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# Variability in the natural Uranium Isotopic composition of the Arctic Ocean

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**Abstract.** The Arctic Ocean plays a significant role in the Global Thermohaline Circulation and is strongly sensitive to ongoing changes. Due to climate change, rising riverine freshwater inputs into the Arctic Ocean and rising instability of the Arctic Ice covering have been observed. The resulting changes in the ocean mixing and circulation can give valuable insights on future impacts. Here, we study whether over the past twenty years additional freshwater input is traceable in the isotopic composition of uranium, using the natural uranium $^{234}U/^{238}U$ isotope composition. Water samples taken at six stations across the Arctic Ocean are analyzed using high-precision MC-ICP-MS. For each station, a depth profile of $^{234}U/^{238}U$ ratios, as ‰ deviation from secular equilibrium ($\delta^{234}U$), is obtained and compared to a previous study by Andersen et al. (2007), which included similar stations taken 20 years prior. Surface water $\delta^{234}U$ reveals overall 1-2‰ lower values as compared to 20 years ago, thus increased freshwater contributions, which are rich in $^{234}U$, are not traced within the upper 30m of the Arctic Ocean. Instead a stronger downward mixing over the past 20 years is detected. Below 300m $\delta^{234}U$ is homogeneous within measurement uncertainty and yields a $\delta^{234}U$ value of 146.94‰ ± 0.09‰ (2σ mean, N=88, HU1-equilibrium scale), which is identical to the global ocean mean value of 146.75 ± 0.28 by Kipp et al. (2022) and confirms that the interior Arctic U isotope composition is in steady state and set by the long-term global ocean circulation. In addition, we determined the Arctic Ocean $^{238}U/^{235}U$ ratio obtaining a mean $\delta^{238}U$ value of -0.348‰ ± 0.046‰ (N=68) that is also identical within uncertainty to the global Ocean mean $\delta^{238}U$ value of −0.379‰ ± 0.023‰ (Kipp et al., 2022) indicating absence of local redox conditions on the Arctic Ocean U cycle.

## 1 Introduction

The Arctic Ocean has a special role in earth's climate. Even though it is the world's smallest ocean, its high global relevance cannot be underestimated due to its role in the Global Thermohaline Circulation. It receives water from the Atlantic and Pacific Oceans and distributes water that has been impacted by freezing processes and riverine inputs into the Nordic Sea where deep convection takes place (Rudels and Carmack, 2022). Unique in its high amount of riverine inputs, the Arctic Ocean receives 10% of global river discharge from Siberian and North-American rivers (Karcher and Oberhuber, 2002, Andersen et al., 2007). Attention on the Arctic Ocean has recently grown as the ongoing global climate warming has caused it to actively change (Lee

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et al., 2023). These changes include the rising instability of the Arctic sea ice cover (Nghiem et al., 2006), increased riverine input into the Arctic Ocean (Peterson et al., 2002, Rood et al., 2017) and changing carbon fluxes from the thawing of permafrost regions (Guo et al. 2025). In addition, the geochemical cycle of trace elements may be affected by such climate changes as new land surfaces are exposed to chemical weathering, which previously were subject to physical erosion by glaciers (Krickov et al. 2020). Moreover, the changing terrestrial and marine carbon cycle may impact the distribution of chemical constituents and the role of Estuaries (Cai et al. 2021).

A high percentage of the Arctic Ocean is covered by ice, below which the water is strongly stratified. The cold Arctic surface layer is separated from the warm and salty water of Atlantic origin in 200 m to 1000 m depth by a strong halocline (Karcher and Oberhuber, 2002). The circulation of surface waters and the sea ice are dominated by the wind and its resulting Ekman transport, which causes vertical mixing of water (Rudels and Carmack, 2022, Price et al., 1987). This results in a clockwise circulation in the Ameriasian Basin centered around the Beaufort Gyre and a counterclockwise circulation in the Nansen Basin and the western Siberian shelf as shown in Fig. 1. These two systems drive the Transpolar Drift (TPD) at their boundary, which runs from the Siberian shelves towards Fram Strait (Rudels and Carmack, 2022, Karcher and Oberhuber, 2002). Below the surface layer the circulation patterns are topographic flows which show clear basin scale re-circulation patterns in accordance with the Gakkel Ridge separating the Atlantic waters from the Central Arctic Ocean (Rudels and Carmack 2022). Freshwater is added from melt-water, rivers that run into the Siberian and Canadian Shelves and by the Norwegian Coastal Current that is flowing into the Barents Sea (Karcher and Oberhuber, 2002). The river inflow is dominated by four large rivers: Yenisey, Lena and Ob' on the Siberian side and Mackenzie on the North-American side.

While the U concentration of river discharge is sensitively modulated within the redox-condition in estuaries, the $^{234}U/^{238}U$ ratio, commonly expressed as Eq. (1), reflects water-rock interactions in the drainage basin.

$$\delta^{234}\mathrm{U} = \left[ \frac{\frac{\delta^{234}\mathrm{U}}{\delta^{238}\mathrm{U}_{sample}}}{\frac{\delta^{234}\mathrm{U}}{\delta^{238}\mathrm{U}_{standard}}} - 1 \right] \times 1000 \quad (1)$$

Excess $^{234}U$ in freshwater originates from the alpha-recoil process that favors the release of $^{234}U$ over $^{238}U$ in continental rock-water interactions (DePaolo et al., 2016). In 2007, Andersen et al. demonstrated the possibility of tracing the freshwater provenance to the Arctic Ocean Basins, even for individual rivers, via the use of the natural $\delta^{234}U$ ratio. Through the measurement of the U isotopic composition of 28 seawater samples it was found that freshwater influenced surface waters carried elevated $\delta^{234}U$ ratios compared with the underlying deeper Arctic waters. Samples from the Canada Basin revealed a significant freshwater component and provide evidence that the Mackenzie River loses ~65% of its U in the Mackenzie shelf/estuary zone before entering the basin. This is in contrast to samples from the Makarov Basin, which provide evidence

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that all of the freshwater input is derived from the Yenisey River alone, despite the proximity of the Lena and Ob Rivers to the study sites. Andersen et al. (2007) suggested that the differing behavior of U between the Mackenzie and Yenisey Rivers is most likely a consequence of the strong binding of U to dissolved organic matter (DOC) or secondary phases in these rivers. The samples studied by Andersen et al. (2007) had been obtained in 2001. For the upper freshwater influenced layer < 100 m excess $^{234}$U varied between 147 and 157.6‰ for the three studied Arctic basins, with largest increases in the Central Arctic Basin, due to the high excess $^{234}$U values of the Yenisey river with 1590 ‰. In contrast, waters below the freshwater influenced upper most surface layer (>100 m depth) yielded excess $^{234}$U values nearly identical to the global ocean mean value with 147.30 ± 0.27‰ (Arctic Ocean) (Andersen et al. 2007) and 146.75±0.28 (Global Ocean, when normalized to HU1 in secular equilibrium for comparability) (Kipp et al. 2022).

The $^{238}$U/$^{235}$U ratio, expressed as Eq. (2), was recently used as a proxy for the areal extent of anoxic seafloor (Tissot et al. 2015).

$$\delta^{238}\mathrm{U} = \left[ \frac{\frac{\delta^{238}\mathrm{U}}{\delta^{235}\mathrm{U}_{sample}}}{\frac{\delta^{238}\mathrm{U}}{\delta^{235}\mathrm{U}_{standard}}} - 1 \right] \times 1000 \quad (2)$$

Variations in this ratio can reflect changes in global or regional redox conditions, as uranium isotopes fractionate during redox transformations, especially under anoxic conditions. Hence, we further want to test here whether the Arctic Oceans $\delta^{238}$U ratio deviates substantially from the global ocean mean value. In a recent study by Kipp et al. (2022) a most precise and comprehensive measurements of global ocean seawater $\delta^{238}$U was conducted. This study provides a benchmark reference point for both methodological rigor and its implications for using uranium isotopes as proxies. An isotopic homogeneous $\delta^{238}$U value of $-0.379 \pm 0.023$‰ was found, and demonstrates only minor regional variations.

We revisit uranium and its natural isotopes in the central Arctic Ocean to study whether recent changes in the bordering continents freshwater balance have left a trace in its cycling and isotope composition. The primary focus is the $\delta^{234}$U isotope ratio. In 2021, just 20 years after the seawater sampling for the first study in the Arctic Ocean achieving high analytical precision for U-isotopes, a similar sampling was conducted as part of the Ventilation and Anthropogenic Carbon in the Arctic Ocean (VACAO) project of the Deutsche Forschungsgemeinschaft (DFG) (Snoeijs-Leijonmalm et al., 2022). The focus of this program was the rate of Arctic in- and outflow and the downward cycling of carbon. Seawater was collected for multiple tracer studies, including radiocarbon, $^{236}$U, CFCs, $SF_6$ and $^{39}$Ar (Snoeijs-Leijonmalm et al., 2022). Consequently, seawater samples have been available from all three Arctic Ocean basins including the surface water, which allowed to study the change of the U isotopic composition, including $\delta^{234}$U and for the first time $\delta^{238}$U. Therefore, the VACAO project provided the

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opportunity to revisit the observations by Andersen et al. 2007, and thus to study whether changes of the natural U flux have occurred over the past 20 years of global warming and sea ice retreat.

## 2. Methods

The VACAO research cruise was part of the SAS-Oden 2021 expedition into the Arctic Ocean, which is part of the international "Synoptic Arctic Survey" (SAS), The sampling sites analyzed in this study are shown in Fig. 1.

