

We thank Prof Damien Cardinal for his time in completing this review. He has contributed to some very useful points to help improve this manuscript. 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) Use dissolved silicon instead of silica: (3) Inconsistencies were addressed in the manuscript and "silica" replaced to "dissolved silicon" where relevant

(1) Fig. 1 and Table 1 and §2.1 From all these parts, it is unclear what are the sampling stations for N and Si isotopes from which cruises. There are 4 cruises on table 1, but in the paper isotopes data come mostly from only 2?... Please clarify.

(2) Thank you for highlighting this. The data presented in this manuscript for the main section across Fram Strait comes from two cruises (FS2018 & JR17005), with profiles for the Ile de France section from separate oceanographic cruises (FS2017, FS2018, FS2019, data presented in section 4.2.4 on sea-ice). (3) This has now been clarified in the Methods and Table 1 has been edited.

(1) In Fig. 1 mention that sea-ice extent displayed is from summer (I guess). There is no mention of N isotope sampling? (2) Sea ice extent is from September, (3) this has now been added to the Figure caption. (2) Nitrate isotope stations are shown by the blue & green dots on the map, silicon isotope stations are a subset of this. (3) This is now mentioned in the Figure 1 caption.

(1) L49 This sentence is unclear, since Pacific water are entering the Arctic, so the link with DSi export is not straightforward. Put "net supply of DSi" instead of "net export"? Or rephrase more clearly. (3) This sentence was rephrased to read "the excess of DSi in the Arctic Ocean's Si budget is attributed to Pacific water [...] and freshwater sources".

(1) L84 and 87-88, 93 provide st. dev. of these end-members isotopic signatures. (3) Stdvs for end-member isotopic signatures now included in the text.

(1) L86 Wrong reference. It should be Fripiat et al. 2018 instead of Fripiat et al. 2011 (3) Thank you for pointing out this mistake. It has now been edited to the correct reference

(1) L115: Samples have been deep-frozen, also for silicic acid concentration? This is not optimal since silica precipitation can take place during deep freezing. Please comment.

(2) We recognise that measurement from frozen is suboptimal for silicic acid concentrations, but separate non-frozen samples could not be collected for nutrients due to sampling and shipping restrictions. Concentrations were independently checked at the University of Edinburgh from the silicon isotope samples (acidified preserved) during analysis with the HACH reagent method. Both datasets were in very good agreement and frozen samples were not found to have lower DSi concentrations. DSi concentrations from FS2018 also closely align with concentrations measured in the same water masses in JR17005 below the seasonal layer in the upper 500m of the water column, and align with published concentrations in the literature. Through these combined methods of verification, we are confident on the validity on the accuracy of the concentrations measured by DTU.

(1) L120 Even though it is standard protocol, some reference on methods should be provided for nutrient analyses. (3) References to nutrient analysis of JR17005 (Brand et al., 2020) and FS2018 (Hansen and Koroleff, (1999) & Schnetger and Lehnert, (2014)) are now included.

(1) L135 What is the difference between this preconcentration protocol and the one of Reynolds 2006, which has 2 steps too? (2) It follows the same principles as Reynolds et al. (2006) with increased sample volumes (40ml instead of 2ml) and reduced NaOH volumes for co-precipitation (1.1% v/v in 1<sup>st</sup> step, 1% v/v in 2<sup>nd</sup> step instead of 2%).

(1) Fig. 2 Legend is incomplete, e.g. there is no scale for the T°C panels. (3) Temperature scale added to figure 2 and legend has been updated.

(1) Table 2. Add st. dev. on all parameter water mass averages (e.g. NO<sub>3</sub> and DSi concentrations, N\*, Si\*, capital delta). (3) Stdvs for all parameters now included in Table 2.

(1) Fig. 4 and 9. Be consistent with the name of the parameter, certainly no silica concentration is displayed here. Dissolved silicon or silicic acid would be much more appropriate. (3) Figure captions edited to read dissolved silicon instead of silica.

