

We would like to thank the editor and the reviewer for the constructive comments and all the ideas suggested to our manuscript. These comments and suggestions are insightful and very helpful in improving the quality of our paper. We have read all the comments carefully and have made the necessary revisions to the manuscript. **All changes to the text are indicated in blue.** A detailed explanation for each issue raised is provided below:

Reviewer 1

There is growing interest in how the silicon isotope composition (expressed as $\delta^{30}\text{Si}$) of radiolarians can be used to supplement/complement those of the more established proxy archives in diatoms and sponges. As with these two groups, there is a need to understand and account for any post-mortem alteration of the initial isotope signal. Here, Zhang et al compare bulk-assemblage radiolarian $\delta^{30}\text{Si}$ values from water-column plankton tows and underlying core-top sediments. They demonstrate the two are statistically indistinguishable, lending more confidence to the use of radiolarian $\delta^{30}\text{Si}$ as a window on to past Si cycling.

In general, this manuscript is very well written and the figures are clear (with possible exception of Fig.3). The referencing is generally appropriate – with some notable absences (see below). The data are generated by appropriate techniques (though some more details may be warranted). Overall, there is little to criticise in terms of the central conclusion – that the radiolarian $\delta^{30}\text{Si}$ signal is not altered during water column sinking – which is supported by the data (though I have a series of minor comment/suggestions that I detail below). Nevertheless, this is a relatively small dataset and I have the impression that with a slightly expanded dataset much more could be done. I note in the supplement, Fig. S1 contains 22 ‘unpublished’ radiolarian datapoints. By integrating these, and dissolved Si $\delta^{30}\text{Si}$ data, a much more impactful paper would result. Some suggestions are below, but this is ultimately an editorial decision.

Response: We appreciate the reviewer’s insightful comments. Yes, we have included 22 ‘unpublished’ radiolarian $\delta^{30}\text{Si}$ data in Fig. S1 of the Supplement Material. Given that the primary objective of the present study was to ascertain whether and why radiolarian silicon isotopes ($\delta^{30}\text{Si}_{\text{rad}}$) signatures are faithfully transferred from the water column to the sediments, combined $\delta^{30}\text{Si}_{\text{rad}}$ records from paired water column and surface sediment samples are required. However, the radiolarian tests used to obtain these 22 data were extracted from surface sediments, and lack corresponding $\delta^{30}\text{Si}_{\text{rad}}$ data available from the overlying water column (although plankton samples were collected from some of these 22 stations, the quantity of radiolarian tests was insufficient for isotope analysis). As these 22 data were measured within the same analytical batch as samples used in this study, we included them in the Supplement Material to demonstrate that $\delta^{29}\text{Si}$ and $\delta^{30}\text{Si}$ values of radiolarian tests fall on the expected mass-dependent fractionation line $\delta^{29}\text{Si} = 0.51 \times \delta^{30}\text{Si}$ (Reynolds et al., 2006) (Figure S1 in the Supplementary Material), thereby indicating the effective removal of all polyatomic interferences during measurement.

Regarding dissolved Si $\delta^{30}\text{Si}$ data and the constraint of the radiolaria Si isotope fractionation factor, we also collected water samples from the relevant depth range when collecting plankton samples and surface sediments at each sampling station in this study. Considering the relatively low $\text{Si}(\text{OH})_4$ concentrations in these water samples from the upper water column, we plan to use a

Neoma MC-ICP-MS for the silicon isotopic analysis of dissolved silicon because of its enhanced sensitivity and mass resolution. However, isotopic analyses for seawater samples are pending due to ongoing instrument-related issues with the Neoma MC-ICP-MS at the Geochronology and Tracers Facility, British Geological Survey. When these data are obtained, we will prepare a separate manuscript addressing the radiolarian fractionation factor and its implication for nutrient levels in the mid-upper water column.

Minor comments and suggestions

L16-27 – the prominence of the SALH in the introduction is a bit strange to me, considering it’s not the focus of the manuscript (and isn’t returned to)

Response: In the introduction, we referenced the “silica acid leakage hypothesis” (SALH) due to its emphasis on the influence of changes in dissolved silicon (DSi) concentrations between the Southern Ocean and low-latitude regions on atmospheric $p\text{CO}_2$ and climate. While the SALH remains debated, partly due to inconsistent records of siliceous productivity and DSi concentrations in overlying waters across different low-latitude regions during the late Quaternary, we cite it here to highlight the importance of reconstructing past Si nutrient levels throughout the water column. This is expected to facilitate a more comprehensive understanding of the relationships between the Si cycle, biological pump efficiency, and global climate change.

L48: Two papers that deserve citation/discussion here and elsewhere are Closset et al. 2015 (doi: 10.1002/2015GB005180) and Grasse et al. 2021 (doi: 10.3389/fmars.2021.697400) – both present a comparison of plankton tow diatoms and sediment trap (Closset) or core-top (Grasse) material, concluding that the transfer of biogenic silica from surface ocean to depth isn’t associated with a resolvable change in $\delta^{30}\text{Si}$. See also Varela et al. 2004 (doi: 10.1029/2003GB002140; sediment trap data) and Fripiat et al. 2012 (doi: 10.5194/bg-9-2443-2012; water column biogenic silica data).

Response: We appreciate the reviewer bringing these important references to our attention, and [we have included them in the revised version](#). Indeed, the constancy of $\delta^{30}\text{Si}$ of diatom biogenic silica ($\delta^{30}\text{Si}_{\text{BSi}}$) with depth has been documented for suspended particles in the Atlantic sector of the Southern Ocean (Fripiat et al., 2012), as well as in sediment traps in the Southern Ocean south of New Zealand (Varela et al., 2004) and Australian (Closset et al. 2015). Furthermore, a strong agreement has also been observed between $\delta^{30}\text{Si}_{\text{BSi}}$ from seawater samples and those from core-top sediments in the central upwelling region off Peru (Grasse et al. 2021). These findings suggest that the transfer of diatom biogenic silica from the surface ocean to sediments is not associated with a dissolution-driven alteration of $\delta^{30}\text{Si}$.

References:

- Closset, I., Cardinal, D., Bray, S. G., Thil, F., Djouaev, I., Rigual-Hernández, A. S., Trull, T. W.: Seasonal variations, origin, and fate of settling diatoms in the Southern Ocean tracked by silicon isotope records in deep sediment traps. *Global Biogeochemical Cycles*, 29(9), 1495-1510. <https://doi.org/10.1002/2015GB005180>, 2015.
- Fripiat, F., Cavagna, A. J., Dehairs, F., De Brauwere, A., André L., Cardinal, D.: Processes controlling the Si-isotopic composition in the Southern Ocean and application for paleoceanography, *Biogeosciences*, 9(7), 2443-2457, <https://doi.org/10.5194/bg-9-2443-2012>, 2012.

