Resolving heterogeneity in CO2 uptake potential in the Greenland coastal ocean

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Abstract

The oceans play a pivotal role in mitigating climate change by sequestering approximately 25% of annually emitted anthropogenic carbon dioxide (CO2). High-latitude oceans, especially the Arctic continental shelves, emerge as crucial CO2 sinks due to their cold, low saline, and highly productive ecosystems. However, these heterogeneous regions remain inadequately understood, hindering accurate assessments of their carbon dynamics. This study investigates variation in pCO2 levels during peak ice sheet melt, in the Greenland coastal ocean and estimates rates of air-sea exchange across 6° of latitude. The East and West coast of Greenland displayed distinct regions with unique controlling factors. Though, both coasts represent CO2 sinks in summer. Geographical variation in pCO2 and air-sea exchange was linked intricately to freshwater export from the Greenland ice sheet and levels of primary production in these ecosystems. CO2 uptake ranged from 0.17 to -38 mmol m-2 day-1. However, we found that flux estimation faces substantial uncertainties (up to 770%) due to wind product averaging and gas exchange formula selection. Despite these considerations, we report a first order estimate that Greenland coastal ocean takes up -9.5 ± 9.0 Tg C year-1, corresponding to nearly 4% of global coastal CO2 uptake. Obtaining a reliable assessment of air-sea CO2 exchange necessitates data collection across seasons, and, even more so, refinement of the gas transfer velocity estimations in the Arctic coastal zone.

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Key Points:

- The Greenland coastal ocean takes up large quantities of carbon dioxide yet displays considerable spatial heterogeneity.
- Biology and freshwater runoff control pCO_2 levels to varying degrees in different regions.
- Estimation of the true flux magnitude is beset by large uncertainties, particularly in polar coastal regions.

Abstract

The oceans play a pivotal role in mitigating climate change by sequestering approximately 25% of annually emitted anthropogenic carbon dioxide (CO_2). High-latitude oceans, especially the Arctic continental shelves, emerge as crucial CO_2 sinks due to their cold, low saline, and highly productive ecosystems. However, these heterogeneous regions remain inadequately understood, hindering accurate assessments of their carbon dynamics. This study investigates variation in pCO_2 levels during peak ice sheet melt, in the Greenland coastal ocean and estimates rates of air-sea exchange across 6° of latitude. The East and West coast of Greenland displayed distinct regions with unique controlling factors. Though, both coasts represent CO_2 sinks in summer. Geographical variation in pCO_2 and air-sea exchange was linked intricately to freshwater export from the Greenland ice sheet and levels of primary production in these ecosystems. CO_2 uptake ranged from 0.17 to -38 mmol m^{-2} day^{-1}. However, we found that flux estimation faces substantial uncertainties (up to 770%) due to wind product averaging and gas exchange formula selection. Despite these considerations, we report a first order estimate that Greenland coastal ocean takes up -9.5 ± 9.0 Tg C year^{-1}, corresponding to nearly 4% of global coastal CO_2 uptake. Obtaining a reliable assessment of air-sea CO_2 exchange necessitates data collection across seasons, and, even more so, refinement of the gas transfer velocity estimations in the Arctic coastal zone.

Plain Language Summary

The oceans help to limit climate change by absorbing large amounts (1/4) of carbon dioxide (CO_2) that humans emit to the atmosphere. The majority of this CO_2 enters the oceans near the poles due to the special conditions that occur in these regions: namely that there is cold, less salty water, with high quantities of photosynthetic algae in the surface ocean. However, scientists do not completely understand the regional variability of oceanic CO_2 uptake, which limits our ability to accurately predict future climate scenarios. This study measured the partial pressure of CO_2 in the ocean along East and West Greenland to calculate rates of air-sea transfer. CO_2 uptake ranged from 0.17 to -38 mmol m^{-2} day^{-1} and the controlling factors behind this flux of carbon were wind speed, amount of glacial runoff, and the balance between biologic producers/consumers. The Greenland coastal ocean may represent nearly 4% of global coastal ocean CO_2 uptake, indicating the need to better understand carbon sequestration variability in this region.

Introduction

The world's oceans serve as a critical buffer against the escalating impacts of anthropogenic carbon dioxide (CO_2) emissions, absorbing approximately 25% of these emissions annually (Ciais et al., 2013; Sabine, 2004; Takahashi et al., 2009). Oceans at high latitudes uptake more of this CO_2 due to a combination of low temperatures and high productivity (Bates and Mathis, 2009; Takahashi et al., 2009). Although 25% of continental shelves (depth < 200 m) are located in the Arctic, we still have a limited understanding of the carbon dynamics in these high-latitude coastal systems due to the lack of field observations compared to low-latitude coastal environments (Bates and Mathis, 2009).

The driving force behind CO_2 exchange across the sea surface is the difference in partial pressure of CO_2 (ΔpCO_2) between seawater and the overlying air. However, the fluctuation of oceanic pCO_2 is more spatially and temporally variable compared to atmospheric pCO_2, making oceanic pCO_2 the thermodynamic driver of air-sea carbon uptake. Arctic coastal waters, are generally considered as a sink for CO_2 due to their undersaturated pCO_2 conditions throughout the year (Ahmed et al., 2020; Dai et al., 2022; Laruelle et al., 2014, 2018). Some distinct regions, such as the Siberian shelf seas, exhibit CO_2 effluxes due to decomposition of abundant terrestrial organic matter (Anderson et al., 2009). However, other coastal regions, where net-autotrophy dominates, remain large atmospheric carbon sinks (e.g. Meire et al., 2015). Still, the magnitude and relative driving processes in shelf seas remain poorly quantified. The coastal ocean is unique, in that it is influences both by both by the local environment on land, as well as larger oceanic and atmospheric circulation. These coastal systems play a pivotal role in ocean-atmosphere dynamics due to their large exchanges of matter and energy (e.g., heat, CO_2, water) at their interfaces, yet have often been overlooked in air-sea exchange surveys due to their spatial and temporal complexity (Chen and Borges, 2009; Miller et al., 2019).

