

Glacial meltwater and primary production as drivers for strong CO<sub>2</sub> uptake

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# Glacial meltwater and primary production as drivers for strong CO<sub>2</sub> uptake in fjord and coastal waters adjacent to the Greenland Ice Sheet

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## Abstract

The Greenland Ice Sheet releases large amounts of freshwater, which strongly influences the physical and chemical properties of the adjacent fjord systems and continental shelves. Glacial meltwater input is predicted to increase strongly in the future, but the impact of meltwater on the carbonate dynamics of these productive coastal systems remains largely unquantified. Here we present seasonal observations of the carbonate system in the surface waters of a west Greenland tidewater outlet glacier fjord. Our data reveal a permanent undersaturation of CO<sub>2</sub> in the surface layer of the entire fjord and adjacent shelf. The average annual CO<sub>2</sub> uptake for the fjord is estimated to 65 g C m<sup>-2</sup> yr<sup>-1</sup> indicating that the fjord system is a strong sink for CO<sub>2</sub>. Primary production and the high input of glacial meltwater strongly affect the carbonate system in the Godthåbsfjord system. The largest CO<sub>2</sub> uptake occurs near to the ice sheet. High glacial meltwater input during the summer months correlates strongly with high levels of CO<sub>2</sub> undersaturation, which can be explained by the non-linear effect of salinity on surface water pCO<sub>2</sub> resulting from the mixing of glacial meltwater and ambient fjord water. Our findings hence imply that glacial meltwater may form a major driver for CO<sub>2</sub> undersaturation in fjord and coastal waters adjacent to an Ice Sheet.

## 1 Introduction

The Arctic Ocean plays an important role in the global carbon cycle and contributes 5–14 % to the global CO<sub>2</sub> uptake (Bates and Mathis, 2009). Intensive biological activity combined with high seasonality in freshwater input and sea ice cover lead to strong dynamics in the carbonate system (Kaltin and Anderson, 2005). Increasing water temperatures, freshwater input and decreasing ice cover will likely have a profound effect on the Arctic Ocean and will likely amplify the large seasonal and spatial biogeochemical gradients that occur in this area (Bates and Mathis, 2009; Mathis et al., 2011). Yet at present, we have still a limited understanding of the carbon dynamics in these high-

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representative) is the inner part close to the freshwater sources, which is most strongly affected by glacial meltwater.

At each station and sampling time, conductivity and temperature depth profiles were recorded by a CTD instrument (Seabird SBE19+), which was equipped with a fluorescence (Seapoint Chlorophyll Fluorometer) and Photosynthetic Active Radiation sensor (LI-COR 190SA quantum Q PAR sensor). Partial pressure of carbon dioxide ( $p\text{CO}_2$ ) was measured in situ using the HydroC™ Carbon Dioxide Sensor (Contros, Germany) at seven water depths (1, 5, 10, 20, 30, 40 and 50 m). At every depth the HydroC sensor was equilibrated for 2–5 min until a stable reading was obtained.

Water samples were collected at eight water depths (1, 5, 10, 20, 30, 40, 100 and 400 m) using 5 liter Niskin bottles (KC Denmark Research Equipment). Unfiltered water was transferred by gastight tubing (Tygon) to 12.5 mL exetainers (Labco, UK) for dissolved inorganic carbon (DIC) and total alkalinity (TA) analysis. Exetainers were left to overflow and samples were preserved by adding 0.02 % of a saturated  $\text{HgCl}_2$  solution (Dickson and Goyet, 1992). Samples were stored in darkness at 4 °C until further analysis. DIC was measured using an infra-red DIC analyzer (Apollo SciTech), which consists of an acidification and purging unit in combination with a LI-COR-7000  $\text{CO}_2/\text{H}_2\text{O}$  Gas analyser. Relative SDs for DIC were  $\pm 0.1\%$  ( $n = 10$ ). TA was determined using the standard operating procedure for open cell potentiometric titration (Dickson et al., 2007, SOP 3b), using an automatic titrator (Metrohm 888 Titrand), a high accuracy burette ( $1 \pm 0.001$  mL), a thermostated reaction vessel ( $T = 25$  °C) and combination pH glass electrode (Metrohm 6.0259.100). TA values were calculated by a non-linear least-squares fit to the titration data (Dickson et al., 2007, SOP 3b) in a custom-made script in the open source programming framework R (R Core Team, 2013). The relative SD of the procedure was  $\pm 0.2\%$  ( $n = 10$ ). Quality assurance of TA and DIC analysis involved regular analysis of Certified Reference Materials (CRM Batch 126 provided by A. G. Dickson, Scripps Institution of Oceanography).

Water samples for chlorophyll analysis were filtered onto 25 mm GF/F filters (Whatman, nominal pore size of 0.7  $\mu\text{m}$ ). Filters were placed in 10 mL of 96 % ethanol for 18

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to 24 h and chlorophyll fluorescence in the filtrate was analyzed using a fluorometer (TD-700, Turner Designs) before and after addition of 200  $\mu\text{L}$  of a 1 M HCl solution.

Primary production was measured using the  $^{14}\text{C}$  incubation method (Nielsen, 1952). Incubation bottles were filled with 55 mL unfiltered seawater and spiked with 175  $\mu\text{L}$   $\text{NaH}^{14}\text{CO}_3$  ( $20 \mu\text{Ci mL}^{-1}$ ) and incubated for two hours in an ICES incubator (Hydro-Bios, Germany). The samples were filtered onto 25 mm GF/F filters (Whatman) and 100  $\mu\text{L}$  of 1 M HCl was added to remove excess  $\text{NaH}^{14}\text{CO}_3$  and the filters were left open for 24 h in the fume hood. Subsequently, 10 mL of scintillation cocktail (Ultima Gold, Perkin Elmer) was added to the samples before counting them on the scintillation counter (Liquid Scintillation Analyzer, Tri-Carb 2800TR, PerkinElmer). After subtracting fixation rates obtained from the dark incubations, gross primary production rates were calculated based on measured DIC concentrations. Photosynthesis-Irradiance (P-I) curves were obtained for 11 sampling dates at 2 separate depths (5 and 20 m). The light extinction coefficient was calculated from the measured PAR profile. Solar irradiance values were obtained from the meteorological survey in Nuuk (Meteorological station 522, Asiaq Greenland Survey). Using the solar irradiance at each day, the light extinction coefficient and the P-I curves at the monthly sampling dates, the daily productivity was calculated over the entire year. This approach assumes that light extinction and P-I curves remain the same in the two-week period before and after the sampling dates.

