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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 $p\text{CO}_2$ levels to varying degrees in different regions
- Estimation of the true flux magnitude is beset by large uncertainties, particularly in polar coastal regions

Supporting Information:

Supporting Information may be found in the online version of this article.

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Resolving Heterogeneity in CO_2 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 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 $p\text{CO}_2$ 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 $p\text{CO}_2$ and air-sea exchange was linked intricately to freshwater export from the Greenland ice sheet and levels of primary production in these ecosystems. Air-sea exchange of CO_2 ranged from 0.23 to $-64 \text{ mmol m}^{-2} \text{ day}^{-1}$. However, we found that flux estimation faces substantial uncertainties (up to 672%) due to wind product averaging and gas exchange formula selection. Upscaling only heightens this uncertainty leading to wide ranging estimates of Greenland coastal CO_2 uptake between -16 and $-26 \text{ Tg C year}^{-1}$ (This study, Dai et al., 2022, <https://doi.org/10.1146/annurev-earth-032320-090746>; Laruelle et al., 2014, <https://doi.org/10.1002/2014gb004832>). 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 is annually emitted to the atmosphere. The majority of oceanic CO_2 uptake occurs 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. Air-sea exchange of CO_2 ranged from 0.23 to $-64 \text{ mmol m}^{-2} \text{ day}^{-1}$ and the controlling factors behind this flux of carbon were wind speed, amount of glacial runoff, and the balance between biologic producers/consumers. However, several wide-ranging estimates of uptake in the Greenland coastal ocean have been proposed, indicating the need to better understand carbon sequestration variability in this region.

1. 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 global CO_2 emissions annually (Ciais et al., 2013; Sabine et al., 2004; Takahashi et al., 2009). Oceans at high latitudes uptake more CO_2 due to a combination of low temperatures and high productivity (Bates & Mathis, 2009; Takahashi et al., 2009). Although 25% of continental shelves (depth $< 200 \text{ m}$) 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 & Mathis, 2009).

The driving force behind CO_2 exchange across the sea surface is the difference in partial pressure of CO_2 ($\Delta p\text{CO}_2$) between seawater and the overlying air. However, the fluctuation of oceanic $p\text{CO}_2$ is more spatially and temporally variable compared to atmospheric $p\text{CO}_2$, making oceanic $p\text{CO}_2$ the thermodynamic driver of air-sea

carbon uptake. Arctic coastal waters are generally considered as a sink for CO₂ due to their undersaturated *p*CO₂ 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 CO₂ 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 influenced 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, CO₂, water) at their interfaces, yet have often been overlooked in air-sea exchange surveys due to their spatial and temporal complexity (Chen & Borges, 2009; Miller et al., 2019).

Understanding the intricate dynamics of CO₂ 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 the entire Greenland coastal ocean. Knowing that the coasts of Greenland are a heterogeneous landscape, it is crucial we gain a better understanding of the spatial variation in *p*CO₂ levels and carbon fluxes in this region to obtain reliable assessments of the CO₂ uptake by the Greenlandic coastal area. Additionally, a large-scale perspective on the drivers of surface *p*CO₂ 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 glacial meltwater and biological activity will drive heterogeneity in *p*CO₂ 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 CO₂ fluxes in this region. This study aims to fill in this gap in knowledge by exploring CO₂ fluxes in Greenlandic fjords and shelf waters spanning 6° of latitude. Given the region's sensitivity to climate change and its substantial CO₂ uptake relative to other marine areas, a realistic assessment of exchange rates is imperative. By delving into the drivers influencing air-sea CO₂ 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.

2. Methods

2.1. Sampling

Two research cruises were conducted along the coasts of Greenland where oceanic partial pressure of CO₂ (*p*CO₂) 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 24 August 2018 (Figure 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 *p*CO₂ profiles were measured using a Contros Hydro-C CO₂ sensor (Meire et al., 2023). The Hydro-C sensor was lowered from the ship side and equilibrated for 5–10 min until a stable reading was acquired at 8 depths between 1 and 60 m (1, 5, 10, 20, 30, 40, 50, 60 m). Motors were shut off at each station to limit surface mixing. An annual calibration has been conducted to ensure the accuracy of the Contros Hydro-C CO₂ sensor using a certified 400 ± 2% ppm CO₂ standard gas. The sensor has been in use for over 8 years and consistently demonstrates long-term stability, with recorded values ranging between 397 and 401 ppm each year. Given that these measurements fall within the known range of the standard gas, no further correction of the output has been deemed necessary. This stability indicates minimal drift over time, ensuring reliable data collection in the field. 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 (Ahmed et al., 2020; Macovei et al., 2021; Miller et al., 2019). Therefore, membrane-based sensors at 1 m allowed a robust examination of surface layer *p*CO₂ 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 *p*CO₂. Meanwhile the eastern cruise conducted 89 CTD casts, with *p*CO₂