Understanding the intricate dynamics of CO_2 exchange in Greenland's coastal ocean is imperative given the heterogeneous nature of its coastline. A few previous studies have estimated air-sea exchange in Greenland fjords (Meire et al., 2015; Rysgaard et al., 2012; Sejr et al., 2011). However, these studies estimate fluxes from only two fjord systems, and have previously been used to upscale for the entire Greenland coastal ocean (eg. Laruelle et al., 2013). Knowing that the coasts of Greenland are a heterogeneous landscape, it is crucial we gain a better understanding of the spatial variation in pCO_2 levels and carbon fluxes in this region to obtain reliable assessments of the CO_2 uptake by the Greenlandic coastal area. Additionally, a large-scale perspective on the drivers of surface pCO_2 levels is necessary to gauge if the combined effects of climate change will result in a positive or negative feedback on coastal carbon uptake. We expect that a glacial meltwater and biological activity will drive heterogeneity in pCO_2 levels because carbon dynamics are controlled by a combination of abiotic and biotic factors (Henson et al. 2023). However, the relative contribution of these drivers as well as the magnitude of this heterogeneity have not been examined for CO_2 fluxes in this region. This study aims to fill in this gap in knowledge by exploring CO_2 fluxes in Greenlandic fjords and shelf waters spanning 6° of latitude. Given the region's sensitivity to climate change and its substantial CO_2 uptake relative to other marine areas, a realistic assessment of exchange rates is imperative. By delving into the drivers influencing air-sea CO_2 exchange in these coastal waters, we strive to contribute valuable insights toward a more comprehensive understanding of the role played by high-latitude coastal systems in the global carbon cycle.

Methods

Sampling

Two research cruises were conducted along the coasts of Greenland where oceanic partial pressure of CO_2 (pCO_2) and other environmental parameters were measured. The first cruise, along the western coast of Greenland (69–75°N) was conducted during 12–30 August 2016. Meanwhile, the second cruise along the eastern coast of Greenland (68–74°N) was conducted between July 30 and August 24, 2018 (Fig. 1). Additional stations in Young Sound, East Greenland were sampled in 2018 as part of the Greenland Ecosystem Monitoring program (Christensen et al., 2017; Sejr et al., 2011). Seawater pCO_2 profiles were measured using a Contros Hydro-C CO_2 sensor (Meire et al., 2023). The Hydro-C sensor was equilibrated for 5-10 min until a stable reading was acquired at 8 depths between 1–60m (1m, 5m, 10, 20m, 30m, 40m ,50m, 60m). The relative standard deviation of the Contros pCO_2 measurement has been estimated to be 1% (Fietzek et al., 2014). While some in the research community argue that membrane-based sensors are less precise, we posit that measurement from underway systems in highly stratified waters poses as much or more error when calculating air-sea exchange (Macovei et al., 2021; Miller et al., 2019). Therefore, membrane-based sensors at 1m allowed a robust examination of surface layer pCO_2 values in coastal Greenland waters. At each station, depth profiles of temperature and salinity, were obtained using CTD instruments (SBE19 + V2 and SBE25 on West and East coast cruises respectively), equipped with additional sensors for chlorophyll-a fluorescence, photosynthetically available radiation (PAR), and dissolved oxygen (Bendtsen et al., 2017; Carlson et al., 2020; Holding et al., 2021). Dissolved oxygen sensors were calibrated using Winkler titrations using water samples collected in Niskin bottles. During the western cruise, 56 CTD casts were conducted with 53 stations measured for pCO_2. Meanwhile the eastern cruise conducted 89 CTD casts, with pCO_2 profiles measured at 63 stations. Through visual examination of satellite imagery, stations in the very inner fjord arms, were categorized based on the prevalent glacial output (marine- or land-terminating, "MTG" and "LTG" respectively). When a station was located in the larger, more outer fjord system with influence from many different sources of freshwater is was categorized as just "fjord," and when a station was located outside the fjord mouth, they were assigned as "shelf" stations (Supplemental Fig. 1).

Calculation of air-sea exchange

The net air-sea exchange of CO_2 was calculated using the diffusive boundary model (Sejr et al., 2011):

$$(1)\quad F = k_{660}s(\Delta pCO_2)$$

where F is the flux of CO_2, k_{660} is the gas transfer velocity normalized to 20˚C and a salinity of 35 PSU, s is the solubility of CO_2 in seawater, and ΔpCO_2 is the difference between atmospheric and surface water pCO_2 values (at 1m depth). Atmospheric pCO_2 levels were obtained from the SeaFlux data product (Fay et al., 2021; Gregor, 2023) in August of 2016 and 2018 at each measurement location. The solubility of CO_2, *s*, was calculated according to Weiss (1974) using surface water temperature and salinity from CTD casts. Gas transfer velocities (k_{660}) were normalized according to: $k_{660} = k(660/Sc)^{-0.5}$ where the Schmidt number (Sc), was calculated following Wanninkhof (1992). The gas transfer velocity, *k*, is a product of turbulence and is often estimated based on empirically derived relationships with wind speed in oceanic environments. A variety of formulas have been proposed based on different datasets from varying regions. We chose the formulation by Nightingale et al. (2000) as it was derived for coastal environments and has been used in previous assessments of CO_2 fluxes around the Greenland coast (Meire et al., 2015; Rysgaard et al., 2012; Sejr et al., 2011)

$$(2)\quad k = 0.333U_{10} + 0.222U_{10}^{2},$$

where U_{10} is wind speeds (m s^{-1}) at 10 m above sea surface. Wind speeds were obtained from the Copernicus Arctic Regional Reanalysis (Copernicus Climate Change Service, 2021) and averaged over the months of August 2016 and 2018.

