Supplementary material

S1. Evaluating the freshwater sources from Total Alkalinity (JR302 cruise)

We present here another method to estimate the MW and SIM fractions by using Total Alkalinity (TA) instead of $\delta^{18}O$ in the mass balance calculations presented in section 2.3 (e.g. Sutherland et al., 2009, Jones et al., 2008). During the JR302 cruise, TA samples were collected and measured according to Dickson et al. (2007). Water was collected using silicone tubing into either 500 ml or 250 ml Schott Duran borosilicate glass bottles and poisoned with saturated mercuric chloride solution (50 µL for 250 ml bottles and 100 µL for 500 ml bottles) after creating a 1 % (v/v) headspace. Samples were sealed shut with Apiezon L grease and electrical tape and stored in the dark at 4 °C until analysis. JR302 TA samples were analysed on board using two VINDTA 3C systems (Mintrop, 2004). Measurements were calibrated using certified reference material (batches 135 and 136) obtained from Prof. A. G. Dickson (Scripps Institute of Oceanography USA). The precision of the replicate and duplicate measurements was 2.0 $\mu mol.kg^{-1}$ (King and Holliday, 2015).

As for $\delta^{18}O$, we assume that the saline end-member is only composed of AW. The end-members we use in our calculations are S_{AW}=35, TA_{AW}=2305 µmol/kg (average from JR302 measurements), S_{MW}=0, TA_{MW}=800 µmol/kg and S_{SIM}=4, TA_{SIM}=263 µmol/kg (Sutherland et al., 2009, Jones et al., 2008). Notice that TA_{MW} is difficult to estimate as the meteoric water alkalinity may change on an annual basis, due to local variations in currents, continental sources having a wide range of values, dependent on geology and hydrology, so that a typical range for the alkalinity of the fresh water in this region is 600-1000 $\mu mol.kg^{-1}$ (Cooper et al., 2008). We also expect local precipitation and snow on sea ice to have very low TA values. Moreover, there are few TA measurements for freshwater from Greenland ice sheet and glaciers, for which we expect to have lower TA values compared to MW originating from the Arctic rivers.

Figure S1 compares the mass balance calculations from TA and $\delta^{18}O$ for the 113 samples between 0 and 200 m for which both sets of measurements have been done during the JR302 cruise. For MW and SIM fractions, the correlation coefficients are higher than 0.7, indicating a general agreement between the two methods. Nevertheless, for (near-)surface samples (0-70 m), the noise is generally larger for MW and SIM fractions calculated from TA measurements. Biological activity in shallow waters and coastal environment affects TA which changes fraction calculations by ~1%, while $\delta^{18}O$ computation is not sensitive to biological processes. Furthermore, exchanges with particles will also modify TA. Then, a problem common to the two methods is the uncertainty we have when choosing the end-members for mass balance calculations (e.g. the Greenland ice-sheet melt water and runoff has a range of properties that will be different from the MW from the Arctic or local spring snow). To illustrate these previous points, we will now discuss the two methods with some examples.