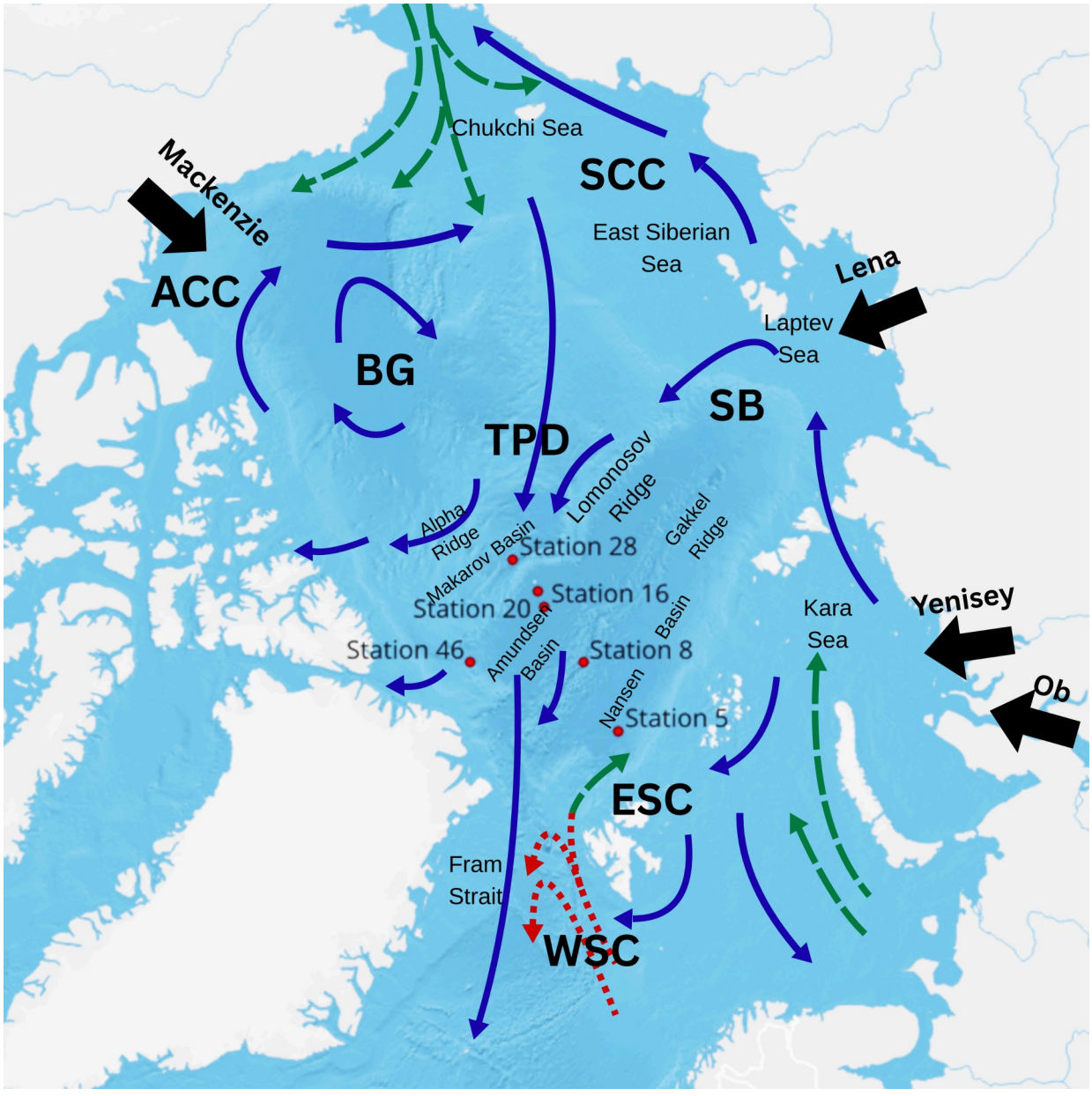

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**Figure 1: Map detailing the physiography and surface circulation of the Arctic Ocean as described in Rudels and Carmack (2022) and the location of the sampled stations. The Transpolar Drift (TPD), the Beauford Gyre (BG), the Siberian branch of the Transpolar Drift (SB) and the Alaskan Coastal Current (ACC) are cold Arctic currents indicated by solid blue arrows. The West Spitsbergen Current (WSC) is a warm Atlantic current indicated by dotted red arrows. The dashed green arrows indicate low salinity transformed currents like the East Spitsbergen Current (ESC).**

The samples were originally taken for $^{14}C$ analysis conducted according to (Bryant et al., 2013): Aluminum foil bags for gas sampling were modified in a way that water can be stored. Before sampling, the bags were flushed with pure Nitrogen before they were evacuated. Each bag was filled with about 500 ml of seawater using a 45 µm filter to avoid sampling of particulate material. The bags were frozen at -20 °C to prevent biological activity. Once in Heidelberg, the bags were kept in a freezer at -28°C until preparation of $^{14}C$ measurements, of which the data is available at https://doi.pangaea.de/10.1594/PANGAEA.994605. For $^{14}C$ measurements 50 ml of water was removed with a syringe from the bag and inserted in evacuated glass bottles, which were sealed with septum butyl stoppers and any remaining air was flushed with nitrogen. The remaining water from these analyses was used here. For numerous stations, additional aliquots (B-samples) were stored in this state and used for U-isotope analysis. Otherwise, samples labeled here C and D-samples, were taken from the original frozen samples. A full list of the samples is given in Table 1. Eleven samples were taken along the depth profiles of each of stations 5, 8, 16, 20, 28 and 46, resulting in a total of 65 samples. The temperature and the salinity, given here in practical salinity units, of the samples were determined during the cruise using a CTD system, from which the density and depth were inferred.

The sample preparation follows a modified method from the U/Th ion exchange purification protocol for corals of the Institute of Environmental Physics, Heidelberg (Kerber et al. 2023). For each sample 50 ml of seawater were used, except for two samples with less available water marked accordingly in Table 1. Before any further preparation, all samples were weighted. All samples were then spiked with an in-house triple spike (TriSpike).

Seawater samples were mixed with 8 ml of 7N $HNO_3$ and evaporated at 65 - 75 °C until dryness. The dry sea salt samples were re-dissolved in 20 ml 7N $HNO_3$. Uranium was extracted from the samples using 500 µl UTEVA resin packed in Triskam columns. For the extraction, the columns were cleaned and preconditioned with Milli-Q, 3N HCL, and 7N $HNO_3$. The dissolved samples were completely loaded onto the columns, after which the columns were rinsed with 3 ml of 7N $HNO_3$ to remove the salt matrix. Next, uranium was eluted from the columns with 5 ml of 1N HCl and 1-2 ml of Milli-Q. The uranium fractions were then once more evaporated at 65 - 75 °C and re-dissolved in 1 ml 7N HNO3 and the column chemistry was repeated using the same columns. For the second columns, the samples were eluted with 1 ml 3N HCl and 5 ml of 1N HCl and dried down in the same manner as before. In a final step, the samples are dissolved in 1.5 ml 1% $HNO_3$+0.05% HF. For the isotopic analysis, the samples are centrifuged to remove potential residue particles from the ion exchange resin and transferred into corning tubes for subsequent isotope analysis. The extraction chemistry does not cause measurable fractionation in $\delta^{234}U$ within the reached precision of 0.2-0.5‰ (Appendix A).

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Isotope measurements were conducted on a multiple collector inductively coupled plasma mass spectrometer (MC-ICP-MS, ThermoFisher NeptunePlus) at the Institute of Environmental Physics, Heidelberg. MC-ICP-MS applications are the state of the art for $^{234}U/^{238}U$ measurements, with recent advances reaching reproducibility of ±0.08‰ (Hu et al., 2025). The samples were introduced into the spectrometer using, an Aridus II desolvating system. For the highly abundant $^{238}U$, a $10^{10}$ Ω resistor is used, allowing signals up to 500 V. The $10^{11}$ Ω resistors, used for signals up to 50 V, are applied for $^{236}U$ and $^{235}U$. For the low abundant isotopes $^{234}U$ and $^{233}U$, $10^{13}$ Ω resistors yield the best results. Since a second $10^{13}$ Ω resistor was not available for all measurements, a $10^{11}$ Ω resistor was applied on $^{233}U$ for a significant part of the measurements. To reliably measure the already rare isotope $^{234}U$ in blanks, a SEM multiplier was used.

The measurement procedure involves Standard-Sample-Standard bracketing in which a HU-1 standard solution is measured under the same settings as the sample solutions. Following each sample and standard, the machine is cleaned in a diluted HF-wash solution, consisting of 1% $HNO_3$ and 0.05% HF, after which a process blank from the same solution is measured for about 70 seconds. At the start and end of each sequence, a spiked CRM-112A solution is measured. All Samples and Standards are measured for 60 cycles resulting in a measurement time of 6 min. The Blanks are measured for 10 cycles (70 sec) and the cleaning in the HF-wash lasts for 30 cycles (2 min). In the beginning and end of each sequence, half-masses are measured on a unspiked CRM-112A standard for 5 cycles, lasting 2 min, to quantify peak-tailing and hydrite-formation. Aside from blank correction, bracketing and tailing and hydrite correction, the obtained data is also 2σ outlier tested over the different scans, corrected for spike impurities and for the gain offset between each cup relative to the center cup C. Mass fractionation correction is conducted according to the exponential law (Albarède et al., 2015) using the measured $^{238}U/^{235}U$ to correct the $^{234}U/^{238}U$ ratio since $^{238}U/^{235}U$ can be measured with significantly higher accuracy than $^{236}U/^{233}U$ in the current setup. For $\delta^{238}U$ the measured $^{236}U/^{233}U$ ratio is used. Given the minor deviation of the natural $^{238}U/^{235}U$ ratio used here as reference for $^{234}U$ mass bias corrections and the final $^{235}U/^{238}U$ isotope ratio of the sample, a final correction of -0.5‰ is applied to the mean offset in the seawater $^{235}U/^{238}U$ ratio of -0.379. Note, we here assumed HU-1 to be in secular equilibrium, which causes our lab internal CRM-112A measurements (-36.92‰ ± 0.29‰, Greve et al. 2025) to fall significantly above the reference value of -38.5‰. To normalize the results to the reference value of CRM-112A an offset of -1.20‰ to our measurements needs to be applied (Chen et al. 2013). Most recently, Hu et al. (2025) has confirmed the feasibility of routine sub-epsilon precision measurements of the $^{234}U/^{238}U$ ratio of secular equilibrium material previously demonstrated by Tissot et al. (2015). However, for our study such ultra-high precision measurements are solely feasible for multiple repeated analysis such as the interior Arctic Ocean seawater samples. In all other cases difference between samples are by far larger in the order of several ‰. From repeat extraction and measurement of bulk seawater samples and VACAO samples with spare material, we estimate a reproducibility of an individual measurement to 0.5‰ for $\delta^{234}U$ and 0.15‰ in $\delta^{238}U$ (Appendix A, Appendix C). When repeating seawater measurements for more than 20 times a precision of 0.09 ‰ for $\delta^{234}U$ can be achieved.