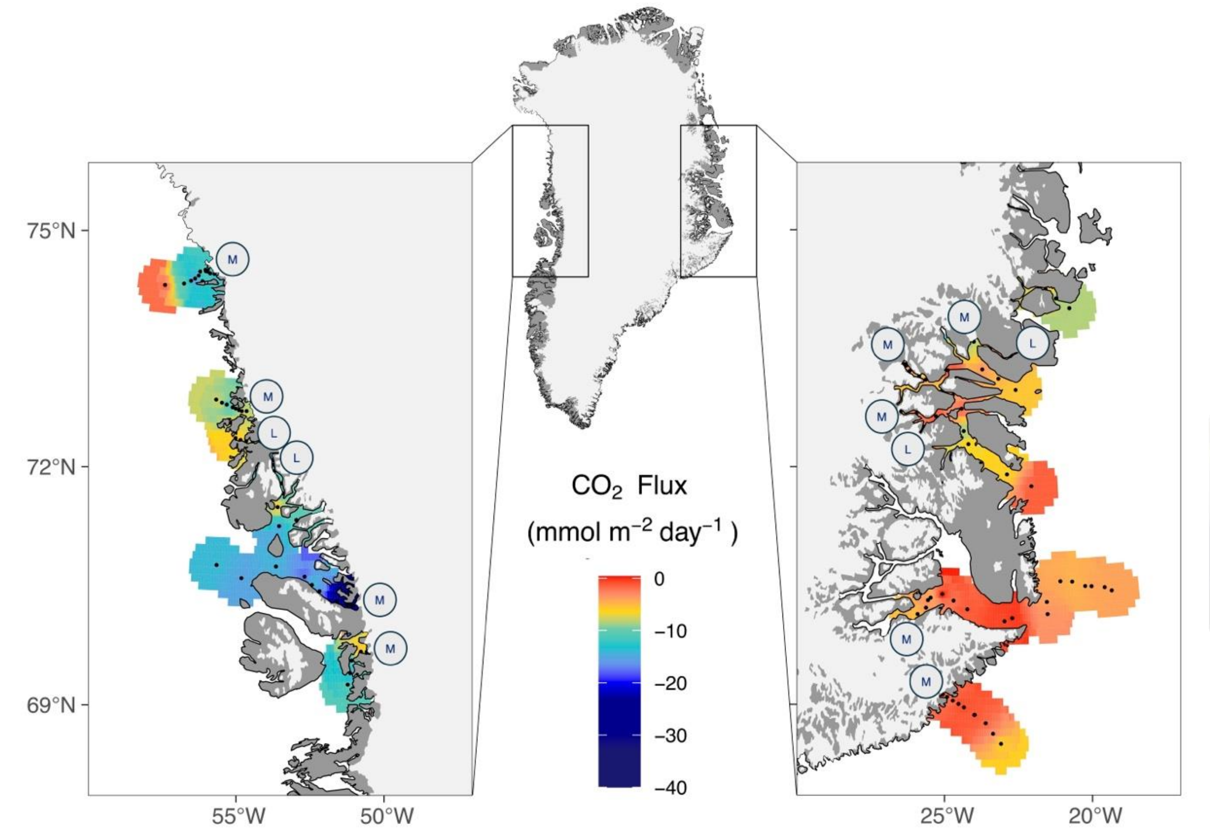

Figure 1: Map of air-sea exchange of CO_2 in Greenland coastal waters. Station locations are pictured as black points. Data was interpolated using an inverse distance weighted matrix limited to a maximum distance of 39 km. Marine- and Land-terminating glaciers are labeled with M and L respectively.

Statistical Modelling

In order to examine the impact of environmental parameters on surface layer pCO_2 levels, statistical models were applied to our data. First, general linear models were fit to identify relationships between predictors and pCO_2 in the upper 20 m. Model parameters were chosen based on the reduction of Akaike's information criterion (AIC) and verified using likelihood ratio tests. Multicollinearity was evaluated and limited using calculated variance inflation factors.

Spatial autocorrelation is often a feature of large-scale biodiversity and ecological data (Kreft and Jetz, 2007), making it necessary to check for a spatial structure of our data. Plotting residuals and conducting a Moran's I test confirmed there is spatial structure to our data (Moran I = 0.36, p-value < 5e-5). Spatial autocorrelation can inflate type 1 errors and potentially influence parameter estimation (Kreft and Jetz, 2007), making it necessary to correct for spatial autocorrelation. Generalized least squares models (GLS) with spatial structures as well as spatial autoregressive models were fit to model pCO_2. Final spatial linear model (SLM) selection was based on the reduction of spatial autocorrelation in the residuals and the minimization of AIC values. A spatial GLS model with a rational quadratics spatial correlation structure performed the best for modeling response data. A likelihood ratio test confirmed that adding a rational quadratic spatial correlation structure was significantly better than the base model (p-value < 5e-4). In order to construct this model, a spatial weights matrix was fit using k = 1 nearest neighbors. The correlogram of the fitted model illustrated that the problem with spatial autocorrelation had been successfully accounted for.

Results

Heterogeneity and drivers of pCO_2

Comparison between the east and west coasts of Greenland revealed differences extending beyond surface waters. Depth profiles of pCO_2 depicted distinct signals in both regions. Western Greenland exhibited more variable pCO_2 values with depth, displaying high undersaturation (94-364 µatm) in surface waters and CO_2 accumulation (291-522 µatm) already at a depth of 50 meters (Fig. 2, Supplemental Fig. 2). Conversely, the eastern coast displayed more uniform pCO_2 levels across the mixed layer, with values slightly below atmospheric levels (221-399 µatm at 1 m and 330-414 at 50 m; Fig. 2).

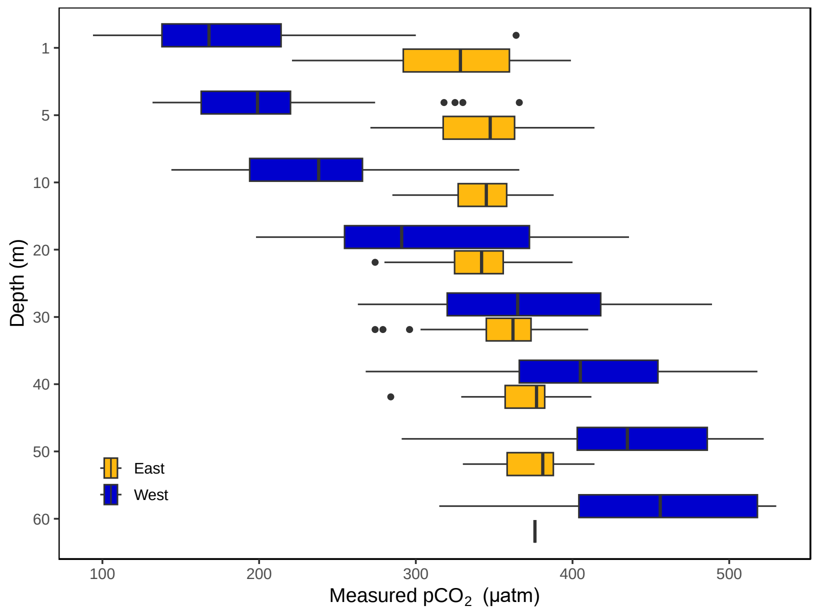

Figure 2. Boxplots of pCO_2 profiles on the East and West coast of Greenland.