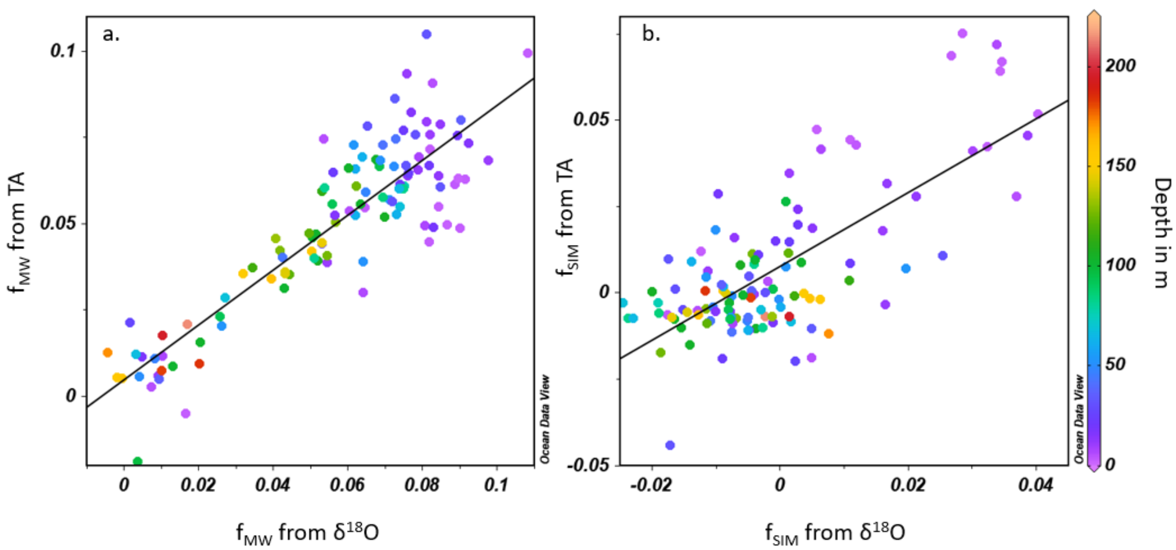

Figure S1. Comparison of a. f_{MW} and b. f_{SIM} calculations from TA (Y-axis) and $\delta^{18}O$ (X-axis) (113 samples between 0 and 200 m, see colour). The linear regressions (black lines) are a. f_{MW} ($\delta^{18}O$) = 0.79 f_{MW} (TA) (r=0.88) and b. f_{SIM} ($\delta^{18}O$) = 1.07 f_{SIM} (TA) (r=0.73).

<u>Example 1. South of Cape Farewell (Section SE)</u>

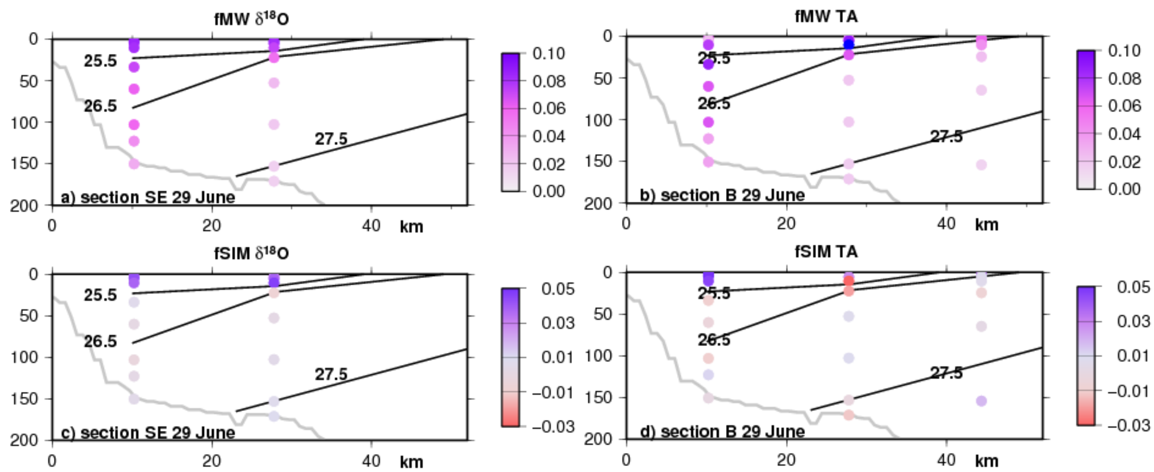

Figure S2. Spatial distribution of freshwater fraction estimates from (left) $\delta^{18}O$ and (right) TA for the B section of JR302, south-east of Cape Farewell. Top f_{MW} fraction, bottom f_{SIM} fraction. The shelf break is located 38 km from shore. The isopycnal contours for 25.5, 26.5 and 27.5 kg/m^3 are also sketched, and the light grey contour indicates the bottom depth from ETOPO1.

The trend of the FW distribution is similar for the two methods, with a freshening increasing towards the coast and the surface. We also notice a good agreement at subsurface from 75 m to 200 m. Nevertheless, the signal seems slightly noisier using TA measurements (see surface samples of the two most inner stations as well as the two high SIM fractions of 0.03 at 100 m and 0.02 at 150 m for the two stations between 60 and 80 km from the coast). We suggest that a part of the noise can be due to biological activity affecting TA.