Grasse, P., Haynert, K., Doering, K., Geilert, S., Jones, J. L., Brzezinski, M. A., Frank, M.: Controls on the silicon isotope composition of diatoms in the peruvian upwelling. *Frontiers in Marine Science*, 8, 697400, <https://doi.org/10.3389/fmars.2021.697400>, 2021.

Varela, D. E., Pride, C. J., Brzezinski, M. A.: Biological fractionation of silicon isotopes in Southern Ocean surface waters. *Global biogeochemical cycles*, 18(1). <https://doi.org/10.1029/2003GB002140>, 2004.

L85 and introduction: In general, there is a growing awareness that ‘bulk’ assemblage $\delta^{30}\text{Si}$ data have disadvantages as a paleo-archive, and that where possible single-species records are much stronger. Therefore it would be good to see some justification for why this was not attempted here.

Response: We thank the reviewer for raising this important point.. We agree with the reviewer that $\delta^{30}\text{Si}$ records derived from single-species shells are generally preferred for paleo-archive studies, as the potential for different vital effects on $\delta^{30}\text{Si}$ fractionation may exist between various radiolarian species (e.g. Doering et al., 2021). However, obtaining sufficient material for silicon isotopic analysis from individual radiolarian taxa in our samples proved to be a significant challenge, necessitating our focus on the bulk radiolarian species record for this study.

We did, in fact, attempt to isolate single-species tests via manual picking under a microscope, but encountered several obstacles. Firstly, for silicon isotope analysis using a Thermo Fisher Scientific Neptune Plus MC-ICP-MS at the Geochronology and Tracers Facility, British Geological Survey, 1 to 1.5 mg of purified radiolarian tests is typically required. Radiolarian tests from sediments in the tropical Ocean are notably lightweight, averaging 0.063 to 0.136 mg/shell (Moore, 1969; Takahashi, 1982). Consequently, a mean of approximately 7,000 to 15,000 individual tests are required to achieve the ~1 mg of material needed for the silicon isotope analysis. Given the high diversity of radiolarians in Holocene sediments, particularly in low-latitude oceans (Moore, 1969; Takahashi, 1982; Boltovskoy et al., 2010) and in our samples, which average over 100 species per sample, coupled with the observation that most species typically constitute less than 10% of the total radiolarian community (Boltovskoy et al., 2010; Chen et al., 2008), this means that 70,000 to 150,000 bulk radiolarian tests are potentially required for each sample to obtain sufficient single-species material. Regrettably, the limited volume of the plankton samples made it difficult to obtain such a large number of radiolarian shells. Secondly, manually picking 7,000 to 15,000 individual tests out of 70,000 to 150,000 bulk radiolarian tests is highly time-consuming, rendering this approach impractical for the study of single-species radiolarian $\delta^{30}\text{Si}$. Furthermore, even with meticulous hand-picking under a microscope, the electrostatic adsorption of these light tests to transfer tools, such as brushes, inevitably results in test loss during transfer to storage vials. This further compounded the difficulty of accumulating the required quantity of monospecific tests for the isotope analysis.

It was these insurmountable challenges encountered during our attempts at hand-picking that prompted us to develop the method for extracting and purifying bulk radiolarian tests from the sediments for the study of the radiolarian $\delta^{30}\text{Si}$ (Zhang and Swann, 2023). This method enabled us to effectively obtain sufficient pure radiolarian tests to conduct the comparative analysis of radiolaria $\delta^{30}\text{Si}$ compositions using water column and surface sediment samples in this current study.

Based on the reviewer’s comment, we have included a brief explanation in the revised manuscript regarding the absence of silicon isotopic analysis for individual radiolarian taxa:

“Given the lightweight nature of shells and the high diversity of radiolarians, particularly in low-latitude oceans (Moore, 1969; Takahashi, 1982; Boltovskoy et al., 2010), combined with the observation that most species typically comprise less than 10% of the total radiolarian assemblage (Boltovskoy et al., 2010; Chen et al., 2008), obtaining sufficient material from individual radiolarian taxa for silicon isotopic analysis remains a considerable challenge.”

By the way, we have measured bulk radiolarian $\delta^{30}\text{Si}$ ($\delta^{30}\text{Si}_{\text{rad}}$) from a sediment core collected in the northern South China Sea (SCS), spanning a period from ~17 ka to the present. The results indicate that $\delta^{30}\text{Si}_{\text{rad}}$ values, and the calculated silicic acid utilisation efficiency, were generally lower during the last deglacial period compared to the Holocene. This suggests a potential increase in silicic acid concentrations in the mid-upper water column during the last deglacial, potentially stemming from a deepened mixed layer induced by an intensified winter monsoon in the northern SCS (e.g. Steinke et al., 2011), or from an enhanced influx of silica-rich Antarctic Intermediate Water (AAIW) into the northern SCS (e.g. Huang et al., 2014; Yang et al., 2017). Such conditions, characterized by increased silicic acid levels in the mid-upper water column, could account for the previously observed higher productivity in the northern SCS during the last glacial period (e.g. Lin et al., 1999; Higginson et al., 2003). This finding may provide compelling evidence for the effectiveness of bulk radiolarian $\delta^{30}\text{Si}$ as a proxy for reconstructing past changes in silicic acid levels or availability within the water column. A manuscript detailing these findings is currently in preparation.

References:

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- Takahashi, K.: Vertical flux, ecology and dissolution of Radiolaria in tropical oceans: implications for the silica cycle. PhD thesis. Woods Hole Oceanographic Institution and Massachusetts Institute of Technology, 1982
- Steinke, S., Glatz, C., Mohtadi, M., Groeneveld, J., Li, Q., Jian, Z.: Past dynamics of the East Asian monsoon: No inverse behaviour between the summer and winter monsoon during the Holocene, *Global and Planetary Change*, 78(3-4), 170-177, <https://doi.org/10.1016/j.gloplacha.2011.06.006>, 2011.

Yang, Y., Xiang, R., Liu, J., Fu, S., Zhou, L., Du, S., Lü, H.: Changes in intermediate water conditions in the northern South China Sea using *Globorotalia inflata* over the last 20 ka. *Journal of Quaternary Science*, 32(7), 1037-1048, <https://doi.org/10.1002/jqs.2974>, 2017.

Zhang, Q., Swann, G. E. A.: An effective method to extract and purify radiolaria from tropical marine sediments, *Front. Mar. Sci.*, 10, 1150518, <https://doi.org/10.3389/fmars.2023.1150518>, 2023.