The concentrations of pCO_2 in the water column are influenced by a combination of biological and physical/chemical factors. The main drivers affecting pCO_2 are depicted in Figure 3. Apparent Oxygen Utilization (AOU), also known as the deviation from O_2 saturation, serves as a valuable indicator of biological activity (e.g. Sejr et al., 2014). Positive AOU values imply bacterial O_2 consumption, while negative values signify photosynthetic O_2 production. Along the western coast of Greenland, AOU showed a strong correlation with measured pCO_2. Primary producers' biological incorporation of carbon decreased pCO_2 levels in the surface layer (Fig. 3a). Conversely, deeper waters exhibited net respiration, where O_2 consumption and CO_2 production took place concurrently. Along the Eastern coast of Greenland, lower levels of net production and net respiration were observed (Fig. 3b), leading to a decoupling between AOU and measured pCO_2. This region, known for its lower productivity (Henson et al., 2023), exhibited less variation in pCO_2 levels.

Figures 3c and 3d highlight the primary physical and chemical drivers of pCO_2 in this region: freshwater influx. Glacial meltwater input has been documented to reduce seawater pCO_2 through both thermodynamic effects and dilution (Meire et al., 2015). The relationship between freshwater input and pCO_2 is apparent in both eastern and western Greenland, where the lowest pCO_2 levels correspond with lower salinities. However, a stronger relationship was observed, in eastern Greenland where low levels of biological production, and fewer high salinity values make this relationship more apparent.

These relationships were confirmed using statistical models. This study applied a generalized least squares (GLS) model incorporating a rational quadratic spatial correlation structure to examine the dynamics of pCO_2 in relation to environmental predictors. Fit model parameters are shown in Table 1. Two interactions helped to improve model fit: the interaction between coast & AOU as well as the interaction between coast & temperature, illustrating the different effects of biology in each coast. Both biological and physical factors emerged as robust predictors for surface layer pCO_2 levels. The spatial model illustrates that in western Greenland, fluctuations in pCO_2 were primarily driven by AOU (biological processes). Conversely, in eastern Greenland, salinity and temperature exerted the most substantial influence on pCO_2 levels. This spatial linear model duly accounted for the spatial autocorrelation of measured pCO_2 values. The estimated range (distance at which spatial correlation becomes negligible) is approximately 1.15 longitudinal degrees, and the nugget parameter (unaccounted variability) was 0.43.

Table 1. Fitted GLS model with rational quadratic spatial structure.

	Estimate	T statistic	p-value
Intercept	108.3	4.214	<0.0001
AOU	0.016	0.230	0.8180
Salinity	6.718	10.56	<0.0001
Temperature	12.66	8.719	<0.0001
Coast (west)	-53.96	-2.318	<0.0001
AOU:Coast (west)	0.770	7.068	<0.0001
Temperature:Coast (west)	-11.72	-6.140	<0.0001

Model performance underwent rigorous assessment via cross-validation, utilizing subsets of the dataset to train and validate the model (Table 2). Our spatial linear model (SLM) accurately predicted 83% of the variance in pCO_2 within the test dataset, indicating that a relatively simple model can accurately estimate coastal pCO_2 levels. This robust predictive capacity was further supported by the evaluation metrics: the root mean standard error (RMSE) and mean absolute error (MAE). These metrics indicated an average and absolute difference of 36.2 and 27.8 µatm, respectively, between the predicted and actual pCO_2 values. Therefore, knowledge of the spatial heterogeneity in temperature, salinity, and AOU (or magnitude of auto/heterotrophy) could be used to estimate or explain variability in surface water pCO_2 around coastal Greenland.

Table 2. GLS model cross validation

R-squared	RMSE	MAE
0.836	36.2 µatm	27.8 µatm

Spatial variability in pCO_2 could also be explained by circulation-driven glacial effects. The entry of glacial meltwater into fjord waters occurs at varying depths, contingent upon whether the fjord contains an outlet glacier that is marine- or land-terminating (MTG and LTG, respectively). This inflow dramatically influences the circulation within fjords (Mortensen et al., 2014). Surface runoff from land-terminating glaciers resulted in a warmer, fresher surface layer (Henson et al., 2023). In these turbid waters on the western coast of Greenland, oxygen production, indicative of primary production, was limited compared to near marine terminating glaciers (p-value < 5e-5) where subglacial discharge drives upwelling of nutrient-rich bottom water. Meanwhile, AOU did not vary spatially on the east coast (Fig. 4a). Similar to AOU, pCO_2 values in western Greenland were elevated near LTGs compared to near MTGs, with even higher values in shelf waters (Fig. 4b). Eastern Greenland did not demonstrate a difference in pCO_2 values between stations near LTGs and MTGs (p-value = 0.997). However, the stations close to glacial output exhibited lower pCO_2 values than mid-fjord or shelf waters (Fig. 4b).