Bacterial production was measured using the  $^3\text{H}$ -thymidine method (Fuhrman and Azam, 1982). Triplicate samples (10 mL) were incubated at in situ temperature. After an incubation of 6 to 8 h, bacterial activity incubations were stopped by adding 500  $\mu\text{L}$  of 100 % trichloroacetic acid (TCA). The samples were subsequently filtered through 25 mm cellulose ester filters (pore size 0.2  $\mu\text{m}$ , Advantec MSF). Equations from Søgaard et al. (2010) were used to calculate bacterial production. For the calculation of the bacterial carbon demand from the bacterial production, a bacterial growth efficiency of 0.5 was used according to Rivkin et al. (2001).

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The difference in  $p\text{CO}_2$  between surface water and atmosphere drives the air–sea  $\text{CO}_2$  exchange (ASE). Air–sea  $\text{CO}_2$  fluxes were calculated using the relation:

$$\text{ASE} = K_{\text{av}} \alpha \Delta p\text{CO}_2 \quad (1)$$

where  $\Delta p\text{CO}_2$  is the difference in  $p\text{CO}_2$  of the surface water (here at 1 m water depth) and the atmospheric  $p\text{CO}_2$ . Negative values of ASE imply an uptake by the sea and positive values an efflux to the atmosphere. The atmospheric  $p\text{CO}_2$  was measured monthly at GF3 using an infrared  $\text{CO}_2$  monitor (EGM-4 PP systems). The mean atmospheric  $p\text{CO}_2$  was  $400 \mu\text{atm}$  for 2013. The quantity  $\alpha$  is the solubility of  $\text{CO}_2$  in seawater ( $\text{mol m}^{-3} \text{atm}^{-1}$ ).  $K_{\text{av}}$  ( $\text{m s}^{-1}$ ) is the gas transfer coefficient calculated using both the formulation of Nightingale et al. (2000) and of Wanninkhof and McGillis (1999). These formulations depend on the wind speed data ( $\text{m s}^{-1}$ ), at 10 m a.s.l., obtained from the Nuuk weather station in the fjord system (Meteorological station 522, Asiaq Greenland Survey). The monthly mean wind speed varied from 5 to  $9 \text{ m s}^{-1}$  for 2012 and 2013 but during storms wind speed peaks up to  $30 \text{ m s}^{-1}$  were recorded.

Processing of data was done in the open-source programming language R (R Core Team, 2013). The AquaEnv R-package (Hofmann et al., 2010) was used for acid-base speciation and  $\text{CO}_2$  system calculations. Interpolation of the data and contour plots were produced using the Akima package (Akima et al., 2006).

### 2.3 Biogeochemical model

To analyze the impact of the glacial meltwater input on the seasonal carbon dynamics of the fjord system, a simplified biogeochemical model was constructed. The model describes how the  $p\text{CO}_2$  dynamics is influenced by the circulation in the fjord (including the input of glacial meltwater), air–sea exchange of  $\text{CO}_2$  and net ecosystem production. This biogeochemical model is constrained by data from monthly measurements of 2013 and the seasonal length section data from the fjord system, using an inverse modelling procedure.

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The biogeochemical model fjord system is composed out of three separate, connected boxes, representing the outer, central and inner part of the fjord system and one large “open sea” box, representing the outer shelf; the latter was added to obtain full mass balance closure. Each box represents the upper 40 m of the water column since this depth range is most strongly affected by primary production (due to light availability) and the glacial meltwater imprint. The model includes a water mass balance in addition to the mass balances of three state variables (salinity, DIC and TA) for each box (Table 1). Once DIC and TA are known, all relevant parameters of the carbonate system (including  $p\text{CO}_2$ ) can be calculated.

### 2.3.1 Water mass balance

Figure 2 shows a simplified circulation model for the fjord system. Intrusion of coastal water into the fjord, leads to an input of saline water ( $F_i$ ) in each of the three zones. The inner zone of the fjord experiences an input of glacial meltwater ( $Q_g$ ). The combination of saline seawater intrusion and freshwater from the glacier results in a return flow ( $Q_i$ ) in the surface water which is eventually discharged onto the Greenland shelf. The resulting water mass balance equations are shown in Table 1 (Eqs. 1–3, Table 1). Based on the water mass balance equations, a mass balance can be constructed for salinity in the different zones (Table 1).

The magnitude of the different flows in the fjord system is unknown and these flows are expected to vary strongly throughout the year due to the strong seasonality in the glacial meltwater input as well as seasonal inflows of coastal water (Mortensen et al., 2011). The water mass balance provides 3 independent equations, which allows to constrain the three return flows,  $Q_i(t)$ , when the seawater inputs,  $F_i(t)$ , and glacial meltwater input,  $Q_g(t)$ , are known.

The variation of the glacial meltwater input with time,  $Q_g(t)$ , was estimated from salinity observations at station GF10 close to the outlet glaciers. The total annual meltwater input into Godthåbsfjord was constrained to be  $20 \text{ km}^3 \text{ yr}^{-1}$  as derived from regional climate model simulations for Godthåbsfjord (Langen et al., 2014; Van As et al., 2014).

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The relative contribution of freshwater ( $x$ ) at station GF10 was estimated from a two end-member mixing model ( $S = x \cdot S_{FW} + (1 - x) \cdot S_{SW}$ ) (Sect. 2.3.3.). The contribution  $x$  was calculated at each month and fitted with a smoothing spline. Assuming that  $x$  scales with  $Q_g(t)$ , the temporal variation of  $x$  was used to predict the temporal variation of  $Q_g$  (and ensuring that integrated annual meltwater was equal to  $20 \text{ km}^3 \text{ yr}^{-1}$ ).