Section 2.3: I would suggest more detail is needed here. Specific suggestions include:

- **Define what is ‘sufficient’ radiolarian tests (L80; what is the typical mass of Si processed)**

Response: We are sorry for the lack of clarity regarding the typical mass of processed Si. For silicon isotope analysis using a Thermo Fisher Scientific Neptune Plus MC-ICP-MS at the Geochronology and Tracers Facility, British Geological Survey, 1 to 1.5 mg of purified radiolarian tests is typically required. Accordingly, it was expected that at least approximately 1 mg of purified radiolarian tests would be extracted and purified from the samples used in this study. In this study, we successfully extracted over 1.5 mg of pure radiolarian tests for isotope analysis from all surface sediment samples. However, due to limited sample volumes, the quantity of pure radiolarian tests obtained from plankton samples was approximately 1 mg.

In the revised manuscript, we have substituted “sufficient radiolarian tests” with “approximate 1 to 1.5 mg of radiolarian tests”.

- **Give a brief overview of Zhang and Swann (L81). Is there potential for larger diatoms or sponge spicules to ‘contaminate’ the sample?**

Response: Following the reviewer’s suggestion, we have added a brief overview of Zhang and Swann (2023) in the revised manuscript as follows: Overall, the procedure for extracting and purifying radiolarian tests from marine sediments in Zhang and Swann (2023) comprises four stages: chemical treatment, initial sieving and differential settling, subsequent sieving, and finally density separation (Figure A). In the first stage, raw samples were treated with ~30% H₂O₂ and 15% HCl to remove the organic matters and calcareous components, and to facilitate particle dispersal. Following chemical treatment, the particles were rinsed and filtered using a 53 µm sieve to remove fine detritus, small diatoms, seaweed spines, and some sponge spicules. The filtered particles then underwent differential settling two to three times, followed by sonication, to further isolate radiolarians from large diatoms. Subsequently, particles were filtered three times using a 53 µm sieve by half immersing the sieve in a container filled with distilled deionized water (DDW) and gently tapping the base of the sieve for 10-15 minutes. This process aimed to remove all monoaxonic spicules and a portion of the small non-monoaxonic spicules. Finally, the retained fraction was further refined by density separation, using specific gravities from 2.1-2.0 g/cm³ at 0.5-unit interval, to remove any remaining coarse detritus and non-monoaxonic sponge spicules.

Larger diatoms and sponge spicules can also be effectively separated from radiolarian tests following the method of Zhang and Swann (2023). The majority of larger diatoms can be removed through differential settling (Figure A). Residual diatoms can then be broken down by sonication treatment for no more than 10 minutes, to avoid the breakage of radiolarian tests. These fragmented diatoms can then be further removed through sieving or differential settling. Although most sponge spicules are greater than 53 µm in length, the diameter (cross section) of monoaxonic sponge spicules is generally several micrometers. Therefore, monoaxonic spicules may pass

through a 53 μm sieve during wet sieving (Figure A), provided they are repeatedly suspended and settle non-horizontally in the water during the sieving process. This can be achieved by half immersing samples in a container filled with DDW, and gently tapping the base of the sieve to maintain particle suspension. Non-monoaxonic sponge spicules can be further refined via density separation (Figure A), using specific gravities from 2.1-2.0 g/cm^3 at 0.5-unit interval. This is based on findings that the mean density of sponge spicules is higher than that of tropical radiolarians in late Quaternary sediments (Zhang and Swann, 2023).

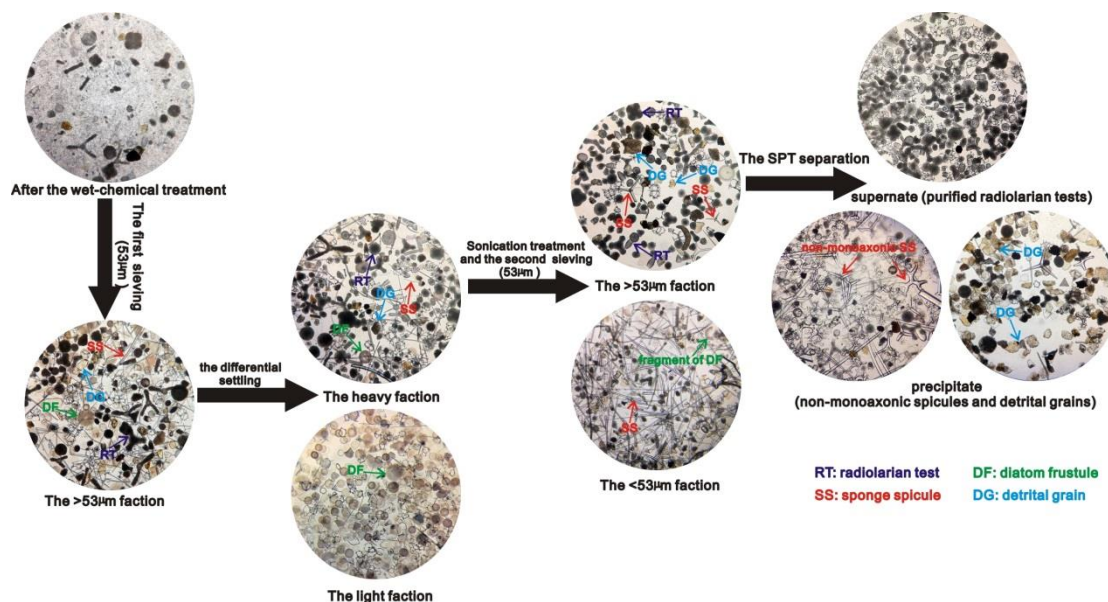


Figure A the main process for separating and purifying the radiolarian tests from tropical marine sediments (Zhang and Swann, 2023). All photos taken under the inverted microscope at 100x magnification.

Microscopic examination of purified radiolarian tests, across several fields of views at x100 magnification using inverted light microscopy, revealed a negligible quantity of larger diatoms and sponge spicules (<1%) remaining with the purified radiolarian tests (as shown in Videos 1 and 2 in the supplementary material of Zhang and Swann (2023)). Consequently, the potential for larger diatoms or sponge spicules to contaminate purified radiolarian samples is considered minimal or negligible.

References:

Zhang, Q., Swann, G. E. A.: An effective method to extract and purify radiolaria from tropical marine sediments, *Front. Mar. Sci.*, 10, 1150518, <https://doi.org/10.3389/fmars.2023.1150518>, 2023.

● Confirm that Na (used in dissolution) was successfully removed by the ion exchange chromatography

Response: Prior to isotopic analysis, all samples are purified using cation exchange chromatography. At a pH of 2 and 8, Si species are either neutral ($\text{Si}(\text{OH})_4$) or anionic (H_3SiO_4^-) and will therefore pass freely through a cation exchange resin whilst all major cations, including Na^+ , remain trapped on the column.

In the revised manuscript, the statement “Subsequent purification was achieved via ion exchange chromatography at a pH of between 2-8 (Georg et al., 2006; van den Boorn et al., 2006).” has been revised to “Subsequent purification was achieved via ion exchange chromatography at a

pH of 2-8 to ensure complete removal of cations, such as magnesium (Mg) and/or sodium (Na) (Georg et al., 2006; van den Boorn et al., 2006).”

