{29}\text{Si}$ to $\delta^{30}\text{Si}$ (by simply assuming mass dependency) results in better accuracy, *per se*.

Are there any seawater $\delta^{18}\text{O}$ data that could be used together with salinity values to determine the contribution of meteoric and sea-ice sourced freshwater to the samples? This might help to deconvolve the runoff vs. sea ice contributions to the system.

It would be useful to add + or – signs before silicon isotope values for clarity.

Please be consistent with the use of high/low and heavy/light when referring to delta values or 'isotopic compositions/isotopic values/isotopically' (etc.) respectively.

I'm not sure that the authors need to include the dashed lines (showing the products of uptake e.g., biogenic silica) in Figure 6, given that solid phase data are not included in the paper. These could be removed for clarity.

Note that Lena is spelled incorrectly in the caption to Figure 8.

References:

Brzezinski, M. A., Closset, I., Jones, J. L., De Souza, G. F., & Maden, C. (2021). New constraints on the physical and biological controls on the silicon isotopic composition of the Arctic Ocean. *Frontiers in Marine Science*, 931.

Giesbrecht, K. E., & Varela, D. E. (2021). Summertime biogenic silica production and silicon limitation in the Pacific Arctic Region from 2006 to 2016. *Global Biogeochemical Cycles*, 35(1), e2020GB006629.

Giesbrecht, K. E., Varela, D. E., de Souza, G. F., & Maden, C. (2022). Natural variations in dissolved silicon isotopes across the Arctic Ocean from the Pacific to the Atlantic. *Global Biogeochemical Cycles*, 36(5), e2021GB007107.

Hawkings, J. R., Wadham, J. L., Benning, L. G., Hendry, K. R., Tranter, M., Tedstone, A., ... & Raiswell, R. (2017). Ice sheets as a missing source of silica to the polar oceans. *Nature communications*, 8(1), 1-10.

Hopwood, M. J., Carroll, D., Dunse, T., Hodson, A., Holding, J. M., Iriarte, J. L., ... & Meire, L. (2020). How does glacier discharge affect marine biogeochemistry and primary production in the Arctic?. *The Cryosphere*, 14(4), 1347-1383.

Meire, L., Meire, P., Struyf, E., Krawczyk, D. W., Arendt, K. E., Yde, J. C., ... & Meysman, F. J. R. (2016). High export of dissolved silica from the Greenland Ice Sheet. *Geophysical Research Letters*, 43(17), 9173-9182.

Ng, H. C., Cassarino, L., Pickering, R. A., Woodward, E. M. S., Hammond, S. J., & Hendry, K. R. (2020). Sediment efflux of silicon on the Greenland margin and implications for the marine silicon cycle. *Earth and Planetary Science Letters*, 529, 115877.

Ward, J. P., Hendry, K. R., Arndt, S., Faust, J. C., Freitas, F. S., Henley, S. F., ... & Tessin, A. C. (2022). Stable silicon isotopes uncover a mineralogical control on the benthic silicon cycle in the Arctic Barents Sea. *Geochimica et Cosmochimica Acta*.

Ward, J. P., Hendry, K. R., Arndt, S., Faust, J. C., Freitas, F. S., Henley, S. F., ... & Airs, R. L. (2022). Benthic Silicon Cycling in the Arctic Barents Sea: a Reaction-Transport Model Study. *Biogeosciences Discussions*, 1-34.