## Meteoric water and glacial melt Glacial Meltwater in the Southeast

## Amundsen Sea: A timeseries from 1994-2020

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- 10 **Abstract.** Ice sheet mass loss from Antarctica is greatest in the Amundsen Sea sector, where 'warm' modified Circumpolar Deep Water moves onto the continental shelf and deep seawater melts and thins the bases of ice shelves hundreds of meters below the sea surface. We use nearly 1000 paired salinity and oxygen isotope analyses of seawater samples collected on seven expeditions from 1994 to 2020 to produce a time series of glacial meltwater inventory foron the Southeast Amundsen Sea continental
- shelf. Water column salinity-δ<sup>18</sup>O relationships yield freshwater endmember δ<sup>18</sup>O values from <u>-</u> 31.3±0.6-30.2%<sub>0</sub> to -28.4±0.8‰, consistent with the isotopic composition of local glacial ice. Meteoric water inventories, consisting of an estimated >92% glacial meltwater by mass, account for 7.6±0.5 m to 9.2±0.6 m of‰, demonstrating that regional freshwater content is dominated by deep glacial melt. The meltwater fractions display temporal variability in the basal melting, with 800 m water column, and exhibit greater meltwater inventories from 7.7 m to 9.2 m. This result corroborates recent studies
- suggesting interannual variability than trend over the study period, based on the available data. in basal melt rates of West Antarctic ice shelves and is consistent with the Amundsen region's influence on ocean salinity and density downstream in the Ross Sea.

#### 1 Introduction

Four decades of observations <a href="mailto:showhave shown">showhave shown</a> significant and increasing <a href="mailto:glacialiee">glacialiee</a> mass loss from Antarctica (Rignot et al., 2011; Velicogna et al., 2014; Rignot et al., 2019a). (Rignot et al., 2011; Velicogna et al., 2014; Rignot et al., 2019). A Special Report on the Ocean and Cryosphere in a Changing Climate (SROCC) projected 0.61 m to 1.10 m of sea level rise (SLR) by 2100 under RCP8.5 forcing, with uncertainty largely hinging on the <a href="mailto:future of the">future of the</a> Antarctic ice sheet (IPCC, 2022). Over the past two decades, losses from the West Antarctic Ice Sheet (WAIS) have comprised 84±12% of the total Antarctic contribution to SLR (5.5±2.2 mm from 1993-2018 cm, since 1901; WCRP Global Sea Level Budget Group, 2018), with glaciers flowing into the Amundsen Sea Sector (particularly the Pine Island and Thwaites glaciers) dominating the overall negative mass balance of the ice sheet (Rignot et al., 2019b; Shepherd et al., 2019).

dominating the overall negative mass balance of the ice sheet (Shepherd et al., 2019). High ice shelf basal melt rates in the Southeast (SE) Amundsen Seathere have been linked to the flow of 'warm' and salty modified Circumpolar Deep Water (mCDW) onto the continental shelf, separated by cooler but fresher waters above by a thermocline between 300 m and 700 m (Dutrieux et al., 2014; Jacobs et al., 2011). mCDW flows from the continental shelf break towards SE Amundsen Sea ice shelves via "central" and "eastern" glacially-carved bathymetric troughs (Nakayama et al., 2013). This 'warm' mCDW penetrates-and its penetration into sub-ice shelf cavities (Jacobs et al., 1996; Paolo et al., 2015; Pritchard et al., 2012)(Jacobs et al., 1996; Paolo et al., 2015; Pritchard et al., 2012) where it can access ice shelf grounding lines (Rignot and Jacobs, 2002). To access the Pine Island Ice Shelf (PIIS) 45 grounding line, mCDW passes between the bottom of the ice shelf at ~350 m and a seafloor ridge at ~700 m (Jenkins et al., 2010). Basal melt is driven by total heat transport, dependent more on the thickness of the mCDW layer transported on-shore than its temperature (Dutrieux et al., 2014; Jenkins et al., 2018), with the thickness controlled by local wind forcing of a shelf break undercurrent, in turn influenced by the Amundsen Sea Low. Despite the strong sensitivity of these ice shelves to ocean forcing, and evidence of increasing mass loss in this region. however estimates of Antarctic SLR contributions from basal melt remain poorly constrained (van der Linden et al., 2021, 2023) (van der Linden et al., 2021, 2023). While there is evidence for accelerating ice mass loss throughout West Antarctica (e.g. Shepherd et al., 2019) driven largely by ice shelf basal melt (Paolo et al., 2015; Pritchard et al., 2012), some studies have shown greater interannual variability in basal melt rates than increase (Paolo et al., 2018; Holland et al., 2019; Adusumilli et al., 2020; Flexas et al., 2022), and some have even suggested a slowing of basal melt rates (Paolo et al., 2022) and grounding line retreat (Christie et al., 2023). Here we use a method independent of satellite-based measurements to assess the glacial meltwater (GMW) inventory in the coastal water columns of the SE Amundsen Sea.

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Southern Ocean water masses have typically have been differentiated and defined by measurements of temperature and, salinity, and less often by including oxygen isotopes ( $\delta^{18}$ O; Jacobs et al., 1985, 2002; Meredith et al., 2008, 2010, 2013; Brown et al., 2014; Randall-Goodwin et al., 2015; Silvano et al., 2018; Biddle et al., 2019). (Jacobs et al., 1985). Salinity- $\delta^{18}$ O relationships can be used to infer the source region and concentration influence of highly  $\delta^{18}$ O-depleted glacial meltwater on seawater properties (Jacobs et al., 1985; Hellmer et al., 1998; Jacobs et al., 2002; Meredith et al., 2008; Randall-Goodwin et al., 2015).zero-salinity glacial freshwater on seawater properties. Where ice shelf basal melting is deep, meltwater oxygen isotope depletion far exceeds that of local ocean surface precipitation. A spatial and temporal array of T, S and  $\delta^{18}$ Osamples can thus be utilized to track GMW content<del>quantity</del> and distribution, especially in nearshore waters adjacent to<del>coastal regions where ice</del> shelf melting ice shelves is strong. Prior studies have used  $\delta^{18}O$  measurements to estimate meteoric water abundance (precipitation and GMW) waters in the Amundsen Sea water column (Biddle et al., 2019; Jeon et al., 2021; Randall-Goodwin et al., 2015) and elsewhere around Antarctica (Meredith et al., 2010, 2018; Silvano et al., 2018) but so far have revealed little about temporal variability or possible trends in GMWglacial meltwater content. Here, we use nearly 1000 seawater isotope samples collected during seven austral summers from 1994 to 2020 (Figure 1 Figure 1) to investigate meteoric

<u>water</u>meltwater sources, water column inventories, and <u>their</u> interannual variability in the SE Amundsen Sea.

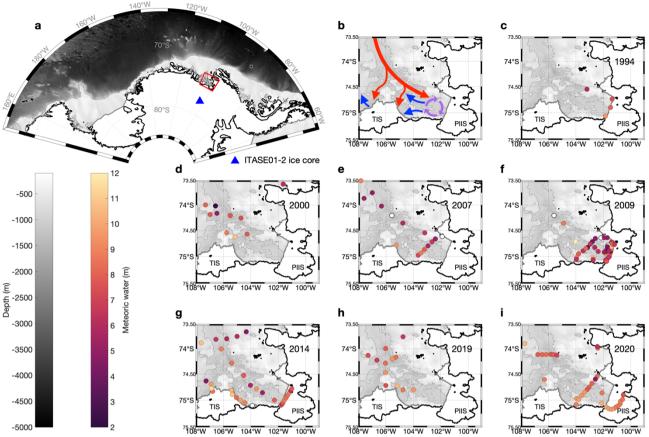


Figure 1: Study area bathymetry, circulation, and  $\delta^{18}$ O sampling locations each of 7 years between 1994 and 2020. 800 m isobaths are shown as thin gray lines. (a) SE Amundsen study area, and location of the ITASE01-2 ice core. (b) Location of the Pine Island Bay gyre (purple), pathways of warm deep mCDW (red) toward the ice shelves and pathways of shallower meltwater rich waters (blue) from beneath Pine Island Ice Shelf (PIIS; Nakayama et al., 2019; Wåhlin et al., 2021). (c-i) Colored dots show sample locations, with colors representing vertically integrated glacial meltwater inventories between 0 m and 800 m from 1994 to 2020. Thick gray lines indicate seaward boundaries of Thwaites Ice Shelf (TIS) and the PIIS. Calving fronts are referenced to 2000 (Schaffer et al., 2014; Fretwell et al., 2013), a relatively stable location before a ~20 km retreat following calving events between 2017 and 2020 (Joughin et al., 2021). Stations where sampling did not extend to the seafloor show only partial water column inventories, and stations shown as white dots (2007, 2009, 2014) had only one depth sampled. In 2000, 2007, and 2019, access to sampling along the front of PIIS calving front was precluded by fast ice.: 618O sampling locations each year, (a) SE Amundsen study area, and location of the ITASE01 2 ice core. (b) detailed bathymetry of study location (shading), with isobaths at 500 m and 1000 m (lines), (e-i) Colored dots show sample locations, with colors representing vertically integrated glacial meltwater inventories between 0 m and 800 m from 1994 to 2020. Gray lines indicate seaward boundaries of Thwaites Glacier Tongue (TGT) and the Pine Island Ice Shelf (PIIS). Often limited by fast ice, station locations tend to show higher GMW inventories along or near those calving fronts. The former is referenced to 2000 (Schaffer et al., 2014; Fretwell et al., 2013), a relatively stable location before a ~20 km retreat following calving events between 2017 and 2020 (Joughin et al., 2021a). Stations where depth sampling did not extend to the seafloor show only partial water column inventories.

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#### 2 Data and Methodsmethods

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### 1.12.1 Sample collection and analysis

We compile data from samples Samples were collected during 7 field seasons from sites in the SE Amundsen Sea from 1994 to 2020 (Figure 1 Figure 1, Table 1 Table 1). Salinity profiles were obtained using calibrated conductivity cells on SBE911 conductivity-temperature-depth (CTD) instruments CTDs, monitored with shipboard bottle sample analyses using with Guildline AutoSal and PortaSal salinometers calibrated with IAPSO seawater salinity standards. In . For most years (1994, 2007, 2009 and, 2014,), δ18O was measured using an Isotope Ratio Mass Spectrometer (IRMS; Micromass Optima Multiprep or a Finnigan MAT252 HDO). All samples collected in 2019 and 2020, and some in 2007 and 2009, were measured with a Picarro L2140-i Cavity Ring Down System (CRDS).

Equivalence has been demonstrated between CRDS and IRMS measurements (Walker et al., 2016; **Appendix** A8). A7). In all cases, values are reported as per milmille (‰) deviations(δ), relative to Vienna Standard Mean Ocean Water (VSMOW2; Coplen, 1994).

Table 14: Summary of  $\delta^{18}O$  data sources, sampling intervals, methods & applications

Year	Cruise	Sample collection dates	# Samples	δ <sup>18</sup> O Technique(s)
1994	NBP94-02	14 Mar. 1994 15 Mar. 1994	26	IRMS CO <sub>2</sub> equilibration
<u>1994</u>	NBP94-02 (Hellmer et al., 1998)	<u>14 Mar. 1994 –</u> 15 Mar. 1994	<u>26</u>	IRMS CO <sub>2</sub> equilibration
<del>2000</del>	NBP00-01	<del>16 Mar. 2000 - 20</del> <del>Mar. 2000</del>	<del>62</del>	IRMS CO <sub>2</sub> equilibration
<u>2000</u>	NBP00-01 (Jacobs et al., 2002)	<u>16 Mar. 2000 –</u> <u>20 Mar. 2000</u>	<u>62</u>	IRMS CO <sub>2</sub> equilibration
2007	NBP07-02	24 Feb. 2007 – -27 Feb. 2007	74	IRMS CO <sub>2</sub> equilibration, CRDS
2009	NBP09-01	16 Jan. 2009 – -29 Jan. 2009	175	IRMS CO <sub>2</sub> equilibration, CRDS
2014	iSTAR2014	5 Feb. 2014 20 Feb. 2014	213	IRMS CO <sub>2</sub> -equilibration
<u>2014</u>	iSTAR2014 (Biddle et al., 2019)	<u>5 Feb. 2014 –</u> <u>20 Feb. 2014</u>	<u>213</u>	IRMS CO <sub>2</sub> equilibration
2019	NBP19-01	12 Jan 2019 – 14 Jan 2019	107	CRDS
2020	NBP20-02	5 Feb. 2020 – 8 Mar. 2020	280	CRDS

Some of the 2009 samples were processed at Rutgers University in 2010 using a Micromass IRMS; the remainder in 2020 using a Picarro CRDS system at Stanford University. While the latter samples had not been opened since collection, a substantial number were compromised by evaporation during 1140 years of storage. The 2009 samples analyzed in 2020 were scrutinized visually and newly measured sample densities compared with those derived from the CTD measurements. Data from compromised

- samples were discarded (Appendix A7). A subset of 100 samples from 2019 and 2020 were processed 120 concurrently using CO<sub>2</sub> equilibration on a Finnigan MAT252 IRMS and CRDS via vaporizer to ensure data comparability between instrumentation (Appendix A8). Measurements (CRDS) measurements for all years achieved a precision of 0.04% for IRMS and (0.02% for CRDS, based on replicate analyses.
- 125 After a review of the literature, we considered a possible salt effect in measured seawater  $\delta^{18}$ O, as suggested by a small number of studies Previous studies (Lécuyer et al., 2009; Skrzypek and Ford, 2014; Benetti et al., 2017). As no salt effect offset was applied to the previously published data in this study (1994, 2000, 2014) we have not applied any offset to data from other years. The mCDW  $\delta^{18}$ O value (**Table 2**) for 2014 is significantly higher than other years (**Appendix A2**) – likely due to a calibration offset but may also point to sample storage issues. The mCDW and meteoric water 130 endmembers are defined from observations each year, minimizing the impact of interlaboratory offsets on the results (Appendix A2, Data and Methods 2.2). have observed a possible salt effect offset in measured seawater δ<sup>18</sup>O using both CRDS and IRMS equilibration methods. Benetti et al. (2017) note this salt effect offset is likely to vary between laboratories and analytical methods. As no salt correction was applied to the originally published (1994, 2000, and 2014) data, we have not applied an offset to 135 samples from 2007, 2009, 2019, and 2020. Here, we compare calculated GMW fractions (Data & Methods), rather than direct isotope measurements, minimizing any potential issues arising from offsets between laboratories (Appendix A2).

