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Biogeosciences Discussions

Enhancement of the North Atlantic CO_2 sink by Arctic Waters

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Abstract

The North Atlantic north of 50°N is one of the most intense ocean sink areas for atmospheric CO_2 considering the flux per unit area, 0.27 Pg-C yr^{-1}, equivalent to -2.5 mol C m^{-2} yr^{-1}. The Northwest Atlantic Ocean is a region with high anthropogenic carbon inventories. This is on account of processes which sustain CO_2 air-sea fluxes, in particular strong seasonal winds, ocean heat loss, deep convective mixing and CO_2 drawdown by primary production. The region is in the northern limb of the Global Thermohaline Circulation, a path for the long term deep sea sequestration of carbon dioxide. The surface water masses in the North Atlantic are of contrasting origins and character, on the one hand the northward flowing North Atlantic Drift, a Gulf Stream offspring, on the other hand southward moving cold low salinity Polar and Arctic Waters with signatures from Arctic freshwater sources. We have studied by observations, the CO_2 air-sea flux of the relevant water masses in the vicinity of Iceland in all seasons and in different years. Here we show that the highest ocean CO_2 influx is to the Arctic and Polar waters, respectively, -3.8 mol C m^{-2} yr^{-1} and -4.4 mol C m^{-2} yr^{-1}. These waters are CO_2 undersaturated in all seasons. The Atlantic Water is a weak or neutral sink, near CO_2 saturation, after poleward drift from subtropical latitudes. These characteristics of the three water masses are confirmed by data from observations covering 30 years. We relate the Polar and Arctic Water persistent undersaturation and CO_2 influx to the excess alkalinity derived from Arctic sources, particularly the Arctic rivers. Carbonate chemistry equilibrium calculations indicate clearly that the excess alkalinity may support a significant portion of the North Atlantic CO_2 sink. The Arctic contribution to the North Atlantic CO_2 sink which we reveal is previously unrecognized. However, we point out that there are gaps and conflicts in the knowledge about the Arctic alkalinity budget and that future trends in the North Atlantic CO_2 sink are connected to developments in the rapidly warming Arctic. The results we

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020


Biogeosciences Discussions

present need to be taken into consideration for the question: Will the North Atlantic continue to absorb CO_2 in the future as it has in the past?

1 Introduction

The oceans take up about a quarter of the annual anthropogenic CO_2 emissions (Friedlingstein et al., 2019). The North Atlantic north of 50°N is one of the most intense ocean sink areas for atmospheric CO_2 considering the flux per unit area (Takahashi et al., 2009). The reasons are strong winds and large natural partial pressure differences, $\Delta pCO_2=(pCO_{2sw} - pCO_{2a})$, between the atmosphere and the surface ocean. The ΔpCO_2 in seawater is a measure of the escaping tendency of CO_2 from seawater to the overlying air. The ΔpCO_2 is proportional to the concentration of undissociated CO_2 molecules, $[CO_2]aq$, which constitutes about 1 % of the total CO_2 dissolved in seawater (the remainders being about 90-95 % as $[HCO_3^-]$ and 4-9 % as $[CO_3^{2-}]$). The seawater pCO_2 depends sensitively on temperature and the TCO_2/Alk ratio, the relative concentrations of total CO_2 species dissolved in seawater ($TCO_2 = [CO_2]aq + [HCO_3^-] + [CO_3^{2-}]$) and the alkalinity, Alk, which reflects the ionic balance in seawater. Large ΔpCO_2 has been attributed to, a) a cooling effect on the CO_2 solubility in the poleward flowing Atlantic Water, b) an efficient biological drawdown of pCO_2 in nutrient rich subpolar waters and c) high wind speeds over these low pCO_2 waters (Takahashi et al., 2002). Evaluations of ΔpCO_2 based on observation and models have indicated that the Atlantic north of 50°N and northward into the Arctic takes up as much as 0.27 Pg-C yr^{-1}, equivalent to -2.5 mol C m^{-2} yr^{-1} (Takahashi et al., 2009;Schuster et al., 2013;Landschützer et al., 2013;Mikaloff Fletcher et al., 2006). Estimates of long term trends for the North Atlantic CO_2 sink due to changes in either ΔpCO_2 or wind strength are conflicting, particularly the Atlantic Water dominated regions (Schuster et al., 2013;Landschützer et al., 2013;Wanninkhof et al., 2013). The drivers of seasonal flux variations are inadequately understood (Schuster et al., 2013) and a mechanistic understanding of high latitude CO_2 sinks is considered incomplete (McKinley et al., 2017). It is common to many large scale flux evaluations, modelled or from observations, that they are based on regions defined by geographical borders, latitude and longitude, e.g. between 49°N and 76°N for the high latitude Sub Polar North Atlantic (Takahashi et al., 2009;Schuster et al., 2013). The influence of oceanographic property differences within this region on CO_2 fluxes has thus not been apparent, primarily due to Arctic latitude data limitations. The ability of current generation Earth System Models to predict trends in North Atlantic CO_2 has recently been questioned and suggested that their

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

inadequacies may be caused by biased alkalinity in the simulated background biogeochemical state (Lebehot et al., 2019).