- **Give approximate mass resolution (m/Dm, L93)**

Response: In high-resolution (HR) mode, resolution (R) = $m/\Delta m$, where Δm is derived from the rising edge of the peak, measured at 5% and 95% relative peak height. On the Neptune Plus at the British Geological Survey, R typically equates to between 9,000–10,000, which is sufficient for partial (or pseudo) resolution of each of the silicon (Si) isotopes from their respective interferent/s.

In the revised manuscript, we have included the information of resolution “In high-resolution mode, the instrument typically exhibited a mass-resolution between approximately 9,000 to 10,000, and a sensitivity of 4-5 V/ppm.”

- **Give details of how Mg measured/which ratio(s) (presumably in ‘dynamic’ mode), and a reference to Cardinal et al. 2004 (doi: 10.1039/b210109b) is probably appropriate.**

Response: The samples were doped with ~300ppb magnesium (Mg, Alfa Aesar SpectraPure). Spiking with an external element of known isotopic composition ($^{24}\text{Mg}/^{25}\text{Mg} = 0.126633$) allows the data to be corrected for the effects of instrument-induced mass bias. Simply, any deviation of the measured $^{24}\text{Mg}/^{25}\text{Mg}$ value from the known value is attributed to the effects of mass bias. The isotopes of Si are assumed to be similarly affected, and consequently, an exponential drift correction is applied. The collector configuration used is illustrated below:

Sequence \ Detector position	Low 4	Low 3	Axial	High 3	Integration time/seconds	Settle time /seconds
1		^{28}Si	^{29}Si	^{30}Si	16.8	3
2	^{24}Mg		^{25}Mg		8.4	3

Yes, we agree that the reference to Cardinal (Cardinal. D et al. J. Anal. At. Spectrom., 2003, 18, 213–218) (not Cardinal et al., 2004) should be cited here.

In the revised manuscript, the statement “Finally, all samples are spiked with approximately 300 ppb magnesium (Mg, Alfa Aesar SpectraPure, $^{24}\text{Mg}/^{25}\text{Mg} = 0.126633$) to enable correction of the data for instrument-induced mass bias.” has been revised to “Finally, all samples are spiked with approximately 300 ppb magnesium (Mg, Alfa Aesar SpectraPure, $^{24}\text{Mg}/^{25}\text{Mg} = 0.126633$) to enable correction of the data for instrument-induced mass bias (Cardinal et al., 2003). Any deviation of the measured $^{24}\text{Mg}/^{25}\text{Mg}$ value from the known value is attributed to the effects of mass bias. The isotopes of Si are assumed to be similarly affected, and consequently, an exponential drift correction is applied (Cardinal et al., 2003).”

References:

Cardinal, D., Alleman, L. Y., De Jong, J., Ziegler, K., & Andr   L.: Isotopic composition of silicon measured by multicollector plasma source mass spectrometry in dry plasma mode. Journal of Analytical Atomic Spectrometry, 18(3), 213-218, <https://doi.org/10.1039/B210109B>, 2003.

- **Give approximate sample introduction rate, concentration, and instrument sensitivity**

Response: Yes, we have included the information of sample concentration, introduction rate, and instrument sensitivity in the revised manuscript. The statement “Silicon isotope analysis was performed in dry plasma mode using the high mass-resolution capability of a Thermo Fisher

Scientific Neptune Plus MC-ICP-MS (multi collector inductively coupled plasma mass spectrometer) at the Geochronology and Tracers Facility, British Geological Survey.” has been revised to “Silicon isotope analysis was performed in dry plasma mode using the high mass-resolution capability of a Thermo Fisher Scientific Neptune Plus MC-ICP-MS (multi collector inductively coupled plasma mass spectrometer) at the Geochronology and Tracers Facility, British Geological Survey. Samples were typically prepared to yield a Si concentration of approximately 2 ppm and introduced to the MC-ICP-MS via an Aridus de-solvating unit, incorporating a PFA nebulizer with an uptake rate of 50 µL/min. In high-resolution mode, the instrument typically exhibited a mass-resolution between approximately 9,000 to 10,000, and a sensitivity of 4-5 V/ppm.”

● **Confirm what one analytical replicate represents (just one standard-sample-standard bracket, or (as is usual) three or four?)**

Response: “Analytical replicates” in our study refer to the repeated analysis of the standard-sample-standard bracket, typically performed for three times. As we usually do not have sufficient sample, specifically for radiolarian tests from plankton samples, it is difficult to conduct a full procedural replicate that encompass both chemical processing and analysis. The samples selected for replication following the standard sample bracketing procedure are the ones that have a higher Si concentration and which can, therefore, be diluted to run multiple times.

To clarify this point, in the revised manuscript, the statement “Analytical replicates were conducted where sample volume allowed...” has been revised to “Analytical replicates of the standard sample bracketing procedure were conducted where sample volume allowed...”.

L114: It’s not clear what volume the 28025-102443 individuals refer to – in a 1m² water column? It is also not clear how these numbers are derived – presumably because the volume of water passing through the nets (L66) is known? This could be clarified.

Response: We are sorry for the confusion caused by our lack of clarity. The Hydro-Bios MultiNet used for collecting plankton samples has an aperture area of approximately 0.25 m² (a square aperture with 0.5 m sides). During sampling, we typically employ a retrieval rate of ~0.1 m/s for the MultiNet to ensure adequate filtration of the seawater. Therefore, the figures of 28,025–102,443 individuals represent the number of radiolarian shells in a 0.25 m² water column sampled from 0–100 m.

In the revised manuscript, we have included the aperture area of the Hydro-Bios MultiNet. The statement “Plankton samples were collected from the 0-100 m and 100-300 m water layers at each station using a Hydro-Bios MultiNet with a 63 µm mesh size...” has been revised to “Plankton samples were collected from the 0-100 m and 100-300 m water layers at each station using a Hydro-Bios MultiNet with an aperture area of approximately 0.25 m² and a 63 µm mesh size...”.

In this study, 1/8 of each plankton sample was prepared as radiolarian slides for light microscope observations. More than 500 specimens were quantitatively identified and counted on each slide under the microscope at x100 magnification. The total number of radiolarian individuals in each sample was calculated from the count data as follows:

$$T = A * V_t / V * S * N$$

Where T is the total number of radiolarian individuals; A is the number of radiolarian shells counted from V fields of view on the slide; V_t is the total number of view fields on the radiolarian

slide; V is the number of view fields examined under the microscope for the radiolarian individual count; S is the number of radiolarian slides; and N is the aliquot size of the sample (eight for the plankton sample, and one for the sediment sample in this study).