#### 1.2 Meteoric waters defined by the δ<sup>18</sup>O – salinity relationship

- The zero-salinity δ<sup>18</sup>O endmember, defined by data below 200 m, reflects pure meteoric freshwater in the form of ice shelf basal melt (Data and Methods). Freshwater endmembers (intercepts) revealed by salinity- $\delta^{18}$ O relationships over the seven sampled summers differ by <2%, ranging from -28.4% to-30.2% (Figure 2, Table 2). These measurements are consistent with the nearest ice-core (ITASE01-2, from 77.84°S, 102.91°W, Figure 1a; Schneider et al., 2006; Steig et al., 2005) with a mass-averaged δ<sup>18</sup>O value of 29.8±1.9%. Ice cores further east have less negative δ<sup>18</sup>O values (-20%: Thomas et al., 2009), while those further west are more negative (-40%; Blunier & Brook, 2001). Local precipitation, in the form of snow collected in early 2019 at 72°S, vielded a 8<sup>18</sup>O value of ~-15\%, consistent with previous observations at sea level from this latitude (Masson Delmotte et al., 2008). The zero-salinity endmembers extrapolated from the regional salinity and δ<sup>18</sup>O observations indicate that freshwater added to the water column is dominated by locally derived GMW, as intimated earlier for the 1994 data 150
- (Hellmer et al., 1998).

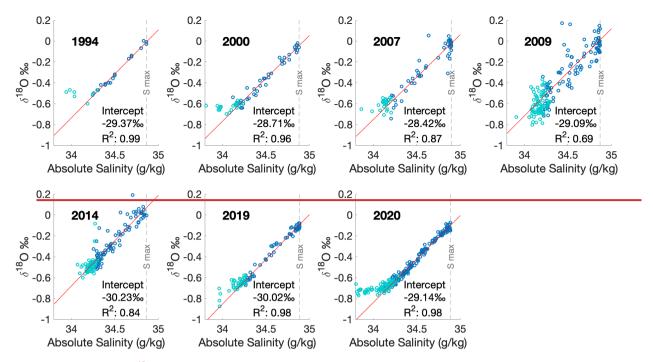


Figure 2: Salinity vs δ<sup>18</sup>O plots for each year. Dark blue circles represent samples deeper than 200 m, fit by linear regression shown as red lines (with R<sup>2</sup> from 0.69 in 2009 to 0.99 in 1994) that project zero salinity glacial meltwater endmember intercepts. Light gray dashed vertical lines indicate the modified Circumpolar Deep Water (mCDW) salinity maxima (Table 2). The most negative upper water column seawater δ<sup>18</sup>O measurements tend to reach minima between 0.8‰ and 0.5‰. The larger scatter in 2009 and 2014 results likely from sample storage issues (Appendix A6).

## **1.32.2** Three-endmember mixing model

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We adapt an approach from Östlund & Hut (1984) <u>asthat has been</u> applied <u>by others</u> in the Peninsula-Bellingshausen-Amundsen region of West Antarctica (Biddle et al., 2019; Jeon et al., 2021; Randall-Goodwin et al., 2015; Meredith et al., 2010) and near <u>the Totten Ice Shelfice shelf</u> (Silvano et al., 2018). We use a 3-endmember Our mixing model (Equations 1-3) to determine water source fractions in the <u>field area. The model</u> assumes <u>thethat</u> observed  $\delta^{18}$ O and salinity values are the result <u>fromof</u> mixtures of mCDW, sea ice <u>melting/freezingmelt</u>, and meteoric waters contributing a range of  $\delta^{18}$ O and salinity signatures. Meteoric waters <u>deeper than 200 m deep below the surface</u> are dominated by ice shelf basal melt, as are surface waters near the ice shelves. Shallower but moving toward shallower depths in the water column, <u>canwill</u> also include <u>ice front/wall melt</u>, and iceberg melting. Near; near-surface waters are also influenced by sea ice melt and formation, and are likely towill contain <u>bothsome amount of local and advected sea level</u> precipitation.

Equations 1-3: Three-endmember mixing model. The 3-endmember mixing model uses the absolute salinity and  $\delta^{18}O$  of mCDW, sea ice melt, and meteoric water endpoints to solve for the relative <u>fractionsabundance</u> of the three water <u>sources</u> in each sample analyzed.

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f_{sim} + f_{met} + f_{mcdw} = 1 
175 \quad f_{sim} * S_{sim} + f_{met} * S_{met} + f_{mcdw} * S_{mcdw} = S_{obs} 
f_{sim} * \delta_{sim} + f_{met} * \delta_{met} + f_{mcdw} * \delta_{mcdw} = \delta_{obs} 
f = \text{fraction of water source} 
S = \text{salinity} 
180 \quad \delta = \delta^{18}O 
sim = \text{sea ice melt} 
met = \text{meteoric water} 
mcdw = \text{modified circumpolar deep water} 
obs = \text{observed sample} 
(1) 
(2) 
(3)
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The primary fraction of interest is the meteoric water fraction comprised of ice shelf melt. The meteoric water endmember is defined as the zero-salinity  $\delta^{18}$ O intercept of data below 200 m (). This intercept reflects the average  $\delta^{18}$ O of pure meteoric freshwater in the form of ice shelf basal melt (**Results 3.1**, **Appendix A1**). Below 200 m the  $\delta^{18}$ O-salinity plots yield mixing lines with mCDW as the saltiest, least-depleted component, and meteoric water as the most highly depleted freshwater endmember, 190 indicative of basal meltwater. To minimize issues that could arise from inter-laboratory calibration offsets (Data and Methods 2.1, Appendix A2), we define individual mCDW and meteoric water endmembers separately for each year (Table 2). Model outputs (mCDW, meteoric water, and sea ice melt fractions) critically depend on appropriate endmember inputs, which will affect resulting water source fractions and trends. The primary fraction of interest is that of ice shelf basal and wall meltwater 195 fractions, defined at the zero-salinity intercepts on  $\delta^{18}$ O-salinity plots of all samples taken each year (Figure 2). Below the surface mixed layer, more generally influenced by sea ice melt and local precipitation, the 8<sup>18</sup>O-salinity plots yield mixing lines with mCDW as the saltiest, least-depleted component, and glacially-derived meteoric water as a highly depleted freshwater endmember. The zero-200 salinity intercepts extrapolated from these lines represent the mean  $\delta^{18}$ O properties of basal ice being melted by warm mCDW. This approach differs somewhat from other studies (Biddle et al., 2019;

The mCDW is the warmest, saltiest, and least δ<sup>18</sup>O-depleted water mass in the region and comprises the vast majority of the overall water column. Interannual changes in mCDW inflow will result from variable wind forcing (Dotto et al., 2019; Holland et al., 2019; Kim et al., 2021), combined with onshelf lateral and vertical mixing. While mCDW incorporates a range of salinities, temperatures, and δ<sup>18</sup>O values, in the Amundsen Sea it is the warmest, saltiest, and deepest water mass. In the 3-endmember mixing model, mCDW and meteoric waters are defined by the mixing line of data >200m; with mCDW separately for each year (Table 2) based on the δ<sup>18</sup>O valuevalues at the salinity maximum (Biddle et al., 2017) and meteoric water the 0-salinityzero-salinity intercept on the mixing line (Results 3.1; ).mCDW glacial meltwater mixing line (Figure 2). Sea ice endmember isotopic values adopted from previous studies in the Amundsen and Bellingshausen region (Meredith et al., 2008, 2010, 2013; Randall-Goodwin et al., 2015; Biddle et al., 2019)(Randall-Goodwin et al., 2015; Meredith et al., 2008)

Meredith et al., 2010) that estimate meteoric water content from approximate glacier 8<sup>18</sup>O values.

are based on the  $\delta^{18}$ O of surface water with an offset to account for isotopic fractionation ( $\sim +2.1\%$ ) due to freezing.

Table 22: Salinity and  $\delta^{18}$ O values used in the 3-endmember mixing model. mCDW and meteoric components are defined independently using the mCDW-GMW mixing line produced from  $\geq 200$  m depth) salinity and  $\delta^{18}$ O observations for each year, as the salinity maximum and  $\frac{0}{2}$  salinity maximum and  $\frac{0}{2}$  salinity intercept, respectively (Results 3.1; (Figure 2; Appendix A1). A1). Sea ice melt uses single values for each year. Salinities are reported as absolute salinity (g/kg).

Year	mCDW salinity (g/kg)	mCDW δ <sup>18</sup> O (‰)	Meteoric water (GMW) δ <sup>18</sup> Ο (‰)	Sea ice melt salinity (g/kg)	Sea ice melt δ <sup>18</sup> O (‰)
1994	34.86±0.010	-0.01±0.01	-29.4±1.4		
2000	34.86±0.011	$-0.05\pm0.01$	$-28.7 \pm 0.6$		
2007	34.89±0.009	$-0.02\pm0.02$	$-28.4\pm0.8$		
2009	34.87±0.002	$0.01 \pm 0.02$	-29.1±0.6	7	2.1
2014	34.85±0.015	$0.08\pm0.01$	-31.3±0.5		
2019	34.88±0.006	$-0.09\pm0.01$	$-30.0\pm0.4$		
2020	34.87±0.017	$-0.10\pm0.01$	$-29.1\pm0.2$		

#### 3 Results

## 3.1 Meteoric waters defined by the $\delta^{18}O$ – salinity relationship

Freshwater endmembers (zero-salinity δ¹8O intercepts) over the seven sampled summers differ by
≤3‰, ranging from -28.4‰ to -31.3‰ with a standard deviation of 1.0‰ (, Table 2). These measurements are consistent with the nearest ice-core (ITASE01-2, from 77.84°S, 102.91°W, Figure 1a; Schneider et al., 2006; Steig et al., 2005Deep introduction of GMW is consistent with subglacial melt) with a mass-averaged δ¹8O value of -29.8±1.9‰. Ice cores further east have less negative δ¹8O values (~-20‰; Thomas et al., 2009), while those further west are more negative (~-40‰; Blunier & Brook, 2001). Intercept uncertainty from analytical precision and environmental variability (Appendix A3) ranges from ±0.2‰ in 2020 to ±1.4‰ in 1994, varying inversely with the number of data points available.

Local precipitation, in the form of snow collected in early 2019 at 72.5°S, yielded a δ<sup>18</sup>O value of ~- 15‰, consistent with previous observations and model outputs at sea-level from that latitude (Masson-Delmotte et al., 2008). Endmember extrapolations from the regional salinity and δ<sup>18</sup>O measurements indicate that freshwater added to the water column is dominated by locally derived GMW, as intimated earlier from 1994 data (Hellmer et al., 1998).

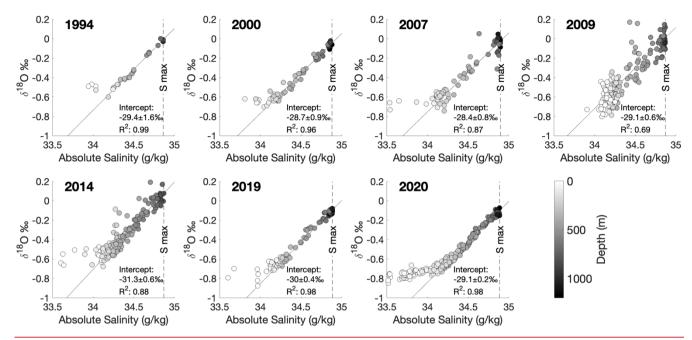


Figure 2: Salinity vs δ<sup>18</sup>O plots for each year, shaded by depth. Linear regressions (solid) gray lines (with R² from 0.69 in 2009 to 0.99 in 1994) project to zero-salinity glacial meltwater endmember intercepts using data >200m. Dashed vertical lines indicate the mCDW salinity maxima (Table 2). Data diverge from the mCDW-meteoric water mixing line in the upper water column, where sea ice melt freshens the resultant mixture but has an enriching effect on δ<sup>18</sup>O (Table 2). Years with greater divergence at the surface have more sea ice melt (Appendix A6). The most negative upper water column seawater δ<sup>18</sup>O measurements tend to reach minima between -0.9% and -0.6%.

Samples below 200 m show a strong  $\delta^{18}$ O-salinity relationship, forming a mixing line between mCDW and a (glacial) meteoric freshwater endmember introduced at depth. Closer to the surface (from 10 m in 2009 to 160 m in 2000) data diverge from the mixing line due to the net influence of sea ice melt and local precipitation, moving the  $\delta^{18}$ O of the mixture in a more positive direction. Below 200m the  $\delta^{18}$ O-salinity relationship is strongly linear; greater scatter in 2009 and 2014 likely results from sample storage issues (**Appendix A7**).

In 2009 and 2014, samples were collected in bottles with taped (2009) or parafilm-wrapped (2014), threaded caps, and stored for several years before analysis. Samples from 2019 and 2020 were collected in glass serum vials capped with rubber stoppers and aluminum seals (**Appendix A9**), which internal lab data demonstrate the maintenance of seawater  $\delta^{18}$ O sample integrity for 5+ years. Scatter occurs mostly in the positive  $\delta^{18}$ O direction (particularly in 2014), indicative of sample evaporation.

## 1.43.2 Vertical distribution of meteoric water illustrates basal melt

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A 3-endmember mixing model of mCDW, sea ice melt, and meteoric water <u>iscan be</u> used to determine the constituent freshwater components of seawater (<u>Equations 1</u>-3) at all depths sampled in the water column (<u>Figure 3</u>Figure 3). <u>By using We compare the derived GMW fractions that are</u>

ealculated from separately defined mCDW and meteoric waterGMW values for each year. Using mCDW and GMW endmember values based on annual data from each year individually, minimizing the potential impact of mean the calculated GMW fractions are extremely unlikely to be impacted by analytical calibration offsets between laboratories on the calculated meteoric water fractions. (Data and Methods 2.1; Appendix A2).

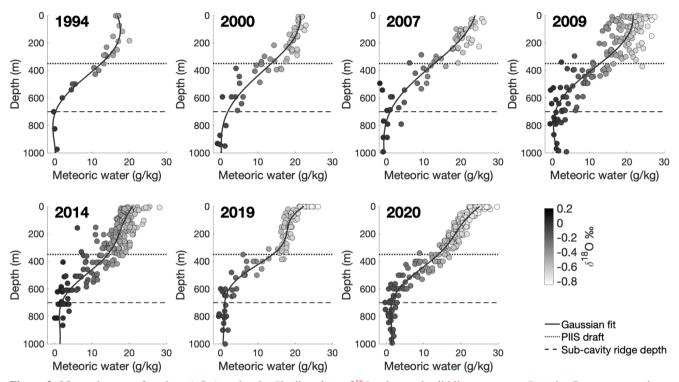


Figure 3: Meteoric water fractions (g/kg) vs depth. Shading shows  $\delta^{18}$ O value, and solid lines represent Gaussian Process regression fits. Dotted and dashed lines show the depth of the PIIS draft and sub-cavity ridge (Jenkins et al., 2010). Slightly negative meteoric water values at depth occur when the sample exceeds the mCDW endmember value (Table 2; Data and Methods 2.2), the result of environmental, sampling and analytical uncertainties (discussed in further detail in Results 3.4).: Glacial meltwater fractions (g/kg) in the SE Amundsen Sea water columns. Red lines represent Gaussian Process regression fits. Small negative GMW values at depth in some years are mathematical artifacts that occur when the sample  $\delta^{18}$ O is higher than that of the mCDW endmember (Table 2, Data and Methods).