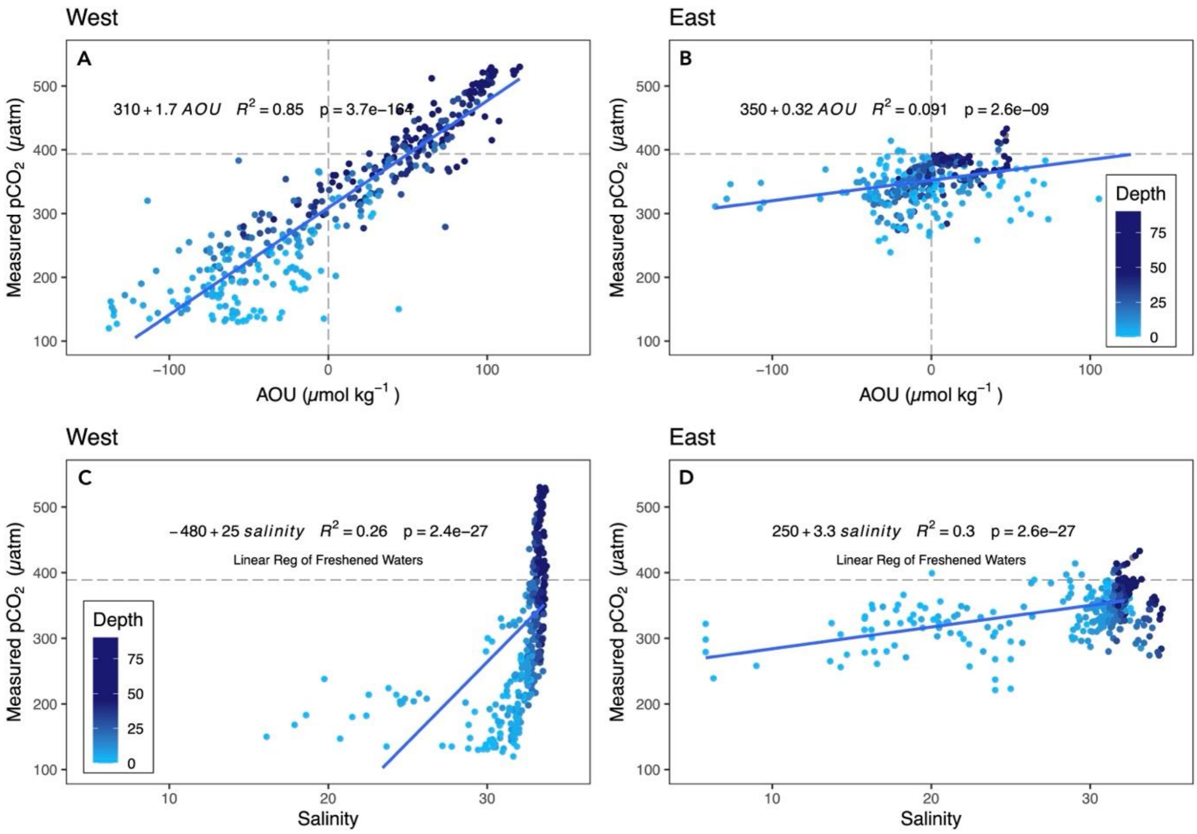

Figure 3. Relationships between apparent oxygen utilization (AOU) and pCO_2 (a, b) and salinity and pCO_2 (c, d) for East and West coasts. Linear regression fits use all data in a & b while c & d fit regression for all data below salinity of coastal seawater endmembers (Henson et al. 2023). Atmospheric pCO_2 concentrations and the equilibrium between net auto- and heterotrophy are depicted with horizontal and vertical gray dashed lines respectively.

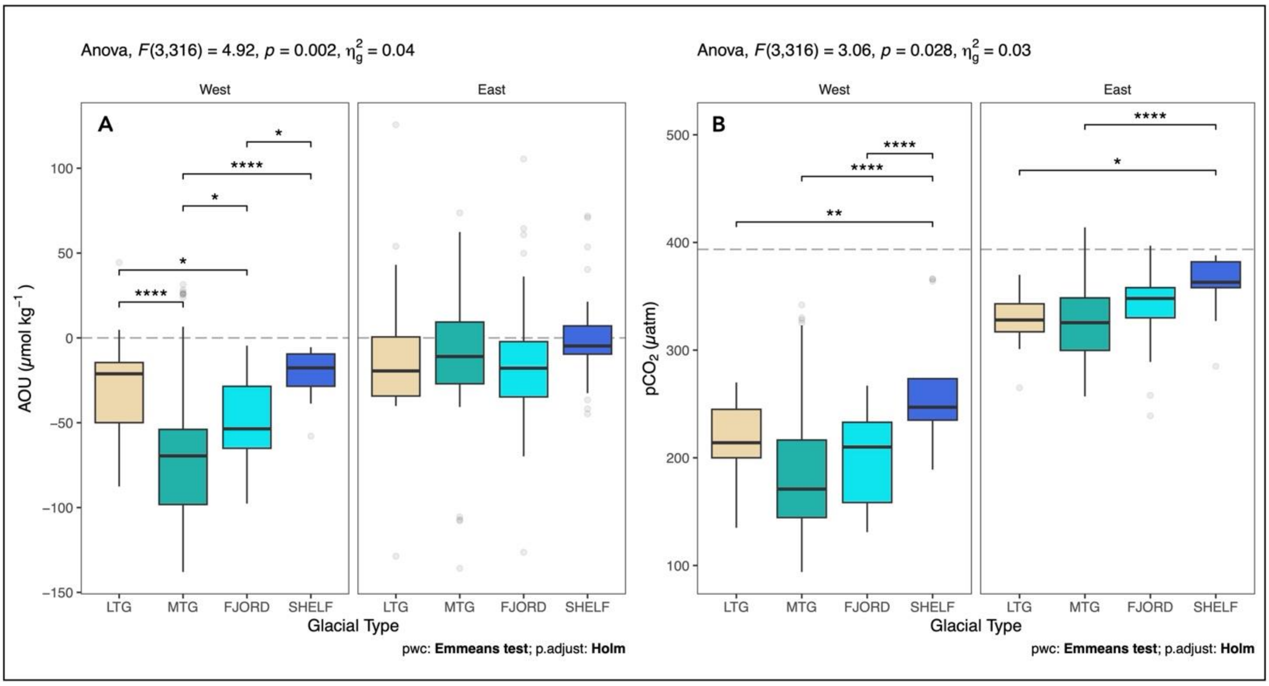

Figure 4. Boxplots and two-way ANOVA analysis of AOU with fjord location (a) and pCO_2 (b) with fjord location for surface layer samples (above 20 m depth). Significant interactions between coast and fjord location on AOU and on pCO_2 are described above each plot. All pairwise comparisons were analyzed between the different fjord location groups organized by coast. Statistical differences are depicted as brackets above boxplots with significance codes. Non-significant pairwise comparisons are not pictured. Equilibrium between net auto- and heterotrophy (a) as well as atmospheric pCO2 concentrations (b) are depicted as gray dashed lines.

CO_2 Fluxes

The air-sea exchange of CO_2 is a product of wind speed driven turbulence and ΔpCO_2. Understanding the interplay between these factors is crucial in understanding the dynamics of CO_2 fluxes in marine environments. In our investigation, we delve into the impact of wind speed on this exchange, examining fluxes calculated from both monthly and three-hour wind speed averages. The monthly wind speeds in August ranged between 1.7 to 6.5 m s^{-1} across our stations within Greenland fjords while three-hour averages ranged from 0-16.6 m s^{-1}. Notably, we found a strong correlation (R^2 = 0.77; P = 1.1e-36) between these two flux calculations. However, the air-sea CO_2 flux derived from three-hour wind averages exceeded the estimates from monthly wind speeds by an average of 10%. Disparities in these calculations are attributed to the non-linear relationship between gas transfer velocity and wind speed (Eq. 2), leading to significant exchanges during periods of high wind speed events. As a result, if 1hr or 10 min average wind speeds were available, one could expect an even larger difference to monthly averages.

