

We thank Reviewer #1 for their time in reviewing our manuscript and their useful discussion concerning our findings. Here we include responses to all of the comments as follows: (1) Reviewer's comment (2) Author's comment (3) Suggested change to 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 d³⁰Si(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 d³⁰Si 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.]

(2) Samples shown in Figure 4 are for FS2018 cruise only, while the dataset in Figure 6 includes both FS2018 and JR17005. The majority of the d³⁰Si dataset from within the mixed layer is from JR17005. We have focused our measurements in the core of AW & PSW water masses (-5° < >5° longitude) as the physical circulation of central Fram Strait is strongly eddying and highly temporally variable, impeding straightforward interpretation of biogeochemical data. (3) The equivalent section of Figure 4 for JR17005 is now included in supplementary material S2. Samples from within the mixed layer are now highlighted in a different colour in Figure 6 for clarity. (2) As we have no measurements for either d¹⁵N & d³⁰Si from within the mixed layer for later summer PSW as nutrient concentrations were depleted to nearly 0 μM, (3) this aspect of the discussion has been toned down in the manuscript. The data link provided in the manuscript for biogeochemical data and d¹⁵N data of JR17005 have now been updated to the correct year.

(1) Related to this question, I'm also intrigued as to why the observational data consistently fall 'below' the d³⁰Si(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) Following suggestions from all reviewers, we have updated the parameters of our Rayleigh model in Figure 6 and the heavier prediction was related to incorrect assumptions

about endmember compositions. This is addressed in more details in the general comment to all reviewers.

(1) 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.

(2) We agree with Reviewer #1 that nutrient regimes and subsequent limitation in the Arctic Ocean are strongly varying, and would like to emphasise that the conclusions drawn in this paper relating to N availability are relating to the Eurasian Arctic nutrient regime only, not to Arctic biological systems with high nutrient concentrations like ones from Pacific origins. (3) Certain aspects of the paper have been rephrased to clearly state that N-limitation and subsequent modulation of DSi uptake is observed in the Eurasian Arctic although we recognise that this does not apply to different nutrient regimes of the Arctic Ocean where biogeochemical cycles are vastly different. These aspects of the paper have been rephrased to account for the difference in regimes.

(1) Giesbrecht et al., 2022, also present $d^{30}\text{Si}(\text{OH})_4$ 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) Watermass endmember data from Giesbrecht et al. (2022) are now included in Figure 8.

(1) 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 $d^{30}\text{Si}$, 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.

(2) Thank you for this comment. We agree with the reviewer's comment that much remains to be understood regarding estuarine processes of glacial geochemical cycling and export to the ocean, and that it would be useful to understand the role of ASi in Arctic river particulates. (3) References to Hopwood et al. (2020) & Meire et al. (2016) have been added to improve the discussion from line 57 onwards regarding the characterization of the export of DSi and ASi into the open ocean. (2) We recognize that findings from Ng et al. (2020) show significant fluxes of DSi into shelf waters, however, it is hard to make this comparison to the data presented in Figure 8 without direct measurements from isotopic signatures from the shelf waters themselves and any discussion resulting from this would be speculative. As such, their research is now acknowledged in this paragraph but we refrain from making any direct comparison to this study.

(1) 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.

(2) We thank the reviewer for bringing the two Ward manuscripts to our attention as we were not aware of their publishing at the time of writing our manuscript. They greatly add to our understanding of benthic efflux in the Arctic Ocean and (3) their measurements from the Barents Sea are now referred to on line 211 and included in the discussion from line 307 onward. (2) With regards to calculating fluxes of benthic DSi in this manuscript, the light $d^{30}\text{Si}$ referred to from line 214 onwards are primarily observed in the deep basin of Fram Strait. Benthic cycling is vastly different in the Arctic Ocean on the shallow shelves compared to the deep basins (März et al., 2015), and we feel that as the model results from Ward et al. (2022) characterize benthic cycling on the shallow Barents sea shelf, these should not be directly applied to our measurements as they can be unrepresentative of the

biogeochemical cycling of the deep Fram Strait. As our measurements are also sparse in resolution above the sediment interface and not easily distinguished from advective signals, we maintain our opinion that our dataset precludes accurate quantification of recycling processes in the deep Fram Strait and we therefore refrain from doing so in this manuscript.

(1) 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. (2) Detection limit for nutrient analysis was $0.1\mu\text{M}$ and $0.03\mu\text{M}$ for silicate and nitrate respectively with accuracy with respect to CRMS of 2.75% and 0.91% for JR17005. For FS2018, analytical precision is of 2% and the detection limit was of $0.4\mu\text{M}$ for nitrate and $0.1\mu\text{M}$ for silicate. (3) Detection limit and precision data is included in method section.

(1) 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*.

(2) Following from another comment from Reviewer #3, we realise the writing of the method section read as if the $\delta^{30}\text{Si}$ isotope of samples was not measured during analysis and thank the reviewers for highlighting this. To clarify, all three isotopes were measured during analysis and a strong relationship was found between measured $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ ($\delta^{29}\text{Si} = 0.5131 * \delta^{30}\text{Si}$, $R^2 = 0.99$), but larger variability and standard deviations were measured on the $\delta^{30}\text{Si}$, hence the conversion from $\delta^{29}\text{Si}$ to $\delta^{30}\text{Si}$. (3) The method description has now been amended for clarity on our method of analysis and the $\delta^{29}\text{Si}$ vs $\delta^{30}\text{Si}$ relationship of our measurements is now included in the supplementary material S1. (2) We agree with the comment on the conversion of both isotopes and (3) the sentence was edited to now read " $\delta^{29}\text{Si}$ were converted to $\delta^{30}\text{Si}$ to improve reliability and global comparability of datasets" instead.

(1) 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. (2) We recognise that seawater $\delta^{18}\text{O}$ data could bring important hydrographical information, however, for the FS2018 cruise data was not available at the time of writing this manuscript, but we refer to the long term dataset from Dodd et al. (2012) to evaluate PSW composition.

(1) It would be useful to add + or – signs before silicon isotope values for clarity. (2) + / - signs were not originally added as all $\delta^{30}\text{Si}$ seawater measurements considered in this study were positive, (3) but these have now been added to silicon isotope values for clarity.

(1) 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. (3) Manuscript amended to use heavy/light only when referring to delta values for consistency

(1) 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. (3) The products line have been removed from Figure 6 in the main body of text for clarity, and solid phase data where available is now included in the supplementary section S3 alongside collection and method of analysis.

(1) Note that Lena is spelled incorrectly in the caption to Figure 8. (3) Spelling corrected in Figure 8 caption.

References

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