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## 3 Results

In this study 61 arctic seawater samples were processed and multiple samples were processed more than once to estimate the accuracy of $\delta^{238}U$ and $\delta^{234}U$ measurements. In total, 127 measurements were conducted for $^{234}U/^{238}U$ and 95 for $^{238}U/^{235}U$. The different overall numbers of analysis stems from the fact that we started processing for $^{238}U/^{235}U$ following the first about 30 analysis of $^{234}U/^{238}U$. The individual repeated measurements at a single depth below 300 m were used to determine the average depth value for $\delta^{234}U$ (N=88) and $\delta^{238}U$ (N=68) and its external variability. The full data of the isotopic measurements can be found in Table 1 and depth averaged values are plotted in Fig. 2.

The dataset includes samples which have been diluted for repeat measurements and have therefore a lower intensity during the measurements than the samples prepared according to protocol. This has been tested (Appendix B) and has been found to be a reasonable method for repeat measurements. Samples that have been diluted are marked in Table 1. In the same manner, for some samples less than 50 ml of sample material were available, which is also noted in Table 1.

| Name | Depth [db] | $\delta^{234}U$ [‰] | $\delta^{234}U$ [‰]* | $\delta^{238}U$ [‰] | T [°C] | Salinity | Note |
|---|---|---|---|---|---|---|---|
| Station 5 | | | | | | | |
| VACAO1 5-4B | 3989 | 147.27 ± 0.23 | 146.07 ± 0.23 | / | -0.66 | 34.941 | |
| VACAO1 5-4D | 3989 | 147.41 ± 0.27 | 146.21 ± 0.27 | -0.403 ± 0.017 | | | |
| VACAO1 5-4D | 3989 | 146.24 ± 0.26 | 145.04 ± 0.26 | -0.384 ± 0.016 | | | |
| VACAO3 5-3 B | 3000 | 146.94 ± 0.30 | 145.74 ± 0.30 | -0.209 ± 0.020 | -0.75 | 34.935 | |
| VACAO3 5-3C | 3000 | 146.79 ± 0.31 | 145.59 ± 0.31 | -0.345 ± 0.016 | | | |
| VACAO3 5-3C | 3000 | 146.47 ± 0.29 | 145.27 ± 0.29 | -0.489 ± 0.023 | | | |
| VACAO5 5-4B | 2500 | 146.90 ± 0.28 | 145.70 ± 0.28 | / | -0.76 | 34.928 | |
| VACAO5 5-4D | 2500 | 146.60 ± 0.23 | 145.40 ± 0.23 | -0.451 ± 0.020 | | | |
| VACAO7 5-4B | 2000 | 147.91 ± 0.25 | 146.71 ± 0.25 | / | -0.73 | 34.921 | |
| VACAO7 5-4D | 2000 | 146.57 ± 0.32 | 145.37 ± 0.32 | -0.170 ± 0.017 | | | |
| VACAO7 5-4D | 2000 | 146.94 ± 0.27 | 145.74 ± 0.27 | -0.360 ± 0.024 | | | |
| VACAO9 5-3B | 1500 | 146.79 ± 0.42 | 145.59 ± 0.42 | -0.174 ± 0.022 | -0.59 | 34.915 | |
| VACAO9 5-3D | 1500 | 146.49 ± 0.31 | 145.29 ± 0.31 | -0.283 ± 0.013 | | | |
| VACAO11 5-4B | 1000 | 147.65 ± 0.24 | 146.45 ± 0.24 | / | -0.20 | 34.905 | |
| VACAO11 5-4D | 1000 | 147.19 ± 0.34 | 145.99 ± 0.34 | -0.455 ± 0.018 | | | |
| VACAO13 5-3B | 700 | 147.32 ± 0.25 | 146.12 ± 0.25 | -0.183 ± 0.019 | 0.42 | 34.903 | |

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| | | | | | | | |
|---|---|---|---|---|---|---|---|
| VACAO15 5-4B | 500 | 147.35 ± 0.34 | 146.15 ± 0.34 | / | 1.18 | 34.913 | |
| VACAO15 5-4B | 500 | 147.25 ± 0.32 | 146.05 ± 0.32 | / | | | |
| VACAO17 5-4B | 300. 376 | 147.18 ± 0.31 | 145.98 ± 0.31 | / | 1.60 | 34.879 | |
| VACAO19 5-3B | 100 | 147.66 ± 0.26 | 146.46 ± 0.26 | -0.201 ± 0.015 | -0.64 | 34.517 | |
| VACAO19 5-3C | 100 | 148.13 ± 0.27 | 146.93 ± 0.27 | -0.420 ± 020 | | | |
| VACAO19 5-3C | 100 | 147.49 ± 0.26 | 146.29 ± 0.26 | -0.394 ± 0.018 | | | |
| VACAO21 5-4B | 28.8 | 148.66 ± 0.30 | 147.46 ± 0.30 | / | -1.54 | 34.027 | |
| VACAO21 5-4B | 28.8 | 147.54 ± 0.23 | 146.34 ± 0.23 | -0.258 ± 0.020 | | | |
| VACAO21 5-4B | 28.8 | 147.36 ± 0.39 | 146.16 ± 0.39 | -0.377 ± 0.020 | | | [1] |
| Station 8 | | | | | | | |
| VACAO1 8-9B | 2940 | 145.93 ± 0.35 | 144.73 ± 0.35 | / | -0.74 | 34.932 | |
| VACAO1 8-9D | 2940 | 146.96 ± 0.27 | 145.76 ± 0.27 | -0.236 ± 0.015 | | | [2] |
| VACAO3 8-9B | 2891 | 147.72 ± 0.26 | 146.52 ± 0.26 | / | -0.74 | 34.931 | |
| VACAO3 8-9D | 2891 | 146.99 ± 0.29 | 145.79 ± 0.29 | -0.367 ± 0.015 | | | |
| VACAO7 8-9C | 2001 | 146.51 ± 0.35 | 145.31 ± 0.35 | -0.416 ± 0.021 | -0.66 | 34.920 | |
| VACAO7 8-9C | 2001 | 146.65 ± 0.29 | 145.45 ± 0.29 | -0.406 ± 0.017 | | | |
| VACAO7 8-9D | 2001 | 146.60 ± 0.25 | 145.40 ± 0.25 | / | | | |
| VACAO9 8-9B | 1501 | 146.94 ± 0.26 | 145.74 ± 0.26 | / | -0.50 | 34.914 | |
| VACAO9 8-9B | 1501 | 146.44 ± 0.35 | 145.24 ± 0.35 | -0.342 ± 0.027 | | | |
| VACAO11 8-9B | 1001 | 146.78 ± 0.31 | 145.58 ± 0.31 | -0.460 ± 0.016 | -0.17 | 34.901 | |
| VACAO11 8-9B | 1001 | 146.48 ± 0.25 | 145.28 ± 0.25 | -0.353 ± 0.022 | | | [1] |
| VACAO13 8-9D | 699.5 | 146.63 ± 0.33 | 145.43 ± 0.33 | -0.350 ± 0.016 | 0.20 | 34.893 | |
| VACAO15 8-9D | 500.5 | 146.78 ± 0.29 | 145.58 ± 0.29 | -0.306 ± 0.016 | 0.65 | 34.887 | |
| VACAO17 8-9B | 300 | 146.88 ± 0.30 | 145.68 ± 0.30 | / | 1.09 | 34.864 | |
| VACAO17 8-9B | 300 | 145.78 ± 0.27 | 144.58 ± 0.27 | -0.344 ± 0.035 | | | [1] |
| VACAO19 8-9B | 100 | 147.63 ± 0.27 | 146.43 ± 0.27 | / | -0.89 | 34.391 | |
| VACAO19 8-9D | 100 | 146.57 ± 0.28 | 145.37 ± 0.28 | -0.539 ± 0.057 | | | |
| VACAO21 8-9D | 30 | 148.85 ± 0.31 | 147.65 ± 0.31 | -0.451 ± 0.024 | -1.67 | 33.472 | |
| Station 16 | | | | | | | |
| VACAO1 16-6B | 4428 | 146.32 ± 0.26 | 145.12 ± 0.26 | / | -0.62 | 34.942 | |
| VACAO1 16-6D | 4428 | 146.84 ± 0.38 | 145.64 ± 0.38 | -0.246 ± 0.019 | | | |