The values of the three remaining seawater inputs ( $F_i(t)$ ) in the different zones were obtained by an inverse modelling approach using the monthly salinity data obtained at the three representative stations (GF3, GF7 and GF10). To this end, we estimated the salinity changes  $dS_i/dt$  in the different zones by fitting a cubic smoothing spline (Hastie and Tibshirani, 1990) through the monthly salinity data and subsequently taking the derivative. If we implement both the observed salinities and the salinity changes, we obtain a linear set of three equations (Eqs. 4–6, Table 1), which allows to estimate the seawater inputs ( $F_i(t)$ ).

### 2.3.2 Dissolved inorganic carbon balance

The different flows as derived from the water mass balance were used to parameterize the transport terms in the mass balances for TA (Eqs. 7–9, Table 1) and DIC (Eqs. 10–12, Table 1). Alkalinity was assumed to behave conservatively within the fjord, and hence was only influenced by transport (Thomas and Schneider, 1999). In contrast, the DIC mass balance accounted for transport processes, but also air–sea exchange of CO<sub>2</sub> (ASE) and net community production (NCP) (Table 1). Air–sea CO<sub>2</sub> fluxes were calculated according to Eq. (1) using the formulation by Nightingale et al. (2000) for the gas transfer coefficient. The  $p\text{CO}_2$  concentration of the surface water was calculated from salinity, temperature, TA and DIC using the R package AquaEnv (Hofmann et al., 2010). The net community production (NCP) was calculated as the difference between the primary production (PP) and bacterial carbon demand (BCD) and values for the rate of these processes were determined based on monthly rate measurements. Accordingly, NCP was imposed as a forcing function upon the model.

### 2.3.3 End members composition

The model uses two end-member types of water as input, freshwater from glacial meltwater and saline water. The composition (S, DIC, TA) of both end-member types of water was determined based on collected data. The largest fraction of the freshwater input is from glacial origin and so we used the properties for glacial meltwater for the freshwater end-member. The values were estimated from twenty icebergs samples collected in the fjord. Salinity, DIC and TA were measured after thawing the ice in the laboratory in gas-tights bags ( $S_{FW} = 0$ ,  $DIC_{FW} = 80 \pm 17 \mu\text{mol kg}^{-1}$ ,  $TA_{FW} = 50 \pm 20 \mu\text{mol kg}^{-1}$ ). For the seawater end-member, we used the properties of deep fjord water (water at 400 m depth) which is shown to be comparable to the properties of the water on the shelf ( $S_{SW} = 33.65$ ,  $DIC_{SW} = 2150 \mu\text{mol kg}^{-1}$ ,  $TA_{SW} = 2220 \mu\text{mol kg}^{-1}$  and  $T_{SW} = 2^\circ\text{C}$ ).

### 2.3.4 Numerical solution

A numerical solution procedure for the resulting differential equations was implemented in the open-source programming language R following Soetaert and Meysman (2012). The set of differential equations was integrated using the R-package deSolve (Soetaert et al., 2010). The calculation of the carbonate system (and hence  $p\text{CO}_2$ ) at each time step of the numerical simulation was performed via the operator splitting approach as detailed in Hofmann et al. (2008). The resulting  $p\text{CO}_2$  concentration then can be employed in the kinetic rate expression for the air–sea  $\text{CO}_2$  exchange. The model was run over a full seasonal cycle (representing the year 2013) and with a spin-up period of 2 years. The goodness of fit (GOF) between model simulation output and observational data was calculated as the sum of squared residuals.

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## 3 Results

### 3.1 Spatial variability in the fjord system

The hydrography of Godthåbsfjord is strongly affected by the seasonal input of glacial meltwater. Figure 3 shows the spatial distribution of salinity in the upper 40 m of the water column during four cruises in 2013 (February, May, August and October). The length transects range from the west Greenland continental slope to the inner part of the fjord where six glaciers discharge. Low freshwater runoff during winter and spring months coincides with high salinities  $\sim 33$  in the upper 40 m of the water column throughout the fjord (Fig. 3a and b). Increased input of glacial meltwater during summer creates a strongly stratified system, where the surface water shows a distinct layer in the central and inner part of the fjord (Fig. 3c). Due to distribution of freshwater sources, the impact is most pronounced at the inner fjord stations (GF9 to GF13) where in August salinity drops to  $\sim 8$  in a shallow surface water layer of 10 m depth. In the central fjord (GF5 to GF8), summer salinity decreases to 17 and upper layer of low-saline upper water layer extends deeper to 15–20 m depth. In the outer part of the fjord (the outer sill region, GF1 to GF4) the salinity decrease with depth is less pronounced as the freshwater is mixed deeper into the water column by strong tidal mixing. Still, even at the shelf stations (FB1 to FB4), a weak imprint of glacial meltwater can be observed. Decreased input of glacial meltwater during the autumn months coincides with a gradual increase in salinity in the surface layer and a less steep halocline (Fig. 3d).

Strong seasonality was also observed in the fluorescence data (Fig. 4). Evidence of a spring bloom is indicated by high chlorophyll *a* concentrations on the shelf (FB4 to FB1) and in the central fjord (GF5 to GF8) observed during the May cruise (Fig. 4b). In contrast to the May situation, where the chlorophyll *a* is evenly distributed in the upper 40 m of the water column, distinct chlorophyll maxima were observed in August (Fig. 4c). During the August cruise, high fluorescence values of  $\sim 6 \mu\text{g L}^{-1}$  were observed at the outer shelf stations at approximately 30 m depth (FB4 and FB3.5). Clear chlorophyll *a* maxima also occurred in the central and inner part fjord with values of

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these three stations, an area-averaged annual CO<sub>2</sub> uptake in the entire fjord system is estimated to be ~ 65 gC m<sup>-2</sup> yr<sup>-1</sup>.

The mean CO<sub>2</sub> uptake was also calculated for the seasonal transects in May, August and October based on the measured *p*CO<sub>2</sub> surface data and using the daily wind speed values during the month that spans the sampling date. Confirming the pattern observed at the three monitoring stations, the uptake is higher close to the glaciers and lower in stations downstream the fjord, but rises slightly again over the shelf (Fig. 8c). Lower surface *p*CO<sub>2</sub> in the central and inner fjord led to almost a doubling of the CO<sub>2</sub> uptake in the inner part of the fjord compared to the outer part. Anova analysis indicates that uptake is significantly different between stations and between different months (*P* < 0.001).