Based on comments from both reviewers, the original statement “To determine the species composition of radiolarians, in both plankton samples and surface sediments, all samples were wet-sieved through a 63 µm sieve and prepared into radiolarian slides following the method described by Zhang et al. (2014). Radiolarian species were then identified and counted under a Nikon optical microscope at x100 or x200 magnification, with more than 500 specimens identified on slides using the publications of Chen and Tan (1996) and Tan and Chen (1999). Relative abundances of various species were then calculated based on individual count of each species and the total number of radiolarian specimens observed under the microscope.” has been revised to:

“To determine the species composition of radiolarians, in both plankton samples and surface sediments, all samples were wet-sieved through a 63 µm sieve and prepared into radiolarian slides following the method described by Zhang et al. (2014). Briefly, samples were treated with a sufficient volume of 5% HCl solution for 15 minutes to eliminate calcareous organisms. Subsequently, the residual was processed using a sonic oscillator for one minute, and subjected to differential settling to remove impurities potentially adhering to the radiolarian tests. Following these procedures, all residual material was strewn onto microscope slides and permanently mounted with Canada Balsam. Radiolarian species were then identified and counted under a Nikon optical microscope at x100 or x200 magnification, with more than 500 specimens identified on slides using the publications of Chen and Tan (1996) and Tan and Chen (1999). Radiolarian diversity was determined by the species richness in each sample. The total number of radiolarian individuals in each sample was estimated from the count data as follows:

$$T = A * (V_t/V) * S * N \quad (1)$$

Where T is the total number of radiolarian individuals; A is the number of radiolarian shells counted from V fields of view on the slide; V_t is the total number of view fields on the radiolarian slide; V is the number of view fields examined under the microscope for the radiolarian individual count; S is the number of radiolarian slides; and N is the aliquot size of the sample (eight for the plankton sample, and one for the sediment sample in this study). Relative abundances of various species were then calculated based on individual counts of each species and the total number of radiolarian specimens on each slide observed under the microscope.”

L126: Can an approximate detection limit be given for these elements?

Response: In this study, the detection limit of routine EDS analysis using IT-200 EDS detector for major elements is approximately 1000pm or 0.1 %. However, elemental concentrations generated by the EDS software exceeding 0.1% do not necessarily confirm their presence within the analysed sample. As shown in Figure 4 of the manuscript, some elements (e.g. Na, exceeding 0.1%) within EDS (A” and B”) spectrum images are flagged in red. This indicates that the software does not have a 99% statistical confidence in the presence of these elemental signatures within the analysed sample. This typically occurs for two reasons: either 1) the calculated wt% of these elements is very low, accompanied by a high standard deviation; or 2) the x-ray energy level peak for these elements does not stick out above the background noise peaks.

In the revised manuscript, we have included the approximate detection limit of the EDS analysis using IT-200 EDS detector for these major elements. The sentence “The concentrations of

other elements (Fe, Al, Mg, Sr, Na, K and Cl) were all below the minimum detectable limit for the EDS detector (Figure 4)” has been revised to “other elements (Fe, Al, Mg, Sr, Na, K and Cl) flagged in red within the EDS spectrum images (Figure 4) indicate that either their concentrations were below the minimum detectable limit (~ 0.1 %) for the EDS detector, or the software lacked a 99% statistical confidence in the presence of these elemental signatures within the analysed sample”.

Discussion section: In general, there is no discussion of any spatial pattern in radiolarian assemblage. But I feel there is probably useful insight here. For example, Station 28 is located away from the cluster of other stations, and visually in Fig. 3 looks different. What physicochemical parameters influence the community assemblages? As an aside, I note that the assemblage data is not made available. Could the species in Fig 3 be amalgamated at a higher taxonomic level in order to make the similarities and differences clearer? And/or condensed, via an appropriate multivariate statistical approach, to 2 axes?

Response: Thanks for the important point raised by the reviewer. Yes, spatial patterns within the radiolarian assemblage are not discussed in this study. This is because, whilst samples were collected from various stations in the South China Sea (SCS) in the current study, the primary objective of the present study was to 1) determine the depth interval from which the radiolarian community contributes to the $\delta^{30}\text{Si}_{\text{rad}}$ signature in the water column, and 2) ascertain whether and why radiolarian silicon isotopes ($\delta^{30}\text{Si}_{\text{rad}}$) signatures are faithfully transferred from the water column to the sediments. Consequently, our focus has been on comparing prominent radiolarian compositions at various water depths at each station, and $\delta^{30}\text{Si}_{\text{rad}}$ values between the water column and sedimentary record at individual stations. Variations in radiolarian assemblages across different regions may potentially result in spatial differences in $\delta^{30}\text{Si}_{\text{rad}}$; however, such considerations were beyond the immediate scope of this investigation. A separate manuscript focusing on the radiolarian fractionation factor and its implication for nutrient levels in the mid-upper water column, will address the spatial distribution of $\delta^{30}\text{Si}_{\text{rad}}$ and examine the primary factors influencing this, including the regional differences in radiolarian assemblages.

The reviewer is correct in noting the difference between the radiolarian assemblage at Station 28 and those at some of the other 6 stations. This discrepancy is likely attributed to variations in regional environmental conditions, primarily including sea surface temperature (SST) and nutrient levels, such as dissolved silica or silicic acid in seawater. Station 28 is situated in the southern SCS, whereas the remaining 6 stations are located in the northern SCS. Overall, the radiolarian assemblage at station 28 exhibits a higher relative abundance of typical warm-water tropical species in radiolarian assemblages, such as *Botryocyrtis scutum* and *Pterocorys hertwigii* at station 28. This is likely due to the higher mean annual SST in the southern SCS, which forms part of the Western Pacific Warm Pool, compared to the northern SCS. Conversely, the radiolarian assemblage at station 28 has a lower relative abundance of *Didymocyrtis tetrathalamus* t., a species indicative of nutrient-depleted Western Equatorial Pacific water intrusion into the northern SCS (e.g. Anderson et al., 1990; Zhang et al., 2015). This suggests a reduced influence of Western Pacific waters on the southern SCS compared to the northern SCS. These observed differences in specific species and radiolarian assemblages between stations in the southern and northern SCS are approximately consistent with previous investigations into variations in radiolarian

assemblages within surface sediments and the water column, and their correlation with primary environmental parameters in the SCS (Chen et al. , 2008; Zhang et al., 2005, 2009).

The primary objectives of radiolarian assemblage data presented in this study are: 1) to identify the primary contributors to $\delta^{30}\text{Si}_{\text{rad}}$ compositions in the water column at each station; 2) to ascertain the potential for substantial dissolution-induced alteration of the radiolarians by comparing the prominent radiolarian composition between the water column and sediments at each station; and 3) to provide a crucial foundation for explaining why $\delta^{30}\text{Si}_{\text{rad}}$ signatures is faithfully transferred from the water column to the sediments. We contend that amalgamating the species presented in Fig. 3 of the manuscript at a higher taxonomic level would diminish the clarity and effectiveness of this data in addressing these objectives. Therefore, we have retained the species-level information shown in Fig. 3, as it provides a more effective means of fulfilling the stated objectives for radiolarian assemblage data in this study. A correlation analysis provides a clear understanding of the similarities and differences in dominant radiolarian species between different water depth ranges and the sedimentary record at each station, as well as between various sampling stations. This analysis indicates a variation in correlations between the prominent radiolarian composition from station 28 and those from northern SCS, with Pearson correlation coefficients ranging from 0.27 to 0.83 (Table A shown on the next page).