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| | | | | | | | |
|---|---|---|---|---|---|---|---|
| VACAO3 16-6B | 4000 | 147.19 ± 0.32 | 145.99 ± 0.32 | / | -0.67 | 34.942 | |
| VACAO5 16-6B | 3500 | 147.26 ± 0.36 | 146.06 ± 0.36 | / | -0.71 | 34.936 | |
| VACAO5 16-6D | 3500 | 146.22 ± 0.35 | 145.02 ± 0.35 | -0.015 ± 0.022 | | | |
| VACAO5 16-6D | 3500 | 147.10 ± 0.32 | 145.90 ± 0.32 | 0.066 ± 0.044 | | | |
| VACAO7 16-6B | 3000 | 146.80 ± 0.36 | 145.60 ± 0.36 | / | -0.73 | 34.930 | |
| VACAO7 16-6B | 3000 | 146.78 ± 0.20 | 145.58 ± 0.20 | -0.467 ± 0.022 | | | [1] |
| VACAO7 16-6D | 3000 | 146.87 ± 0.25 | 145.67 ± 0.25 | -0.338 ± 0.018 | | | |
| VACAO7 16-6D | 3000 | 146.97 ± 0.28 | 145.77 ± 0.28 | -0.384 ± 0.019 | | | |
| VACAO9 16-6B | 2000 | 146.90 ± 0.38 | 145.70 ± 0.38 | / | -0.61 | 34.921 | |
| VACAO9 16-6D | 2000 | 146.80 ± 0.39 | 145.60 ± 0.39 | -0.325 ± 0.019 | | | |
| VACAO9 16-6D | 2000 | 146.64 ± 0.27 | 145.44 ± 0.27 | -0.352 ± 0.015 | | | |
| VACAO11 16-6B | 1000 | 147.13 ± 0.31 | 145.93 ± 0.31 | / | -0.06 | 34.903 | |
| VACAO11 16-6D | 1000 | 146.82 ± 0.32 | 145.62 ± 0.32 | 0.162 ± 0.025 | | | |
| VACAO11 16-6D | 1000 | 146.23 ± 0.28 | 145.03 ± 0.28 | -0.015 ± 0.032 | | | |
| VACAO14 16-6B | 700 | 147.08 ± 0.35 | 145.88 ± 0.35 | / | 0.34 | 34.891 | |
| VACAO15 16-6B | 500 | 147.24 ± 0.30 | 146.04 ± 0.30 | / | 0.92 | 34.896 | |
| VACAO15 16-6D | 500 | 147.87 ± 0.33 | 146.67 ± 0.33 | -0.360 ± 0.019 | | | |
| VACAO17 16-6B | 300 | 147.17 ± 0.24 | 145.97 ± 0.24 | / | 1.23 | 34.874 | |
| VACAO17 16-6D | 300 | 147.11 ± 0.32 | 145.91 ± 0.32 | / | | | |
| VACAO 19 16-6B | 100 | 148.72 ± 0.30 | 147.52 ± 0.30 | / | -1.50 | 34.241 | |
| VACAO19 16-6C | 100 | 147.79 ± 0.40 | 146.59 ± 0.40 | -0.263 ± 0.019 | | | |
| VACAO21 16-6B | 29.8 | 149.92 ± 0.23 | 148.72 ± 0.23 | / | -1.68 | 33.173 | |
| VACAO21 16-6D | 29.8 | 149.02 ± 0.30 | 147.82 ± 0.30 | -0.359 ± 0.018 | | | |
| Station 20 | | | | | | | |
| VACAO1 20-3D | 4326 | 147.46 ± 0.31 | 146.26 ± 0.31 | 0.006 ± 0.026 | -0.63 | 34.942 | |
| VACAO3 20-3D | 3998 | 146.69 ± 0.32 | 145.49 ± 0.32 | -0.184 ± 0.019 | -0.67 | 34.942 | |
| VACAO5 20-3D | 3500 | 146.50 ± 0.30 | 145.30 ± 0.30 | 0.063 ± 0.018 | -0.71 | 34.937 | |
| VACAO5 20-3D | 3500 | 146.83 ± 0.22 | 145.63 ± 0.22 | -0.319 ± 0.018 | | | |
| VACAO7 20-3D | 2500 | 146.83 ± 0.25 | 145.63 ± 0.25 | -0.212 ± 0.021 | -0,71 | 34,924 | |
| VACAO9 20-3D | 2000 | 146.92 ± 0.36 | 145.72 ± 0.36 | -0.355 ± 0.021 | -0.56 | 34.927 | |
| VACAO9 20-3D | 2000 | 146.13 ± 0.24 | 144.93 ± 0.24 | -0.383 ± 0.016 | | | |

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| | | | | | | | |
|---|---|---|---|---|---|---|---|
| VACAO11 20-3D | 1500 | 147.05 ± 0.26 | 145.85 ± 0.26 | -0.312 ± 0.018 | -0.41 | 34.916 | |
| VACAO13 20-3C | 1000 | 147.09 ± 0.33 | 145.89 ± 0.33 | -0.394 ± 0.020 | -0.02 | 34.898 | |
| VACAO13 20-3C | 1000 | 147.41 ± 0.50 | 146.21 ± 0.50 | / | | | [1] |
| VACAO13 20-3D | 1000 | 146.94 ± 0.42 | 145.74 ± 0.42 | / | | | |
| VACAO15 20-3D | 700 | 146.97 ± 0.41 | 145.77 ± 0.41 | / | 0.47 | 34.888 | |
| VACAO19 20-3D | 247 | 147.46 ± 0.30 | 146.26 ± 0.30 | -0.427 ± 020 | 1.41 | 34.858 | |
| VACAO21 20-3D | 30 | 150.42 ± 0.05 | 149.22 ± 0.05 | / | -1.70 | 32.741 | [1] |
| Station 28 | | | | | | | |
| VACAO1 28-3B | 4017 | 147.28 ± 0.27 | 146.08 ± 0.27 | / | -0.25 | 34.955 | |
| VACAO1 28-3C | 4017 | 147.18 ± 0.31 | 145.98 ± 0.31 | -0.397 ± 018 | | | |
| VACAO1 28-3C | 4017 | 147.05 ± 0.31 | 145.85 ± 0.31 | -0.303 ± 0.026 | | | |
| VACAO1 28-3D | 4017 | 147.32 ± 0.26 | 146.12 ± 0.26 | / | | | |
| VACAO3 28-3D | 3000 | 146.74 ± 0.31 | 145.54 ± 0.31 | -0.261 ± 020 | -0.35 | 34.955 | |
| VACAO3 28-3D | 3000 | 147.31 ± 0.36 | 146.11 ± 0.36 | -0.183 ± 0.018 | | | |
| VACAO7 28-3B | 2000 | 147.68 ± 0.35 | 146.48 ± 0.35 | / | -0,40 | 34,947 | |
| VACAO7 28-3C | 2000 | 146.23 ± 0.39 | 145.03 ± 0.39 | -0.203 ± 0.019 | | | |
| VACAO7 28-3D | 2000 | 146.94 ± 0.27 | 145.74 ± 0.27 | -0.305 ± 0.019 | | | |
| VACAO9 28-3D | 1500 | 147.36 ± 0.46 | 146.16 ± 0.46 | -0.444 ± 0.024 | -0.44 | 34.924 | |
| VACAO9 28-3D | 1500 | 147.11 ± 0.35 | 145.91 ± 0.35 | -0.476 ± 0.017 | | | |
| VACAO11 28-3B | 1000 | 147.60 ± 0.26 | 146.40 ± 0.26 | / | -0.15 | 34.890 | |
| VACAO11 28-3C | 1000 | 146.12 ± 0.33 | 144.12 ± 0.33 | -0.409 ± 0.029 | | | [1] |
| VACAO11 28-3D | 1000 | 146.74 ± 0.32 | 145.54 ± 0.32 | -0.499 ± 0.032 | | | |
| VACAO13 28-3D | 700 | 146.60 ± 0.42 | 145.40 ± 0.42 | -0.380 ± 0.020 | 0.15 | 34.877 | |
| VACAO13 28-3D | 700 | 147.43 ± 0.36 | 146.23 ± 0.36 | -0.376 ± 0.021 | | | |
| VACAO15 28-3B | 500 | 147.14 ± 0.33 | 145.94 ± 0.33 | / | 0.54 | 34.87 | |
| VACAO15 28-3C | 500 | 146.80 ± 0.28 | 145.60 ± 0.28 | -0.317 ± 0.023 | | | |
| VACAO15 28-3D | 500 | 146.67 ± 0.36 | 144.47 ± 0.36 | -0.595 ± 0.020 | | | |
| VACAO19 28-3B | 100 | 149.19 ± 0.30 | 147.99 ± 0.30 | / | -1.50 | 33.72 | |

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| | | | | | | | |
|---|---|---|---|---|---|---|---|
| VACAO24 28-3D | 30 | 150.54 ± 0.28 | 149.34 ± 0.28 | -0.372 ± 0.021 | -1.56 | 30.291 | |
| Station 46 | | | | | | | |
| VACAO1 46-2C | 3301 | 146.63 ± 0.25 | 145.43 ± 0.25 | -0.347 ± 0.022 | -0.72 | 34.939 | |
| VACAO1 46-2D | 3301 | 146.63 ± 0.30 | 145.43 ± 0.30 | / | | | |
| VACAO3 46-2C | 3000 | 147.29 ± 0.46 | 146.09 ± 0.46 | / | -0.71 | 34.929 | |
| VACAO3 46-2D | 3000 | 146.66 ± 0.38 | 145.46 ± 0.38 | / | | | |
| VACAO5 46-2C | 2500 | 147.10 ± 0.28 | 145.90 ± 0.28 | -0.255 ± 029 | -0.66 | 34.923 | |
| VACAO5 46-2D | 2500 | 147.34 ± 0.31 | 146.14 ± 0.31 | -0.304 ± 0.024 | | | |
| VACAO5 46-2D | 2500 | 146.98 ± 0.38 | 145.78 ± 0.38 | -0.393 ± 0.019 | | | [1] |
| VACAO 7 46-2B | 2000 | 147.69 ± 0.33 | 146.49 ± 0.33 | / | -0.47 | 34.930 | |
| VACAO 11 64-2B | 999 | 147.45 ± 0.29 | 146.25 ± 0.29 | / | -0,03 | 34.891 | [2] |
| VACAO 13 46-2C | 700 | 147.60 ± 0.39 | 146.40 ± 0.39 | / | 0,31 | 34.875 | |
| VACAO 13 46-2D | 700 | 147.11 ± 0.31 | 145.91 ± 0.31 | -0.539 ± 0.013 | | | |
| VACAO 15 46-2B | 500 | 147.47 ± 0.28 | 146.27 ± 0.28 | / | 0,75 | 34,870 | |
| VACAO 15 46-2C | 500 | 146.94 ± 0.28 | 145.84 ± 0.28 | -0.251 ± 0.019 | | | |
| VACAO 15 46-2C | 500 | 146.39 ± 0.33 | 145.19 ± 0.33 | -0.320 ± 0.019 | | | |
| VACAO 15 46-2D | 500 | 147.38 ± 0.37 | 146.18 ± 0.37 | -0.140 ±0.021 | | | |
| VACAO 17 46-2C | 300 | 146.86 ± 0.32 | 145.66 ± 0.32 | -0.232 ± 0.020 | 0.90 | 34.826 | |
| VACAO 17 46-2D | 300 | 147.24 ± 0.29 | 146.04 ± 0.29 | -0.321 ± 0.014 | | | |
| VACAO 19 46-2C | 100 | 148.84 ± 0.30 | 147.64 ± 0.30 | -0.313 ± 0.021 | -1.42 | 33.636 | |
| VACAO 19 46-2C | 100 | 148.53 ± 0.25 | 147.33 ± 0.25 | -0.447 ± 0.018 | | | |