### 3.4 Model results: driving factors of the carbon dynamics

To resolve the importance of the different driving forces of CO<sub>2</sub> uptake in the fjord, a simplified biogeochemical model was used to simulate the DIC and *p*CO<sub>2</sub> in the region closest to the glacier (GF10, zone 3) (Fig. 2). This zone is most strongly affected by glacial meltwater input and primary production, and hence, shows the highest excursions in the parameters of the carbonate system. Figure 9 shows the simulated annual cycle of DIC and *p*CO<sub>2</sub> at GF10 compared with the measured data. Simulations were performed (1) with and without the effect of net community production on carbon dynamics and (2) with a constant temperature throughout the year (the average mean winter temperature in upper 40 m 0.5 °C) or with a variable temperature based on the observations. Simulations that include NCP manage to reproduce the DIC and *p*CO<sub>2</sub> evolution better as quantified by goodness of fit. NCP has especially a strong effect on the evolution of the *p*CO<sub>2</sub>. The inclusion of seasonal temperature variation had overall a moderate effect on the simulation output. Higher temperatures during summer and autumn (1.5 to 3 °C, Fig. 7a) led to a reduction of *p*CO<sub>2</sub> undersaturation in the simulation with variable temperature.

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different composition. When mixing is conservative, the concentration of a chemical compound obeys the relation

$$[E]_{\text{Mix}}(M_1 + M_2) = [E]_1 M_1 + [E]_2 M_2 \quad (2)$$

where  $[E]$  is the concentration of the compound  $E$  and  $M$  are the masses of the water parcels. Salinity, TA and DIC are conservative quantities with respect to mixing (Wolf-Gladrow et al., 2007). However, the fact that TA and DIC mix conservatively, does not imply that  $p\text{CO}_2$  will behave conservatively upon mixing. In other words, if two water parcels, initially in equilibrium with the atmosphere, mix, it does not imply that the resulting mixture will also be in equilibrium with the atmosphere. In fact, mixing of fresh water and saline water induces a strong  $p\text{CO}_2$  undersaturation. Figure 11a shows the undersaturation created when two water masses at equilibrium are mixed, one with low salinity, low TA and low DIC (representative for a water parcel from glacial origin) and one with high salinity, high TA and high DIC (representative for saline fjord water). The mixture of these two parcels will be undersaturated in  $p\text{CO}_2$  due to the non-linear effect of salinity on  $p\text{CO}_2$ . Consequently this water parcel will take up  $\text{CO}_2$  when in contact with the atmosphere (Fig. 11a). Note that the strongest undersaturation is obtained when the resulting mixture has a salinity of  $\sim 8$  and that  $p\text{CO}_2$  undersaturation can exceed  $200 \mu\text{atm}$  below the atmospheric level (Fig. 11a). This salinity effect on pH and  $p\text{CO}_2$  dynamics has been described previously for estuarine systems (Mook and Koene, 1975; Whitfield and Turner, 1986) but as yet, the mechanism has not been invoked for glacier affected systems. In fjord systems affected by glacial melt, meltwater (with low TA, DIC and salinity) mixes with ambient fjord waters (with high TA, DIC and salinity). This mechanism hence could constitute an important driver for  $p\text{CO}_2$  undersaturation when sufficiently large amounts of meltwater are discharged into the fjord, so that salinity levels are sufficiently reduced. The undersaturation that has been previously observed in other systems affected by glacier melt water (Evans et al., 2014; Sejr et al., 2011; Torres et al., 2011) might possibly be explained by this same mechanism.

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### 4.3.1 Phase I: autumn and winter period

During autumn, glacier melting decreases and freshwater runoff to the fjord slowly diminishes (Figs. 3 and 7). Combined with dense coastal inflows to the fjord, this leads to a gradual salinity increase in the upper water layer and flushing of accumulated freshwater out of the Godthåbsfjord system (Fig. 7) (Mortensen et al., 2011, 2013). Weakening of the surface stratification combined with strong winter storms lead to a stronger mixing of the upper water layers. DIC and  $p\text{CO}_2$  concentration increases slowly due to advection of water masses with high DIC and  $p\text{CO}_2$  (upwelling of fjord deep water) and continued air–sea exchange to reach an average  $p\text{CO}_2$  of  $\sim 350 \mu\text{atm}$  (Figs. 7, 8 and 10).

### 4.3.2 Phase II: spring bloom

At the start of April, a strong spring bloom is observed in the inner part of the fjord, leading to high chlorophyll *a* concentrations, high net primary production and strong  $\text{CO}_2$  uptake (Fig. 7). The high biological carbon uptake decreases the DIC and lowers the  $p\text{CO}_2$  to  $250 \mu\text{atm}$  in the surface waters of the inner fjord (GF10) by the middle of May (Figs. 7 and 10). The strong effect of primary production on the  $p\text{CO}_2$  is observed in the entire fjord system in May (Figs. 4 and 5). Low surface water  $p\text{CO}_2$  concentrations occur in almost the entire fjord system and on the shelf. During this period, the impact of the glacial meltwater on the fjord system is not pronounced, and the upper water column is still well mixed (Mortensen et al., 2011, 2013). Consequently low  $p\text{CO}_2$  occur at nearly constant salinity and the undersaturation is almost homogenous in the water column. This matches with the evenly distribution of the fluorescence in the upper 40 m (Figs. 3–5). Only in the inner part of the fjord a clear gradient in  $p\text{CO}_2$  with water depth can be observed. Continued inflow of dense water into the fjord system leads to upwelling in the inner part of the fjord, bringing up the deep water masses rich in DIC and  $p\text{CO}_2$  (Figs. 3 and 5) (Mortensen et al., 2011).





the inner part of the fjord. Consequently both glacial meltwater and primary production can be considered as crucial drivers for the CO<sub>2</sub> uptake in coastal areas affected by glacial meltwater.