[In the revised manuscript, we will include the detailed radiolarian assemblage data \(original radiolarian count data\) for the samples used in this study as supplementary material.](#)

Reference:

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- Zhang L L, Chen M H, Lu J, et al. Polycystine radiolarian fauna and their distribution in the upper water column of the southern South China Sea. *Journal of Tropical Oceanography*, 2005, 24, 55–64 (in Chinese with English abstract).
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Table A Correlations between plankton samples and surface sediments for the prominent radiolarian species

Sample	S7 (0-100m)	S7 (100-300m)	S7 SS	S8 (0-100m)	S8 (100-300m)	S8 SS	S10 (0-100m)	S10 (100-300m)	S10 SS	S11 (0-100m)	S11 (100-300m)	S11 SS	S12 (0-100m)	S12 (100-300m)	S12 SS	S13 (0-100m)	S13 (100-300m)	S13 SS	S28 (0-100m)	S28 (100-300m)	S28 SS
S7 (0-100m)	1																				
S7 (100-300m)	.77**	1																			
S7 SS	.79**	.87**	1																		
S8 (0-100m)	.62**	.67**	.75**	1																	
S8 (100-300m)	.61**	.80**	.80**	.91**	1																
S8 SS	.65**	.77**	.84**	.90**	.93**	1															
S10 (0-100m)	.47*	.58**	.51**	.63**	.72**	.70**	1														
S10 (100-300m)	0.34	.58**	.55**	.53**	.68**	.58**	.74**	1													
S10 SS	.54**	.78**	.79**	.69**	.84**	.84**	.72**	.82**	1												
S11 (0-100m)	.54**	.72**	.68**	.55**	.62**	.64**	.47*	.48*	.71**	1											
S11 (100-300m)	.55**	.71**	.71**	.50**	.70**	.73**	.56**	.53**	.75**	.71**	1										
S11 SS	.52**	.67**	.60**	.77**	.83**	.83**	.79**	.66**	.83**	.69**	.79**	1									
S12 (0-100m)	.50**	.62**	.62**	.66**	.68**	.74**	.64**	.55**	.79**	.84**	.64**	.80**	1								
S12 (100-300m)	.56**	.77**	.71**	.69**	.80**	.74**	.73**	.70**	.80**	.71**	.68**	.86**	.75**	1							
S12 SS	.41*	.60**	.60**	.71**	.78**	.78**	.78**	.70**	.74**	.60**	.64**	.85**	.74**	.85**	1						
S13 (0-100m)	.58**	.81**	.77**	.72**	.81**	.84**	.66**	.65**	.86**	.87**	.72**	.82**	.89**	.82**	.75**	1					
S13 (100-300m)	.49*	.70**	.66**	.62**	.71**	.69**	.62**	.69**	.86**	.81**	.75**	.83**	.84**	.79**	.66**	.84**	1				
S13 SS	.40*	.55**	.60**	.84**	.81**	.84**	.78**	.64**	.74**	.54**	.57**	.88**	.72**	.81**	.86**	.74**	.73**	1			
S28 (0-100m)	0.27	0.38	.41*	.50**	.57**	.42*	.61**	.76**	.62**	.49*	0.36	.63**	.53**	.70**	.62**	.54**	.63**	.63**	1		
S28 (100-300m)	0.27	.46*	.45*	.48*	.58**	.42*	.58**	.83**	.67**	0.37	0.33	.55**	.41*	.71**	.54**	.48*	.56**	.56**	.85**	1	
S28 SS	0.30	0.38	0.36	.61**	.65**	.51**	.72**	.58**	.48*	0.30	0.380	.70**	.48*	.74**	.72**	.47*	.47*	.73**	.71**	.64**	1

** Correlation is significant at the 0.01 level (2-tailed)

* Correlation is significant at the 0.05 level (2-tailed)

S=station

SS=surface sediments

L134-140: In general, it's the fractionation between dissolved Si and biogenic silica that is relevant, not the absolute $\delta^{30}\text{Si}$ value. So it's a shame not to see any dissolved Si $\delta^{30}\text{Si}$ data from the water samples (perhaps this is coming in a later publication?). An existing dataset of $\delta^{30}\text{Si}$ exists for the SCS (Cao et al. 2012, doi: 10.1016/j.gca.2012.08.039). The overlap isn't perfect in terms of location or seasonality but its surprising that it's not mentioned here, given the importance of water $\delta^{30}\text{Si}$ in setting radiolarian $\delta^{30}\text{Si}$, and that it would allow the authors to place constraints on the fractionation of Si isotopes by radiolarians.

Response: The reviewer is correct. The fractionation between $\delta^{30}\text{Si}_{\text{rad}}$ and dissolved silicon isotopes ($\delta^{30}\text{Si}_{\text{DSi}}$), expressed as the apparent Si isotope fractionation factor ($\Delta^{30}\text{Si}_{\text{rad}} \sim {}^{30}\epsilon = \delta^{30}\text{Si}_{\text{rad}} - \delta^{30}\text{Si}_{\text{DSi}}$), is an important parameter for understanding to what extent the radiolarians fractionate the $\delta^{30}\text{Si}_{\text{DSi}}$ during Si absorption, and a potential proxy for reconstructing the silicon cycle in mid-upper depth waters. Previous published data show that the $\delta^{30}\text{Si}(\text{OH})_4$ values in the upper water column (above 100 m) of the northern South China Sea (SCS) range from 1.33 to 2.94 ‰, with a mean of 2.3 ‰ (Cao et al., 2012). Based on $\delta^{30}\text{Si}_{\text{rad}}$ compositions in plankton samples and surface sediments from our study, and a mean $\delta^{30}\text{Si}(\text{OH})_4$ values in the northern SCS from Cao et al., (2012), the radiolarian fractionation factors can be calculated to range from -0.45 to -0.74‰, with a mean of -0.56‰ in the SCS. The mean $\Delta\delta^{30}\text{Si}_{\text{rad}}$ value calculated in our study is more positive than the radiolarian fractionation factor (-1.5‰) applied by Hendry et al. (2014), but is close to the factor (-0.8‰) reported by Abelmann et al. (2015) and that (-0.62‰, mixed radiolaria) reported by Doering et al. (2021).

