We thank Prof Damien Cardinal and Reviewer #3 for their additional comments on our 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.

## **Response to Reviewer #3**

(1) The authors should clarify, if they produced duplicate and triplicate measurements and mention that in the method part. It is very uncommon for stable Si isotopes in seawater if all the samples were only measured once (using a standard-bracketing method with 3-4 replicates) as they are normally not volume limited. It is very important to have at least parts of the samples measured as full replicates including the MAGIC precipitation step and column chemistry and run in different analytical sessions (eg. De Souza et al., 2012; Grasse et al., 2013; Liguori et al., 2021). This is especially important in this study, as 1)  $\delta^{30}$ Si in the study underlies many different processes, 2) samples with low DSi concentrations are difficult to measure correctly due to high matrix/Si ratio. (2) With the addition of the international seawater standard ALOHA for seawater silicon isotope measurements, it is not uncommon to not run full method replicates on each individual seawater samples (Giesbrecht et al., 2022; Ng et al., 2020; Cao et al., 2012). This is because running several ALOHA standards also provides full replicates of the full preparation method and analytical procedure over the entire measurement period. In the case of this study, those very good intrarun reproducibility where 1SD = 0.08‰ and 0.05‰ for ALOHA<sub>1000</sub> and ALOHA<sub>300</sub> across 58 and 30 individual measurements respectively, including full method repeats. The reproducibility of the full chemical and analytical procedure, which includes chemical preparation and isotopic measurements in separate analytical sessions, was additionally estimated on a subset of duplicate samples (n = 8). The mean absolute difference between duplicate samples analyzed in this way was 0.04‰ (1 SD). (3) The above sentence is now included on L223.

(1) 3) The authors did not measure  $\delta^{30}$ DSi directly, but instead only measured  $\delta^{29}$ Si. (2) As already stated in the supplementary material and explained in the previous round of reviews,  $\delta^{30}$ DSi was also measured, but we have chosen to convert  $\delta^{29}$ Si to  $\delta^{30}$ Si as a precaution against any interferences that could have affected  $\delta^{30}$ Si in low concentration samples.

(1) Please report the uncertainties as 2SD, which is a common practice in reporting seawater Si isotopes data, and which will be useful for lab comparison. (2) Uncertainties for Si isotopes remain commonly reported in 1SD in seawater, modern and past work (Brzezinski & Jones, 2015; Closset et al, 2022; Dumont et al, 2020). Thus for consistency with N isotope datasets and as it does not impede comparison with other datasets, we have kept our measurements reported to 1SD.

(1) The authors stated "AW follows the traditional isotopic effect of 5‰ and PSW follows the particularly low isotopic effect of 2‰", but it is not obvious in Figure 6. If the authors plot fractionation curves with  $\varepsilon$ =2‰ on the left upper plot, they should also fit the dataset as much as the curves with  $\varepsilon$ =5‰. The authors should do statistical analyses for example curve fitting to support this statement. Same goes to the statement "In PSW... A shift towards a mostly linear trend in summer is observed, suggesting open system kinetics below the mixed layer", it is difficult to see this trend from the right upper plot in Fig.6. There are also some triangle datapoints visually follow the exponential trend. So please use statistical analyses to support this statement. This is very important for the follow up discussion to be convincing. (2) This discussion refers to N isotopic trends previously described for the published dataset in Tuerena et al., 2021, with justification and statistical analysis therein. (3) "As described in Tuerena et al. (2021a) for this dataset" added to L384 and reference to the same paper on L628.

(1) The authors should also be careful with some terms/concepts that are discussed in the manuscript, for example: " $\delta^{30}$ Si(OH)4 in PSW does not show a good fit with either of the Rayleigh models". (3) Sentence on L530 was changed to read " $\delta^{30}$ Si(OH)<sub>4</sub> in PSW does not show a good fit with either of the fractionation models"

(1) In the paragraph Line 458 to 463, there's a logic gap from previous discussion to "this suggests that....PSW carries a remote signal of partial DSi uptake, and is controlled by mixing and dilutive effects rather than local biological processes". In fact, this is a conclusion to draw after the discussion in section 4.2. So, it came a bit too early to place this conclusion already in section 4.1.1, or more discussions/arguments are needed here. (3) L533-534 rephrased to read "This suggests that unlike nitrate, DSi in PSW is not primarily controlled by biological processes, and its variations are more likely to be driven by physical mixing and dilutive effects instead." instead, following the logic of line 530-533 above.

## References

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## **Response to Damien Cardinal**

(1) L 100-103, L260 and all over the manuscript. The st dev provided in the revised version were needed are welcome. It is however not obvious that isotopic signatures are significantly different (e.g. 5.5 +/- 0.4 vs. 5.1 +/- 0.2 p: mil). Simple stats should be reported to limit the discussion to significant differences only. Note that this remark applies to the whole manuscript, often – but not always – average and SD are provided, but never p-value and significance of the differences when comparing concentration or isotopic signatures of water masses (t-test is probably appropriate most of the time). (3) t-test values were added to L268, L278, L332, L346-347.

(1) L124-125 Unclear / meaningless. Need to rephrase. (3) L124-125 We rephrased this statement to "This heavy isotopic enrichment is attributed to physical processes (Liguori et al., 2020) and biological modification within surface waters (Giesbrecht et al., 2022; Varela et al., 2016)."

(1) L150-168 It'd be good to put the answer to my comment on frozen samples and DSi measurements in the revised paper to inform the readers that this has been handled and also to underline that it's not a standard protocol. (3) Added to L167 "While measurement from frozen is suboptimal for silicic acid concentrations, separate non-frozen samples could not be collected for nutrients due to sampling and shipping restrictions. DSi concentrations were independently checked at the University of Edinburgh from the silicon isotope samples (acid preserved) during analysis with the HACH reagent method. Both datasets from frozen and acidified 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."

(1) Fig. 6 Incomplete caption. To which depths these lines were drawn (it's mentioned that red dots are from the mixed layer, but what is the depth range of the black data?). Refer in the caption to how f has been calculated (table 2). (3) Depth range of the black data plotted is now included in the caption of Fig.6: "Nitrate utilisation vs  $\partial^{15}N$ -NO<sub>3</sub> for AW (left, depth <600m) and PSW (right, depth < 150m). Bottom panels: DSi utilisation vs  $\partial^{30}Si(OH)_4$  for AW (left, depth <600m) and PSW (right, depth < 150m)." (3) Also added to figure caption: "f is the fraction of nutrient remaining, calculated from the nutrient concentrations of water masses AW and PSW below the MLD (Table 2)."

(1) Fig. 8 + L460 + L 615 + section 4.2.6 starting L715: A fig. d30Si vs 1/DSi would really help to look at mixing. It could ideally be in a second panel in Fig. 8 since the current panel (and the text) clearly shows that PSW are largely scattered and are not explained by the Rayleigh system displayed.

(2) While we agree that d30Si vs 1/DSi can be a useful way to display data and look at mixing, in the case of looking at a wide area over the Arctic Ocean, where both mixing and uptake come into play and the range of DSi concentrations is large, we do not believe that displaying the data in this way significantly improves the discussion from section 4.2.6 onwards, as shown on the plot below. As such, it was not included in the manuscript.



(1) Section 4.2.6 and Fig. S4. This figure is very informative, I'd put it in the main manuscript. With this figure, there could be a brief discussion on what model is the more likely to represent the data. It is quite clear that the open model is more coherent since (i) r2 is much better than Rayleigh and, (ii) the slope is more consistent with global epsilon (-1pmil). (3) Figure S.4 is now included in the main text as Figure 11, with brief discussion on the fit of models from L809-813.