Evidence of highly  $\delta^{18}$ O-depleted freshwater is found at depths above ~700 m (the depth of PIIS sub-ice shelf ridge; Jenkins et al., 2010), with highest concentrations found at depths shallower than 350 m – above which glacial meltwater has been observed to flow out from beneath the ice shelf (Biddle et al., 2017; Naveira Garabato et al., 2017). Made less dense by the addition of GMW, such outflows rise through denser waters above, along ice shelf calving fronts and strongly influencing surface waters in this region Evidence of highly  $\delta^{18}$ O-depleted freshwater is found at depths shallower than 800 m, increasing rapidly upward from ~600 m to the surface or its mixed layers (**Figure 3**). The mean depth of the Pine Island Ice Shelf calving front was about 400 m in 2009 (Jenkins et al., 2010). Outflows from such depths, lightened by the addition of GMW, rise through denser waters above, along ice shelf

calving fronts (Dierssen et al., 2002; Mankoff et al., 2012; Thurnherr et al., 2014; Fogwill et al., 2015).

Depending on the extent of mCDW heat remaining after basal melting, that will contribute to the formation of coastal polynyas (Mankoff et al., 2012) and surface layer δ<sup>18</sup>O minima. Where GMW ends up in coastal water columns depends on its production rate, impact on the density of circulating water, and access of return flows to the ice shelf calving fronts.

Sea ice melt and mCDW fractions are discussed in **Appendix A6**.

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## 1.53.3 Average meteoric water GMW inventory over the last two decades

Average meteoric water column inventories (**Table 3**) in the study area were estimated by depth integrating the Gaussian Process fit of the calculated meteoric water fractions (solid lines in **Figure 3**). The average meteoric water column inventory was relatively low in 1994 and higher from 2000-2020. Strongly influenced by the low meteoric water inventory in 1994, a linear regression of the mean meteoric water inventories suggests an increase of 0.03±0.02 m/y (p-value 0.28). Meteoric water column inventories have uncertainties of <0.8 m, based on analytical precision and environmental variability of the model inputs (**Results 3.4**). Assuming 1 m water equivalent (2 years) of precipitation (Boisvert et al., 2020; Donat-Magnin et al., 2021), the meteoric water inventories are likely to consist of >92% GMW (**Appendix** Error! Reference source not found.).

Regional average GMW column inventories were estimated by depth integrating the Gaussian Process fit of the calculated meteoric water fractions (red lines in Figure 3). The average GMW inventory was relatively low in 1994 and high in 2000 and 2020, with an uncertainty of <0.7 m in an overall average of 8.60 m; ~1% the upper 800 m water columns. Though largely a product of the low GMW inventory in 1994, a linear regression of the mean GMW inventories produces a modest increase of 0.02 m/y. Observations indicate the mCDW responsible for basal melt does not exhibit significant seasonal variability in this region (Mallett et al., 2018), and A related study showed mostly invariable overall melt rates during austral summer (Kimura et al., 2017), when the samples used in this study were collected (Table 1).

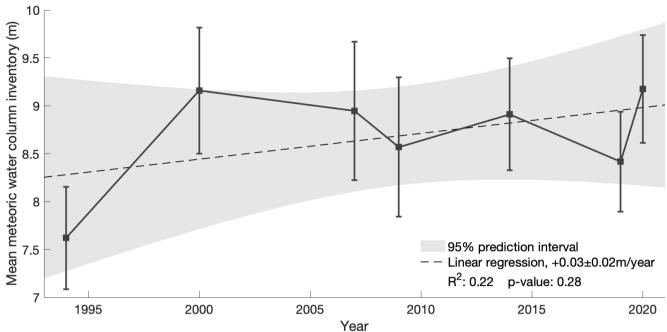


Figure 44: Average meteoric water GMW-column inventory in study area. Depth-integrated meteoric water content GMW volume from the Gaussian Process fit (see-Figure 3Figure 3) between the sea surface and 800 m-depth. Error bars show the uncertainty (Table 3) in mean GMW column inventory associated with data volume, analytical precision, and uncertainty in endmember values (Data and Methods 2.2). A linear regression of the mean values shows an average increase of 0.03±0.02 m/year (p-value 0.28). Grey shading shows the 95% prediction interval for the linear regression.

## **1.63.4** Uncertainty and sensitivity analyses

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## 3.4.1 Analytical precision and environmental variability

Comparing GMW content and δ<sup>18</sup>O-salinity relationships between different groups of stations, we found that sample location variability had an insignificant impact on calculated average meteoric water content, except for 2014, when the stations along the edge of TGT had considerably higher GMW content. In the interest of improving interannual comparability, 2014 stations closest to TGT were excluded from the average GMW content analyses (Appendix A4).

We <u>ranused</u> 10,000 Monte Carlo simulations where observations were perturbed around their measured value by on each data point to test for sensitivity to variance in mCDW properties, endmember uncertainty <u>associated with</u>, and analytical precision (0.04% δ¹8O for IRMS; 0.02% δ¹8O for CRDS; 0.002 g/kg salinity) and water source endpoints (mCDW, sea ice melt, meteoric water). mCDW and meteoric water endpoints were defined using perturbed observational data for each simulation, and around the selected endpoint by in the 3-endmember GMW fraction model. Each observed value and water mass endpoint was varied around the reported value by the analytical and environmental

uncertainty associated with that tracer (Appendix A3). Zero-salinity intercept (meteoric endmember) uncertainty associated with environmental variability (Table 3, Appendix A3). analytical precision ranges from  $\pm 0.48\%$  in 2020 to  $\pm 1.62\%$  in 1994, varying inversely with the number of data points available for the extrapolation. Uncertainty in mean meteoric water GMW-fractions ranges from 1. was  $\pm 1$  g/kg in 2019 to .5—1.7 g/kg in 2007/, corresponding to  $\pm 0.5$ —0.7 m ( $\pm 6$ —8%) 2009, and uncertainty in mean meteoric column inventories ranges from 6-9%.

water column GMW inventory (Gaussian Process fit, red lines in **Figure 3**; Appendix A3) with 2009 exhibiting the greatest uncertainty, and 2020 the least. Calculated water GMW fractions are most strongly influenced by changes made to the mCDW endmember (comprising ~99% of an 800m water column on average; ~95% in surface waters rich in meteoric water and sea ice melt). Meteoric water fractions vary inversely with the magnitude of the meteoric water endmember δ<sup>18</sup>O. 1994 has the fewest samples, but the strongest fit (), and exhibits similar uncertainty to other years (**Figure 4**).

Table 3: Meteoric water column inventory analytical and environmental uncertainty. Depth-integrated meteoric water content using the Gaussian Process fit (Figure 3) between the sea surface and 800 m depth. Uncertainties are associated with number of data points, analytical precision, and environmental variability in endmember values (Data and Methods 2.2).

Year	Meteoric water fraction uncertainty (g/kg)	Meteoric water column inventory (m)	% meteoric water column inventory uncertainty
	uncertainty (g/kg)		
<u>1994</u>	<u>1.5</u>	$7.6 \pm 0.5$	<u>7.0%</u>
<b>2000</b>	<u>1.7</u>	$9.2 \pm 0.7$	<u>7.2%</u>
2007	1.7	$8.9 \pm 0.7$	8.1%
2009	1.7	8.6±0.7	8.5%
2014	1.5	$8.9 \pm 0.6$	6.6%
2019	1.1	8.4±0.5	6.1%
2020	1.2	9.2±0.6	6.1%

## 3.4.2 Spatial variability

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This study relied on the compilation of data collected for 6 independent studies over 7 different cruises. To determine the impact of inconsistency in sampling locations each year, two different spatial sensitivity analyses were used. First, we conducted 10,000 simulations, for each year selecting random sets of 3 stations within the field area. In each case, mCDW and meteoric water endmembers, and mean meteoric water column inventories were calculated using only those data. Uncertainty is represented as the standard deviation of those results (**Table 4**).

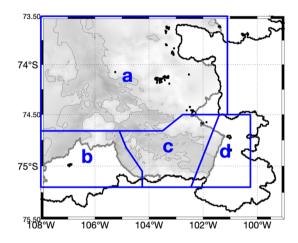
Table 4: Results of randomized spatial sensitivity analysis. Uncertainty is represented by the standard deviation of the results of 10,000 simulations for each year, calculating results using data from only 3 randomly selected stations.

	mCDW		mCDW	<u>Meteoric</u>	<u>Average</u>	<u>Meteoric</u>
	<u>absolute</u>	<b>mCDW</b>	<u>potential</u>	<u>water δ<sup>18</sup>O</u>	<u>meteoric</u>	water fraction
	salinity	$\delta^{18}O$ (% vs	temperature	(% vs	water column	uncertainty
<b>Year</b>	(g/kg)	VSMOW)	(°C)	VSMOW)	inventory (m)	<u>(g/kg)</u>

1994*	<u>34.86±0</u>	$-0.01 \pm 0.01$	$1.08\pm0.01$	$-29.4\pm0.6$	$7.6 \pm 0.2$	<u>0.1</u>
<b>2000</b>	$34.86 \pm 0.012$	$-0.06\pm0.02$	$1.03\pm0.07$	$-28.7\pm1.6$	$8.8 \pm 0.5$	<u>0.8</u>
<b>2007</b>	$34.89\pm0.009$	$-0.03\pm0.04$	$1.18\pm0.03$	$-27.8\pm3.4$	$9.0 \pm 0.7$	<u>1.1</u>
2009	$34.87 \pm 0.002$	$-0.01\pm0.06$	$1.17 \pm 0.01$	$-28.9\pm4.1$	$8.8 \pm 0.7$	<u>1.8</u>
<u>2014</u>	$34.85\pm0.015$	$0.05 \pm 0.03$	$1.10\pm0.04$	$-31.2\pm3.3$	$8.5 \pm 0.8$	<u>1.1</u>
<u>2019</u>	$34.88 \pm 0.006$	$-0.10\pm0.01$	$1.08\pm0.06$	$-29.6\pm1.6$	$8.5 \pm 0.4$	<u>0.6</u>
2020	$34.87 \pm 0.017$	$-0.12\pm0.02$	$1.03\pm0.01$	$-29.0\pm1.4$	$8.8 \pm 0.6$	<u>0.7</u>

\$\frac{8}{48}\$ 1994 has only 4 sampling locations, and the strongest fit of any year () its uncertainty may be artificially decreased

Additionally, we conducted a spatial sensitivity analysis by separately analyzing different spatial groups of stations across each year (Figure 5, Table 5), running 10,000 Monte Carlo analyses for each group as described in Results 3.4.1. Uncertainty is represented as the standard deviation of those results (Table 5, Table A2).



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Figure 5:Boundaries of geographic groupings used for spatial sensitivity analysis. Blue lines show boundaries of geographic areas analyzed separately. Gray shading shows bathymetry, with isobaths drawn at 800m. More detailed maps for each year are in Appendix A4.1.

Table 5: Summarized results of meteoric water column inventory (m) from spatial sensitivity analysis (Figure 5). Results by year in rows, and by area group in columns. See Appendix A4.1 for further detail.

	<u>a</u>	<u>b</u>	<u>c</u>	<u>d</u>
<u>1994</u>	6.3±0.5*	Ξ.	Ξ	$8.0 \pm 0.6$
2000	$9.2 \pm 0.7$	Ξ	Ξ	Ξ
<u>2007</u>	8.4±0.8	10.3±0.8*	$9.0\pm0.7$	Ξ
2009	7.0±0.6*	Ξ	$8.7 \pm 0.8$	$8.5 \pm 0.7$
<u>2014</u>	$8.3 \pm 0.6$	$10.1 \pm 0.7$	$7.9\pm0.5$	$7.5\pm0.5$
<u>2019</u>	8.2±0.6	$10.0 \pm 0.7$	$8.2 \pm 0.5$	Ξ
<u>2020</u>	$8.7 \pm 0.6$	Ξ	$8.7 \pm 0.5$	$8.5 \pm 0.5$
. = -			4.0000	

<sup>\*</sup> Data from only one station. For 1994, 2007, and 2009, result is based on only 8, 4, and 4 samples, respectively.

Both sets of spatial sensitivity analyses show little spatial variability in calculated endmember values or mean meteoric water column inventories, possibly excepting 2009, where there are known sample quality concerns. Mean meteoric water column inventories are remarkably consistent spatially, except

those calculated from stations in Group b alongside the TIS, which showed significantly higher meteoric water inventories than the rest of the study area. Although only 1 and 2 stations alongside the TIS were sampled in 2007 and 2019, the average column inventories are consistent with the 2014 data, where there were 8 stations in Group b. The higher inventories in Group b are suggestive of an accumulation of basal melt, and consistent with findings from another study showing that basal melt from beneath PIIS ends up along the eastern edge of TIS (Wåhlin et al., 2021). The relative insensitivity of sampling location (except for those alongside TIS) in calculated mean meteoric water column inventories suggests that precise reoccupation may be unnecessary, and potentially that a relatively small number of stations/samples could be used to reliably assess mean meteoric water column inventory in this region.

#### 4 Discussion

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## 4.1 The utility of zero-salinity $\delta^{18}$ O intercepts

The meteoric water endmember has been described as the least well-constrained component (Meredith et al., 2008, 2010, 2013; Randall-Goodwin et al., 2015; Biddle et al., 2019) of the three-endmember mixing model employed here, leading previous studies to use plausible mean meteoric δ¹8O values, falling between the δ¹8O value of glacial melt and local precipitation. However, δ¹8O-salinity plots presented here () show mixing diagrams of different water sources in the SE Amundsen Sea and suggest that the meteoric endmember for waters below the surface mixed layer can be well-constrained. mCDW is the saltiest, and least-depleted water, while glacial meltwater is fresh and the most depleted in δ¹8O. Data from deeper than 200 m fall along a mixing line between mCDW and a meteoric freshwater source with a δ¹8O value indicative of local glacial freshwater (Steig et al., 2005; Schneider et al., 2006), with lower uncertainty than the mass-weighted standard deviation of the average δ¹8O of the ITASE01-2 ice core ().

The 2014 data here was previously used in another study (Biddle et al., 2019), where they selected - 25% as a meteoric water  $\delta^{18}O$  endmember. In comparison to the meteoric endmember used in this study (-31.3% for 2014), a -25% endmember results in overestimating mean (800 m water column) meteoric water content by 2 m (23%), and underestimating mean sea ice melt by 2.9 m – changing the signal from one of net sea ice melt (this study) to one of net formation (Biddle et al., 2019). The -25% endmember was selected as a "mean" meteoric water endmember midway between the  $\delta^{18}O$  of GMW and precipitation as defined in another study (Randall-Goodwin et al., 2015). We estimate that >92% of the meteoric water in the study area is GMW (**Discussion 4.2**, **Appendix** Error! Reference source not found.). Determining the meteoric water endmember using the better-constrained zero-salinity  $\delta^{18}O$  intercept of the sample data mixing line (**Data and Methods 2.2**) provides more accurate meteoric water and sea ice melt fractions. The length of the intercept extrapolation emphasizes the importance of careful sample collection, storage, and high-precision analyses.