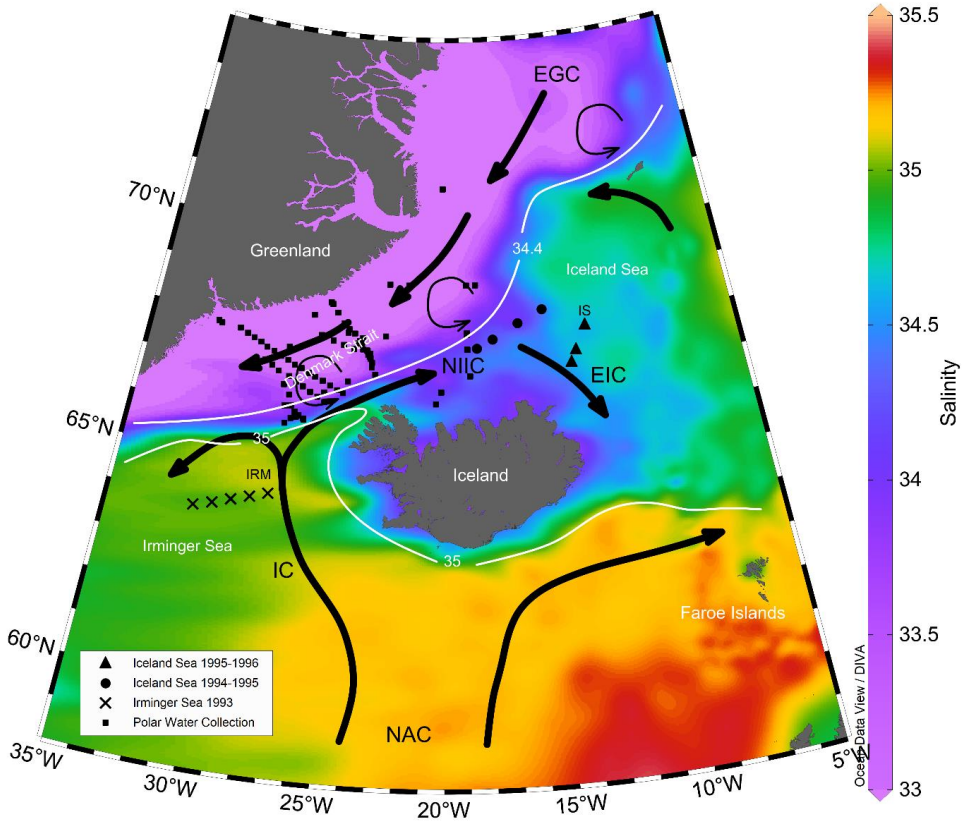
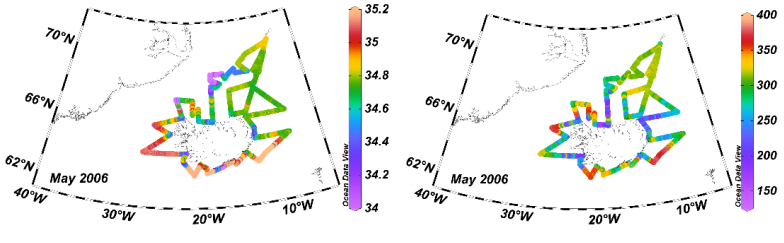


Figure 1. Mean July to September surface salinity in the vicinity of Iceland. *The S=35 isohaline marks the bounday between northward flowing Atlantic Water and southward flowing cold Arctic Water and low salinity Polar Water. Stations in Irminger Sea marked X and stations in Iceland Sea marked ●for 1994-1995 and ▲for 1995-1996 observations. Collection of Polar Water stations 1983-2012 marked ▪. IRM and IS mark the location of time series stations. NAC: North Atlantic Current, IC: Irminger Current, NIIC: North Iceland Irminger Current, EIC: East Icelandic Current, EGC: East Greenland Current. Map based on the NISE dataset (Nilsen et al., 2008) and drawn using the Ocean Data View program (Schlitzer, 2018).*

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

The high latitude North Atlantic Ocean in the vicinity of Iceland, is a region of contrasting surface properties (Fig. 1). The northward flowing North Atlantic Current carries relatively warm and saline Atlantic Water, derived from the Gulf Stream, as far as the Nordic Seas and the Arctic Ocean north of Svalbard. The Irminger Current branch carries Atlantic Water to south and west Iceland and a small branch, the North Icelandic Irminger Current that transports 1 Sv (1 Sv= 10^6 m^3 s^{-1}), reaches the Iceland Sea (Stefánsson, 1962;Våge et al., 2011). The rapid East Greenland Current (EGC) (Håvik et al., 2017) flows southward from the Arctic to the North Atlantic, carrying Polar Water cold and with low salinity, S<34.4, due to ice melt and the large freshwater input to the Arctic from rivers that contribute about 11% of the global riverine discharge (Sutherland et al., 2009;McClelland et al., 2012). In between these extremes there are large areas of the Greenland and Iceland Seas that contain predominantly the intermediate, Arctic Water which is a product of heat loss and freshwater export from the EGC (Fig. 1) (Våge et al., 2015). The north- and southward flowing currents are separated by the Arctic Front outlined in Figure 1 by the salinity=35 contour generally oriented SW-NE. Deep water formation in the high latitude North Atlantic produces cold dense waters which, together with a similar product in the Labrador Sea, are source waters for the Global Thermohaline Circulation linking the regional air-sea CO_2 flux to a route for the long term deep ocean sequestration of anthropogenic CO_2 (Broecker, 1991). The region of our study affects large scale ocean-atmosphere CO_2 exchange processes in the North Atlantic. Here we evaluate regional, seasonal and interannual air-sea carbon dioxide fluxes for the main surface waters characteristic of this region (Fig. 1). We base this work on extensive observations which cover regional water masses, all seasons and include different states of the North Atlantic Ocillation, NAO (Flatau et al., 2003). We employ two different observation approaches for flux estimates. Firstly, repeat station hydrography with emphasis on the seasonal flux patterns in Atlantic Water and in Arctic Water (Fig. 1). Secondly, underway ship records of surface pCO_2 where the emphasis was on the different surface water masses (Fig. 2).