In August, both the East and West coasts of Greenland showed pCO_2 values in surface water below atmospheric levels, indicating this region acts as a CO_2 sink. However, considerable geographic variability existed in ΔpCO_2 values, leading to varied flux magnitudes. Fluxes in West Greenland ranged from -37.52 to -1.80 mmol m^{-2} day^{-1} while those in East Greenland ranged from -13.24 to 0.17

mmol m^{-2} day^{-1}, highlighting differences in sink capacity and controlling factors between regions. Despite regional variations, certain trends emerged. The highest CO_2 uptake was observed near the Greenland ice sheet, whereas lower uptake, driven by smaller ΔpCO_2, was recorded in shelf waters (Fig. 1). These glacial fjords therefore shape a gradient between sources of freshening and the open ocean, displaying a transition from more negative to less negative fluxes.

Discussion

Greenland coastal pCO_2 is highly heterogeneous

Our observation of low surface water pCO_2 in coastal Greenland waters affirm the previous conclusions of Sejr et al. (2011, 2014), Meire et al. (2015, 2023), and Rysgaard et al. (2012) that these waters are large CO_2 sinks. We measured ΔpCO2 values ranging from -295 to 5 µatm with an average of -127 µatm. Only a single measurement was above atmospheric levels and was located in Kangertittivaq. These measurements, while displaying wide variation, represent similar values to summer measurements in other regions of coastal Greenland. Sejr et al. (2011) observed averages between -77 to -134 µatm in Young Sound, Rysgaard et al. (2012) observed ΔpCO2 values between -20 and -330 µatm in offshore areas of Southern Greenland and Meire et al. (2015) measured between -115 and -323 µatm within the Nuup Kangerlua fjord system. These seem analogous to the variability and levels of ΔpCO_2 that we observed on the east and west coast. Additionally, when comparing to syntheses of global shelf seas, our measured ranges of ΔpCO_2 fall within low-end of measurements at the same latitude (Dai et al., 2022).

Although pCO_2 values have been reported before, this is the first study to examine CO_2 uptake potential in more than one fjord system. We measured pCO_2 levels across 6° of latitude in both East and West Greenland and discovered quite different ecosystems comparing the two coasts. The observed spatial variability of pCO_2 emerges as a consequence of multifaceted oceanographic factors (Meire et al., 2015; Sejr et al., 2011). Therefore, understanding spatial flux variability requires an understanding of the environmental drivers of marine carbon dynamics.

Autotrophy/Heterotrophy balance dictates pCO_2 in West Greenland

Both biological and physical parameters were found to be significantly related to surface water pCO_2 within SLM models (Table 1). As a result, oxygen saturation in combination with salinity and temperature can be used to predict surface layer pCO_2 levels in coastal Greenland waters. This is because biological processes dictate levels of dissolved inorganic carbon (DIC) within the water column (Henson et al., 2023). The uptake of DIC by primary production reduces its concentration in seawater, while bacterial decomposition acts as a source of DIC. This direct consumption and production of DIC helps to explain the strong spatial link between pCO_2 levels and AOU in western Greenland fjords (Fig. 3a). Similarly, limited biological productivity in eastern Greenland resulted in less variability in pCO_2 profiles with depth (Fig.2). Western Greenland is known to be more productive ecosystem compare to eastern Greenland (Henson et al., 2023; Vernet et al., 2021). Primary production in the photic zone consumes DIC at the expense of deeper waters. In areas exhibiting elevated photosynthesis, increased amounts of particulate organic matter sink below the photic zone and undergo remineralization (Arendt et al., 2010; Henson et al., 2023). Nevertheless, the specific impact of the biological pump compared to the advection of shelf water on pCO_2 levels remains uncertain.

Glacial meltwater lowers pCO_2 levels along the coasts of Greenland

Our other primary environmental driver of pCO_2 variability in these ecosystems is meltwater runoff from the Greenland ice sheet. Freshwater input to coastal waters has two main effects: it alters pelagic biological production by modifying water mass circulation within the fjords and induces direct physical and chemical changes.

Fjords influenced by land-terminating glaciers (LTGs) showcase distinctive characteristics marked by pronounced thermohaline stratification due to glacial river runoff (Henson et al., 2023). Heightened turbidity and stratification-driven nutrient limitation restrict primary production (Henson et al., 2023; Meire et al., 2017) and therefore CO_2 uptake in surface waters. Conversely, subglacial meltwater from marine-terminating glaciers (MTGs) generates buoyant plumes near glacier termini. These plumes entrain nutrient-rich water, stimulating phytoplankton blooms, and contribute to the reduction of surface DIC in fjords where MTGs are present (Chierici and Fransson, 2009; Henson et al., 2023; Meire et al., 2015, 2017; Rysgaard et al., 2012). However, these buoyant plumes may also entrain DIC-rich bottom water, elevating pCO_2 below the photic zone. Therefore, freshwater-driven circulation dynamics play a pivotal role in dictating the location and intensity of biological DIC consumption, thereby impacting the magnitude of air-sea exchange. Indeed, a recent study found that neighboring fjords with different glacial termini showed double the carbon uptake near MTGs compared to near LTGs (Meire et al., 2023). This relationship is evident in West Greenland, where the spatial distribution of AOU closely corresponds to the distribution of surface layer pCO_2 values (Fig. 4). However, this association seems absent in Eastern Greenland where no apparent difference in biological activity or pCO_2 levels near the termini of MTGs or LTGs are observed. This discrepancy might be attributed to the source history of water masses along the East Coast. Seawater entering fjords from the East Greenlandic current consists of cold, nutrient-poor water originating from the Arctic outflow (Henley et al., 2020). This combined with relatively lower glacial discharge (Velicogna et al., 2020) results in reduced upwelling of already nutrient-limited water and therefore restricted primary production along the East Coast, even near marine-terminating glaciers.

Freshwater input also has direct impacts on coastal carbon dynamics. A distinct relationship between pCO_2 and salinity was clearly visible (Figure 3c, 3d). Lowest observed pCO_2 values and therefore the strongest carbon sinks had low salinities and were located near the glacial termini (Figure 3c, 3d, 4b). Greenland's coastal waters are profoundly impacted by freshwater inputs. Within these fjords, August freshwater fractions were primarily of glacial origin while sea ice melt was limited (Henson et al., 2023). When compared to regions like Svalbard and Hudson Bay, Greenland stands out for its unique freshened waters characterized by lower salinity levels and ^{18}O isotopic signatures (Azetsu-Scott et al., 2010; Ericson et al., 2019; Fransson et al., 2015; Granskog et al., 2011; Henson et al., 2023). This freshwater has physical and chemical effects on the carbon dynamics of seawater. Glacial fjords, particularly affected by freshening, experience enhanced CO_2 undersaturation due to the non-linear influence of salinity on pCO_2 (Meire et al., 2015; Rysgaard et al., 2012). The combination of low temperatures, pCO_2 undersaturation, and the non-linear relationship with salinity amplifies the potential for CO_2 uptake in surface waters within fjords where freshening occurs.