Part of the low  $p\text{CO}_2$  created by glacial meltwater and biological processes in the fjord is however compensated by higher temperatures in summer and autumn which reduce the CO<sub>2</sub> solubility in water, therefor counteracting the established undersaturation (Fig. 9) (Shadwick et al., 2011; Takahashi et al., 2002). Higher temperatures reduce  $p\text{CO}_2$  up to 50  $\mu\text{atm}$  (Fig. 9), reducing the CO<sub>2</sub> uptake with 10–20 % compared to constant temperature simulation.

## 5 Summary and outlook

Our observations show that Godthåbsfjord is a strong sink for CO<sub>2</sub> due to high biological carbon uptake and undersaturation induced by the input of glacial meltwater. During winter, absence of significant glacial meltwater and biological consumption brings the fjord waters near equilibrium with the atmosphere due to air–sea exchange. A strong bloom during spring leads to a decrease in DIC and  $p\text{CO}_2$  indicating the importance of biological processes. During summer, primary production continues to play a central role in the carbon dynamics. But the input and mixing of glacial melt water also plays a crucial role during the summer months. The non-linear effect of salinity on surface water  $p\text{CO}_2$  from the mixing of glacial meltwater and saline fjord water creates a strong  $p\text{CO}_2$  undersaturation and CO<sub>2</sub> uptake, a mechanism that was yet undescribed for glacial systems.

The processes driving the DIC and  $p\text{CO}_2$  dynamics in Godthåbsfjord most likely also apply for other fjord systems and coastal settings that are affected by glacier meltwater. Consequently coastal areas of Greenland and other glacier-influenced areas probably constitute a much larger sink compared to other coastal areas and play a more important role in the high-latitude carbon cycle. Increased melting is anticipated as a result of climate change and will likely accelerate processes affecting carbon dynamics; it will in-

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crease the freshwater volume mixing in fjord systems and consequently likely enhance the sink for CO<sub>2</sub> in fjord systems affected by glacial melt.

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**Table 1.** Mass balance equations of the biogeochemical model.  $V_i$  and  $A_i$  are respectively the volumes and areas of the different zones.  $\Delta p\text{CO}_2$  is the difference in  $p\text{CO}_2$  of the water (modelled) and the atmospheric  $p\text{CO}_2$  (400  $\mu\text{atm}$ ) with negative values implying an uptake by the sea.  $\alpha$  is the  $\text{CO}_2$  solubility ( $\text{mol m}^{-3} \text{atm}^{-1}$ ).  $K_{\text{av}}$  ( $\text{m s}^{-1}$ ) is the gas transfer coefficient calculated using the formulation of Nightingale et al. (2000). NCP is the net community production.

Water mass balance		
Zone 1:	$Q_1(t) = Q_2(t) + F_1(t)$	1
Zone 2:	$Q_2(t) = Q_3(t) + F_2(t)$	2
Zone 3:	$Q_3(t) = Q_g(t) + F_3(t)$	3
Salinity mass balance		
Zone 1:	$\frac{dS_1}{dt} = \frac{1}{V_1}(Q_2(t)S_2 + F_1(t)S_{\text{SW}} - Q_1(t)S_1)$	4
Zone 2:	$\frac{dS_2}{dt} = \frac{1}{V_2}(Q_3(t)S_3 + F_2(t)S_{\text{SW}} - Q_2(t)S_2)$	5
Zone 3:	$\frac{dS_3}{dt} = \frac{1}{V_3}(Q_g(t)S_{\text{FW}} + F_3(t)S_{\text{SW}} - Q_3(t)S_3)$	6
Total alkalinity (TA) mass balance		
Zone 1:	$\frac{d\text{TA}_1}{dt} = \frac{1}{V_1}(Q_2(t)\text{TA}_2 + F_1(t)\text{TA}_{\text{SW}} - Q_1(t)\text{TA}_1)$	7
Zone 2:	$\frac{d\text{TA}_2}{dt} = \frac{1}{V_2}(Q_3(t)\text{TA}_3 + F_2(t)\text{TA}_{\text{SW}} - Q_2(t)\text{TA}_2)$	8
Zone 3:	$\frac{d\text{TA}_3}{dt} = \frac{1}{V_3}(Q_g(t)\text{TA}_{\text{FW}} + F_3(t)\text{TA}_{\text{SW}} - Q_3(t)\text{TA}_3)$	9
Dissolved inorganic carbon (DIC) balance		
Zone 1:	$\frac{d\text{DIC}_1}{dt} = \frac{1}{V_1}(Q_2(t)\text{DIC}_2 + F_1(t)\text{DIC}_{\text{SW}} - Q_1(t)\text{DIC}_1) - \frac{A_1}{V_1}K_{\text{av}}\alpha\Delta p\text{CO}_2(t) - \text{NCP}_1(t)$	10
Zone 2:	$\frac{d\text{DIC}_2}{dt} = \frac{1}{V_2}(Q_3(t)\text{DIC}_3 + F_2(t)\text{DIC}_{\text{SW}} - Q_2(t)\text{DIC}_2) - \frac{A_2}{V_2}K_{\text{av}}\alpha\Delta p\text{CO}_2(t) - \text{NCP}_2(t)$	11
Zone 3:	$\frac{d\text{DIC}_3}{dt} = \frac{1}{V_3}(Q_g(t)\text{DIC}_{\text{FW}} + F_3(t)\text{DIC}_{\text{SW}} - Q_3(t)\text{DIC}_3) - \frac{A_3}{V_3}K_{\text{av}}\alpha\Delta p\text{CO}_2(t) - \text{NCP}_3(t)$	12