<u>Example 2. East of Cape Farewell (section E)</u>

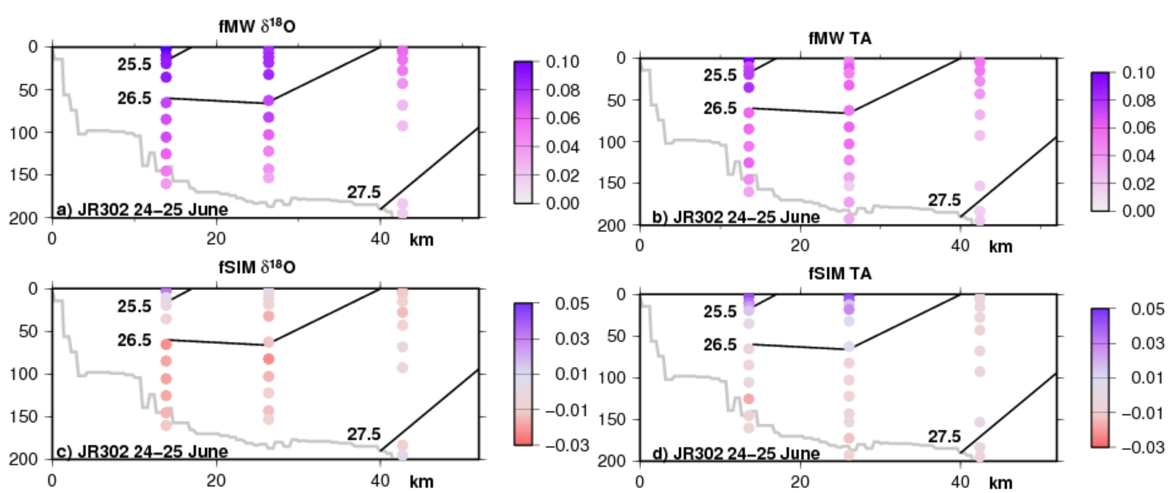

Figure S3. Spatial distribution of f_{MW} estimates from (left) $\delta^{18}O$ and (right) TA for the E section of JR302, east of Cape Farewell. Top f_{MW} fraction, bottom f_{SIM} fraction. The shelf break is located 44 km from shore. The isopycnal contours for 25.5, 26.5 and 27.5 kg/m^3 are also sketched, and the light grey contour indicates the bottom depth from ETOPO1.

As for the example 1, we observe more noise near the surface for the calculations from TA, probably partly due to the influence of biological activity on the shelf on TA. Beyond that, we note that the two methods lead to different interpretations of the FW origins regarding water masses with salinity lower than 32 (first 50 m below the surface). Indeed, isotope measurements indicate that the two inner stations are dominated at the surface by MW inputs (fractions between 0.08 and 0.11) with little influence of SIM (mostly 0.01 but with a value of 0.03 for the most inner surface sample). However, a mass balance calculation from TA indicates that the first inner station is dominated by MW at the surface, while the FW distribution at the second station is balanced between the MW and the SIM inputs (even slightly higher SIM fractions compared to the MW). We suggest that this disagreement between the two methods could originate from the choice of the endmembers. On the shelf, the two stations could be influenced by MW inputs originating from different places (such as local input from Greenland ice sheet, local spring snow melt or as with an arctic origin). The different sources have different TA and $\delta^{18}O$ values, not considered in our calculations. This example illustrates the limitations of these two methods.

<u>Example 3. South of Cape Farewell (Section S)</u>

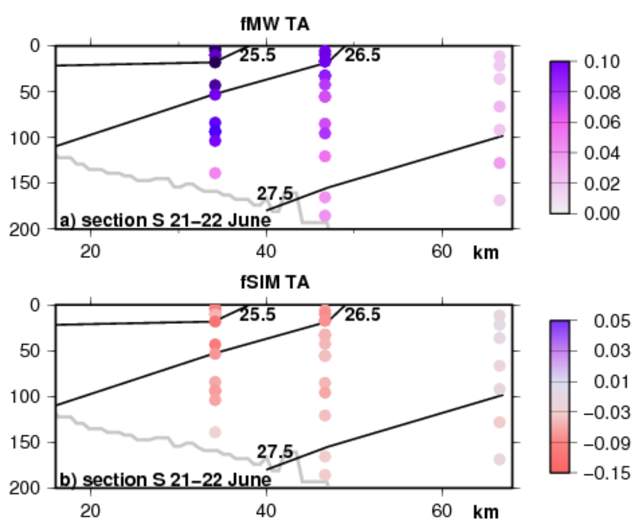

Figure S4. Spatial distribution f_{MW} and f_{SIM} estimates from TA measurements. The shelf break is close to 50 km from shore. The isopycnal contours for 25.5, 26.5 and 27.5 kg/m^3 are also sketched, and the light grey contour indicates the bottom depth from ETOPO1.

There were no $\delta^{18}O$ measurements available for the two stations at 14 and 47 km on the southern JR302 section. The calculations based on TA measurements show very strong MW fractions (from 0.10 to 0.20) with strong negative values for SIM (from -0.08 to -0.03) in the upper 100 m (notice the different scale used on Fig. S4 compared with the other figures). It is clear that this signal is unrealistic. Furthermore, the strong anomaly cannot be only explained by the biological activity on alkalinity. We suggest that in this case, the dissolution of particles coming from the sea ice melting have resulted in a large increase in TA. For these very unusual TA values, it is obvious that the method we used does not estimate realistic SIM and MW fractions. Thus one needs to be aware that an effect that particle dissolution or biological activity could also have occured for some of the other samples with a smaller magnitude, affecting mass balance calculations when using the TA method.