However, the absence of such data in this study is for the following reasons: 1) the primary objective of the present study was to determine the depth interval from which the radiolarian community contributes to the $\delta^{30}\text{Si}_{\text{rad}}$ signature in the water column, and to ascertain whether and why radiolarian silicon isotopes ($\delta^{30}\text{Si}_{\text{rad}}$) signatures are faithfully transferred from the water column to the sediments. Consequently, our focus has been on comparing prominent radiolarian compositions at various water depths at each station, and $\delta^{30}\text{Si}_{\text{rad}}$ values between the water column and sedimentary record at individual stations. The calculation of $\Delta^{30}\text{Si}_{\text{rad}}$ is not essential for this specific investigation; 2) although Cao et al. (2012) have provided a $\delta^{30}\text{Si}(\text{OH})_4$ dataset for the northern SCS, as the reviewer points out, the spatial and temporal overlap with our sampling regime is not sufficient to permit an accurate constraint of $\Delta^{30}\text{Si}_{\text{rad}}$. Actually, at each sampling station, when collecting plankton samples and surface sediments, we have also collected water samples from the relevant depth range for the $\Delta^{30}\text{Si}_{\text{rad}}$ constraint. Considering the typically low $\text{Si}(\text{OH})_4$ concentrations in these water samples from the upper water column, we plan to use a Neoma MC-ICP-MS for the silicon isotopic analysis of dissolved silicon because of its enhanced sensitivity and mass resolution. However, isotopic analyses for these seawater samples are pending due to ongoing instrument-related issues with the Neoma MC-ICP-MS at the Geochronology and Tracers Facility, British Geological Survey. Once these data are obtained, a more robust mean fractionation factor can be determined; and 3) A separate manuscript detailing the radiolarian fractionation factor and its implication for nutrient levels in the mid-upper water column is to be prepared in the near future.

Reference:

Abelmann, A., Gersonde, R., Knorr, G., Zhang, X., Chaplign, B., Maier, E., Esper, O., Friedrichsen, H., Lohmann, G., Meyer, H., Tiedemann, R.: The seasonal sea-ice zone in the glacial Southern Ocean as a carbon sink, *Nat. Commun.*, 6(1), 8136, <https://doi.org/10.1038/ncomms9136>, 2015.

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L165: Can an indication of the timespan covered by the upper 1cm be given? I presume there are some constraints on sedimentation rates and bioturbation in this well studied region.

Response: Thanks for the reviewer's comment. Sheng et al., (2024) provide a comprehensive quantification of the spatial distribution of sedimentation rate (SR) and sediment budget over the entire South China Sea (SCS), using ^{210}Pb measurements from 409 sediment cores, AMS ^{14}C data from 112 gravity cores, and 33 sediment trap observations. The results show that Holocene sedimentation rates in the SCS exhibit regional variability. Specifically, under depositional conditions unaffected by turbidity currents and other anomalous processes, the mean sedimentation rate (SR) exceeds 25 cm/ka on upper continental slopes (<1000m), approximates 19 cm/ka in deeper water areas ($\geq 1000\text{m}$) along the lower continental slope slopes, and approximately 5-10 cm/ka near the deep-sea basin (>3000m) (Sheng et al., 2024). Based on these findings by Sheng et al., (2024), the estimated age of the uppermost 1cm of sediment used in this study ranges from less than approximately 40 to 130 years. During the collection of surface sediments used in this study, the retrieved box-core samples were carefully inspected to avoid sampling at any stations exhibiting apparent bioturbation or disturbance from turbidity currents.

In the revised manuscript, we have included information regarding the timespan of the uppermost 1cm sediment at the end of the section "2.1 Sample material", as follows: No discernible evidence of bioturbation or disturbance from turbidity currents was observed in the sediment samples at these stations. Based on regional variability in Holocene sedimentation rates in the SCS (Sheng et al., 2024), the estimated age of the uppermost 1cm of sediment used in this study ranges from <40 to 133 years.

References:

- Sheng, J., Qiao, S., Shi, X., Liu, J., Liu, Y., Liu, S., Wang, K., Mohamed, C., Khokiattiwong, S., Kornkanitnan, N.: Modern sedimentation and sediment budget in the South China Sea and their comparisons with the eastern China seas, *Marine Geology*, 475, 107348, <https://doi.org/10.1016/j.margeo.2024.107348>, 2024.

Section 4.2: The radiolarian assemblages are similar between water column and core-top, so the inference is that 'dissolution is expected to have limited impacts on these radiolarian shells' (L209) – but is this necessarily true? Is it possible weakly silicified parts of the tests are dissolving? This would be interesting to know, as it has different implications for *how* the $\delta^{30}\text{Si}$ is preserved: if no dissolution occurs, then there's no real potential for altering the isotopic signature (which therefore means the conclusions here are not transferable to other settings where more dissolution does occur). But if dissolution does occur and the $\delta^{30}\text{Si}$

remains the same, then either a) different parts of the tests have the same $\delta^{30}\text{Si}$ or b) a coincidental balance of heavier and lighter parts dissolved. To begin to address this, it would be good to see an independent constraint on the (radiolarian) biogenic silica preservation efficiency, either from the literature, from a comparison of export vs. sediment-trap/burial fluxes, or even a theoretical predicted efficiency based on sinking speeds, water column depth, and dissolution kinetics. Finally, It would be good to see an attempt to engage with what may happen with progressive dissolution in the upper centimeters of the sediment – if there is preferential dissolution of some species here, might that introduce a bias (an apparent fractionation) to the bulk-assemblage $\delta^{30}\text{Si}$ data?

Response: Thanks for reviewer's insightful comments. Yes, the observation that the radiolarian assemblages are similar between water column and surface sediments does not preclude the possibility that weakly silicified parts of the radiolarian tests have been dissolved. Although we have realized that some degree of dissolution is inevitable during the transport of radiolarians from the water column to the sediment, there is currently a paucity of established proxies for identifying weakly silicified parts of the radiolarian tests, or for tracing subtle dissolution effects. Moreover, no quantitative data are available in the literature regarding the preservation efficiency or burial fluxes of polycystine radiolarians from the South China Sea (SCS). Existing data of biogenic silica preservation efficiency (~1-3%) are not directly applicable to our study, as diatoms, a primary component of biogenic silica, exhibit a significantly higher degree of dissolution than polycystine radiolarians (e.g. Tréguer et al., 1995; Ragueneau et al., 2000). Consequently, obtaining effective preservation efficiency data suitable for assessing the dissolution of polycystine radiolarians is a significant challenge.