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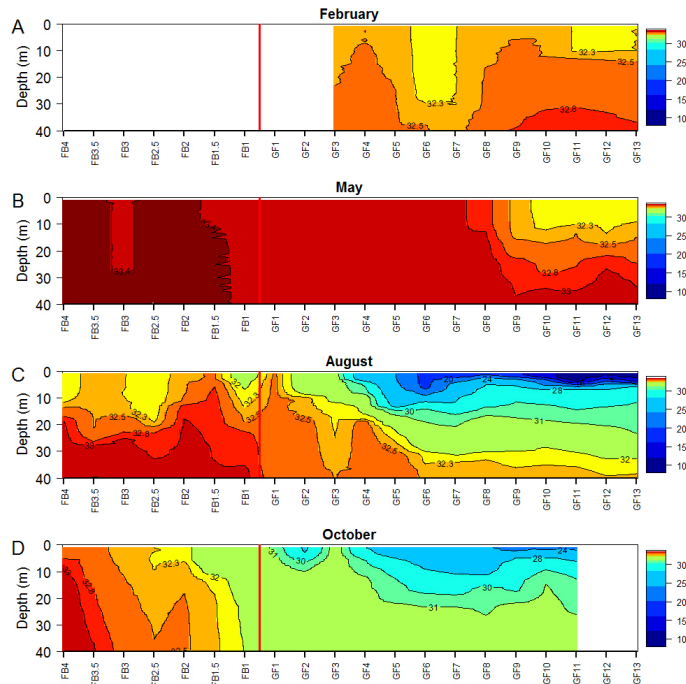






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**Figure 3.** Transects of salinity from shelf (left) to the glaciers (right) during February **(a)**, May **(b)**, August **(c)** and October **(d)** 2013. The red line indicates the mouth of Godthåbsfjord area.

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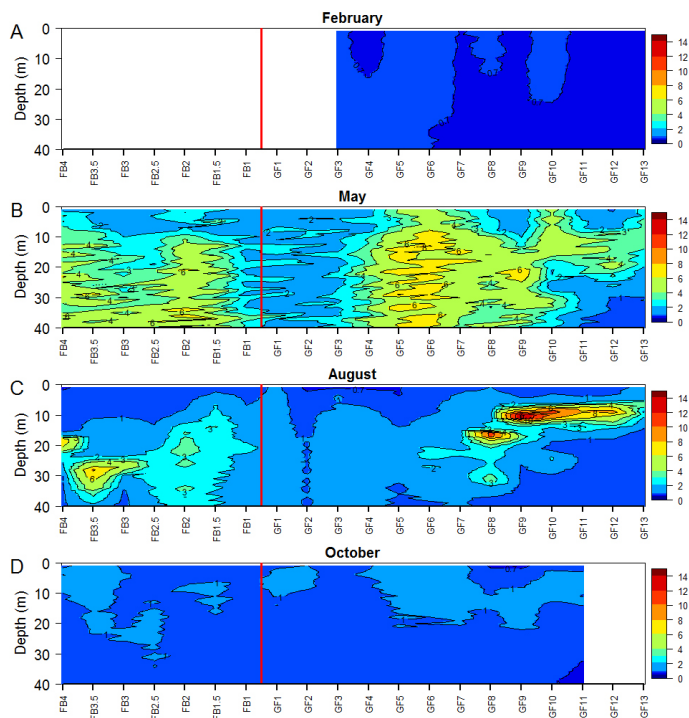
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**Figure 4.** Transects of fluorescence (calibrated vs. chlorophyll *a* in  $\mu\text{g L}^{-1}$ ) from shelf area (left) to glaciers (right) during February (a), May (b), August (c) and October (d) 2013. The red line indicates the mouth of Godthåbsfjord area.

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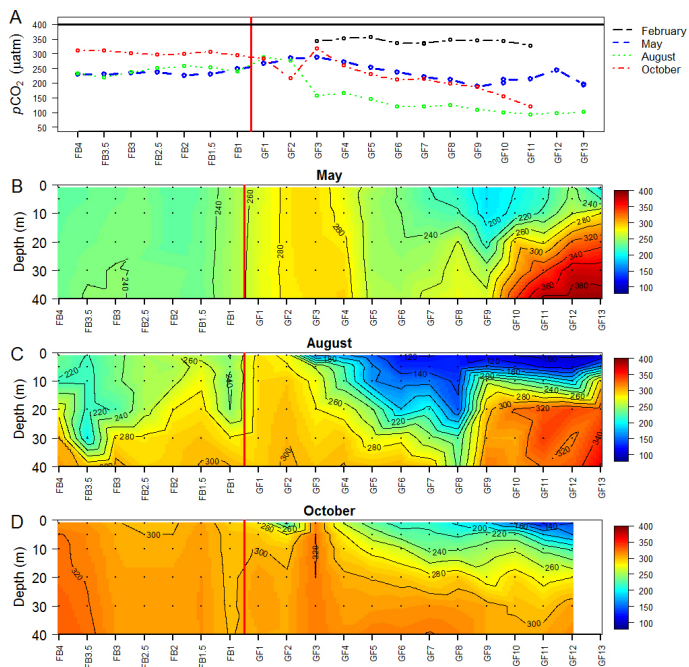
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**Figure 5.** Partial CO<sub>2</sub> pressure data ( $p\text{CO}_2$  in  $\mu\text{atm}$ ) at 1 m depth for the four cruises (a). The full line indicates the average atmospheric concentration (400  $\mu\text{atm}$ ) measured during the year 2013.  $p\text{CO}_2$  data for the May (b), August (c) and October (d) cruise in the upper 40 m water column from shelf area (left) to glaciers (right). The red line indicates the mouth of Godthåbsfjord area.