https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

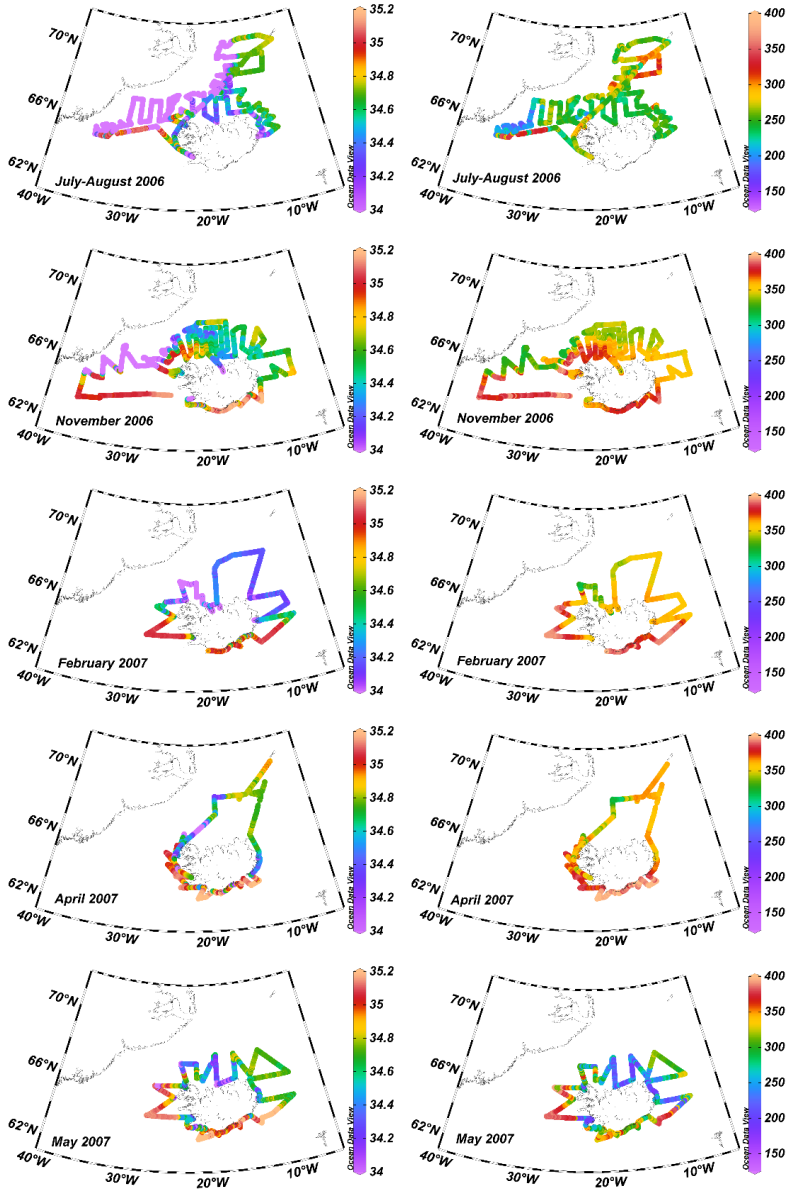


Figure 2. Cruise tracks where surface layer salinity and pCO_2 were recorded underway. Left sea surface salinity, right pCO_2sw(µatm) along the cruise tracks. Maps drawn using the Ocean Data View program (Schlitzer, 2018).

We describe long term carbon chemistry characteristics of water masses in mid winter when physical forces prevail over biological processes. For the Irminger Sea and Iceland Sea from

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

time series observations (Olafsson et al., 2010) and for the EGC Polar Water from a collection of pCO_2 data assembled in the period 1983 to 2012.

2 Methods

2.1 Data acquisition

Seasonal carbon chemistry variations in the relatively warm and saline (S>35) Atlantic Water were studied 1993-1994 on 15 cruises from February 1993 to January 1994 to 5 stations on a 167 km long transect over the core of the Irminger Current and into the northern Irminger Sea (Fig. 1 and Tables S1 and S2). In order to close the full annual cycle, to 23 February 1994 we use data from the previous year and date. In 1994-1996 the study centered on the colder and less saline Arctic Water of the Iceland Sea and was conducted on 22 cruises with sampling dates from 11 Feb 1994 to 12 Feb 1996, two years. In 1994 on 4 stations on a 168 km long transect into the Iceland Sea Gyre and in 1995 on 3 stations across the East Icelandic Current (Fig. 1 and Tables S1 and S3 in the supplement). On each cruise the station work was completed in 1-2 days. For both regions, the timing of cruises was with the period of the phytoplankton spring bloom in mind (Takahashi et al., 1993b). The work was conducted on vessels operated by the Marine Research Institute (MRI) in Reykjavik, Iceland, R/V Bjarni Seamundsson and R/V Arni Fridriksson. Three times in 1994 a fishing vessel M/V Solrun, was hired. In August 1994 the stations were completed on the Norwegian vessel R/V Johann Hjort.

Discrete surface layer, 1m, 5m and 10m, pCO_2 samples were collected into 500 ml volumetric flasks from water bottles on a Rosette and Sea Bird 911 CTD instruments. The samples were preserved with mercuric chloride and analysed ashore by equilibration at 4°C with a gas of known CO_2 concentration followed by gas chromatography with a flame ionization detector. The instrument was calibrated with N_2 reference gas and 3 standards, 197.85 ppm, 362.6 ppm and 811.08 ppm, calibrated against standards certified by NOAA-CMDL at Boulder, CO, USA. So were also calibrated the standards used for the underway measurements (Chipman et al., 1993). Quality assurance and sample storage experiments indicated an overall precision of the pCO_2 determinations better than ±2 µatm (Olafsson et al., 2010).