As shell fracture of microfossils preserved within sediments is commonly attributed to partial dissolution (e.g. Murray and Alve, 1999; Ryves et al., 2001), the proportion of fractured radiolarian shells may be indicative of the potential for radiolarian shell dissolution during sinking. Moreover, the preservation of delicate radiolarian skeletal structures, as assessed by SEM images, may also serve to determine whether the radiolarian shells have undergone significant dissolution. Therefore, in the revised manuscript, we have added the content regarding the assessment of radiolarian shell preservation in both the water column and sediments. This allows us to examine whether significant dissolution of radiolarian shells occurs during sinking within the water column. We have therefore made following essential revisions to the manuscript:

In the revised manuscript, the title of section 2.2 “Radiolarian composition analysis” has been revised to “Radiolarian composition and preservation analysis”, and we have included a second paragraph regarding assessing the preservation of radiolarian shells: “To assess the preservation of radiolarian shells both within the water column and seabed sediments, the proportion of fractured radiolarian shells was quantified in each studied sample under a Nikon optical microscope at x100 magnification. Further scanning electron microscopy (SEM) was employed to examine the potential for dissolution-induced alteration of the radiolarian skeleton.”.

Following the first paragraph of section 2.3, we have included the result of radiolarian preservation analysis and a new Figure 4 (see below): “The proportion of fractured shells was low in the studied samples (Figure 4), generally ranging from 2 to 5% (mean = 3%) in plankton samples and from 3 to 9% (mean = 6%) in surface sediments. SEM images reveal a typical morphology of the pores on the radiolarian shell (Figure 4E), along with a high degree of integrity in the delicate skeletal structures (Figure 4F). These observations suggest good preservation of

radiolarian shells, with no significant dissolution evident in either the water column or the sediments.”

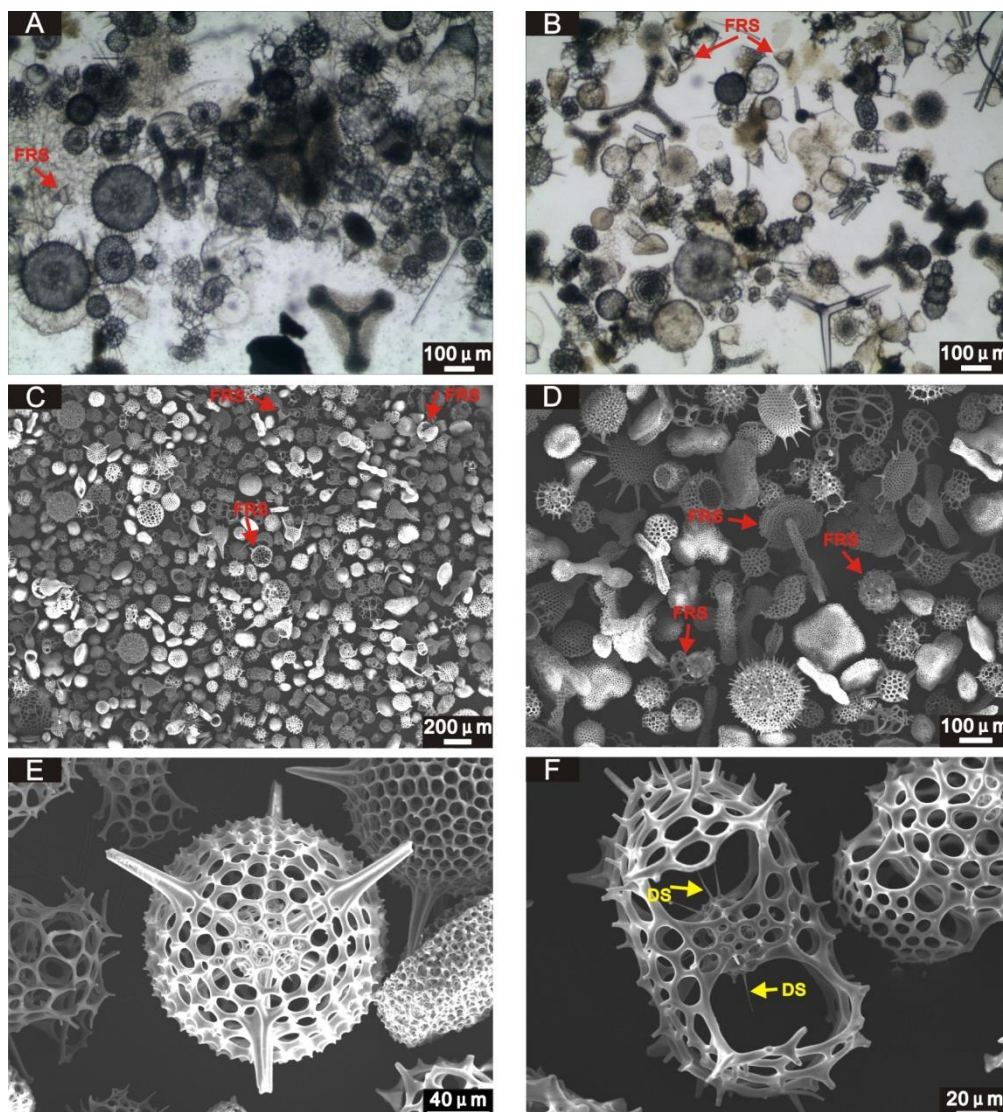


Figure 4 Observations of radiolarian shell preservation at station 12 (water depth: 3497 m) in the plankton sample (100-300 m) (A and C) and surface sediments (B, D, E, and F) using optical and the scanning electron microscope. FRS=Fractured radiolarian shells; DS=Delicate skeletons

The first paragraph of Section “4.2 Transfer of radiolarian $\delta^{30}\text{Si}$ signatures into the sediment record” has been revised to “At each sampling station, $\delta^{30}\text{Si}_{\text{rad}}$ compositions (mean = 1.73‰) in the surface sediment closely resemble those (mean = 1.74‰) in the overlying water column evidenced by the paired t-test ($p=0.75$), indicating a faithful transfer of the $\delta^{30}\text{Si}$ signal incorporated into radiolarian skeletons from the water column to sediments. This suggests that dissolution has a minimal impact on the $\delta^{30}\text{Si}_{\text{rad}}$ signatures as radiolarian shells sink through the water column and become incorporated into the sediment record. One of two possibilities may account for this observation: 1) the radiolarian shells may not have undergone substantial dissolution during sinking; 2) the radiolarian shells have experienced substantial dissolution, but this process may not significantly alter their isotope composition. Considering minor differences in the mean proportion of fractured radiolarian shells between the plankton sample (~3%) and surface sediments (6%), the well-preserved state of radiolarian shells (Figure 4), and the

approximate correspondence between prominent radiolarian species identified in plankton samples and those present in surface sediments at each sampling station (as detailed in section 4.1), we propose that radiolarian shells have not experienced substantial dissolution or remineralisation during their transfer from the water column to the sediments.”

At the beginning of the second paragraph of section “4.2 Transfer of radiolarian $\delta^{30}\text{Si}$ signatures into the sediment record”, we have added “The susceptibility of radiolarian shells to dissolution varies considerably with different taxonomic groups due to the differences in the chemical constituent of their skeletons.”

References:

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