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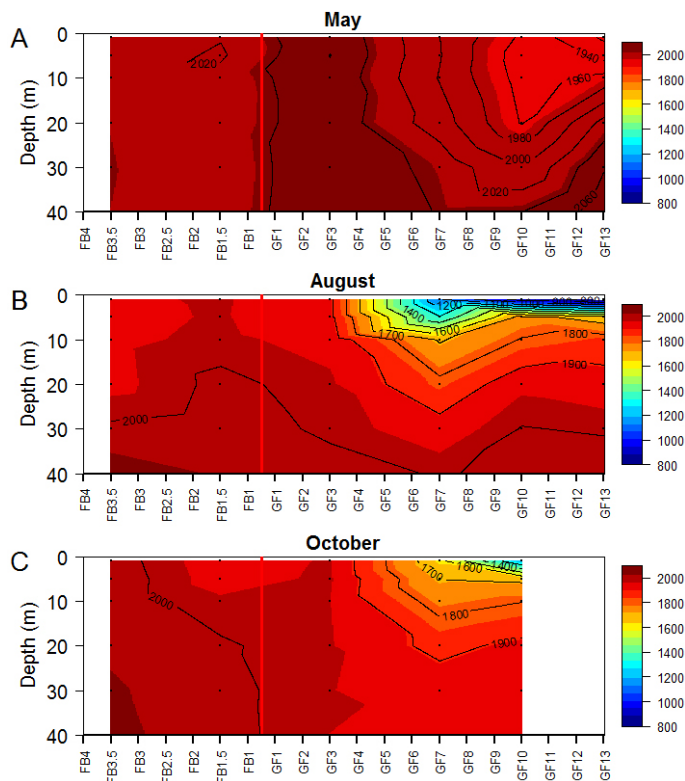
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**Figure 6.** Dissolved inorganic carbon (DIC in  $\mu\text{mol kg}^{-1}$ ) data for May (a), August (b) and October (c) 2013 along a transect from the shelf (left) to the glaciers (right). The red line indicates the mouth of Godthåbsfjord area.

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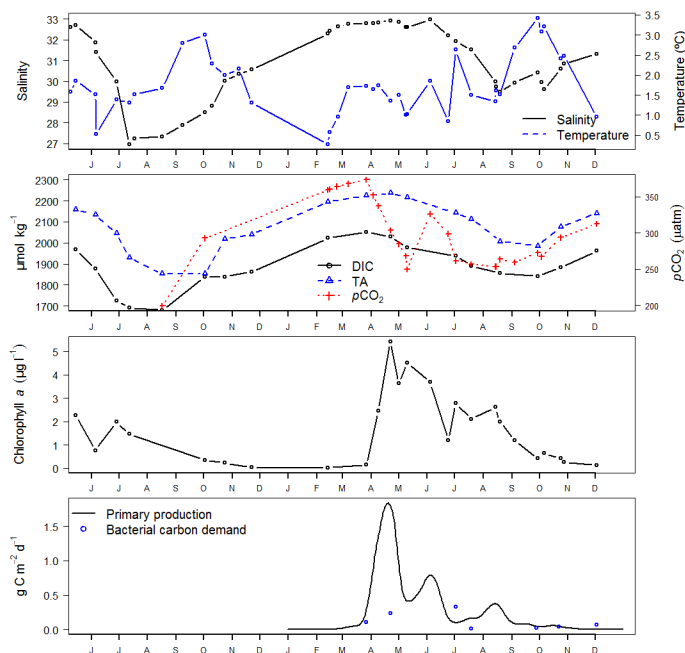
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**Figure 7.** Time series of average (0–40 m) salinity and temperature (°C) **(a)**, DIC, TA ( $\mu\text{mol kg}^{-1}$ ) and CO<sub>2</sub> partial pressure ( $p\text{CO}_2$ ,  $\mu\text{atm}$ ) **(b)** chlorophyll *a* concentration ( $\mu\text{g L}^{-1}$ ) **(c)** and primary production and bacterial carbon demand ( $\text{g C m}^{-2} \text{d}^{-1}$ ) **(d)** from June 2012 to December 2013 for station GF10.

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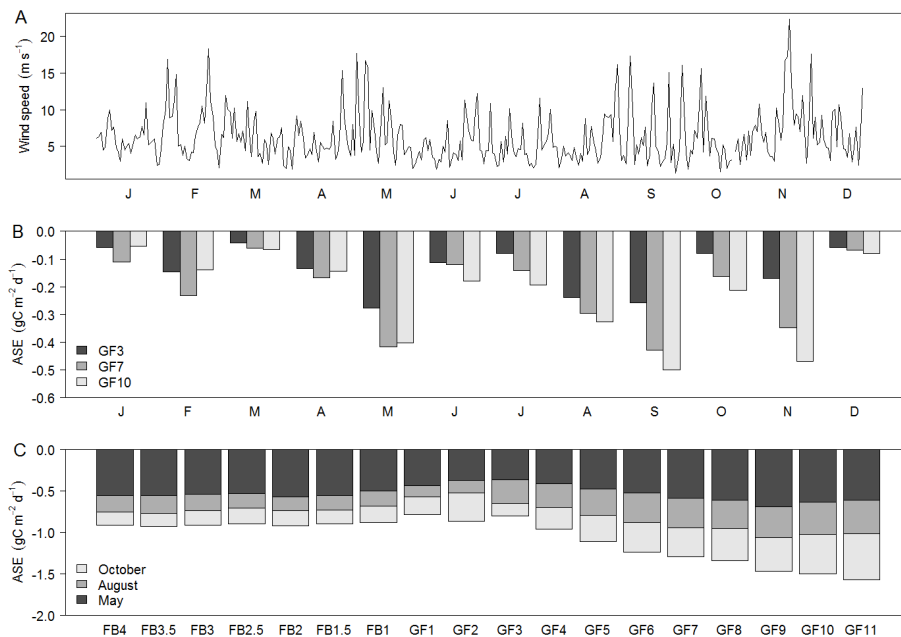
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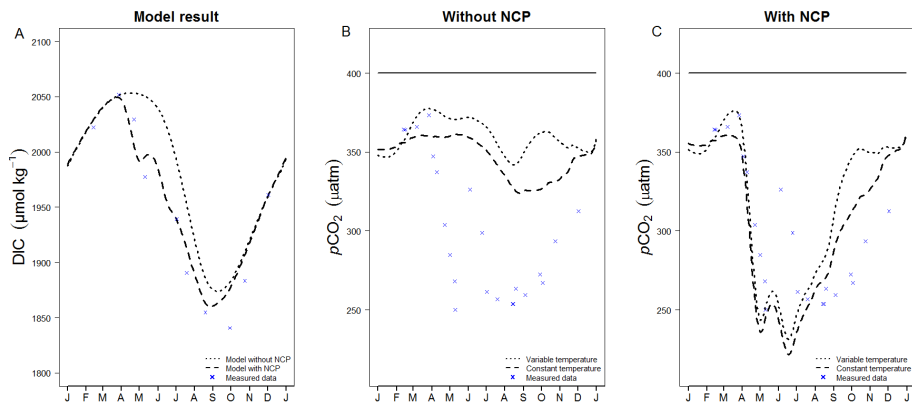
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**Figure 8.** (a) Mean daily wind speed ( $\text{m s}^{-1}$ ) at the meteorological station in Nuuk. (b) Time series of mean monthly air–sea CO<sub>2</sub> flux (ASE,  $\text{gC m}^{-2} \text{d}^{-1}$ ) at three stations (GF3, GF7 and GF10) in the fjord. (c) Mean air–sea exchange (ASE,  $\text{gC m}^{-2} \text{d}^{-1}$ ) from 3 cruises in fjord system from shelf (Fyllas Banke, left) to inner fjord glaciers (right).