(1) L253-254. It is claimed here that AW follow more an open system with small fractionation. This is not so obvious from Fig. 6, especially for spring where almost no point fits the open model (grey line) except at low utilization (f close to 1) where both models cannot be differentiated. In summer, data are more consistent with open model, but then, why summer (i.e. more stratified I guess) would be more behaving as an open system? It would have been expected more from spring.

(2) Regarding the first part of this comment: Thank you for pointing this out, which we agree with. We have updated parameters of the Rayleigh model based on reviewers suggestions (see general comment to all reviewers for details of this). Based on the updated model, AW falls between closed and open conditions in AW, with a shift from more closed conditions in spring to more open conditions in summer. An increase in stratification would indeed lead to closed system fractionation. On the other hand, a shift from utilisation of “new” nitrate to regenerated nitrate is expected as nitrogen becomes depleted over the growth season and becomes more heavily recycled within the water column, leading the apparent fractionation trends to shift from closed to open system instead, as observed in our study.

(1) L262-263 The linearity between d<sub>30</sub>Si et DSi utilization is consistent with an isotopic fractionation highlighted L257, so, why say at the end of the § that it is mixing that control d<sub>30</sub>Si? Could mixing behavior be displayed on Fig. 6 to decipher? (2) This was a typo in the text and the sentence should have referred to the mixing behaviour of PSW instead. (3) This sentence has now been amended in the manuscript.

(1) Fig. 6 is there a justification having spring and summer displayed in different panels? Since they could represent the same growth season / isotopic system, data could be merged? (2) Following suggestions from all reviewers we have updated our Rayleigh model and data is now separated by water masses instead to reflect different nutrient sources. This is further commented on in the general comment to all reviewers.

(1) Fig. 8. Mention from which depths the data have been taken. Is it only surface samples? Here also, the legend seems to be incomplete / erroneous. In the caption panel, the triangles are different from the graph (e.g. there are different colours, and different shapes with triangle tips up / down not consistent with the main graph). Consequently, I don't understand how the linear trendline for

AW has been drawn? How these AW and PSW trendlines compared with Rayleigh models displayed in Fig. 6?

(3) Figure 8 and its figure caption has now been updated for clarity (Figure 8 included below for reference). (2) The AW and PSW trendlines in Figure 8 follow observations from the Rayleigh models in Figure 6. Namely that DSi utilisation in AW follows closed system kinetics from isotopically light Atlantic DSi sources, while fractionation in PSW does not show a good fit with Rayleigh models.  $\delta^{30}\text{Si}$  in PSW appears to be controlled by a mixture of AW and Arctic-sourced nutrients instead, plotting between the AW and Arctic trendlines.