**Tabel 1: Full $\delta^{234}U$ and $\delta^{238}U$ Data of VACAO samples, including measurement reproducibilty. Temperature and Salinity from (Snoeijs-Leijonmalm et al., 2022). [1] Sample diluted with up to 0.3 ml more acid than described in protocol to enable repeat measurement. [2] only 40 ml of sample were available for chemistry. * $\delta^{234}U$ minus 1.2‰ to compare data to CRM-112A scale (Kipp et al. 2022) (see text for details).**

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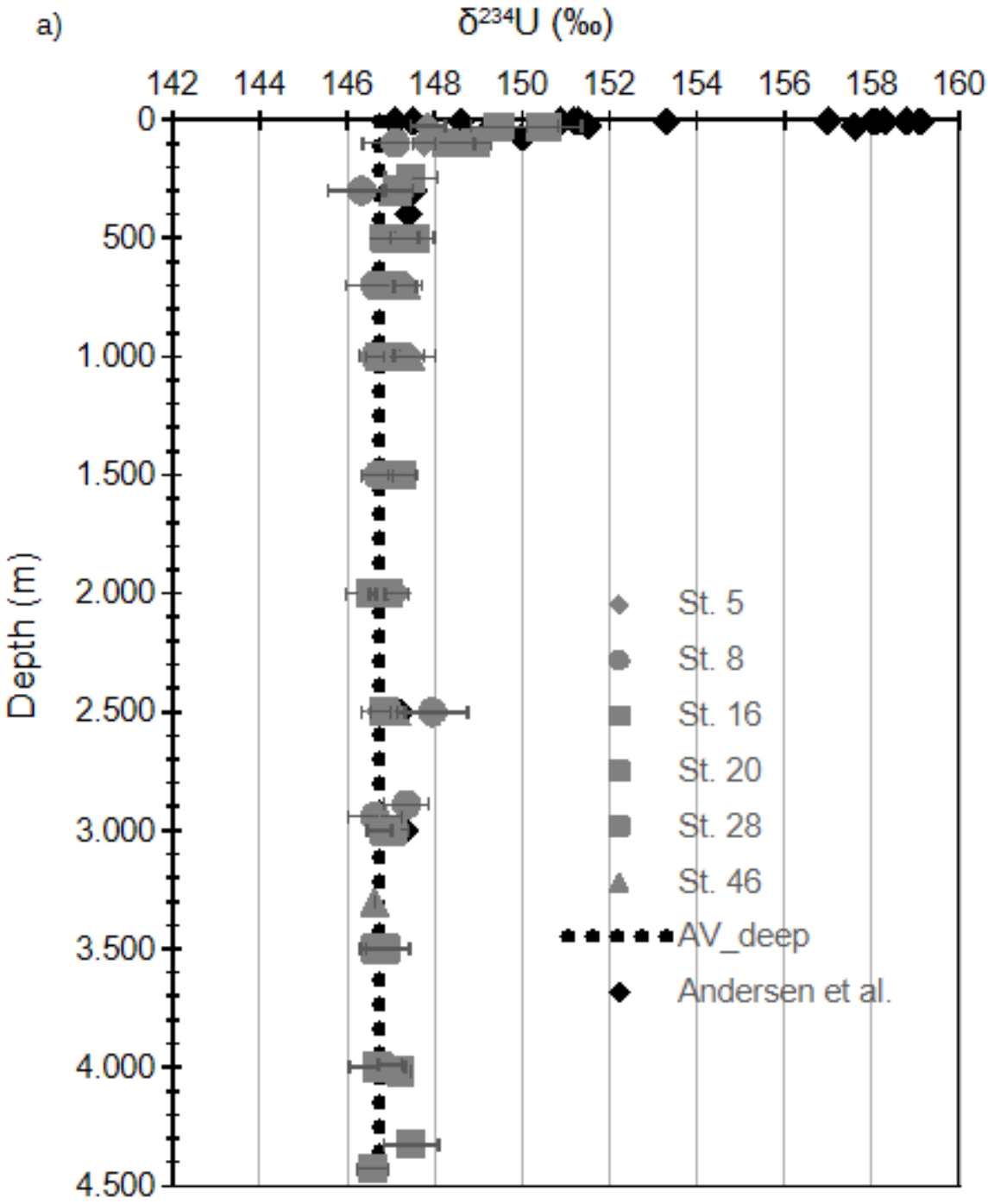

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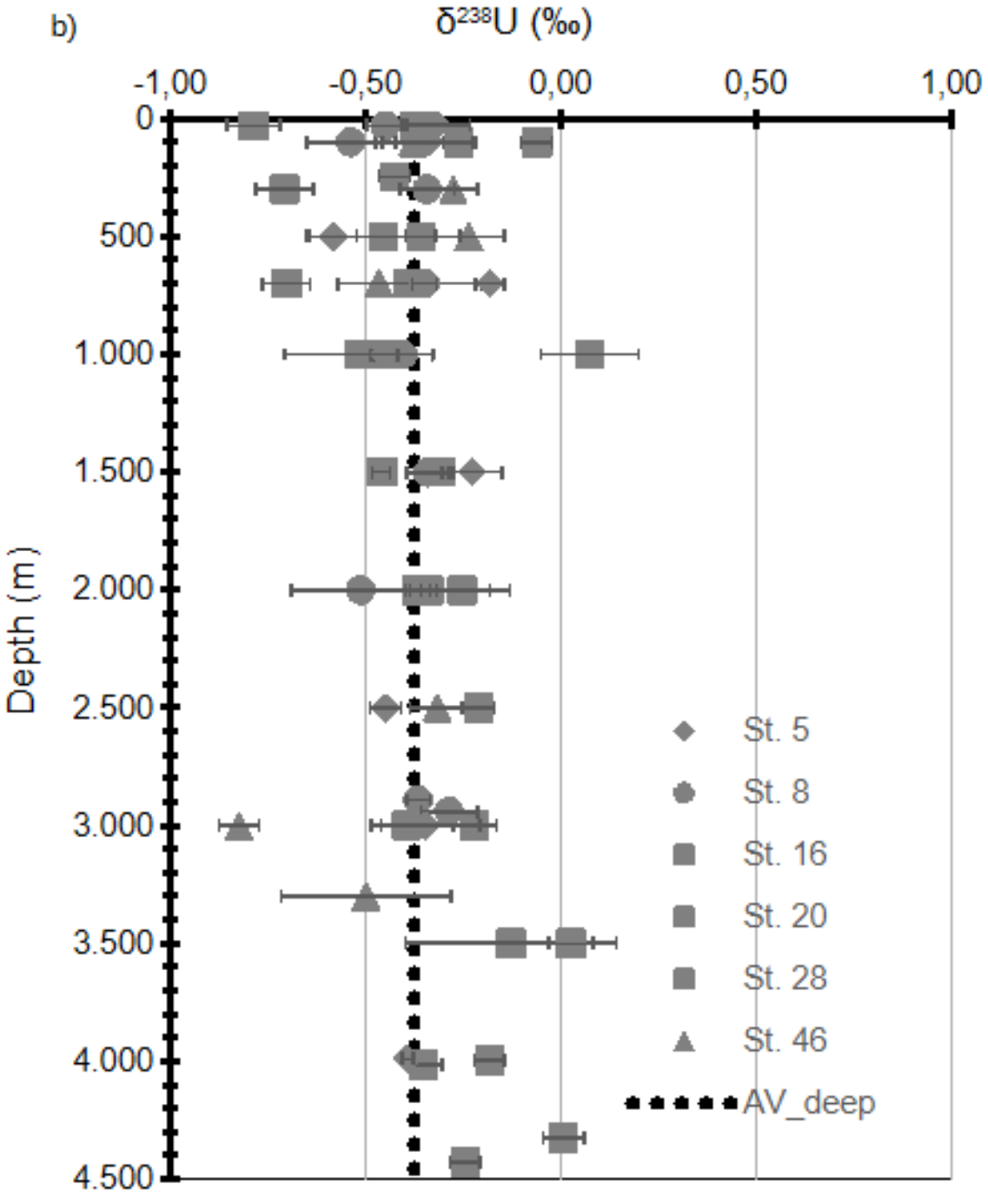

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**Figure 2: Depth average $\delta^{234}U$ and $\delta^{238}U$ values of all VACAO Stations compared to the global Ocean mean values as determined by Kipp et al. (2022). In (a) measurements of $\delta^{234}234U$ conducted in 2001 by Andersen et al. (2007) are further reported. Highest excess $^{234}U$ values are observed in the surface layer were salinities are reduced due to the presence of freshwater. Below the surface layer >300 m all $\delta^{234}U$ values agree with each other yielding an isotope composition identical to the global Ocean mean value (dashed line). For $\delta^{238}U$ (b) all measurements vary around the global Ocean mean value, but partly show large deviations of individual measurements from this mean. No systematic trend is visible for $\delta^{238}U$. The global Ocean mean seawater value of $\delta^{234}U$ is given here normalized to HU-1 in secular equilibrium as 146.75 ± 0.28‰ (Kipp et al., 2022). The global Ocean Mean $\delta^{238}U$ value used here is -0.379‰ ± 0.023‰ (Kipp et al., 2022).**

Compared to the global mean seawater value of 146.75±0.28‰ normalized to HU-1 in secular equilibrium (Kipp et al. 2022) all stations showed distinctly high $\delta^{234}U$ levels at the surface, which steadily lower with depth until matching the seawater value (Fig. 2a). Overall, deviations from the global Ocean mean value were insignificant for $\delta^{234}U$ at depth >300 m (Fig. 2a), while $\delta^{238}U$ values varied wildly around the global Ocean mean value (Fig. 2b). Averaging all $\delta^{234}U$ values below 300 m yielded a $\delta^{234}U$ of 146.94±0.09‰ (2σ mean, N=88), which is identical within uncertainty to the global ocean mean value determined by Kipp et al. (2022) and the one determined by Andersen et al. (2007) for the interior Arctic Ocean of 147.3±0.27 (2σ mean, N=6). For the average $\delta^{238}U$ below 300 m depth we obtained -0.348±0.046‰, which is also within uncertainty identical to the global Ocean mean value determined by Kipp et al. (2022). However, the individual variance of $\delta^{238}U$ largely exceeded the analytical uncertainty, which we interpret as related to fractionation during the chemical U extraction, since we have spiked samples in many cases only after the chemical removal of the salt, which does not affect the $\delta^{234}U$ ratio for which we have used the $^{235}U/^{238}U$ ratio as fractionation reference.