As we anticipate increased melting in the future, one might expect a larger ΔpCO_2 and air-sea exchange. However, while increased melting may augment carbon uptake through freshening-driven CO_2 drawdown, future climate change may in some circumstances have the reverse effect. As temperatures increase, surface water warming could limit gas dissolution and decrease ΔpCO_2 between the ocean and the atmosphere. Additionally, the transition from MTGs to LTGs due to glacial retreat may lead to a reduction in primary production, limiting air-sea CO_2 exchange (Meire et al., 2023).

Air-sea exchange and sources of uncertainty

Flux magnitudes are a product of not only ΔpCO_2 but also gas transfer velocity. This diffusion of gases is related to wind-driven turbulence at the air-sea interface. The interplay between wind speed and ΔpCO_2 values plays a pivotal role in shaping these fluctuations (Ho et al., 2011). Spatial variability in summer fluxes around Greenland, therefore, are dictated not only by carbon dynamics but also wind regimes and fjord morphology. Notably, regions experiencing extreme CO_2 uptake, such as Uummannaq Fjord, exhibit a convergence of both larger ΔpCO_2 values and higher wind speeds in August 2016. In contrast, narrow and elongated fjords such as those present along the east coast of Greenland (Kong Oscar Fjord and Kejser Franz Joseph Fjord) exhibited lower CO_2 uptake, a product of both smaller ΔpCO_2 but also more calm summer wind conditions.

In fact, large uncertainty remains in estimating flux magnitudes based upon the source of wind data as well as the choice of gas transfer velocity parameterization. For instance, Sejr et al. (2011) reported larger flux magnitudes in Young Sound, registering below -25 mmol m^{-2} day^{-1}, compared to our range of -4 to -10 mmol m^{-2} day^{-1} despite relatively similar ΔpCO_2 measurements between 2006-2009 and during this study conducted in 2018. The discrepancies in flux calculations primarily stem from differences in wind speed measurements. Sejr et al. (2011) relied on hourly wind speeds from a nearby weather station, recording higher wind speeds compared to estimates derived from the Copernicus Arctic Reanalysis (2021). The non-linear relationship with wind speed results in intermittent spikes of gas transfer during periods of high wind speeds, enabling a large portion of monthly air-sea exchange to occur within a single storm.

The calculation of gas transfer velocities relies on empirically derived relationships. However, existing studies have primarily quantified k_{600} values in regions that are rather dissimilar to Arctic coastal ecosystems, making the choice of parameterization for coastal Greenland waters difficult. Even more worrying is the fact that the equation chosen to derive k_{600} significantly influences the magnitude of the fluxes we calculate (Fig. 5a). Currently, it remains unclear whether k_{600} values derived from open-ocean, coastal, or estuarine systems best represent Arctic fjords. In our study, this choice results in substantial variations, changing the maximum measured air-sea exchange from -24.8 to -61.6 mmol m^{-2} day^{-1} and altering median values from -2.5 to -15.9 mmol m^{-2} day^{-1}. Comparing Greenland coastal fluxes using Borges et al. (2014) and Wanninkhof & McGillis (1999) parameterizations results in an average flux difference of 772%. Uncertainties based on gas parameterization increase with both faster wind speeds and larger air-sea differences in pCO_2 (Fig. 5b). While Greenland fjords may not exhibit high wind speeds in the summer season, they certainly demonstrate extreme negative ΔpCO_2 values, making this choice of gas transfer equation disproportionally important compared to ecosystems at lower latitudes. These considerable discrepancies underscore the pressing need for better establishment of transfer velocities specific to these coastal Arctic regions.

Despite the uncertainties, we wanted to place this study in context by comparing to other coastal estimates of CO_2 sequestration. Upscaling of coastal carbon fluxes, particularly at high latitudes, is often based upon few datapoints/study sites (Laruelle et al., 2013). This study demonstrates the large heterogeneity of Greenland's coastlines and oceanographic conditions, therefore putting to question the reliability of upscaling efforts based on data from few locations. To compare with these assessments, we estimate coastal Greenland uptake by multiplying mean flux rates for fjord and shelf waters with estuarine and shelf surface areas according to Laruelle et al. (2013), obtaining uptake rates of 0.056 ± 0.052 Tg C day^{-1}. An annual estimate of Greenland coastal uptake was then calculated based upon these August flux rates and then scaled by the yearly-averaged ice cover in the estuarine and shelf regional areas according to Laruelle et al (2014), assuming no gas transfer where sea ice was present (Sejr et al., 2011). Assuming North and South Greenland coasts have 63.5% and 25.4% yearly-averaged ice cover respectively, we obtain the total Greenland coastal uptake rates of -9.5 ± 9.0 Tg C $year^{-1}$. We recognize this annual estimate makes many assumptions, especially as previous studies indicate large seasonality in carbon dynamics and air-sea exchange in Greenland fjords (Meire et al., 2023, 2015; Rysgaard et al., 2012). However, for the sake of comparison and context we present this first order estimate. Both Laruelle et al. (2014) and Dai et al. (2022) report nearly two to three times higher carbon sequestration in this region (-16.37 and -26.44 Tg C $year^{-1}$ respectively). Indeed, this region is beset by wide ranging estimates of air-sea exchange, partly due to the choice of gas-transfer parameterization. Still, based upon this study's, perhaps conservative, CO_2 uptake rates, the Greenland coastal ocean represents 3.8% of global coastal CO_2 uptake (0.25 Pg C $year^{-1}$) and 7.1% of uptake in the Arctic coastal zone (-134 Tg C $year^{-1}$; Dai et al., 2022) despite constituting 2.9% of global coastal ocean area (Laruelle et al., 2013). This considerable percentage of global carbon storage in such a relatively small region reiterates the need to better resolve this highly variable region with regards to carbon dynamics and air-sea exchange.