### 4.2 Basal meltwater and precipitation

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Precipitation grows increasingly depleted in <sup>18</sup>O with latitude (Dansgaard, 1964; Gat and Gonfiantini, 1981; Ingraham, 1998; Masson-Delmotte et al., 2008) and altitude (Dansgaard, 1964; Friedman and 420 Smith, 1970; Siegenthaler and Oeschger, 1980; Ingraham, 1998; Araguás-Araguás et al., 2000; Sato and Nakamura, 2005; Masson-Delmotte et al., 2008). Most (88%) of spatial variation in the  $\delta^{18}$ O of Antarctic precipitation can be explained by linear relationships between latitude, elevation, and distance from the coast, with elevation being the primary driver (Masson-Delmotte et al., 2008). Precipitation collected during the NBP19-01 cruise in the study had a  $\delta^{18}$ O value of -15%, consistent with other data 425 from that latitude and elevation (Gat and Gonfiantini, 1981; Ingraham, 1998; Noone and Simmonds, 2002; Masson-Delmotte et al., 2008). Precipitation collected at Halley Bay (75.58°S, 20.56°W, 30m elevation) has an average composition of -22.0±5.6‰, while that collected at Rothera Point (67.57°S, 68.13°W, 5m elevation) has an average composition of -13.5±3.4‰, and precipitation collected at Vernadsky (65.08°S, 63.98W, 20m elevation) has an average composition of -10.2±3.0% (Global 430 Network of Isotopes in Precipitation (GNIP), 2023).

The nearest Ice cores to our site (ITASE01-2, Steig et al., 2005; Schneider et al., 2006; Siple, Mosley-Thompson et al., 1990) have average  $\delta^{18}$ O compositions of -29.6±1.6‰. Using locally collected salinity and  $\delta^{18}$ O data from deeper than 200 m to calculate a zero-salinity intercept, we identify average freshwater endmembers ranging from -31.3±0.8‰ to -28.4±0.8‰ (average 29.3±0.7‰). The similar zero-salinity intercept, and strong linear salinity-  $\delta^{18}$ O relationship below 200 m demonstrates that glacial freshwater is responsible for the observed freshening signal. We find roughly half of the total meltwater inventory in the upper 200 m, below which inventories yield the same general trend in interannual variability (**Appendix A5**). This indicates the observed variability results from changes in glacial meltwater content, and not from interannual variability in local precipitation.

The Pine Island/Thwaites area receives ~0.5 m/y (water equivalent) of precipitation (Donat-Magnin et al., 2021), and the residence time of deep shelf waters here is ~2 years (Tamsitt et al., 2021). We recalculated our meteoric water column inventories assuming 2 years' worth of local precipitation (δ<sup>18</sup>O -15‰) in the upper 200 m, and find that on average, the addition of precipitation decreases the meteoric water (δ<sup>18</sup>O ~-30‰, **Table 2**) content by 0.55±0.01 m, and increases sea ice melt by 0.57±0.03 m, indicating that >92% of the meteoric water column inventory consists of GMW (**Appendix A5**). A substantial fraction of local (and non-local) precipitation will be deposited on sea-ice, much of which is subsequently advected out of the study area (Assmann et al., 2005), and as a result have no impact on locally measured meteoric water content, suggesting that GMW could comprise an even greater fraction.

While previous studies (Biddle et al., 2019; Meredith et al., 2010) have excluded the upper water column from GMW accounting, due to uncertainty surrounding the impact of local precipitation, our results suggest realistic (glacial) meteoric water content can still be estimated in the upper water column. Nearly half (~4 m) of the water column meteoric water content resides in the upper 200 m, and >92% of water column meteoric water is comprised of glacial meltwater (Appendix A5). Discounting

this upper water column meteoric water unnecessarily hampers the usage of this technique for glacial meltwater accounting. The potential for overestimating (glacial) meteoric water due to precipitation is dwarfed by the potential for underestimating glacial meltwater content by excluding the upper water column.

#### 4.3 Temporal changes in mean meteoric water column inventories

- We have estimate average meteoric water column inventories in the SE Amundsen Sea using seawater oxygen isotopes and salinity in a three-endmember mixing model. In 1994, 2007, 2014, and 2020, there is a tendency for the maximum integrated meteoric water volume to extend westward from the SW corner of the PIIS, and along the eastern TIS (Figure 1), consistent with the gyre-like circulation there (Thurnherr et al., 2014). This pattern of meteoric water distribution is consistent with local GMW patterns previously observed using traditional hydrographic tracers (Thurnherr et al., 2014; Naveira Garabato et al., 2017; Wåhlin et al., 2021).
- Local meteoric water content varies from a low of 7.6±0.5m in 1994 to highs of 9.2±0.6m in 2000 and 2020. Inventories fluctuated over the latter period, without apparent trend, dependent on the spatial and temporal coverage of available datasets. While salinity and δ¹8O alone cannot be used to determine basal melt rates, the average meteoric water inventories are sufficient to identify relatively small changes in melt rates, assuming a constant residence time. The inventories are consistent with other studies suggesting relative stability in recent decade-scale glacial melt rates with significant interannual variability (Paolo et al., 2018; Dotto et al., 2019; Adusumilli et al., 2020; Flexas et al., 2022). A recent modelling study shows an increase in basal melt through the 1990s, followed by relative stability from 2000-2020 (Flexas et al., 2022).
- The mCDW entering the SE Amundsen Sea and accessing the underside of the ice shelves has been shown to exhibit little seasonal variability, with a maximum variance in T of <0.1°C, salinity of <0.05 g/kg, and thickness of <50 m (Mallett et al., 2018). All samples used in this study were collected from 12 January to 15 March, while melt rates for the PIIS and TIS exhibit very little seasonal variability (Kimura et al., 2017). With a residence time of ~2 years (Tamsitt et al., 2021), it is unlikely that the variability in yearly meteoric water column inventories is a product of a seasonal signal.
- Glacial meltwater measured in the SE Amundsen Sea includes mCDW-driven basal melt, local iceberg melt, and meltwater entering the ocean at the grounding zone that is driven by the geothermal heat flux to the base of the ice sheet (~5.3 Gt/y; Joughin et al., 2009). The greatest uncertainty in using average meteoric water inventories as a means for GMW accounting arises from the poorly constrained residence time of regional shelf waters, as there has been little study of this component. With local circulation generally moving waters westward (Nakayama et al., 2013; Thurnherr et al., 2014; Naveira Garabato et al., 2017; Nakayama et al., 2019; Wåhlin et al., 2021), it is likely that the calculated meteoric water fractions in the study area (with the exception of those on the western side of TIS) are primarily comprised of basal melt from PIIS.

500 Assuming a mean residence time of 2 years (Tamsitt et al., 2021) and GMW comprising >92% of total meteoric water column content (Appendix Error! Reference source not found.) is representative of the whole study area (~30,000 km<sup>2</sup> ocean), we estimate GMW inputs of between 106±17 Gt/y in 1994 to 129±17 Gt/y in 2000/2020. Though empirical, these figures are consistent with satellite-based estimates of mass loss from PIIS via basal melt (Rignot et al., 2013, 2019a), demonstrating the utility of 505 geochemical ocean measurements for estimating ice shelf melt rates. to the mCDW endmember, which comprises >95% of the mixture at any given location.

#### 4 Discussion

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In this study, we estimate average water column Glacial Meltwater in the SE Amundsen Sea using seawater oxygen isotopes and salinity in a three-endmember mixing model. We defined mCDW and GMW signatures separately for each data year and compare GMW fractions rather than 8<sup>18</sup>O values. This this limits way any systematic isotopic offsets between laboratories from affecting the year's calculated GMW fractions (Appendix A2). The results of this study show local meltwater content varying from a low of 7.6 m in 1994 to a high of 9.2 m in 2020. Between 2000-2020, inventories fluctuate, without an apparent trend. Several other studies have also shown relatively stable basal melt rates, fluctuating interannually in this region of Antarctica (Paolo et al., 2018; Holland et al., 2019; Adusumilli et al., 2020; Flexas et al., 2022).

Basal melt in the Amundsen Sea sector is driven by the thickness of the mCDW layer transported onshore, rather than its temperature (Dutrieux et al., 2014; Jenkins et al., 2018). The thickness of the 520 mCDW layer is controlled by local wind forcing of a shelf break undercurrent, coupled to the Amundsen Sea Low. Changes in local wind forcing have decelerated the local undercurrent, decreasing the thickness of the mCDW layer and reducing the heat transport onto the shelf and resulting in an overall cooling of Amundsen Sea shelf waters from 2010 through 2016 (Dutrieux et al., 2014; Webber et al., 2017; Jenkins et al., 2018; Dotto et al., 2020). Grounding line retreat of PHS and TWG had been accelerating, but experienced a slowdown between 2010-2015 relative to the preceding period driven by changes to offshore winds (Christie et al., 2023). Our inventories show a local high in 2000, and a local low in from 2009 2019. A recent modelling study showed an increase in PHS and TWG basal melt between the early 1990s and 2000, followed by relative stability thereafter (Flexas et al., 2022). Our results show an increase in average GMW after 1994, followed by interannual fluctuation from 2000-2020, with average GMW inventories remaining well above 1994 levels. Assuming steady state ocean circulation strength, the relatively steady GMW inventories are consistent with the linear, longer-term freshening trend reported downstream in the Ross Sea (Jacobs et al., 2022).

We calculate average GMW inventories from the surface to 800 m. In most years, there is a tendency 535 for the maximum integrated GMW volume to extend westward from the SW corner of the PHS, and along the Thwaites Glacier Tongue (TGT), consistent with the gyre-like circulation in the area between PHS and TGT. This pattern of GMW distribution is consistent with that previously observed using

- traditional hydrographic tracers (Naveira Garabato et al., 2017). Roughly half of the total meltwater inventory is in the upper 200 m, and inventories below that depth yield the same general trend in interannual variability (Appendix A5), indicating that the observed variability is indeed indicative of basal melt, and not merely an artefact of local interannual variability in precipitation. A spatial sensitivity analysis also shows no significant impact of year to year variation in sampling locations on average GMW inventory (Appendix A4).
- The Amundsen Sea sector receives 0.5 m = 1 m (water equivalent) of precipitation each year (Donat-Magnin et al., 2021) and the residence time of deep shelf waters in this region is ~one year (Tamsitt et al., 2021). With a local precipitation δ<sup>18</sup>O of -15‰, even an entire year's precipitation remaining in the upper water column at the time of sampling would only account for ~3% 7% of the overall column inventories (Appendix A5). Furthermore, a substantial fraction of local precipitation will be deposited on sea-ice, much of which is subsequently advected out of the study area (Assmann et al., 2005). While previous studies (Biddle et al., 2019; Meredith et al., 2010) have excluded the upper water column due to uncertainty surrounding local precipitation, our results suggest realistic glacial meltwater content can still be estimated in near surface seawater.

#### 5 Conclusion

- We use used a time-series of seawater  $\delta^{18}$ O and salinity data collected in the SE Amundsen Sea from 555 1994 to 2020, using salinity and δ<sup>18</sup>O-to calculate water column inventories of meteoric (fresh) water through the GMW. The average water column. Freshwater intercepts from  $\delta^{18}$ O-salinity plots produce a well-constrained meteoric water endmember consistent inventory of GMW was lowest in 1994 and highest in 2000 and 2020, with measurements from regional ice cores, and indicative of glacial enough uncertainty in meltwater. While limited by sampling years and number of observations, meteoric water 560 (which we estimate inventories and interannual variability to be >92% GMW) measured in 1994 is lower than measurements made from 2000-2020, where meteoric water content averaged 8.9±0.3m, with<del>render</del> a maximum difference of 0.75m. linear increase of 0.02 m/y statistically insignificant. These results are consistentalign with recent studies showing an increase in basal melt through the 1990s, 565 followed by relative stability and interannual variability from 2000 throughin basal melt rates in this region. Our results also suggest a lower melt period through the 2000s, followed by the highest melt year in 2020. A linear increase of 0.03±0.02 m/y over the study period is insignificant, with interannual variability that is larger than the increasing trend in meteoric water content.
- The WAIS is an important region for understanding future global sea level rise, as changes in winds and particularly with increasing ocean circulation can increase temperatures driving basal melting of ice shelves, melt and increasing the flow of their its ice streams into the sea. Changes in meteoric water inventories in the SE Amundsen Sea study region More recent remote sensing and modelling studies are consistent with satellite-based estimates of annual mass loss from the PIIS. more indicative of interannual variability than acceleration in WAIS melt. The meltwater inventories calculated here from seawater δ<sup>18</sup>O observations also suggests a relatively stable basal melt rate in the SE Amundsen Sea,

with interannual fluctuations potentially masking an increase over 2.6 decades. These results demonstrate the independent utility of seawater  $\delta^{18}O$  and salinity data as an independent method for estimating ice shelf basal melt rates. Regular sampling for  $\delta^{18}O$  in monitoring and helping to better constrain satellite based estimates of basal melt and salinity in this region could reveal if the existing record and its variability will extend into an era when ice shelves are likely to be thinner, with their grounding lines deeper and farther south glacial change. Integration of  $\delta^{18}O$  data into numerical models could also should further our understanding of ocean circulation strength and ice loss along this climatically sensitive sector of the WAIS. Continued sampling for  $\delta^{18}O$  in this region could also reveal if and when our measured rates of meltwater volume, already consistent with downstream freshening, rise significantly above the observed short-term variability.

#### **APPENDIX**

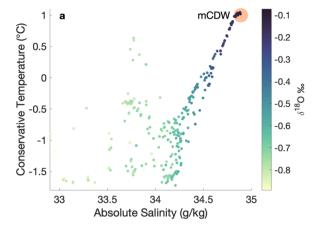
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### 590 A1 Defining mCDW

Modified Circumpolar Deep Water (mCDW) is one of three endmember waters <u>we useused</u> in a mixing model to determine glacial meltwater fractions. As the salinity and  $\delta^{18}O$  of mCDW are well observationally well constrained, <u>with and display</u> interannual variability <u>and, mCDW</u> properties <u>that</u> are defined separately for each year, <u>as in-Figure A1 with 2020 data. Error! Reference source not found. illustrates that process, using 2020 data. Being the warmest, saltiest water on the continental shelf, mCDW appears at the top-right on a T-S diagram (Panel a), where it also identifies waters that are <u>the</u> least-depleted in  $\delta^{18}O$ .</u>

In <u>PanelsPanel</u> b <u>and c</u>, the same <u>2020</u> data <u>showare plotted</u>, <u>with</u> the <u>keystone positions of mCDW iny-axis showing δ<sup>18</sup>O, and the colors indicating</u> temperature/<u>salinity/δ<sup>18</sup>O/depth space.</u> The red <u>and (blue)</u> dashed lines show property <u>mixingmixng</u> lines between mCDW<sub>2</sub>-and glacial meltwater (<u>GMW</u>) and or sea ice melt, with the colder waters being fresher and more depleted in δ<sup>18</sup>O.

Panel c covers the same dataset, but with colors illustrating depth. Most data arclie above  $\sim 800 \text{ m} \times 800 \text{ m}$ , with the least  $\delta^{18}$ O depletion in a few deepdeeper depressions. Waters that fallare diverted off the mCDW-GMW mixing line in the upper 200m have been 200 m of the water column, influenced by sea ice melting/formation and atmospheric processes. Sea ice melt will freshen saltier waters but has a slightly positive (+2.1%)  $\delta^{18}$ O, while GMW has a very negative ( $\sim -30\%$ )  $\delta^{18}$ O. Both freshen seawater, with the sea ice melt slightly counterbalancing  $\delta^{18}$ O ( $\sim 29\%$ ). Its influence makes resultant mixtures fresher, but somewhat counterbalances the strong negative  $\delta^{18}$ O of GMW.