The underway pCO_2 determinations in 2006-2007 covered areas of the East Greenland Current in and northwards from the Denmark Strait, in addition to Atlantic and Arctic Waters. The 6 cruises (Table S4) covered all seasons and all three water masses but with variable areal extensions (Fig. 2 and S1). Seawater was pumped continuously from an intake at 5 m depth at 10 L min^{-1} into a shower-head equilibrator with a total volume of 30 L and a headspace of

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

15 L. Temperature at the inlet and salinity were measured with an SeaBird Model SBE-21 thermosalinograph (Sea-Bird Electronics, Seattle,WA, USA). Underway pCO_2 determinations were carried out with a system similar to the one described by Bates and coworkers (Bates et al., 1998). The mole fraction of CO_2 (V CO_2) in the headspace was determined with a Li-Cor infrared analyzer Model 6251 (Li-Cor Biosciences, Lincoln, NB, USA). The instrument was calibrated against four standards of CO_2 in air certified by NOAA-CMDL at Boulder, CO, USA. and a N_2 reference gas. The standards had CO_2 dry air mole fractions of 122.19, 253.76, 358.41 and 476.81 ppm. The pCO_2 sw determinations were corrected to in-situ seawater temperatures using the equation (Takahashi et al., 1993b):

$$pCO_2 \text{ sw(in situ)} = pCO_2 \text{ sw(eq)} \, e^{0.0423(T_{in\ situ} - T_{eq})} \quad \text{(eq.1)}$$

The sample processing and quality control measures have been described in detail (Olafsson et al., 2010).

Polar Water collection. Discrete samples for carbon chemistry studies were taken on stations (N=97) in the East Greenland Current when opportunites permitted on cruises in the period 1983 to 2012. The data provide pCO_2 for calculation of Delta pCO_2 in Fig. 4.

2.2 CO_2 air-sea flux calculations

In this study, the pCO_2 in seawater samples has been measured by gas-seawater equilibration methods (Olafsson et al., 2010). The bulk flux of the carbon dioxide across the air-sea interface is often estimated from its relationship with wind speed and sea-air partial pressure difference, ΔpCO_2. We determine the flux (F) from ΔpCO_2 and use Eq. 1 and Eq. 2 for estimating the bulk air-sea fluxes of CO_2 (Takahashi et al., 2009)

$$F = k \cdot \alpha \cdot \Delta pCO_2 \quad \text{(Eq 1)}$$

$$F = 0.251\ U^2\ (Sc/660)^{-0.5}\ \alpha\ (pCO_{2\,w} - pCO_{2\,a}) \quad \text{(Eq 2)}$$

There $k=0.251\ U^2\ (Sc/660)^{-0.5}$ is the gas transfer velocity or kinetic component of the expression (Wanninkhof, 2014), α is the solubility of CO_2 gas in sea water (Weiss, 1974) and $\Delta pCO_2 = (pCO_{2sw} - pCO_{2a})$, is the partial pressure difference or thermodynamic component of the expression (Takahashi et al., 2009). For the wind speed, U, we use the CCMP-2 reanalysis wind product (Wanninkhof, 2014;Atlas et al., 2011;Wanninkhof and Triñanes, 2017).

The atmospheric partial pressure values, $pCO_{2\,a}$, used in the ΔpCO_2 calculations are weekly averages from the GLOBALVIEW-CO2 database for the CO2-ICE location which is at Vestmannaeyjar islands, off south Iceland (GLOBALVIEW-CO2, 2013). Mauna Loa values

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

were used for periods where CO2-ICE data was missing (Tans and Keeling, 2019). The dry air V CO_2 mole fraction values were converted to µatm using pCO_2 (µatm)= V CO_2 ($P_a - P_w$) where Pa is the barometric pressure and P_w is the equilibrium water vapour pressure.

For the Irminger Sea seasonal study we use 30 day running means of the squared daily wind speed for the region 63.5°N to 64.5°N and 27°W to 32°W and for the Iceland Sea seasonal study a similar wind product for the region 66.5°N to 68.5°N and 12°W to 19°W. Fluxes were calculated for the periods between cruises from interpolated pCO_2 data and period mean 30 day squared wind running means data. There are thus 14 flux periods covering a year for the Irminger sea and 21 flux periods covering two years in the Iceland Sea (Tables S1 and S2 in the supplement). The annual fluxes were found by summation of the period fluxes (Table 1).

For the underway cruises 2006 to 2007 we used CCMP-2 daily wind fields at 1x1 degree for the region 62°N to 72°N and 5°W to 40°W. This region was further divided into 4 sub-regions by latitude 64.9°N and longitude 20°W. Daily 30 day running means of the squared wind speed from two locations in each sub-region were extracted and their means used for flux calculations when the vessel sailed in the area. Fluxes were calculated for all pCO_2 data from the 6 cruises, in total 42938 measurements.

The flux data from each of the 6 cruises were categorized into the three sea water types using the following criteria:

1) Atlantic Water S>35, Arctic Water S: 34.4-34.9, Polar Water <34.4.
2) Seasonal salinity and temperature variations were taken into account.
3) Waters with runoff influences from Iceland were excluded using salinity and ship position data.

Thus a total of 33352 measurements were used, or 78% of the flux data points. The CO_2 fluxes in the realm of each water mass were assessed for the duration of each cruise by numerical integration. Fluxes in the 5 periods between cruises were assessed by interpolation of temperature, salinity and pCO_2 for each water mass and by using period regional 30 day running means of squared wind speed data. The annual flux for each water mass was assessed by summation.

3 Results

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

Mean monthly reanalysis of winds reveal seasonal variations with strong winds in winter when they may be stronger over the Irminger Sea than the Iceland Sea as in 1993-1994 and in 1994-1995 (Fig. 3).