Consequently, the Arctic Ocean below the freshwater influenced surface layers shows an identical excess $^{234}U$ composition compared to the global Ocean, which implies a well-mixed composition through the influx and outflux of Atlantic and Pacific waters at depth, as well as a balanced influx of $^{234}U$ excess with the decay of $^{234}U$.

The surface layer $\delta^{234}U$ determined here reveal a strong freshwater influence at depth of 30 m, both in terms of salinity, which is reduced and in terms of $\delta^{234}U$, which is elevated by 2-4‰. Andersen et al. (2007) has sampled at 8 m water depth in the Central Arctic, thus freshwater contributions are stronger as indicated by the lower salinities (Fig. 3) and strongly increased surface water $\delta^{234}U$, which is up to 7‰ elevated compared to the interior Arctic Ocean. Due to the different sampled water depth and the resulting freshwater contribution, the temporal difference between the study of Andersen and this study is solely feasible for exactly one measurement conducted in the Makarov Basin in both years and at 30m water depth. The observations in 2001 yield a moderately higher salinity (31.33) compared to 2021 (30.29). Thus, if the salinity would be modulated by freshwater fluxes alone, a higher freshwater contribution would be expected in 2021 compared to 2001 leading to a more elevated $\delta^{234}U$ value. However, here the $\delta^{234}U$ value is higher in 2001 compared to 2021 contradicting a stronger freshwater influence. Within the Nansen, Amundsen, and Makarov Basin the deviation of excess $^{234}U$ is 1-8‰ between 2021 and 2001.

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Note, to achieve comparability of both studies with the global Ocean mean composition, all measurements used here require correcting for an offset of HU-1 and the difference in $^{235}U/^{238}U$ between seawater and CRM-112A. To allow inter-lab comparability outside of the studies cited here we follow the convention of Chen et al. (2013) and apply a correction of HU-1 = -1.2‰. Our measures of CRM-112A imply a difference of HU-1 =1.5‰ to this reference scale, the study by Andersen et al. (2022) yielded a value of HU-1 =1.6‰. Therefore, comparing the two studies to each other and to (Kipp et al., 2022) is reasonable even when accounting for differences in measurement protocol and data treatment.

## 4. Discussion

### 4.1. The surface and subsurface Arctic Ocean $\delta^{234}U$

From the distinctive $\delta^{234}U$ values across depth profiles, conclusions on the origin and mixing of water masses can be made. At the surface of all stations $\delta^{234}U$ levels are significantly elevated compared to the global mean oceanic $\delta^{234}U$ value. This is caused by excess $^{234}U$ fluxes through freshwater inputs of riverine origin as demonstrated by Andersen et al. (2007). At Station 16, 20 and 28 in the Makarov and Amundsen Basin, surface water $\delta^{234}U$ levels are highest (≥149 ‰). Station 8 and 46 near the Lincoln Shelf and on top of the Gakkel Ridge yield $\delta^{234}U$ levels still enriched (>148.5 ‰). Solely Station 5 in the Nansen Basin shows a moderately elevated surface water $\delta^{234}U$ mean value (147.9 ‰, N=3). Consequently, the excess $^{234}U$ enrichment within the surface Arctic Ocean Basins seems to reflect a variable degree of mixing with freshwater and the underlying water of Atlantic origin.

At Station 5, the $\delta^{234}U$ levels are not distinctively higher at 30 m then at 100 m. We, thus, suggest that the upper most water column of Station 5, i.e. the Atlantic waters in the Nansen Basin are well mixed with respect to the riverine contribution of excess $^{234}U$. Water mass mixing in the Nansen Basin is driven by the inflow of Atlantic Water (AW) through the Fram Strait and Barents Sea, and subsequent mixing with colder and fresher Arctic water as well as shelf waters from the Barents and Kara Seas. This mixing is intensified by seasonal processes like sea ice formation and cooling, which drives convection and ventilates deeper layers. This process is leading to a phenomenon called "Atlantification," where the basin is experiencing warming and increased salinity due to the influx of Atlantic waters (De Rovere et al. 2025). The 500 to 1000 m extend of the dense Arctic – Atlantic water is well visible in the Fram Strait in profiles of the anthropogenic tracer $^{236}U$ (Wefing et al. 2022). In a study by Laukert et al. (2016) the Nansen Basin Atlantified waters show a vertical front at $\sigma_\theta$ = 27.95 with most unradiogenic waters in terms of its $^{143}Nd/^{144}Nd$ isotope composition that is most visible in the upper 500 m of the Fram Straight throughflow. The unradiogenic Nd isotope values further extends to the potential T = 0°C between 900 and 1000 m. The recirculation of water of Atlantic origin is also visible in elevated 129I till mid depths of 1000 m (Karcher et al. 2012). Station 5 in the Nansen Basin is within the fast recirculation pathway of Artic-Atlantic waters. Hence, the $\delta^{234}U$ signature of the Arctic

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– Atlantic water mass reflects a large volume of vertically well – mixed water in the Nansen Basin of predominantly Atlantic origin, with an Atlantic $\delta^{234}U$ value that does not significantly differ from the $\delta^{234}U$ value of the global Ocean mean and Arctic interior water.

In all other stations in the Amundsen and Makarov Basin surface waters at 30 m are distinctively enriched in $\delta^{234}U$ compared to water at 100 m or 250 m. Thus, in the Amundsen and Makarov Basin the freshwater influenced surface waters are less vertically mixed with the underlying waters in terms of the U isotope composition. For the stations north of the Gakkel Ridge, $\delta^{234}U$ levels meet the global mean at already 300 m. Even though for Stations 28 and 46, these vertical profiles were not fully resolved due to a lack in sample material, existing samples show the same behaviour. Thus, we assume that these stations have similar mixing behaviours. It is notable, that at Station 20 the $\delta^{234}U$ is still slightly above the global mean at 250 m, indicating that surface waters are here entrained slightly deeper than at the other stations in the Amundsen and Makarov Basin.

Station 20 and 28 are geographically close but separated by the 1600 m deep Lomonosov Ridge (Fig. 1). The very similar $\delta^{234}U$ levels at the surface imply a transport of freshwater influenced seawater from the pole to the Fram Straight via the trans polar drift (TPD) (Fig.1). Station 16 and Station 8 having lower $\delta^{234}U$ surface levels indicates a gradient in lowering $\delta^{234}U$ levels from the Lomonosov Ridge towards the Gakkel Ridge, confirming the dilution along flow of the TPD.

The $\delta^{234}U$ values found in this work within the freshwater influenced surface layer are significantly lower than the previous value of $\delta^{234}U = (156.4 \pm 0.3)$ ‰ found in a depth of 30 m in the Makarov Basin (Andersen et al., 2007). While only Station 28 is located in the Makarov Basin, the trends discussed above indicate, that $\delta^{234}U$ levels in the Makarov Basin are similar to the levels in the Amundsen Basin. The previous samples taken in 2001 showed only little mixing of surface and subsurface waters. Therefore, these results indicate, that the $\delta^{234}U$ values in Arctic surface waters have significantly lowered over the past 20 years, which is either due to enhanced vertical mixing, which increases the volume of water diluting the freshwater derived excess $^{234}U$, or it is due to decreased supply of freshwater derived $^{234}U$. A possible reason for enhanced vertical mixing could be a reduction of sea ice coverage in the Central Arctic Ocean, that would allow for more wind driven surface mixing. Regarding changes in the supply of freshwater recent reassessment of runoff data products and observations shows minor changes in the overall runoff over the past two decades, except for the increase of the Greenland melting. The latter, however contributes solely a minor amount of freshwater to the Arctic basin compared to continental runoff by the major rivers (Winkelbauer et al. 2022). Thus, we favor the increase of vertical mixing as origin of the decline of excess $^{234}U$ in the surface layer.

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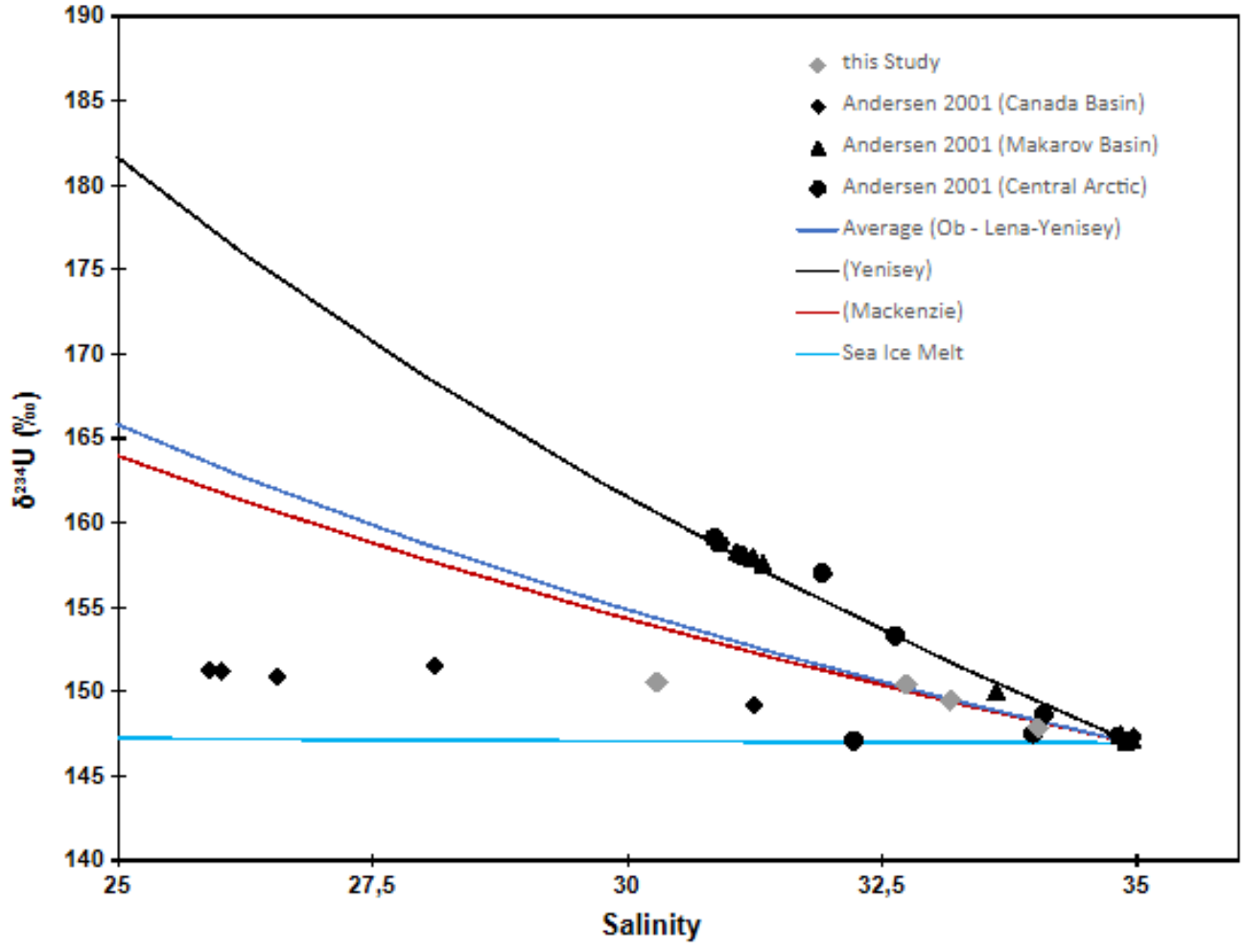