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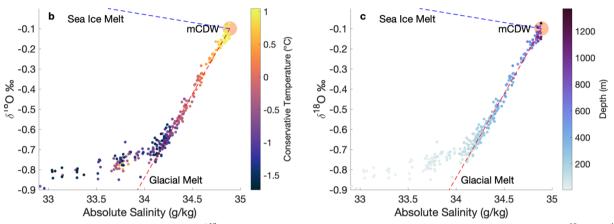


Figure A1: Temperature, salinity, and δ<sup>18</sup>O and depth from 2020 data. Ae) T-S diagram with colorbar showing δ<sup>18</sup>O. b) δ<sup>18</sup>O vs salinity with colorbar showing temperature. Ce) δ<sup>18</sup>O vs salinity with colorbar showing sample depth. The most saline samples are also the warmest, deepest, and least depleted in δ<sup>18</sup>O. Data divergediverges from the mCDW-glacial melt mixing line at depths shallower than 200 m due to the presence of sea ice melt in the admixture. In Panels b and c, dashed lines show the associated property mixing lines for mCDW mixing with sea ice melt, or GMW.

In Figure 2 of the main text,  $\delta^{18}$ O-salinity plots for each year reveal several data points near the the salinity maximum, with some variability in the corresponding  $\delta^{18}$ O. Below Using only data from below 200 m, trendlines that are extrapolated to zero0-salinity intercepts define the mCDW and meteoric water (GMW) endmembers used in the mixing model. mCDW and meteoric water GMW  $\delta^{18}$ O are defined atas the  $\delta$  salinity maximum and 0-salinityzero-salinity intercepts of the trendlines intercept on the trendline, respectively (Table 2Table 2). The mCDW location of mCDW corresponds to conventional measures of the deepest and warmest waters on the continental shelf. The calculated zero0-salinity intercept values are consistent with the properties of locally available GMW.

## A2 Inter-laboratory offsets

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<u>δ</u><sup>18</sup>O When using data from produced by different laboratories are subject, it is important to possible be aware of the potential for systematic offsets. For example, a ~0.1%  $\delta$ <sup>18</sup>O offset between the 2014 data and other years (**Figure A2**) is likely the result of an inter-laboratory calibration offset. On the other hand, greater scatter in the 2009 data suggests that evaporation during sample storage left some samples less depleted in  $\delta$ <sup>18</sup>O. Here, we primarily compare have compared calculated meteoric water fractions rather than  $\delta$ <sup>18</sup>O values, with mCDW and meteoric water GMW signatures defined separately for each year so that any offset will not affect the values of samples from that year relative to their mCDW/meteoric water GMW signatures.

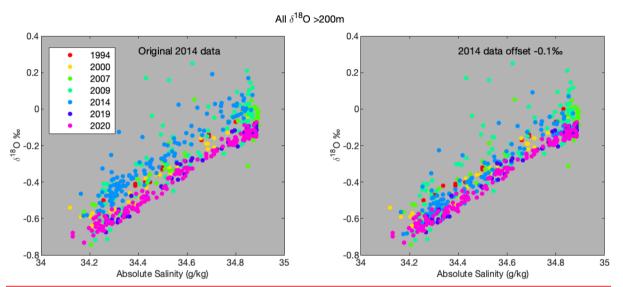


Figure A2:  $\delta^{18}$ O vs Absolute salinity for all years, from data >200 m. Left panel: All  $\delta^{18}$ O vs salinity data, with the 2014 data as published in (Biddle et al., 2019). Right panel: same, with a -0.1% offset correction applied to the 2014 data.

-A sensitivity analysis, wherein all sample data from a given year were offset, and mCDW/meteoric waterGMW signatures were re-calculated using the offset data. The resultoffset data and endmembers were used to calculate meteoric waterGMW fractions in the 3-endmember mixing model, with— sea ice melt values remaining remained static. We found that an inter-lab offset of 5.7%  $\delta^{18}$ O (Figure A3) would bewas necessary to change the calculated meteoric waterGMW fraction by an amount greater than the analytical precision ( $\pm 0.04\%$   $\delta^{18}$ O,  $\pm 0.003$  g/kg for absolute salinity) and environmental uncertainty based on ice core measurements ( $\pm 1.9\%$  for  $\delta^{18}$ O) and year-to-year variability in mCDW values ( $\pm 0.06\%$   $\delta^{18}$ O). Inter) associated with the calculation ( $\pm 1.6$  g/kg). Since, inter-lab offsets should be less would not be expected to be greater than 0.1% (Walker et al., 2016), so any offsets willwould not be significant when comparing calculated meteoric water fractions.

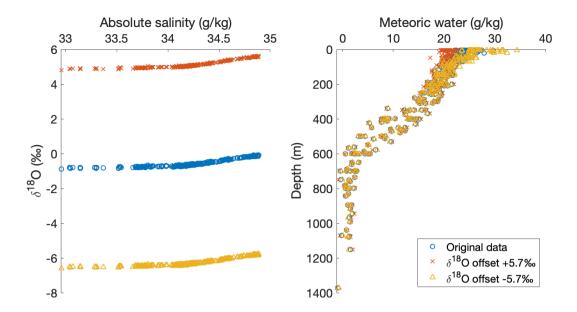


Figure A3: Impact on inter-lab offsets on calculated meteoric (glacial melt) water fraction for NBP20-02 data. The left panel shows the  $\delta^{18}$ O offset ( $\pm 5.7\%$ ) necessary to significantly affect calculated meteoric water GMW fractions, when using mCDW and meteoric water endmembers calculated from that data. The right panel shows the calculated meteoric water fractions produced using the original, and offset data. The calculated meteoric water fractions are impacted very little because two of the three endmembers (mCDW and meteoric water) are defined by the offset data.

## A3 Uncertainty in calculated meteoric water fractions

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Since meltwater fractions are calculated using analytical measures of salinity and  $\delta^{18}$ O, the accuracy and precision of these measurements are important. CTD salinity sensors have a reported precision of ±0.002. The Isotope Ratio Mass Spectrometer (IRMS; 1994 to 2014) measurements have a measured precision of  $\pm 0.04\%$  based on replicates, while the Cavity Ring-Down Mass Spectrometer (CRDS) achieved a precision of  $\pm 0.02\%$ . The meteoric (GMW) endmember is arguably the least-well constrained, with glacial ice in West Antarctica ranging from -20% to -40%, but much of that uncertainty has been eliminated by using the zero-salinity intercept determination on a  $\delta^{18}$ O-salinity mixing line, corroborated by nearby ice core values as discussed in the main manuscript. mCDW is well-constrained, based on many accurate in-situ measurements. Our sea ice melt endmember is adopted from previously published studies in the region (Meredith et al., 2008, 2010, 2013; Randall-Goodwin et al., 2015; Biddle et al., 2019). Since meltwater fractions are calculated using analyticallyderived measures of salinity and  $\delta^{18}$ O, the accuracy and precision of these measurements are important. CTD salinity sensors have a reported precision of ±0.002. The IRMS (1994 to 2014) measurements in this study have a reported precision of ±0.04% based on replicates, while the CRDS achieved a precision of ±0.02‰. Our end-member values for sea ice melt and meteoric water are largely theoretical. The meteoric endmember is arguably the least-well constrained, with glacial ice in West Antarctica ranging from -20% to -40%, but much of that uncertainty has been eliminated by using the 0-salinity intercept determination on a δ<sup>18</sup>O-salinity mixing line, corroborated by nearby ice core values, as discussed in the main manuscript. mCDW is the best constrained endmember and is based on many in-situ measurements.

We use Monte Carlo simulations to estimate uncertainty in our water mass fraction calculations. We ran 10,000 simulations with input values varied randomly within these bounds and represent uncertainty by the standard deviation of the difference between the simulated runs, and the initial runs. 685 Observations<del>run. Each parameter (8<sup>18</sup>O, salinity) was varied around the reported endpoints (**Table 2**)</del> and the measured observational values for each of our samples, by the uncertainty associated with environmental variability, and instrument precision (Table A1). All observations were varied randomly perturbed by analytical precision above, (0.04% 8<sup>18</sup>O; 0.002 g/kg salinity); these perturbations also impacting have an impact the mCDW and meteoric water GMW endmembers for each run. Additional 690 perturbations, since they are calculated from observations >200 m. In addition to perturbations made to the endmember values; sea ice melt based on theoretical values (Rohling, 2013), meteoric water by observations, endmembers are further perturbed. The perturbation used for the glacial meltwater endmember is the standard deviation of the ITASE01-2 ice core,. The perturbations for sea ice melt were selected based on theoretical values (Rohling, 2013), mCDW salinity perturbations were selected based on the data distribution for each year. 8180 was varied by the results of a spatial sensitivity analysis (Appendix A4.2) and  $\delta^{18}$ O was varied by half<del>standard deviation associated with</del> the 95% prediction interval ( $\sim 1\sigma$ ) of produced from the > 200m  $\delta^{18}$ O-salinity relationship at the salinity

maximum. Perturbations used in the uncertainty analysis are summarized in Table A1.; salinity was varied by the standard deviation in salinity of the highest  $\delta^{18}$ O data falling within the 95% prediction interval at the salinity maximum.

**Table A1:** Perturbations for uncertainty analysis. Perturbations are based on analytical precision for observations, the ITASE01-2 ice core for meteoric water, and theoretical values for sea ice melt (Rohling, 2013). mCDW perturbations are based on the 95% prediction interval of the >200m  $\delta^{18}$ O-salinity relationship at the salinity maximum, and salinity perturbations are based on the results of the spatial randomization analysis (**Appendix A4.2**). **Endmember perturbations for uncertainty analysis** 

	Absolute Salinityδ <sup>18</sup> O	
	perturbation	δ <sup>18</sup> OAbsolute Salinity
Parameter Endmember	<u>(g/kg)<del>(‰)</del></u>	perturbation <u>(‰)<del>(g/kg)</del></u>
<u>Observations</u>	<u>0.002</u>	0.04 (0.02 for CRDS)
Meteoric water Glacial		
meltwater (meteoric)	<u>N/A</u> 1.9	<u>1.9</u> N/A
Sea ice melt	2	0. <u>1</u> 11
mCDW, 1994	0.010 <u>*</u>	0. <u>010</u> <del>018</del>
mCDW, 2000	0. <u>011</u> <del>012</del>	0. <u>012</u> <del>011</del>
mCDW, 2007	0. <u>009</u> 018	0. <u>018</u> <del>011</del>
mCDW, 2009	0. <u>002</u> <del>021</del>	0. <u>021</u> <del>008</del>
mCDW, 2014	0. <u>015<del>017</del></u>	0.013
mCDW, 2019	0.006	0. <u>006</u> <del>003</del>
mCDW, 2020	0.017 <del>004</del>	0.004005

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The mean uncertainty in meteoricassociated with GMW water fractions ranges from 1.1 g/kg in 2019

10 to5—1.7 g/kg in 2009,—this corresponding to of ±0.5—0.7 m average meteoric water column inventories uncertainty between ±0.5 m in 2019 inventory. GMW and ±0.7 m in 2009 (Table 3).

Meteoric water and sea ice meltSea Ice Melt fractions vary inversely, while mCDW fractions remain relatively stable. Calculations are most impacted by changes to the mCDW endpoint, as mCDW makes up ~99>951% of the (800m) water column on average; >95% in the meteoric water and sea ice melt rich surface waters, and >98% at all depths below 200 many given location.

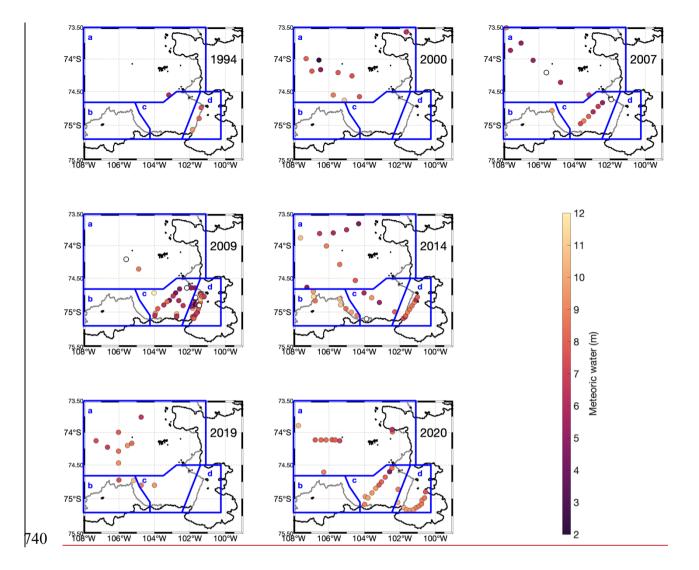
14For a small number of samples from 2007 (3), 2009 (9), and 2014 (2) suggest negative meteoric water the calculated GMW fractions, nine beyond are negative. For the 3 samples in 2007, and 2 in 2009, these figures are within the uncertainty described above. The negative meteoric water GMW fractions result from when high-salinity deep waters with were measured to have a  $\delta^{18}$ O values significantly less negative than the mCDW endpoint in the 3-endmember mixing model, reflective of uncertainty in the data and/or model limitations. Those years also display a used in the GMW calculation. The spatial variability in mCDW, is reasonably well constrained year to year, with a wider spread in mCDW  $\delta^{18}$ O for 2007, 2009, and 2014 than in other years, likely due. The samples in question may have been subject to evaporation during, resulting in a higher  $\delta^{18}$ O, highlighting the importance of appropriate sample collection and storage.

<sup>\*</sup> For 1994, we perturbed salinity by the average salinity standard deviation for the other years, to compensate for the smaller number of samples

# A4 Geographical sensitivity of <u>endmembers and meteoric water column inventories</u> <del>mCDW</del> <del>signature and meltwater inventory</del>

## A4.1 Geographic clustering analysis

We analyzed the spatial sensitivity of results by splitting the study area into four groups and analyzing data from those groups for each year (**Figure A4**, **Table A2**). For each area, mCDW and meteoric water endmembers were defined based on only those data. In 10,000 Monte Carlo simulations, the observations and endpoints were perturbed by uncertainty associated with analytical precision and environmental variability (**Table A1**).