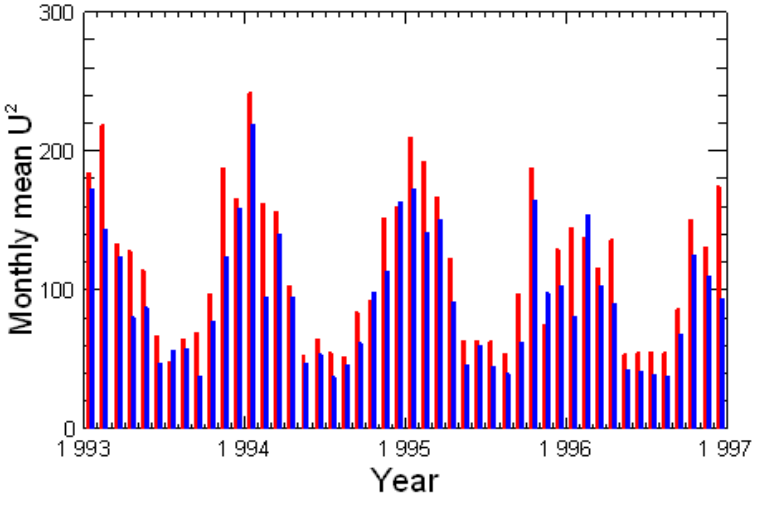


***Figure 3. Monthly means of squared CCMP-2 daily winds, U m/s**. Irminger Sea region (red) and the Iceland Sea region (blue).*

The cyclone tracks in the atmosphere, particularly of the Icelandic Low, influence the regional conditions which may change abruptly as in the 1994-1996 period or become anomalous as in 1995 when cold northeasterly winds persisted north of Iceland under positive NAO conditions (Ólafsson, 1999;Flatau et al., 2003). Both the Irminger Sea and the Iceland Sea seasonal studies reveal the stongest CO_2 undersaturation, with negative ΔpCO_2 of about 100 µatm in May at the time of the phytoplankton spring bloom (Fig.4). The undersaturation diminishes through the summer and autumn followed by a gradual return to winter conditions (Fig. 4b) (Takahashi et al., 1985;Takahashi et al., 1993a).

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

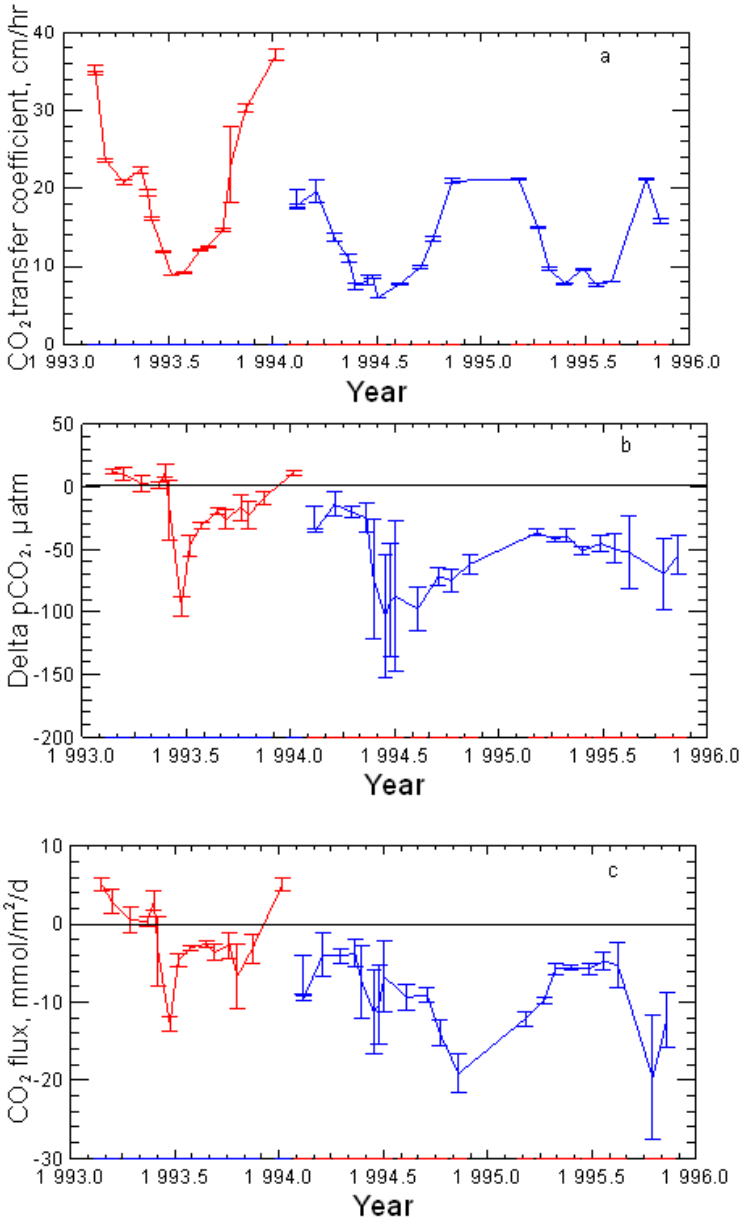


Figure 4. Seasonal variations in the Atlantic Water of the Irminger Sea (red) and in the Arctic Water of the Iceland Sea (blue). *The gas transfer velocity (a) reflects the seasonal wind strength and the error bars its variations during intervals between cruises. Delta pCO_2 (b) records the tendency for CO_2 to be transferred to the atmosphere (positive) or from the atmosphere to the ocean (negative). The CO_2 flux rate (c) reveals that the Arctic Water is a CO_2 sink in all seasons whereas the Atlantic Water is a source in winter and a weak sink at other times of the year. The error bars indicate ± 1 standard deviation from the mean and reflect the variations between the stations observed each cruise.*

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

The CO_2 influx in the spring is, however, relatively small as the wind gas transfer coefficient is then moderate (Fig.4a). In the autumn the winds strengthen with heat loss and vertical mixing while CO_2 undersaturation still persists. In mid winter, February-March, vertical mixing brings richer CO_2 water to the surface of the Irminger Sea leading to supersaturation (Ólafsson, 2003), the flux reverses and the region becomes a weak source for atmospheric CO_2 (Fig. 4c).