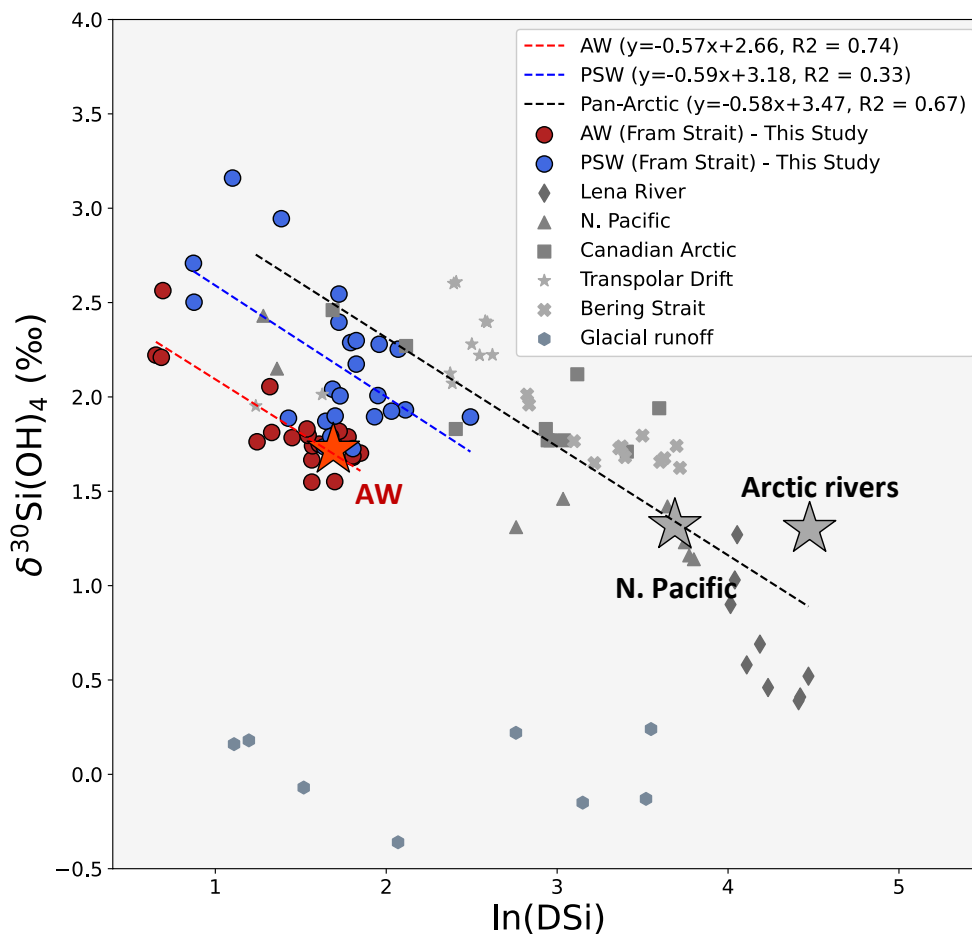


Figure 1: Pan-Arctic trends of  $\delta^{30}\text{Si}(\text{OH})_4$  against  $\ln(\text{DSi})$ . Colored dots show measurements from within AW (red, max. depth = 600m) and PSW watermasses (blue, max. depth = 150m) from this study based on water mass definitions in Table 2. Grey symbol sets are published  $\delta^{30}\text{Si}(\text{OH})_4$  from major DSi sources to the surface Arctic domain and surface water masses. Triangles: N. Pacific (<100m, stations 1-6); Stars: Transpolar drift (<60m, stations 30-38 from Brzezinski et al., 2021). Crosses: Bering Strait (max. depth = 60m, stations 4-6 from Brzezinski et al., 2021). Squares: Canadian Arctic (surface and intermediate water mass signatures of the Canadian Arctic sector, from Table 2 in Giesbrecht et al., 2022). Octogones: Glacial runoff from Greenland and Svalbard glaciers (Hatton et al., 2019). Diamonds: Lena river (Sun et al., 2018). Stars show average endmember composition of AW (red) and Pacific and riverine sources (Grey). Red dotted trendline is the least-squared regression for  $\delta^{30}\text{Si}(\text{OH})_4$  vs the natural logarithm of DSi within AW, blue and grey dotted trendlines are the equivalent for PSW and pan-Arctic (excluding Fram Strait) respectively. These trendlines show fractionation from partial utilisation of DSi consistent with fractionation models.

(1) L397 Weird wording here, probably “while” is not needed...(3) “While” removed from sentence.

(1) L407 who is Francis??? (2) This refers to data from the master’s thesis by A. Francis (University of Edinburgh) on nitrate isotopes in Arctic rivers from ARCTICGRO samples (Francis, 2019). (3) This work is now directly referenced within the manuscript.

## References

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- Francis, A.: Stable Isotope Tracing of Dissolved Nitrogen from Permafrost Degradation in Arctic Rivers, University of Edinburgh., 2019.
- Hansen, H. P. and Koroleff, F.: Determination of nutrients, in *Methods of Seawater Analysis*, edited by K. Kremling and M. Ehrhardt, pp. 159–228, Verlag GmbH., 1999.
- Reynolds, B. C., Frank, M. and Halliday, A. N.: Silicon isotope fractionation during nutrient utilization in the North Pacific, *Earth Planet. Sci. Lett.*, 244(1–2), 431–443, doi:10.1016/j.epsl.2006.02.002, 2006.
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