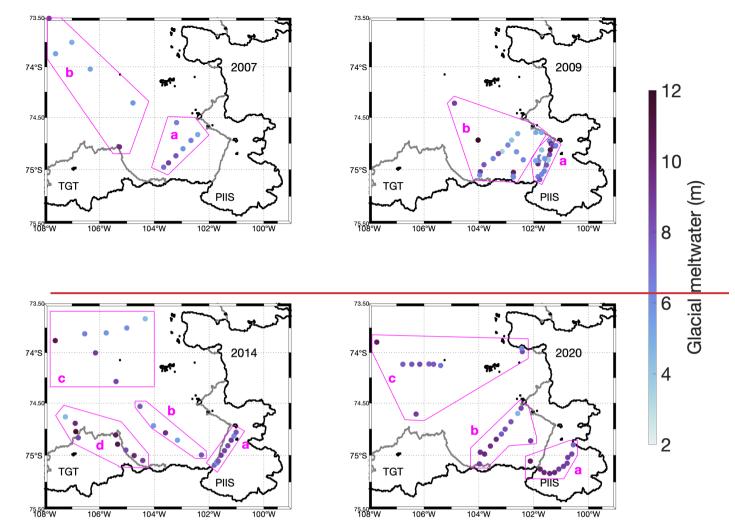


Figure A4: Sampling locations and geographic group boundaries for all years. Dot colors show meteoric water column inventory at individual stations, and outlines showing geographic groupings of stations for geographic sensitivity analysis (Table A2). Some locations provided only partial column inventories. Sampling locations for 4 years and analyzed data groupings. To understand how much year to year differences in sampling location affects mCDW and GMW fingerprinting, and average meltwater inventory calculations, we selected the 4 years with the greatest number of data points and ran these analyses on different geographic groupings of sample data (Table A2).

Table A2: Results of geographic grouping sensitivity analysis. mCDW is defined as the δ18O value at the salinity maximum falling on the linear regression of all salinity-δ18O measurements deeper than 200m in each group of stations; meteoric waterGMW δ18O is defined as the θ salinity intercept on that same line. Uncertainty in mCDW δ18O is represented by half the 95% prediction interval at the salinity maximum (~1 σ), and uncertainty in salinity is the result of the randomization spatial sensitivity analysis, plus variation from perturbation of observations (Appendix A4.2).manufacturer's stated analytical precision. Average meteoric watermeltwater inventory is the depth integration of the Gaussian fit of all calculated meteoric water fractions within each group, with uncertainty represented as the standard deviation in meteoric waterGMW fractions achieved using 10,000 Monte Carlo 1000 Montecarlo simulations perturbing the observations and endpoints by associated analytical and environmental uncertainty (Table A1). For each groupsubset of stations, mCDW and meteoric water endmembers GMW used in meteoric waterGMW calculations endmembers are defined using only those data.

							Average	Mataaria
						Meteoric	<u>meteoric</u> water	<u>Meteoric</u> <u>water</u>
				mCDW	$mCDW \delta^{18}O$	water δ <sup>18</sup> O	<u>water</u> column	fraction
		<u># of</u>	<u># of</u>	absolute	(% vs	(% VS	inventory	uncertainty
Year	Group	<b>Stations</b>	samples	salinity (g/kg)	VSMOW)	VSMOW)	(m)	(g/kg
		1	8	34.83±0.010	-0.02±0.02	-31.9±3.0	6.3±0.5	1.3
<u>1994</u>	<u>a</u> <u>d</u>	<u>3</u>	18	$34.86\pm0.010$	$-0.02 - \pm 0.01$	-28.3±2.0	$8.0\pm0.6$	1.6
2000	<u>a</u>	<u>10</u>	18 62	$\overline{34.88\pm0.012}$	-0.05±0.01	-28.7±0.9	$9.2 \pm 0.7$	1.7
	1		<u>34</u> <u>4</u>	$34.90\pm0.009$	$-0.04 - \pm 0.02$	-26.1±1.4	$8.4 \pm 0.8$	1.8
<u>2007</u>	<u>a</u> <u>b</u>	<u>8</u> <u>1</u>	4	$34.85\pm0.009$	$-0.05 - \pm 0.63$	$-29.7\pm2.7$	$10.3 \pm 0.8$	1.8 1.5
	<u>c</u>	<u>6</u> <u>2</u>	36 4	$34.87 \pm 0.010$	$0.01 = \pm 0.04$	-32.1±1.4	$9.0 \pm 0.7$	<u>1.6</u>
	<u>a</u>	<u>2</u>	<u>4</u>	$34.87 \pm 0.003$	$0.01 - \pm 0.7$	$-43.7\pm4.8$	$7.0\pm0.6$	<u>1.1</u>
<u>2009</u>	<u>c</u>	18 26	<u>61</u>	$34.87 \pm 0.003$	-0.01±0.04	$-28.8\pm1.1$	$8.7 \pm 0.8$	<u>1.8</u>
	<u>c</u> <u>d</u>	<u>26</u>	<u>110</u>	$34.87 \pm 0.002$	$0.02 - \pm 0.03$	$-29.0\pm0.7$	$8.5 \pm 0.7$	<u>1.7</u>
	<u>a</u>	<u>9</u> <u>8</u>	<u>57</u>	$34.86 \pm 0.015$	$0.04 - \pm 0.03$	<u>-27.2±1.2</u>	$8.3 \pm 0.6$	<u>1.8</u>
<b>2014</b>	<u>b</u>	<u>8</u>	<u>61</u>	$34.87 \pm 0.015$	$0.07 = \pm 0.02$	$-32.1\pm1.0$	$10.1\pm0.7$	<u>1.5</u>
2014	<u>c</u> <u>d</u>	5 9 8 2	<u>19</u>	$34.83 \pm 0.015$	$0.06 = \pm 0.02$	$-33.6\pm2.0$	$7.9 \pm 0.5$	<u>1.4</u>
		<u>9</u>	<u>76</u>	$34.83 \pm 0.015$	$0.06 - \pm 0.02$	-31.7±1.1	$\frac{7.5 \pm 0.5}{1}$	1.4
	<u>a</u>	<u>8</u>	<u>68</u>	$34.89\pm0.006$	<u>-0.1±0.01</u>	$-27.6\pm0.6$	$8.2 \pm 0.6$	1.2
<u>2019</u>	<u>b</u>		<u>21</u>	$34.87 \pm 0.006$	$-0.12 - \pm 0.01$	<u>-30.6±0.8</u>	$10.0\pm0.7$	1.1
	<u>c</u>	<u>2</u>	<u>18</u>	$34.85 \pm 0.006$	<u>-0.11±0.02</u>	-31.5±1.0	8.2±0.5	0.9
	<u>a</u>	<u>10</u>	<u>90</u>	$34.89 \pm 0.016$	-0.11±0.01	-28.2±0.4	$\frac{8.7 \pm 0.6}{6.5}$	1.3
<u>2020</u>	<u>c</u> <u>d</u>	11 11	<u>70</u>	$34.86 \pm 0.017$	-0.12±0.01	-29.6±0.5	$\frac{8.7 \pm 0.5}{2.5 \pm 0.5}$	1.2
	<u>d</u>	<u>11</u>	<u>120</u>	$34.85 \pm 0.017$	<u>-0.13±0</u>	$-29.7\pm0.4$	$8.5 \pm 0.5$	<u>1.1</u>

mCDW (as defined in Appendix A1) was broadly shows little geographic sensitivity. In all years, salinity varied by less than observed seasonal salinity variation in mCDW (0.01 g/kg; Mallett et al., 2018). mCDW δ<sup>18</sup>O exhibited less variation than that associated with instrumental precision.

The meteoric water  $\delta^{18}$ O fingerprint calculated for different geographic groupings each year is not geographically sensitive – as would be expected with deep meteoric water (basal meltwater) having a single source. 2009 Group a rendered a significantly different meteoric water endmember, however this number is based on data from just 4 samples (3 from >200 m); given the data limitations and the sample quality issues for 2009, it is unlikely that the -43.7±4.8‰ endmember is representative.

In general, the meteoric water column inventories appear insensitive to geographic groupings. The exceptions are Group a in 1994 and 2009, and Group b in 2007, 2014, 2019. In 1994, Group a contains

only a single station in 1994 (8 samples), and only 4 samples (2 stations) in 2009. Group b consists of those samples collected alongside TIS; locations likely to be dominated by meltwater originating from beneath PIIS (Wåhlin et al., 2021). Surprisingly, given the small number of samples collected near TIS in 2007 and 2019, the meteoric water inventories from Group b stations are consistent.

#### 775 A4.2 Spatial randomization analysis

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A geographic sensitivity Monte Carlo analysis involved calculating results from random sets of 3 stations 10,000 times (**Table A3**). For each group of 3 stations, mCDW and meteoric water endmembers, and average meteoric water column inventories were calculated using only those data. Stations with fewer than 2 samples >200 m were excluded. As the 3-endmember mixing model is most sensitive to the mCDW endmember, a set of stations lacking samples >800m (deeper than the subcavity ridge; 'pure' mCDW) had a random mCDW sample from >800m that year added to the data set for analysis.

Table A3: Results of station randomization sensitivity analysis. mCDW is defined as the δ<sup>18</sup>O value at the salinity maximum on the linear regression of all salinity-δ<sup>18</sup>O measurements deeper than 200 m in each group of stations; meteoric water δ<sup>18</sup>O is defined as the zero-salinity intercept on that same line. Average meteoric water inventory is the depth integration of the Gaussian fit of all calculated meteoric water fractions within each group. In all cases, uncertainty is represented by the standard deviation in the results obtained across 10,000 Monte Carlo simulations for each year. 1994 had only 4 stations, variability represented by this analysis for that year may be artificially low.

<u>YEAR</u>	Mean mCDW salinity (g/kg)	Mean mCDW δ <sup>18</sup> O (‰)	Mean meteoric water δ <sup>18</sup> O (%)	Mean meteoric water inventory (m)	% Meteoric water inventory uncertainty	Meteoric water fraction uncertainty (g/kg)
<u>1994*</u>	34.86±0	$-0.01\pm0.01$	-29.4±0.6	$7.6 \pm 0.2$	<u>2.9%</u>	<u>0.1</u>
<u>2000</u>	34.87±0.03	$-0.06\pm0.03$	$-28.7 \pm 1.6$	$8.8 \pm 0.5$	<u>5.4%</u>	<u>0.8</u>
<u>2007</u>	34.89±0.01	$-0.03\pm0.04$	$-27.9\pm3.4$	$9.0\pm0.7$	<u>7.3%</u>	<u>1.1</u>
<u>2009</u>	34.86±0.06	$-0.01\pm0.08$	$-28.7\pm4.1$	$8.8 \pm 0.7$	<u>8.0%</u>	<u>1.8</u>
<u>2014</u>	34.84±0.03	$0.05 \pm 0.05$	<u>-31.1±3.3</u>	$8.5 \pm 0.8$	<u>9.4%</u>	<u>1.1</u>
<u>2019</u>	34.88±0.01	$-0.10\pm0.01$	<u>-29.6±1.6</u>	8.5±0.4	4.9%	<u>0.6</u>
<u>2020</u>	34.87±0.02	$-0.11\pm0.02$	-29.1±1.4	$8.8 \pm 0.6$	<u>6.3%</u>	<u>0.7</u>

The mCDW properties appear geographically insensitive, though 2009 and 2014 exhibit higher variability than other years, potentially due to sample collection and/or storage issues (**Appendix A2**, **A7**).

Meteoric water endmember properties showed greater spatial variability in 2007, 2009, 2014. In these 3 years, the data show the greatest scatter, and station locations do not always have mCDW samples near the seafloor. The lengthy meteoric water endmember extrapolation benefits from many samples collected below 200 m.

The impact of sample geographic location variability is generally comparable to that of the primary uncertainty analysis (analytical precision and environmental variability), with the exceptions of 2009 and 2014. However, the 2014, spatial uncertainty is somewhat inflated due to the very high (>10m) meteoric water inventories at stations immediately alongside TIS, while the 2009 data are impacted by sample storage issues, and poor depth resolution at some locations.

mCDW  $\delta^{18}$ O (as defined in Section A1, above) was shown to not be geographically sensitive. Surprisingly, in 2007, mCDW defined using only the stations closest to the ice shelf (Group a) produced the least negative  $\delta^{18}$ O values for mCDW. Since the mCDW closest to the ice shelf is the water that will be melting the ice shelf, and samples from both locations fall on the same  $\delta^{18}$ O-salinity mixing line, this is not thought to be a problem for our analysis. In all other years, the greatest variability in mCDW  $\delta^{18}$ O was in 2014, with a total spread of 0.03% (st. dev. 0.01%)—well within ±0.04% (0.02 %) instrument precision for IRMS (CRDS).

As with mCDW, we see little geographical impact on the calculated average meteoric water column inventory. In almost all cases, the small differences in geography are well below the uncertainty of the calculations. The one exception is 2014, where the stations nearest the Thwaites Glacier Tongue (TGT) exhibit significantly higher meteoric water content than the rest of the study area. While this is consistent with known circulation in the area (Jenkins et al., 2010; Thurnherr et al., 2014; Naveira Garabato et al., 2017; Joughin et al., 2021b), we do not have comparable samples from other years, so those stations were excluded for the purposes of interannual comparisons.

#### A5 On measuring precipitation in surface waters

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The size of the meteoric water fraction calculated using the three-endmember mixing model is inversely proportional to the (negative) magnitude of the meteoric water endmember, as a -15% endmember will produce ~1.8x the calculated meteoric water fraction of a -29% endmember. For a given mixture, it would thus take ~2x as much -15% water to produce the 8<sup>18</sup>O signature than for -29% water.

## A5 The impact of precipitation on meteoric water inventories

Sea-level precipitation at this latitude has δ<sup>18</sup>O values of ~-15‰ (based on snow collected during NBP19-01, consistent with expected local values from other studies; Gat and Gonfiantini, 1981;
 Ingraham, 1998; Noone and Simmonds, 2002; Masson-Delmotte et al., 2008). This region of the Amundsen Sea receives ~0.5 m water equivalent of precipitation per year (Donat-Magnin et al., 2021), and mCDW on the shelf has a residence time of ~2 years (Tamsitt et al., 2021). We recalculated water column meteoric water inventories assuming 1 m (2 full years) of local precipitation (-15‰ δ<sup>18</sup>O) in the upper 200 m of the water column at the time of sampling.

We find that adding 1 m of precipitation to the water column decreases the amount of meteoric water (as defined using the zero-salinity intercepts, ) by an average of  $0.55\pm0.01$  m, and decreases sea ice melt by an average of  $0.57\pm0.03$  m (**Table A4**). These results suggest that even with two year's worth of precipitation present in the water column at the time of sampling, the calculated meteoric water inventory could consist of >92% glacial meltwater.

<u>Table Sea-level precipitation at this latitude has δ<sup>18</sup>O values of ~ 15‰ (snow collected at 72°S during NBP19-01)</u>. This region of the Amundsen Sea receives ~ 0.5 m water equivalent of precipitation per year (Donat-Magnin et al., 2020). In an extreme case, we could assume that the entirety of 1 year's 0.5 m of precipitation is present in the upper water column at the time of sampling. Given the mathematical relationships above, that would result in a ~ 0.28 m overestimation of the total glacial meltwater. The lowest (highest) year for average meltwater column inventory 1994, 7.7 m (2020, 9.2 m) corresponds to only a 3.6% (3.0%) overestimation. Even assuming the highest snowfall rates in the region (~1m water equivalent near Abbot and Getz Ice Shelves; Donat-Magnin et al., 2020) this results in an overestimate of <8%. The largest impact of uncertainty around precipitation is thus lower than uncertainty associated with analytical precision and endmember uncertainty.