Table 1 Annual sea–air CO_2 fluxes (mol C m^{-2} y^{-1}) in the three water masses.

Water masses and evaluation methods	**CO_2 flux mol C m^{-2} y^{-1}**
Atlantic water, repeat stations 1993	-0.69±0.16
Atlantic water, Underway Measurements, 2006-2007	0.07 ± 0.15
Arctic water, repeat stations 1994	-3.97±0.48
Arctic water, repeat stations 1995	-3.60±0.31
Arctic water, Underway Measurements, 2006-2007	-2.84 ± 0.19
Polar water, Underway Measurements, 2006-2007	-4.44 ± 0.34

The integrated annual CO_2 flux shows that the Atlantic Water in the Irminger Sea was a weak sink, -0.69±0.16 mol C m^{-2} y^{-1}, in 1993 (Table 1). The more extensive underway area coverage of the Atlantic Water in 2006-2007, confirmed in essence the seasonal pattern and indicated that the Atlantic Water was a neutral sink, 0.07 ± 0.15 mol C m^{-2} y^{-1} for this year (Table 1).

The years of the Iceland Sea observations, 1994-1996, coincided with a large transition in the North Atlantic Ocillation (NAO) from a positive state 1994/1995 to a negative state in 1995/1996 and large scale shifts in ocean fronts (Flatau et al., 2003). Vertical density distribution in the Iceland Sea indicated an enhanced convective activity in 1995 (Våge et al., 2015). Cold northeasterly winds were persistent in the spring of 1995 resulting in record low temperature anomalies for the north Iceland shelf (Ólafsson, 1999). In 1995 the spring bloom associated undersaturaion, ΔpCO_2, was only half of that in 1994, possibly due to a weaker stratification (Fig. 4b). As in the Irminger Sea the spring bloom associated CO_2 influx is small. The largest CO_2 influx was in the fall and early winters of 1995 and 1996 as temperature dropped, winds gathered strength and vertical mixing was enhanced. This

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

compensated for the small spring bloom in 1995 and the annual bulk flues 1994 and 1995 are similar and high despite very different physical conditions (Table 1). The UWpCO_2 surveys had less temporal resolution but confirmed all year undersaturation of the Arctic Water. However, the integrated annual influx, -2.84 mol C m^{-2} y^{-1}, was significantly less than evaluated with repeat station data (Table 1, Fig.5). This may reflect the large underway area coverage compared with the repeated fixed stations.

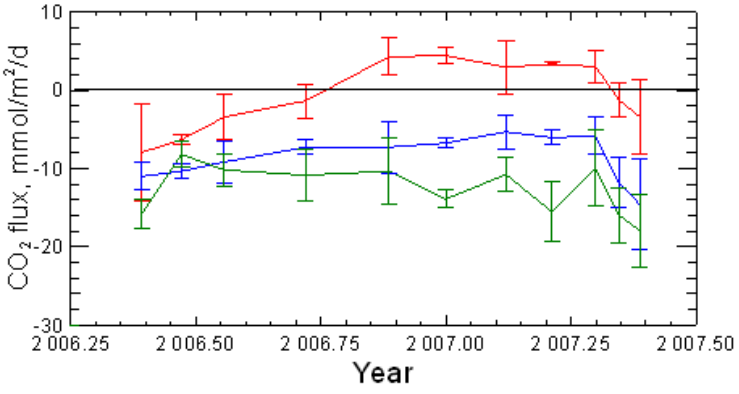


Figure 5. Seasonal air-sea CO_2 flux variations from UWpCO_2 observations*. Atlantic Water (red) is a weak sink in summer and neutral over the year, n=7068. Both Arctic Water (blue) n=16874, and Polar Water (green) n=9410, are strong sinks throughout the year. The error bars indicate ± 1 standard deviation from the mean.*

Ice cover in the East Greenland Current is variable and the ice edge at the seasonal minimum has moved northward and from the Denmark Strait with decreasing Arctic sea ice (Serreze and Meier, 2019). The Polar Water salinity ranges from 34.4 to less than 30 in summer. The lowest salinity water freezes leading to salinity around 34 in winter. We covered the Polar Water in all six UWpCO_2 surveys 2006-2007 (Supplement Fig. S1) and undersaturation characterised this water mass in all cruises. The integrated annual influx, -4.44 mol C m^{-2} y^{-1} (Table 1, Fig.5), shows the Polar Water to be the strongest CO_2 sink, 80 % above the estimated mean for the Atlantic north of 50°N, -2.5 mol C m^{-2} y^{-1} (Takahashi et al., 2009). We evaluate the long term pCO_2 characteristics of the three water mases from other data assembled over about 30 years. A composite picture of seasonal ΔpCO_2 variations in Polar Water in and north of the Denmark Strait (Fig.1) confirms all year undersaturation, deep from biological drawdown in summer, and in mid winter when salinity raises to ~ 34, the ΔpCO_2 levels at about -50 µatm (Fig. 6a). Long term winter ΔpCO_2 in the Irminger Sea and Iceland Sea (Figs. 1 and 6 b) show the Atlantic Water to be near saturation whereas the Arctic Water is undersaturated to about -35 µatm.