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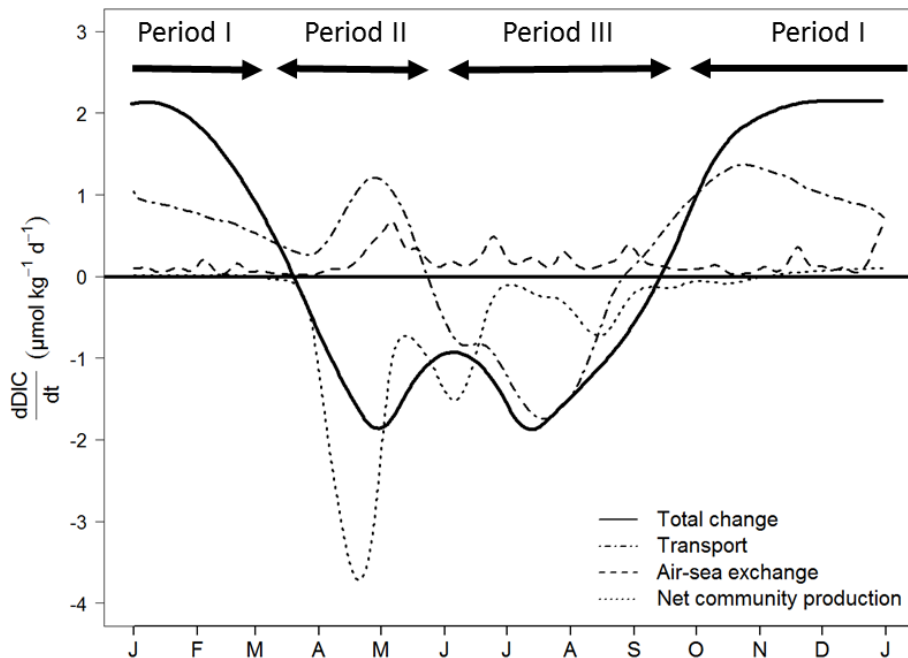


**Figure 9.** Seasonal evolution of DIC in  $\mu\text{mol kg}^{-1}$  (a) and  $p\text{CO}_2$  in  $\mu\text{atm}$  (b and c) calculated by the biogeochemical model together with data from 2013 in station GF10 (blue points indicate measured data averaged over a 40 m box). Simulations of the model are shown with and without net community production (NCP). Simulations of the evolution of  $p\text{CO}_2$  are shown without (b) and with NCP (c) and for a variable temperature and constant temperature ( $0.5^\circ\text{C}$ , the average winter temperature).

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**Figure 10.** Overview of how different processes (Transport, Air–sea exchange and Net community production) contribute to the temporal observed change in DIC ( $\mu\text{mol kg}^{-1} \text{d}^{-1}$ ) for the station close to the ice sheet (GF10). Uptake of CO<sub>2</sub> by the sea from the atmosphere is shown as a positive value.

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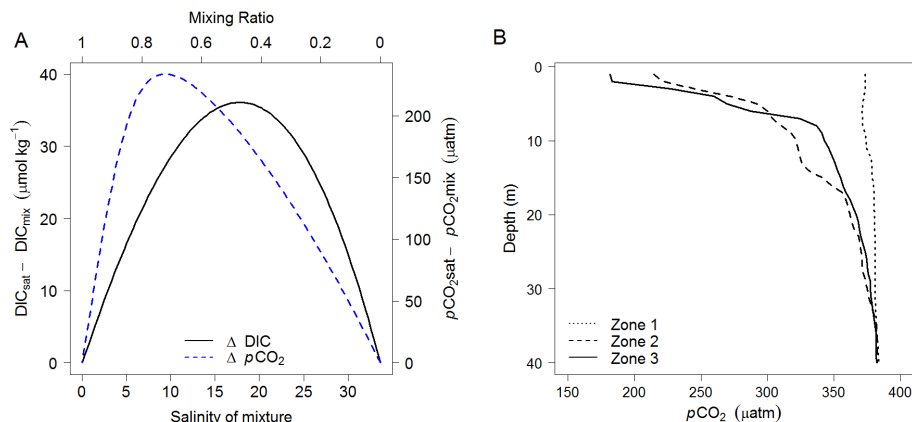
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## Glacial meltwater and primary production as drivers for strong CO<sub>2</sub> uptake

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**Figure 11.** Undersaturation created as  $\Delta\text{DIC}$  (in  $\mu\text{mol kg}^{-1}$ ) and  $\Delta p\text{CO}_2$  (in  $\mu\text{atm}$ ) as a function of salinity of the mixture (and mixing ratio,  $x$ , indicating the fraction freshwater) when two water masses in equilibrium with atmosphere are mixed (a). A first water parcel in equilibrium with the atmosphere with TA of  $50 \mu\text{mol kg}^{-1}$ , DIC of  $81.2 \mu\text{mol kg}^{-1}$ , salinity of 0 and temperature of  $0^\circ\text{C}$  (glacial origin). And a second parcel in equilibrium with the atmosphere with TA of  $2220 \mu\text{mol kg}^{-1}$ , DIC of  $2118 \mu\text{mol kg}^{-1}$ , salinity of 33.65 and temperature of  $0^\circ\text{C}$  (fjord/sea water). Panel (b) shows the estimated  $p\text{CO}_2$  profile calculated from the salinity profiles of August in the three different zones in the fjord system.

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