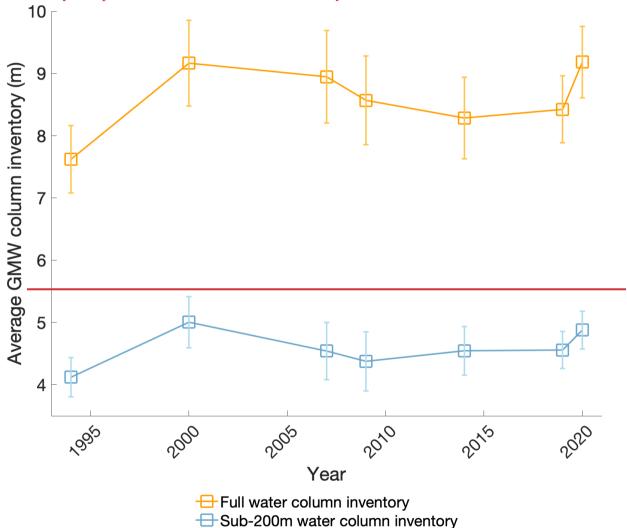


Figure A44: Impact of precipitation on total meteoric water Average meltwater column inventory. Mean meteoric water in study area. As Figure 4 from main text, but also showing meltwater inventory is the integrated mean meteoric water content between the surface 200m and 800m. The upper water column will include meteoric water from both precipitation, and GMW introduced at depth and mixed upward. The three rightmost columns in the table show the impact on meteoric and sea ice melt water column inventories of

recalculating column inventories (meteoric water  $\sim$  30%  $\delta^{18}$ O, **Table 2**) assuming 2 years of precipitation ( $\sim$ -15%  $\delta^{18}$ O) in the water column at the time of sampling.

Impact of 1 m (~2 years) precipitation (-15%  $\delta^{18}$ O)

Year	Mean meteoric water inventory (m)	Change in (glacial) meteoric water (m)	Change in sea ice melt water (m)	Estimated water column glacial meteoric water
1994	7.62	-0.56	-0.61	92.6%
2000	9.16	-0.57	<u>-0.62</u>	93.8%
2007	<u>8.95</u>	<u>-0.56</u>	<u>-0.55</u>	93.7%
2009	<u>8.57</u>	<u>-0.55</u>	<u>-0.55</u>	<u>93.5%</u>
2014	<u>8.93</u>	<u>-0.54</u>	<u>-0.55</u>	<u>94.0%</u>
<u>2019</u>	<u>8.42</u>	<u>-0.54</u>	<u>-0.57</u>	<u>93.6%</u>
2020	<u>9.18</u>	<u>-0.56</u>	<u>-0.54</u>	93.9%

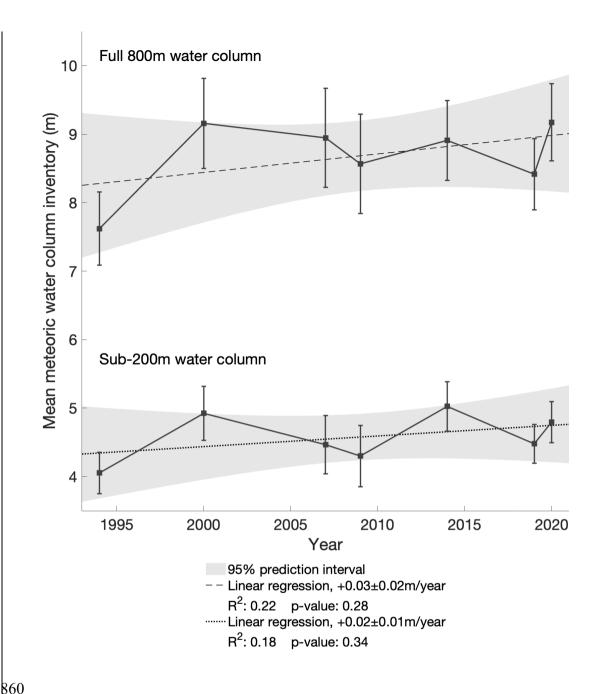


Figure A5: Mean meteoric column inventory for each sampled year. Points Blue bars represent the depth-integrated meltwater volume from the Gaussian Process fit (greyred lines in Figure 3 Figure 3) between 200 and 800 m depth. Error bars show the uncertainty in mean meteoric water GMW column inventory associated with analytical precision and environmental variability (Data and Methods 2.2). The relative year-to-year inventories here show the same general empirical trend (within uncertainty) as Figure 4 Figure 4. 2014 shows the highest sub-200m meteoric water content, owed to the sampling immediately alongside TIS - directly in the pathway of glacial meltwater from PIIS (Wåhlin et al., 2021).

Figure A5 and Table A5 show a comparison of the yearly inventories in the total water column vs the water column deeper than 200 m. Both the full and partial water columns show the same relative trend in meteoric water content, indicating that the observed variability is not an effect of interannual variability in precipitation.

Table A5: Relative Fractions of yearly meteoric water meltwater inventory in the 800mat depth, and 200-800mof full water columns column. Reported column inventories are the depth integration of the Gaussian fit of all measurements in the field area between the specified depths. The Relative fraction is the normalized relative volume of the average inventory from compared year to year.

	0 m – 800 m		200 m – 800 m		Fraction of
	Column Inventory,	<b>Normalized</b>	Column Inventory,	<b>Normalized</b>	total <u>meteoric</u> <u>water</u> GMW in
	<u>meteoric</u>	<u>relative</u> Relative	<u>meteoric</u>	<u>relative</u> Relative	upper
Year	water GMW (m)	fraction	water GMW (m)	fraction	200 m
1994	$7.6\pm0.5$	83 <u>±6</u> .0±5.9	4.1±0.3	8182.3±6.3	45.9%
2000	$9.2 \pm 0.7$	10099.8±7.5	5.0±0.4	98 <del>100</del> ±8.2	45.4%
2007	$9.0\pm0.7$	97 <del>.4</del> ±8 <del>.1</del>	4.5±0.5	89 <del>90.7</del> ±9 <del>.1</del>	49.2%
2009	$8.6 \pm 0.7$	93 <del>±.3±7.</del> 8	$4.4{\pm}0.5$	8687.4±9.4	48.9%
2014	8. <u>9</u> 3±0. <u>6</u> 7	$97\pm690.2\pm7.2$	<u>5.1</u> 4.6±0.4	10090.8±7.8	<u>42.7</u> 45.1%
2019	$8.4{\pm}0.5$	$92\pm691.7\pm5.8$	$4.6 \pm 0.3$	<u>89</u> 91.1±6	45.9%
2020	$9.2 \pm 0.6$	100 <del>.0</del> ±6 <del>.3</del>	$4.9 \pm 0.3$	9597.5±6.1	46.9%

### A6 Sea ice melt and mCDW fractions

## A6.1 Sea ice melt

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While the primary focus of discussion in this paper is meteoric water, the three-endmember mixing model also yields sea ice meltwater fractions. In locations where integrated sea ice melt fractions are negative, net sea ice formation at the time of sampling is indicated. Using a less negative meteoric water δ¹8O endmember (e.g. -25‰ used in Biddle et al., 2019) will result in higher meteoric water fractions and lower sea ice melt fractions, significantly impacting areas/years of net sea ice melt/formation (Figure A7). Since >92% of the meteoric water content in the study area is estimated to be basal melt, using a GMW meteoric water endmember (or close to) will produce more accurate net sea ice melt/formation inventories (and correspondingly more accurate meteoric water inventories).

In 2007, 2009, and 2014 positive ice melt fractions >200 m, likely the resulted from samples compromised before analysis. Evaporation leads to positive fractionation of seawater  $\delta^{18}$ O, leading to a less-depleted  $\delta^{18}$ O observation at time of analysis; less depleted  $\delta^{18}$ O relative to salinities fresher than mCDW will be interpreted by the 3-endmember mixing model as sea ice melt. The stratification in this region makes it unlikely that there are significant sea ice melt fractions below 200 m. As with the  $\delta^{18}$ O-salinity () and meteoric water-depth (**Figure 3**) plots, 1994, 2000, 2019 and 2020 exhibit the tightest distribution, suggesting higher quality data. Grey lines show the Gaussian process fit, and points are shaded to show sample  $\delta^{18}$ O.

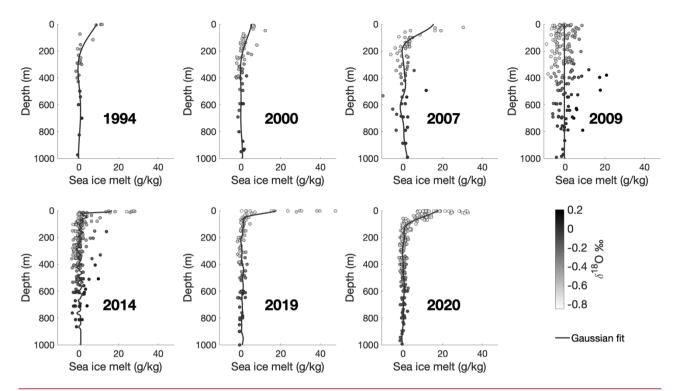


Figure A6: Sea ice melt fractions vs depth. Calculated sea ice melt fractions using salinity and  $\delta^{18}$ O measurements in 3-endmember mixing model. Shading of dots indicates the measured  $\delta^{18}$ O of that sample. In several years (2014-2020) there is a high concentration of sea ice melt near the surface, and very little extending deeper than 200 m. Negative values indicate net sea ice formation. Years with greater sea ice melt (2007, 2020) than sea ice formation show greater divergence from the mCDW-GMW mixing line in surface waters ().

Table A6: Sea ice melt column inventories and uncertainty. Mean sea ice melt column inventories are produced by depth integrating the Gaussian process fit (grey lines Figure A6) between the surface and 800m. Uncertainty as described in Appendix A3.

<b>Year</b>	Mean sea ice melt	Sea ice melt
	<u>column inventory</u>	<u>fraction</u>
	<u>(m)</u>	uncertainty (g/kg)
<u>1994</u>	<u>1.0±0.8</u>	<u>1.9</u>
2000	0.6±0.9	<u>2.1</u>
<u>2007</u>	<u>1.2±0.9</u>	<u>2.2</u>
<u>2009</u>	<u>-0.4±1.0</u>	<u>2.2</u>
<u>2014</u>	<u>0.5±0.9</u>	<u>2.0</u>
2019	<u>1.0±0.7</u>	<u>1.4</u>
<u>2020</u>	<u>1.1±0.8</u>	<u>1.7</u>

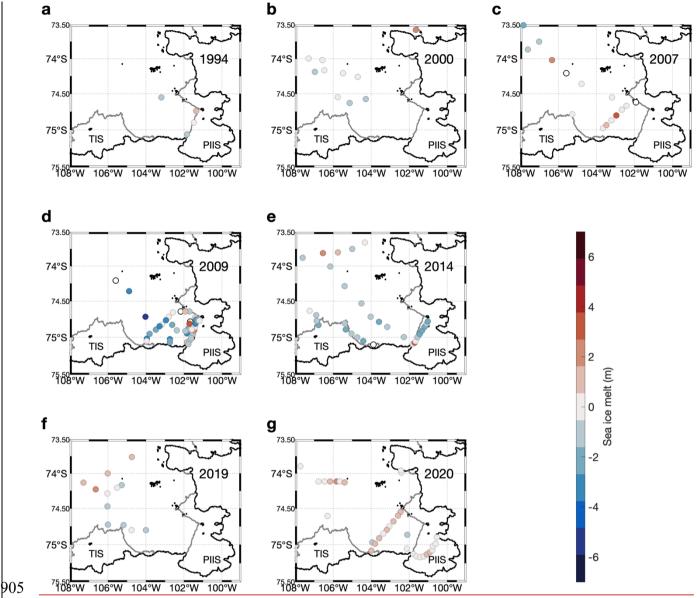


Figure A7: Integrated sea ice melt fractions at sampling locations each year. Negative sea ice melt fractions indicate areas of net sea ice formation. Stations with partial water column sampling show only partial inventories. White dots with black outlines are stations where only one depth was sampled (2007, 2009, 2014). Years with greater sea ice melt (2007, 2020) than formation show greater divergence from the mCDW-GMW mixing line in surface waters ().

# 910 A6.2 mCDW fractions

Waters deeper than ~800m are comprised of pure mCDW; moving toward the surface, meteoric freshwater from basal melt is introduced starting at ~700m. The near surface waters are rich in meteoric water and/or sea ice melt are comprised of >92% mCDW (Figure A8).

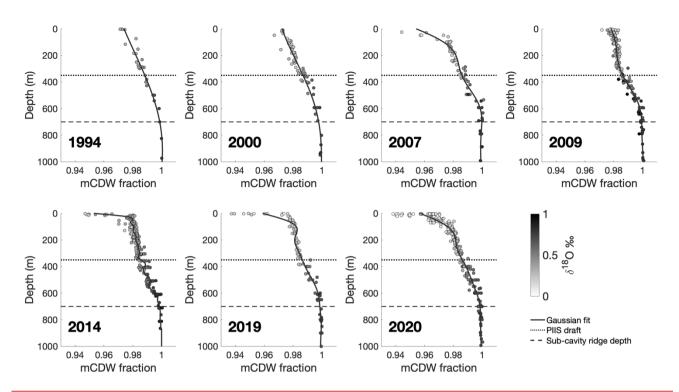


Figure A8: mCDW fractions vs depth. Calculated mCDW fractions using salinity and δ<sup>18</sup>O measurements in 3-endmember mixing model. Shading of dots indicates the measured δ<sup>18</sup>O of that sample. Deep waters (>800m) characterize relatively unadulterated mCDW, while near surface waters contain the highest concentrations of sea ice melt and meteoric water. Dotted horizontal lines show the depth of the PIIS draft, and dashed lines show the depth of the PIIS sub-cavity ridge.

Table A7: mCDW column inventories and uncertainty. Mean sea ice melt column inventories are produced by depth integrating the Gaussian process fit (grey lines Figure A8) between the surface and 800m. Uncertainty as described in Appendix A3.

<b>Year</b>	Mean mCDW	mCDW fraction	
	<u>column</u>	<u>uncertainty</u>	
	inventory (m)	<u>(g/kg)</u>	
<u>1994</u>	781.2±0.3	<u>0.6</u>	
<b>2000</b>	$780.1\pm0.4$	<u>0.6</u>	
<b>2007</b>	$779.8\pm0.3$	<u>0.7</u>	
<b>2009</b>	781.6±0.2	<u>0.6</u>	
<u>2014</u>	780.4±0.5	<u>0.7</u>	
<b>2019</b>	$780.6 \pm 0.2$	<u>0.5</u>	
<u>2020</u>	779.6±0.5	<u>0.8</u>	

## A6A7 2009 Sample quality control

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A <u>subsetportion</u> of the samples for 2009 were analyzed on an IRMS in 2010, while the <u>remainderrest</u> were stored until a 2020 CRDS analysis. At the latter time, 56% of the samples analyzed contained an unknown, <u>clear, needle-shaped</u> precipitate. Several bottles also had a lower\_-than\_-expected sample volume, suggesting evaporation, which would <u>likely have altered thebe expected to alter</u>  $\delta^{18}$ O <u>content viathrough</u> isotopic fractionation. Several steps were taken to ensure the quality of samples analyzed after a decade in storage.