**Figure 3: Surface $\delta^{234}U$ levels plotted against their respective salinity (gray squares) which was determined in [Snoeijs-Leijonmalm et al., 2022]. All other measurements would plot on a salinity of 34.9 and a $\delta^{234}U$ of 146.94. Lines indicate conservative mixing models for freshwater and seawater according to the end-member values of U and $\delta^{234}U$ for Yenisey and Mackenzie and a combination of rivers provided by Andersen et al. (2007). The light blue mixing line represents a strongly U reduced freshwater influence (here about 10% of the Mackenzie U flux). Such an end-member is here assumed to represent a low U, low excess $^{234}U$ freshwater flux from glacier melting and surface runoff. A similar stable isotope composition and change in salt would be related to sea ice melting, which influences solely the salt budget but not the U isotope composition. The previous observations from Andersen et al. (2007) are also plotted (filled symbols). For stations at which the surface $\delta^{234}U$ was measured multiple times, the mean value is plotted.**

To further elucidate the behaviour of $\delta^{234}U$ against salinity we analyse the mixing of seawater with the rivers that flow into the Arctic Ocean and reduce the seawater salinity. The surface $\delta^{234}U$ levels and mean $\delta^{234}U$ found in this study are compared to two-end-member mixing of the Yenisey, Mackenzie River (Fig. 3 black and red curves) and a combination of all Barents Sea Shelf contributions ( Σ(Yenisey+Ob+Lena) (Fig. 3: dark blue). The non-linear behaviour stems from the different U concentrations of the rivers, which carry no salt, but U. To estimate these mixing curves we use the end-member compositions

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of the rivers by Andersen et al. (2007) and the interior Arctic ocean composition from our study, which is identical to the one of Andersen et al. (2007) within uncertainty. First, it is noticeable, that observations within the Central Arctic Ocean and Makarov Basin by Andersen et al. (2007) fingerprint a strong influence of the Yenisey River in 2001, with one exceptional low value plotting close to the curve estimated from melting continental ice or melting sea ice. Thus, the Central Arctic Ocean, i.e. the Amundsen Basin and Makarov Basin seem to be most influenced by the Yenisey River in 2001. In contrast, the samples from the Canada Basin plot significantly below any of the rivers expected mixing lines, which was interpreted by Andersen et al. (2007) as either removal of U from the Estuary of the Mackenzie River causing a strong reduction in its isotope impact or the occurrence of sea ice melt. In 2021 the three Amundsen Basin surface layer measurements, referred to as Central Arctic by Andersen et al. (2007), perfectly plot on the mixing line of either Mackenzie River, or the sum of all Barents Shelf Rivers mixing with seawater. But clearly, the influence of the Yenisey River is not visible at 30 m depth. Interestingly, our observation from the surface water of the Makarov Basin now shows a strongest salt lowering but low $\delta^{234}U$, which highlights either the presence of Greenland meltwater or the melting of sea ice comparable to the previous observations in the Canada Basin. Consequently, the question arises, whether this meltwater/sea-ice-melt signal has been carried from the Canada Basin to the Makarov Basin over the past 20 years. A plausible mechanism to inject a sea ice melt signal into the Makarov Basin would be the recirculation of the Beaufort Gyre. Alternatively, this could also be a local sea ice melting signal similar to the one observed by Andersen et al. (2007) in the Central Arctic (Fig. 3).

While, our data set is very small for the surface Arctic Ocean in 2021, we nevertheless infer that the prominent role of the Yenisey River for the freshwater injection derived excess $^{234}U$ into the Central Arctic Ocean has measurably declined, or the U concentration of the river has decreased. Since our result plot well on a general mixing curve of all rivers, our 30 m water samples seem to integrate the flux of the continental runoff of $^{234}U$, compared to the fingerprinting of individual rivers in the surface layer (~ 8 m) as emphasised by Andersen et al. (2007). Moreover, it seems plausible that the ice melting visible in the Canada Basin in 2001 has propagated via the Beaufort Gyre towards the Makarov Basin over the past two decades, or alternatively the local melting of sea ice and/or the runoff of glacier meltwater has increased, impacting the density of the surface layer. Additionally, our observations from the Nansen Basin confirm enhanced fast-vertical mixing and overall the export of excess $^{234}U$ injected by rivers through the TPD dominates the surface to interior gradient of $\delta^{234}U$.

### 4.2 The $^{234}U/^{238}U$ ratio below 300m

Below 300 m, no distinctive trends in $\delta^{234}U$ are measurable. The general lack of trends in $\delta^{234}U$ data close to the seafloor implies that the influence of a benthic U flux from the sediment into the abyssal waters can be neglected. Samples were taken, however, at least 20 m above the seafloor due to technical limitations (Snoeijs-Leijonmalm et al., 2022), therefore no final conclusion on the amount of pore water-seawater exchange on $\delta^{234}U$ can be drawn from our dataset. The resulting mean $\delta^{234}U$ values of interior Arctic Ocean $\delta^{234}U = (146.94\pm0.09)$‰ is identical to the one of Kipp et al., 2022. Previously, Andersen et al., 2007 determined a $\delta^{234}U$ value for the Makarov Basin of (147.1 ± 0.3) ‰ in 1000 m depth and (147.2 ± 0.3) ‰ in 2500 m

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depth. Inside the uncertainties of both the values from Andersen et al., 2007 as well as the uncertainties on the mean values found in this study, there are no significant differences. This mean showcases that the interior Arctic U isotope composition is in steady state and set by the long-term global ocean circulation, with little local influences from the giant amount of Arctic freshwater inputs visible in the surface layer. Moreover, the interior waters have not significantly changed in $\delta^{234}U$ during the 20 years that passed between the studies.

Since the Arctic Ocean receives about 10% of global river discharge despite comprising only about 1% of global ocean volume (Karcher and Oberhuber, 2002), the lack of accumulated $^{234}U$ in the deep waters observed in this study indicates that riverine waters are rapidly exported from the Arctic Ocean via the TPD. The gradient between surface water with measurable excess 234U and interior Arctic Ocean water is mostly contained to the top 100 m. This finding agrees with an upper layer turnover time of 10 – 20 years maintaining freshwater anomalies (Druffel et al. 2017, Wefing et al. 2022), and turnover time of >500 years in the Arctic interior (Schlosser et al. 1998). This is consistent with the strong export of surface and intermediate waters through the Fram Strait, as well as transport via the TPD (compare Fig. 1). The excess riverine waters are thereby exported into the Atlantic Ocean and enter the Global Ocean Overturning circulation in the Nordic and Labrador Sea.

### 4.3 The $^{238}U/^{235}U$ ratio of Arctic waters

Unlike $\delta^{234}U$, $\delta^{238}U$ shows no distinctive trends at the surface. We observe a large scattering of the data around the global Ocean mean value in our measurements, which does not seem to be related to any physical water mass property of the Arctic Ocean Basin. Our most plausible explanation at present is that our achieved analytical reproducibility is rather low for the $^{235}U/^{238}U$ ratio compared to the one obtained by Kipp et al. (2021). We have spiked the samples foreseen for $^{235}U/^{238}U$ ratio determination in large parts solely after the purification chemistry, because the excellent performance of the ion exchange column and because the large volume samples or salt amounts to spike did make the spike mass concentration challenging. However, changes in the chemical yield, would entrain possible isotope effects and fractionation that cannot be seen in the spiked $^{233}U/^{236}U$ ratio used for mass fractionation correction. The impact of this is invisible in the $\delta^{234}U$ determination as here the conventional $^{235}U/^{238}U$ ratio is used for mass fractionation correction. Consequently, any chemistry derived impact on the $^{235}U/^{238}U$ ratio would equally impact the $^{234}U/^{238}U$ and thus cancel. Therefore, we will not further evaluate in depth the $^{235}U/^{238}U$ ratio determined here. What we can state is that the determined $\delta^{238}U$ values scatter around the global Ocean mean value (-0.379 ± 0.023 ‰) (Kipp et al., 2022), and that within the achieved low reproducibility no significant trends of U isotope fractionation are measurable. With this in mind, the mean $\delta^{238}U$ value of the Arctic Ocean found in the VACAO samples is (−0.348 ± 0.046, N=68) ‰. This value agrees with the global Ocean mean inside its standard error and shows that the Arctic Ocean has no unique tendencies in $\delta^{238}U$. However, future and more precise measurements are needed to exclude the possibility of subtle $\delta^{238}U$ changes in the Arctic Ocean.