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

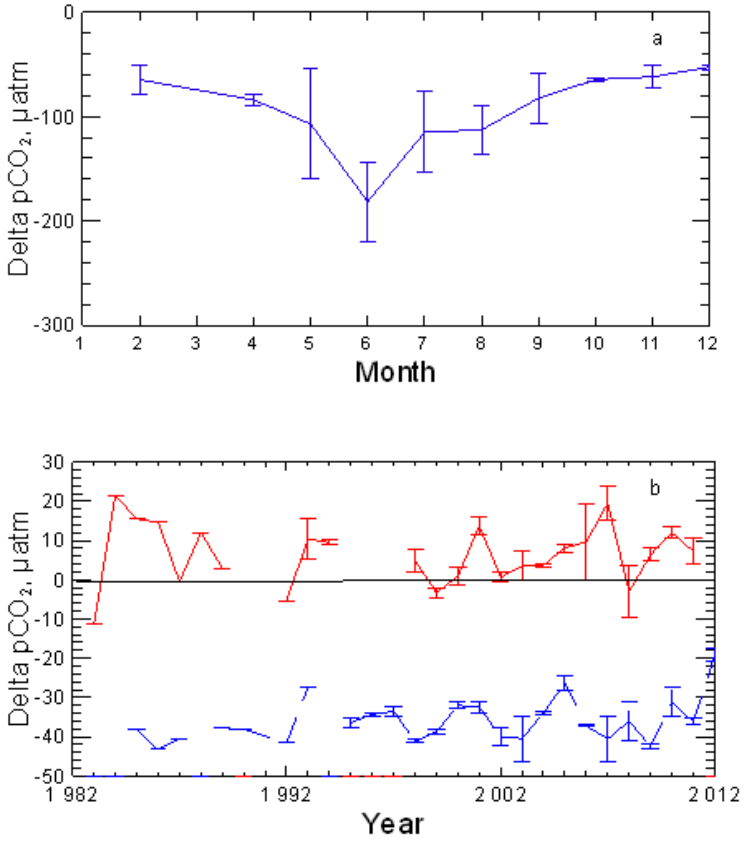


Figure 6. Water mass decadal surface water pCO_2 characteristics. *a) A composite picture of Delta pCO_2 from 97 stations with Polar Water pCO_2 observations (n=280) 1983 to 2012 shows undersaturation at all times of the year. The error bars indicate ± 1 standard deviation from the monthly means. b) Atlantic Water at the Irminger Sea time series station (red) is generally a weak CO_2 source in winter (24 winters, 52 samples), January-March, whereas winter (25 winters, 61 samples) CO_2 undersaturation persits in the Iceland Sea time series site (blue). The error bars indicate ± 1 standard deviation from the station means.*

4 Discussion

The carbonate chemistry of Polar Water differs from that of open ocean waters, e.g. Atlantic Water, in having an increasingly higher alkalinity/salinity and alkalinity/TCO2 ratios as the salinity decreases from about S=34.4. The excess alkalinity has been attributed to the high riverine input from continents to the Arctic (Anderson et al., 2004;Lee et al., 2006). The flow-weighted average alkalinity of 6 major Arctic rivers, discharging 2.245 x 10^3 km^3 yr^{-1}, is 1048 µmol kg^{-1} (Cooper et al., 2008). Linear alkalinity-salinity relationships observed in the Arctic Ocean and the Nordic Seas and their extrapolated intercepts to S=0, have indicated freshwater

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

sources with alkalinity 1412 µmol kg^{-1} (Anderson et al., 2004) and 1752 µmol kg^{-1} (Nondal et al., 2009). Climatological data from the West- Greenland, Iceland and Norwegian Seas show a high S=0 intercept of 1796 µmol kg^{-1} but a lower one for the High Arctic north of 80°N, 1341µmol kg^{-1} (Takahashi et al., 2014). The intercepts may be interpreted as the mean alkalinity of fresh waters added to the Arctic by rivers and melting ice and snow. However, the intercepts indicate considerable variability, they are higher than the average alkalinity of Arctic rivers and the intercepts are high in upstream regions of the East Greenland Current. The excess alkalinity would lower the pCO_2 in seawater (and increase the pH), and thus give it an increased capacity to take up CO_2 from the air (Fig. 7). The thermodynamic driving force for seawater CO_2 uptake, ($pCO_{2sw} - pCO_{2a}$), is enhanced.

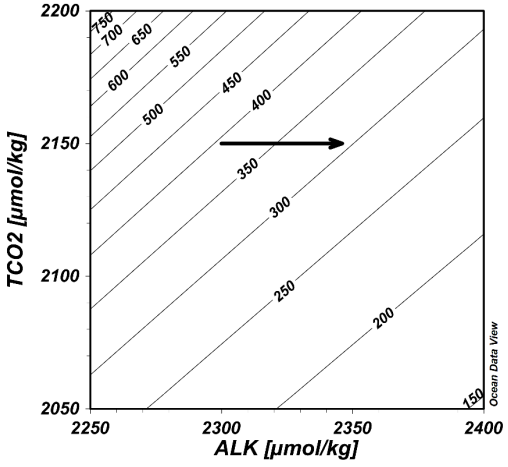


Figure 7. Thermodynamic relations of alkalinity, total inorganic carbon and the equilibrium pCO_2 in seawater (µatm)*. Calculated for S=35 and temperature 5°C, typical for Atlantic Water which reaches the Nordic Seas. The slope of the 350 µatm pCO_2 contour has a value of 0.85 and describes the relative extent of alkalinity and TCO2 additions (ΔTCO_2 /ΔAlk) to maintain unchanged pCO_2. The arrow illustrates that an 46 µmol kg^{-1} alkalinity excess decreases pCO_2 by 88 µatm, causes undersaturation. Figure drawn using the Ocean Data View program (Schlitzer, 2018).*

How large is the effect of Arctic alkalinity inputs on the CO_2 uptake by the Nordic Seas and the North Atlantic? We consider two ways to make this estimate. Firstly, from the river runoff into the Arctic which is estimated to be about 4.2 x 10^3 km^3 yr^{-1}, or 0.133 x 10^6 m^3 s^{-1}