## 930 A6.1A7.1 SEM EDS Analysis of Precipitate

Samples of the precipitate were extracted from multiple sample bottles and analyzed using a Scanning Electron Microscope, equipped with a FEI Magellan 400 XHR SEM with Bruker Quantax XFlash 6 | 60 SDD EDS detector, at the Stanford Nano Shared Facilities (SNSF). Peaks were observed at the spectra associated with Mg, Si, and O, indicating the precipitate is likely some form of Magnesium Silicate Hydroxide (Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>), or Magnesium Silicate Hydrate (Mg<sub>2</sub>Si<sub>3</sub>O<sub>8</sub>•H<sub>2</sub>O). Si(OH)<sub>4</sub> is the simplest soluble form of silica and is found universally in seawater at low concentrations (Belton et al., 2012). The maximum amount of silicate that could be expected in this area of the ocean is ~100 μmol/kg (Rubin et al., 1998). In this case, even if the entire 100 μmol/kg of Si were drawn down to 0, solely into a Magnesium Silicate, with a very high fractionation factor, e.g. the -40‰ reported for diatoms (Leclerc and Labeyrie, 1987) the greatest effect on a sample would be 0.0003‰ – well below the analytical precision of the CRDS (0.025‰) or IRMS (0.04‰). Therefore, it is highly unlikely that the precipitate contributed a detectable fractionation or alteration of seawater δ<sup>18</sup>O in our samples.

# A6.2A7.2 Quality control for evaporation

In all years, the bulk of Assuming accurate 2020 analyses, the  $\delta^{18}$ O data fall in a broadly predictable pattern, less depleted at depths below ~600m, and more depleted near the surface. Values >2 standard deviations from this pattern denote samples that were likely subject to experienced significant evaporation (Figure A9Figure A9)

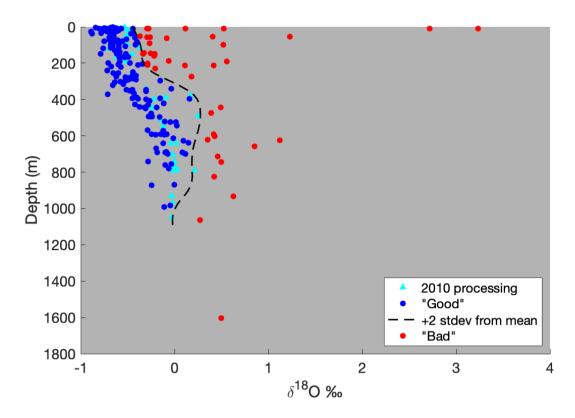


Figure A9:  $\delta^{18}$ O vs depth for all 2009 samples, coded for likelihood of evaporation. The dashed line represents +2 standard deviations from a moving depth-averaged  $\delta^{18}$ O based on the 2010 processing data, beyond which the results are unacceptable. Archivable data will be made available upon publication.

As a secondary check, the  $\delta^{18}$ O of all samples was plotted vs depth with a qualitative indicator of the amount of precipitate found in the sample vial (<u>Figure A10</u>Figure A10) to see if any patterns emerged compared to that of depth-comparable samples processed<del>run</del> in 2010. No clear trend was evident.

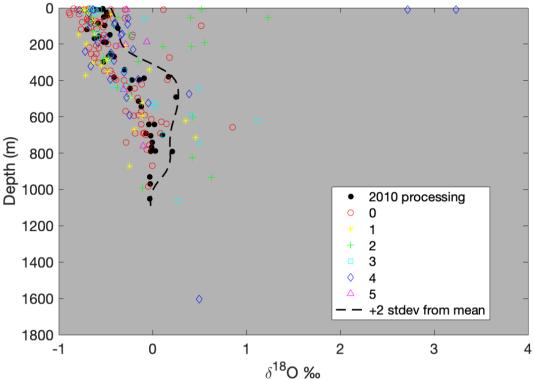


Figure A10:  $\delta^{18}$ O vs depth of all 2009 samples, coded by amount of precipitate present. Each bottle was graded by eye based on the volume of precipitate present with 0 being no precipitate present, and 5 being the most precipitate present. As with Figure A9, Figure S3, the dasheddetted line represents +2 standard deviations from the mean  $\delta^{18}$ O at each depth.

Evaporation is accompanied by isotopic fractionation, with H<sub>2</sub><sup>16</sup>O evaporating evaporates preferentially, leaving the remaining liquid relatively enriched in the H<sub>2</sub><sup>18</sup>O. Evaporation also increases the salinity and thus density of the remaining sample, thereby increasing sample density. We measured the density of each seawater sample 5 times using a calibrated 1ml pipet and mg balance. The theoretical density of each sample was calculated from its associated CTD salinity and temperature, during weight determination. Differences between measured and theoretical densities for each sample are plotted in Figure A11.

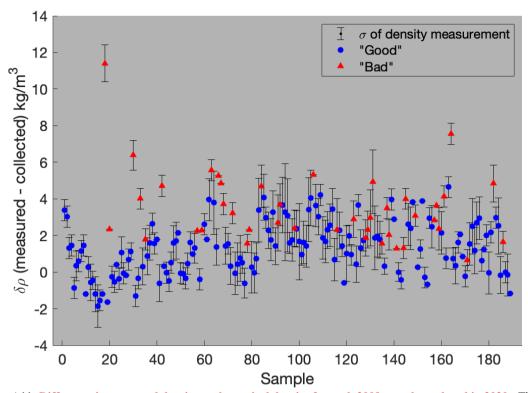


Figure A11: Difference in measured density vs theoretical density for each 2009 sample analyzed in 2020. Theoretical density is based on CTD salinity at each sample location, and measured density calculated from 1ml sample aliquots weighed on a mg scale, with sample coding as in Figure A9. Figure A5. Error bars represent the standard deviation of replicates for each measurement.

While a few samples show clear evidence of evaporation, and correspondingly high  $\delta^{18}$ O values, most show less obvious density <u>anomalies evidence</u>, exposing the limitations of our scale accuracy at that level. 75% of the samples measured showed a higher than expected density and <u>25%the remainder</u> measured a lower than expected <u>density</u>. <u>Figure A11Figure A7</u> displays a significant overlap in measured density space between samples previously identified as "good" or "bad" (<u>Figure A9Figure A5</u>). <u>Figure A11Figure A7</u> shows that there are no samples flagged as compromised (<u>"bad"</u>) from our earlier depth-based analysis (<u>"bad"</u>) with a  $\delta\rho$  greater than 1.3 kg/m³. At an aggressive first pass, we removed all sample data with a  $\delta\rho$  greater than 1.3 kg/m³ and looked at each hydrocast profile individually, using the remaining data. <u>ExcludedWe then added those excluded</u> samples flagged as "good" <u>were returned toback into</u> the dataset, and <u>individual profiles</u> re-scrutinized individual profiles to check for any qualitative <u>anomalies change</u>.

# A6.3 A7.3 Conclusion and final 2009 sample inclusion

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While it is <u>veryextremely</u> unlikely that the precipitate <u>changed contributed to a change in sample values</u>, some samples do appear to have been subject to evaporation. The inclusion of all samples flagged as

"Good" does not qualitatively change <u>ourthe</u> analyses <u>presented in this study</u> when compared with <u>the</u> analyses using only those data <u>processedrun</u> in 2010. We exclude <u>the</u> 41 samples initially flagged as "Bad," (<u>Figure A9</u>Figure A5) and retain the remaining 148 flagged as "Good".

## A7A8 CRDS and IRMS cross-calibration

We <u>processedran</u> 100 samples from 2019 and 2020 concurrently using the Picarro L2140-i CRDS, <u>and</u> on a Finnigan MAT252 IRMS (<u>Figure A12</u>Figure A12) using CO<sub>2</sub> equilibration (Epstein and Mayeda, 1953). Both instruments were independently calibrated using international standards VSMOW, SLAP, and GISP, and all samples were run in duplicate. The data from both machines was comparable, with the Picarro achieving a precision of 0.02<u>%</u>, on replicates, and the IRMS achieving a precision of 0.03‰ on replicates. The On average the offset between the CRDS data and IRMS data <u>averagedwas</u> - 0.02‰, with the CRDS data being more negative, than that from the IRMS. Since the offset between the two machines was less than either instrument's precision, data from the CRDS was used as-is. <u>Values The values</u> reported for the CRDS are the average of 6 individual injections/measurements from each vial; <u>reported</u> precision is <u>reported</u> based on the standard deviation between multiple 6-injection averages from replicate analyses, separated by days, weeks, or months.

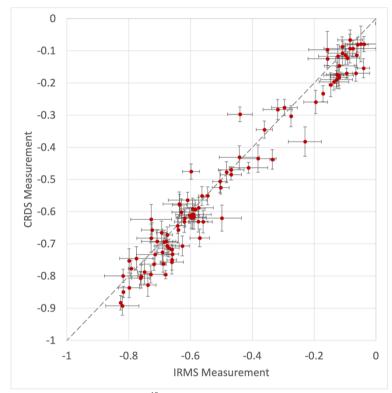


Figure A12: 101 seawater δ<sup>18</sup>O samples collected in 2019 and 2020 analyzed with both CRDS and IRMS. Each point represents the value obtained by measuring the same sample on the IRMS (x-axis) and the CRDS (y-axis). Error Horizontal error bars represent the corresponding standard deviation of the IRMS and CRDS measurements, and vertical error bars represent the standard deviation of the CRDS measurement. The dashed grey line is a 1:1 slope.

### **A8A9** CRDS Methods

- o10 <u>570A large number</u> of the isotope samples for this study (all samples from 2019 and 2020, portions from 2007 and 2009) were run on a Picarro L2140-i CRDS system, rather than a traditional IRMS. Using this system, we were able to achieve an average precision of <0.02‰ on multiple replicate analyses.
- Samples were collected in 10ml or 30ml glass serum vials (Fisher Scientific part number: 06-406D/06-406F), sealed with rubber stoppers (Fisher Scientific part number: 06-406-11B) and aluminum seals (Fisher Scientific part number: 06-406-15).
- Sample vials should be filled to just below the "neck" (narrowest part). Minimizing the headspace of the vial is important for minimizing evaporation, however it is important to leave *some* headspace to allow for expansion/contraction of the sample (if collecting samples larger than 30 ml, slightly more headspace should be left, with 250 ml vials being filled to ~1/2 way up the "shoulder" before the neck). When sealing serum vials with rubber stoppers and aluminum seals, it is important that the tops of the aluminum seals are crimped tightly but remain flat after crimping/capping. An upward "buckling" of the aluminum seal indicates over-crimping and will produce an inferior seal.

Internal lab (Stanford SIL) data shows that samples can be preserved in this manner for up to 5 years without significant degradation, while bottles with threaded caps and parafilm reliably maintain sample integrity below instrumental precision for no more than 1 year.

-The instrument setup used was as follows:

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- 10μl syringe (Trajan Part number: 002982)
- A single standard or unknown run consists of 7 injections (measurements) per sample
- Sample injection volume 2.2 μl
- 3x 5μl rinses with fresh water from inkwell (IW) between each sample
  - 3x 2.2µl rinses with sample before first measurement of new sample or standard vial
  - Rinse only between sample vials, or 1 rinse for every 7 standard or unknown injections.

<u>Several In addition to the above instrument method, several protocols were also followed with regards to sample and instrument handling.</u>

- Fresh vial of internal lab standard (ILS) used each day. The ILS was prepared to have a  $\delta^{18}$ O in the middle of the range expected from the unknowns (i.e.  $\sim$ -0.3%) to minimize memory issues between samples. The IW was also filled with water of approximately this composition.
  - A fresh 2ml vial of ILS was used each day, and discarded at the end of the sequence.

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- Samples were pipetted from sealed 10ml or 30ml serum into 2ml vials (Fisher Scientific part number: 03-391-15) for analysis on the day they were to be analyzed. The 2ml vials used for analysis were found to only reliably preserve sample  $\delta^{18}$ O for <1 week.
- After each sequence, the syringe was cleaned with DI water, and then rinsed thoroughly with water from the Inkwell, to minimize memory/contamination issues of residual water left in the syringe.

- o Treated in this way, syringes can be expected to last <u>for</u> 1500 to 2500 injections
- Fresh vaporizer septa (<u>Trajan Partpart</u> number: <u>0418240</u>) was used every day
- ILS were analyzed no less than every 5th unknown, and no fewer than 3 ILS were measured per run

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- All data were corrected based on the slope of the ILS measurements over the course of the sequence.
- Each run began with no fewer than 10 injections from the IW, to allow the instrument to reach baseline.
- Syringe cleaned thoroughly with DI water each day, and manually rinsed with IW water prior to sequence.
  - No more than 5 unknowns (7 injections each) measured between run of ILS (7 injections)
  - ILS measured at least 3 times during each sequence at the beginning, end, and midpoint. least 3 standards measured during each sequence.

A typical 24h sequence ran 16 unknowns. The sequence was set up, as follows:

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- 15 injections from IW
- ILS (7 injections)
- 4 unknowns (4x7 injections)
- ILS (7 injections)
- 4 unknowns (4x7 injections)

- ILS (7 injections)
- 4 unknowns (4x7 injections)
- ILS (7 injections)
- 4 unknowns (4x7 injections)
- ILS (7 injections)

Overall, this sequence consists of 162 injections, 112 of which contained salt, for a vaporizer load of ~8.6 mg of salt/day.

The instrument vaporizer was cleaned at least every 200mg worth of salt injected.

- @ 35PSU & 2.2μl injections, this is 2597 salty injections, or 371 samples @ 7 injections each.
   (~ every 23 analytical days)
- 080 Finally, analytical data quality control was conducted in the following way
  - The first injection of each sample was discarded, to minimize instrument memory issues
  - If the standard deviation of the remaining 6 injections was >0.04‰, up to one outlier could be removed. Any samples where the standard deviation of measured values was still >0.04‰ were rerun the following day from the same vial, using the same septa.
- If a rerun would not be possible the following day, the vial septa was replaced with a new one.
  - Data from each hydrocast were inspected as a group. Any samples that appeared inconsistent with the rest of the hydrocast (e.g. with regards to salinity, or neighboring  $\delta^{18}$ O values) were rerun. If the rerun occurred within 1 week of the initial run, the same vial was used. Otherwise, a fresh aliquot of sample was drawn from the resealed serum vial.

## 090 Data availability

All data, excluding that flagged as "bad," (Appendix A7) used in this study can be accessed at: <a href="https://doi.org/10.25740/zf704ig7109">https://doi.org/10.25740/zf704ig7109</a>

#### **Author contributions:**

Conceptualization: RBD

095 Methodology: RBD, ANH, DAM Investigation: ANH, DAM, RBD

Visualization: ANH

Funding acquisition: RBD Project administration: RBD

100 Supervision: RBD

Writing – original draft: ANH

Writing - review & editing: ANH, SSJ, RBD, DAM, RAM

Contribution of data: ANH, DAM, RBD, SSJ, RAM

#### 105 Competing interests

The authors declare that they have no conflict of interest.

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