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## 5 Conclusion

Here, measurements of $\delta^{238}U$ and $\delta^{234}U$ in Arctic seawater are presented. In the Arctic seawater samples from the VACAO-Project, enriched $^{234}U$ levels were found in the surface layer, following the Arctic Ocean Surface Circulation. The values found were generally lower than the $\delta^{234}U$ values obtained in the uppermost surface 20 years earlier. Therefore, it is concluded, that $\delta^{234}U$ in the near surface layers of the Arctic Ocean is influenced by a stronger downward mixing over the past 20 year time span in-between the sampling in 2001 and 2021. Moreover, the strong fingerprint of the Yenisey River on upper most Central Arctic Ocean surface water in 2001 was no traceable in our study in 2021. Water influenced sea-ice-melting in the Canada Basin may have propagated to the Makarov Basin, while the freshwater derived excess $^{234}U$ is efficiently exported into the Atlantic Ocean via the Trans Polar Drift (TPD).

Below the surface layer, $\delta^{234}U$ levels match the recently reassessed global mean seawater $\delta^{234}U$ value as well as the Arctic Ocean $\delta^{234}U$ mean values found in 2001 within their uncertainties. Therefore, it is concluded, that the deeper waters are unaffected by the changes at the surface with respect to the resolution of 0.09‰ achieved here.

The behaviour of $\delta^{234}U$ over salinity is similar to the behaviour predicted for the Central Arctic Basins. In this comparison, all our observations closest follow the Arctic seawater – Mackenzie or the sum of Barent Sea river fluxes.
$\delta^{238}U$ in the Arctic Ocean shows no depth-dependent trends and is homogeneous between the different stations. The mean $\delta^{238}U$ value of the Arctic Ocean appears identical to the one of the global Ocean mean, but more precise measures are needed to exclude the possibility of small changes in this ratio.

## Appendix A – Reproducibility of δ234U in CRM 112A Standard:

To test the reproducibility of U-extraction chemistry and isotopic measurements a series of standards has been passed through chemistry. A 1000 ppb CRM-112A standard diluted in 20 ml of 8M $HNO_3$ was prepared, treated and measured according to the full chemical preparation protocol. During the same measurement, a CRM-112A standard that had not undergone extraction chemistry, was measured four times for comparison. Additionally, a CRM 112A standard that is kept in a separate measurement tube was measured once to exclude bias. The results are given in Table A1.

| Sample | $\delta^{234}U$ [‰] | $\Delta\ \delta^{234}U$ [‰] |
|---|---|---|
| CRM-112A after extraction chemistry | | |
| Sample1 | -36.14 | 0.34 |

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| | | |
|---|---|---|
| Sample 2 | -37.07 | 0.18 |
| Sample 3 | -36.01 | 0.26 |
| Mean and 2σ error | -36.41 | 0.67 |
| CRM-112A without extraction chemistry | | |
| Measurement 1 | -35.89 | 0.28 |
| Measurement 2 | -36.45 | 0.53 |
| Measurement 3 | -36.18 | 0.42 |
| Measurement 4 | -36.61 | 0.31 |
| Mean and 2σ error | -36.28 | 0.32 |
| CRM-112A from separate tube | | |
| Measurement 1 | -36.76 | 0.42 |

**Table A1: $\delta^{234}U$ of CRM-112A Standards Before and After Extraction Chemistry. The given Δ δ234U error is the 2σ standard deviation of the 30 measurement cycles conducted for each sample.**

These results show, that the measured CRM-112A samples are mostly identical in their errors. The errors are generally larger than the errors observed in the seawater samples due to a lower U concentration in the standards. There is no systematic difference visible in the CRM-112A samples that have or have not undergone chemistry compared to the other measured CRM-112A. This confirms, that the U-Extraction Chemistry does not cause fractionation in the samples visible at the precision of 0.2-0.5‰. This experiment was not conducted for $\delta^{238}U$, since the measurement setup for $\delta^{238}U$ was not ready at the time of this experiment.

**Appendix B – Measuring diluted samples:**

During the conduction of this study, amounts of prepared sample that were smaller (0.5 ml to 0.8 ml) than the amount of liquid required for a measurement (1 ml) were left over. To test if the leftover samples could be diluted for repeat measurements, small amounts of acid (1% HNO3 + 0.05% HF) were added to leftover samples, creating enough liquid for another measurement. The results are shown in Table B1 and compared to the results from the same station and depth measured according to protocol.

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| Sample | Added Acid [ml] | $\delta^{234}U$ diluted [‰] | $\delta^{234}U$ not diluted [‰] | $\delta^{238}U$ diluted [‰] | $\delta^{234}U$ not diluted [‰] |
|---|---|---|---|---|---|
| 12737 | 0.3 | 147.00 ± 0.58 | 146.96 ± 0.27 | -0.339 ± 0.023 | -0.236 ± 0.015 |
| 12878 | 0.2 | 146.80 ± 0.38 | 147.34 ± 0.31 | -0.393 ± 0.019 | -0.304 ± 0.024 |
| 12504 | 0.2 | 146.48 ± 0.25 | 146.78 ± 0.31 | -0.460 ± 0.016 | -0.353 ± 0.022 |
| 12518 | 0.2 | 146.78 ± 0.27 | 146.80 ± 0.30 | / | / |
| 12515 | 0.2 | 147.07 ± 0.20 | 146.78 ± 0.36 | / | / |
| 12637 | 0.3 | 147.36 ± 0.39 | 147.54 ± 0.23 | -0.377 ± 0.020 | -0.258 ± 0.020 |
| 13157 | 0.2 | 147.41 ± 0.50 | 147.09 ± 0.33 | -0.422 ± 0.028 | -0.394 ± 0.020 |

**Table B1: Comparison of $\delta^{234}U$ and $\delta^{238}U$ measured in diluted and original samples**

It is noticeable, that in no case the difference in $\delta^{234}U$ and $\delta^{238}U$ of the diluted and original samples are outside of their respective statistical uncertainties of 0.5 ‰ and 0.15 ‰. In some cases the dilution can cause a higher uncertainty which needs to be considered when evaluating these samples. Still, remeasuring diluted samples in this fashion is considered a viable option, especially when sample solution is lost due to equipment malfunctioning.

## Appendix C – Reproducibility of $\delta^{234}U$ and $\delta^{238}U$ in Seawater samples:

To compare the reproducibility of measurements, multiple samples taken from two 20l canisters of seawater from the institutes stock were prepared and measured in the same fashion as the samples of this study. The samples contain water taken on the RV METEOR Cruise M122 in the Angolan Margin at two different depths (see [Zabel, 2016]). These samples have been measured along the samples of this study. In this manner, all effects from chemistry as well as daily variations in the measurements are reflected in the data. Additionally, two of the samples were measured twice as a way to additionally check if the results are consistent. The results are given in Table C1. Two of the samples were diluted before measuring. One was diluted to allow the sample to be measured twice and one was diluted to repeat a measurement after instrument issues.

| Sample | Measurement Date | $\delta^{234}U$ [‰] | $\delta^{238}U$ [‰] | Note |
|---|---|---|---|---|

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| | | | |
|---|---|---|---|
| M112 GeoB 20901-1 26m A | 06.08.2024 | 146.49 ± 0.31 | -0.265 ± 0.02 | |
| M112 GeoB 20901-1 26m B | 26.08.2024 | 146.27 ± 0.28 | -0.260 ± 0.016 | |
| M112 GeoB 20901-1 26m C | 03.09.2024 | 146.81 ± 0.30 | -0.311 ± 0.012 | |
| M112 GeoB 20901-1 26m D | 26.09.2024 Day | 146.74 ± 0.27 | -0.735 ± 0.022 | |
| M112 GeoB 20901-1 26m A | 26.09.2024 Night | 146.10 ± 0.42 | -0.245 ± 0.023 | Repeated |
| M112 GeoB 20901-1 26m B | 26.09.2024 Night | 145.92 ± 0.24 | -0.383 ± 0.019 | Repeated, diluted with 0.2ml |
| M112 GeoB 20901-1 1030m A | 06.08.2024 | 146.95 ± 0.29 | -0.411 ± 0.016 | |
| M112 GeoB 20901-1 1030m B | 09.08.2024 | 146.34 ± 0.38 | -0.181 ± 0.023 | |
| M112 GeoB 20901-1 1030m C | 05.09.2024 | 146.49 ± 0.32 | -0.442 ± 0.015 | |
| M112 GeoB 20901-1 1030m D | 26.09.2024 Day | 146.66 ± 0.27 | -0.693 ± 0.029 | diluted with 0.2ml |
| M112 GeoB 20901-1 1030m E | 26.09.2024 Day | 146.08 ± 0.36 | -0.773 ± 0.027 | |

**Table C1: $\delta^{234}U$ and $\delta^{238}U$ measured in reference samples.**

The standard deviation of $\delta^{234}U$ is 0.35‰ in M122 GeoB 20901-1 26m and 0.33 ‰ in M122 GeoB 20901-1 1030m. In the $\delta^{238}U$ data, one measurement run (26th September 2024, Daytime) yields significantly lower values than the rest of the measurements. Therefore, these measurements will be excluded from the evaluation. Without the problematic data, the M122 GeoB 20901-1 26m samples have a standard variation of 0.056 ‰ in $\delta^{238}U$. For the M122 GeoB 20901-1 1030m samples not enough valid data points exist to conduct reliable statistics. The repeated measurements both deviate less than 0.1 ‰ from

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their previous measurements. From the M122 GeoB 20901-1 26m samples, a 2σ error of 0.112 ‰ is calculated. Due to the experimental nature of the $\delta^{238}U$ measurements and the low amount of data used for this statistic, the error is rounded up to 0.15 ‰..

The $\delta^{238}U$ results of A and B samples from the same aluminium foil bag and samples that were measured multiple times are used to assess the full analytical reproducibility of $\delta^{238}U$ measurements. When excluding few individual outliers with >0.3 ‰ difference, the overall reproducibility shows mean differences well below 0.15 ‰, supporting the use of 0.15 ‰ as a conservative estimate of reproducibility in this study.

## Code and data availability

All data and code needed to evaluate and reproduce the results in the paper are present in the paper and/or the appendix Material.

## Author contributions

HFJ: Investigation, Formal Analysis, Writing (original draft preparation), Conceptualization; SG: Investigation, Writing (review and editing), NF: Conceptualization, Supervision, Writing (original draft preparation).

## Competing interests

The authors declare that they have no conflict of interest.

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