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

(0.133 Sv). This is about 11% of the global freshwater input to the oceans (Carmack et al., 2016). Taking the average alkalinity 1048 µmol kg^{-1}, the amount of alkalinity added by rivers to the Arctic and transported to the North Atlantic via the Canadian Arctic Arcipelago and via the Fram Strait and further south with theLabrador and East Greenland Currents, would be 4.4 x 10^{12} mol yr^{-1} (Supplement). The pCO_2 of seawater would be reduced by an addition of this much excess alkalinity. Let us suppose that the pCO_2 in seawater was restored to the original value by absorbing CO_2 from the atmosphere. The carbonate equilibrium relations in seawater give that pCO_2 is unchanged if $\Delta TCO_2 / \Delta Alk = 0.85$ (Fig. 6). This ratio is nearly constant in the temperature and salinity range of the subarctic North Atlantic surface waters. Therefore, the Arctic river alkalinity would contribute to an uptake of CO_2 from the atmosphere of 3.73 x 10^{12} mol CO_2 yr^{-1} or 0.045 Pg-C yr^{-1} (Supplement).

Second estimate. The volume transport of Polar Water, density <1027.8 kg m^{-3}, by the EGC has recently been estimated as 3.9 Sv (Våge et al., 2013). Taking S=33.0 for the mean Polar Water salinity and using Equations 6 and 7 in Nondal et al (2009), the mean Polar Water alkalinity is 2256 µmol kg^{-1} which is 46 µmol kg^{-1} more alkalinity than the Atlantic Water calculated at the same salinity (Nondal et al., 2009). This much excess alkalinity would lower the pCO_2 of Atlantic Water by 88 µatm and increase the pH by 0.10 (Fig. 7). Thus, the excess alkalinity advected to the North Atlantic by the EGC is 5.7 x 10^{12} mol yr^{-1}. Using again 0.85 for the $\Delta TCO_2/\Delta Alk$ changes at a constant pCO_2, we obtain that the contribution of the excess EGC alkalinity to the uptake of CO_2 from the atmosphere would be 34.8 x 10^{12} mol CO_2 yr^{-1}, or 0.058 Pg-C yr^{-1}. The two estimates correspond to 17% and 21 %, respectively, of the net CO_2 uptake of 0.27 Pg-C yr^{-1} for the subarctic oceans north of 50 °N (Takahashi et al., 2009). We did not include in the second estimate any alkalinity contribution with the considerable Canadian Arctic Arcipelago Polar Water freshwater transport (Haine et al., 2015). The effect of excess alkalinity on the oceanic CO_2 uptake flux may therefore be substantially greater than our calculated estimates. We note that the winter undersaturation levels, of -50 µatm and -35 µatm observed in the Polar and Arctic Waters, respectively (Fig. 5), translate to excess alkalinity of 19 µmol kg^{-1} and 21 µmol kg^{-1} for further CO_2 influx downstream.

The difference between the average measured Arctic river alkalinity and the regression based estimates of alkalinity sources suggests that other origins and processes than the rivers contribute to the Polar Water alkalinity exported with currents from the Arctic to the Atlantic Ocean. Photic layer primary production in the absence of calcification may lower the

https://doi.org/10.5194/bg-2020-313
Preprint. Discussion started: 27 August 2020

382 TCO2/Alk ratio and seawater pCO_2 in marginal seas (Bates, 2006), while acidification is
383 increasing in other regions (Anderson et al., 2017;Qi et al., 2017). Furthermore, the sea-ice
384 seasonal formation and melting may affect the TCO2/Alk ratio (Grimm et al., 2016;Rysgaard
385 et al., 2007). The Arctic is complex and complex climate warming related changes are
386 observed in the western Arctic Ocean (Ouyang et al., 2020) and expected in marine
387 freshwater systems of the warming Arctic (Carmack et al., 2016). River water alkalinity
388 increases with an addition of cations derived from the chemical weathering of silicate and
389 carbonate rocks (Berner and Berner, 1987). Accordingly, an increase in Arctic weathering
390 rates, in response to warmer climate and increasing atmospheric CO_2, could increase the river
391 water alkalinity transported into the oceans. Such an increase would lower the pCO_2 in
392 seawater and enhance the oceanic uptake of atmospheric CO_2, providing a negative feedback
393 mechanism to the climatic warming resulting from increased atmospheric CO_2.
394
395 **5 Conclusions**
396 The North Atlantic region we describe has ocean waters advected from southern temperate
397 latitudes and others from the north with Arctic signatures. The Gulf Stream derived Atlantic
398 Water which reaches the northern Irminger Sea and the Nordic Seas, has had a long contact
399 time with the atmosphere to loose heat and reach near CO_2 saturation (Takahashi et al.,
400 2002;Olsen et al., 2006). The Atlantic Water seasonal pCO_2 variations we observe (Fig. 2c),
401 are primarily driven by regional thermal and biological cycles (Takahashi et al., 2002). The
402 southward flowing Arctic and Polar Waters are on the contrary strong and persistent all year
403 CO_2 sinks despite various regional changes upstream in the Arctic Ocean. Downstream from
404 the Polar Water and Arctic Water outflows is the subpolar North Atlantic with high water
405 column inventories of anthropogenic carbon (Khatiwala et al., 2013;Gruber et al., 2019). The
406 high anthropogenic CO_2 regions have been attributed, as mentioned above, to the combined
407 effects of the solubility and biology gas exchange pumps on the CO_2 fluxes (Takahashi et al.,
408 2002). We point here to the Polar Water and Arctic Water CO_2 influx and excess alkalinity as
409 an additional unrecognized source contributing to the North Atlantic CO_2 sink. Climate
410 induced changes in Arctic biogeochemistry and alkalinity budget are likely to affect the
411 sensitivity and future strength of the North Atlantic CO_2 sink downstream.
412
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Preprint. Discussion started: 27 August 2020

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