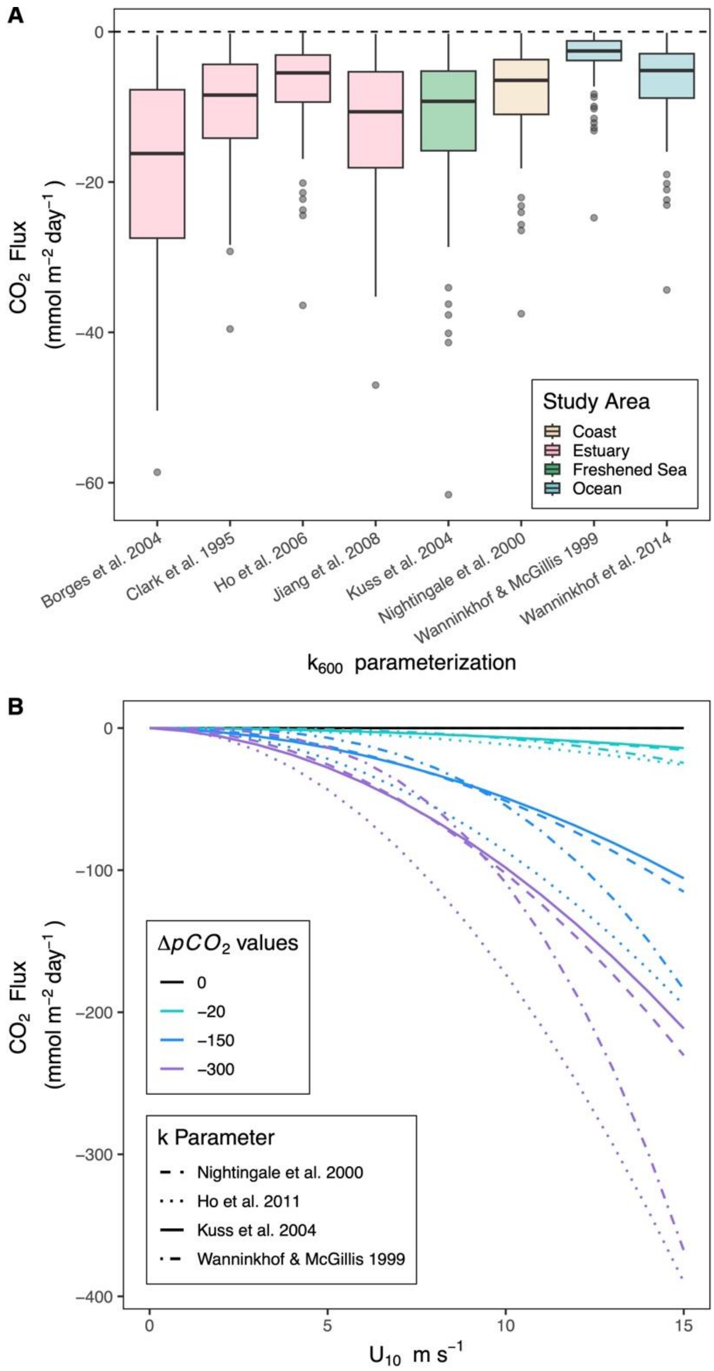

Figure 5. Calculated flux from coastal Greenland waters using different gas transfer velocity parameterizations (Borges et al., 2004; Wanninkhof and McGillis, 1999; Nightingale et al., 2000; Jiang et al., 2015; Kuss et al., 2004; Ho et al., 2011; Wanninkhof, 2014; Clark et al., 1995) (a). Variability in CO_2 uptake explained by wind speed, ΔpCO2 magnitude, and choice of gas parameterization.

Conclusion

The widespread undersaturation of surface water pCO_2 in Greenlandic coastal waters confirms that this region acts as a carbon sink during summer. However, pCO_2 concentrations as well as air-sea exchange rates exhibited considerable geographical variability. Levels of pCO_2 on the West coast of Greenland were controlled by a combination of biological and physical/chemical drivers. Levels of net autotrophy/heterotrophy, indicated by oxygen saturation, combined with freshwater runoff helped to explain spatial variation in West coast pCO_2 levels. In contrast, East Greenland fjords had pCO_2 levels that were dictated mainly by abiotic factors, including temperature and salinity. Freshwater export has two-fold effects, altering primary production by modifying circulation in the surface layer and exerting direct physical/chemical impacts. Therefore, the lowest pCO_2 values and the largest air-sea exchange occurred on the West coast of Greenland where high levels of net production and freshwater export took place concurrently. This resulted in CO_2 uptake reaching -38 mmol m^{-2} day^{-1}. Using these measurements across 6° of latitude, allowed us to create a first order estimate of CO_2 uptake in the Greenland coastal ocean. Using average fluxes in fjords and shelf waters combined with coastal surface area data indicated that the Greenland coastal ocean absorbs -9.5 ± 9.0 Tg C $year^{-1}$, or nearly 4% of global coastal ocean CO_2 uptake. However, quantifying the *true* flux magnitude is beset by significant uncertainties (up to 770%), stemming from the averaging of wind products, as well as the selection of gas exchange formulations. We therefore encourage further studies to refine the estimation of gas transfer velocities in the Arctic coastal zone, as this represents the largest contributor to present uncertainty. Additionally, comprehensive understanding demands data collection across multiple seasons. This expanded temporal scope will provide a more holistic view of the year-round dynamics of CO_2 exchange within Greenlandic fjords, shedding light on how these ecosystems sequester carbon throughout the annual cycle.

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Data Availability

Data relevant to reproduce the results of this study have been made available on Zenodo, an all-purpose open research repository created by Open Access Infrastructure for Research in Europe (OPENAire), a project supported by the European Commission, and by CERN, the European Organization for Nuclear Research. Oceanic pCO2 data are available with the following DOI: https://doi.org/10.5281/zenodo.10679593. Environmental variables recorded by CTD instruments are available for cruises from the West and East coasts respectively at the following DOIs: https://doi.org/10.5281/zenodo.4062024 and https://doi.org/10.5281/zenodo.5572329.

CRediT authorship contribution statement

Henry Henson: Conceptualization, Formal analysis, Data curation, Writing – original draft. Mikael Sejr: Investigation, Writing – review & editing. Lorenz Meire: Investigation, Writing – review & editing. Mie Winding: Investigation, Writing – review& editing. Lise Lotte Sørensen: Supervision, Funding acquisition, Writing – review & editing. Johnna M. Holding: Conceptualization, Investigation, Supervision, Funding acquisition, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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