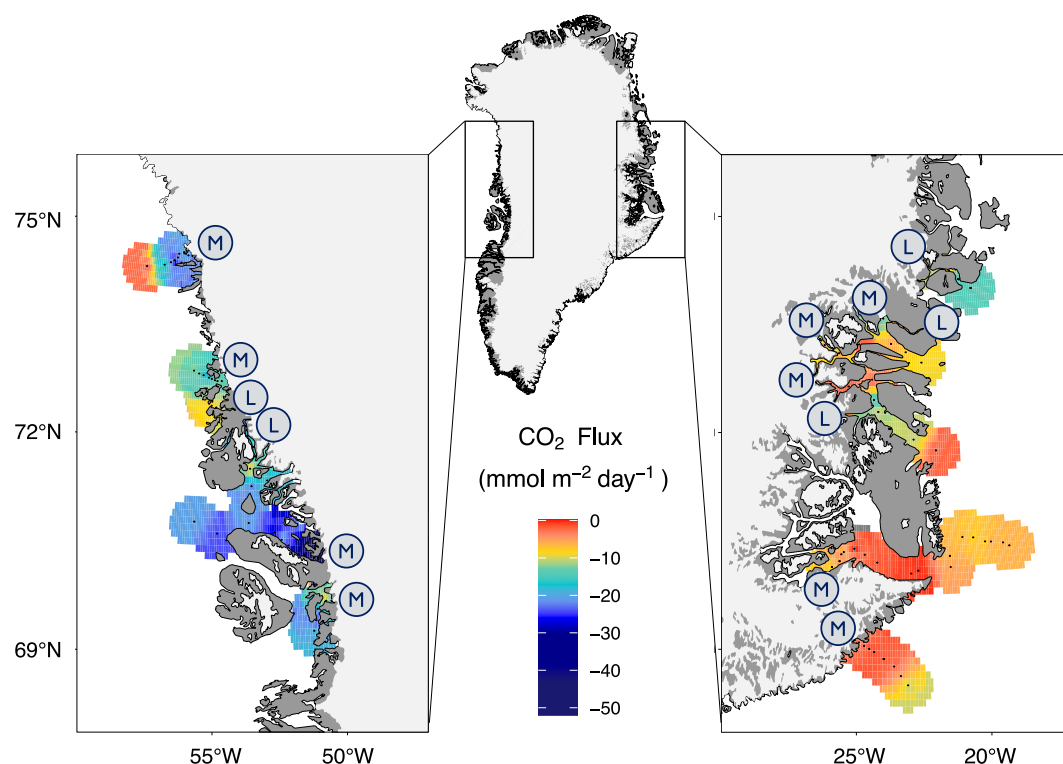


Figure 1. Map of air-sea exchange of CO_2 in Greenland coastal waters. Station locations are pictured as black points. Data was interpolated using an inverse distance weighted matrix limited to a maximum distance of 39 km. Marine- and Land-terminating glaciers are labeled with M and L respectively.

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 it was categorized as just “fjord,” and when a station was located outside the fjord mouth, they were assigned as “shelf” stations (Figure S1 in Supporting Information S1).

2.2. Calculation of Air-Sea Exchange

The net air-sea exchange of CO_2 was calculated using the diffusive boundary model equation:

$$F = k_{660}s(\Delta p\text{CO}_2) \quad (1)$$

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 $\Delta p\text{CO}_2$ is the difference between surface water and atmospheric $p\text{CO}_2$ values (at 1 m depth). Negative fluxes, therefore, represent oceanic CO_2 uptake while positive fluxes represent out-gassing. Atmospheric $p\text{CO}_2$ data 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: $k_{660} = k(660/\text{Sc})^{-0.5}$ 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 data sets 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)

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

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, a high-resolution data set specifically developed for the European Arctic using the HARMONIE-AROME Numerical Weather Prediction system. The data undergoes routine validation against in situ observations and is benchmarked against the global ERA5 reanalysis to ensure its high meteorological quality (Copernicus Climate Change Service, 2021). The CARRA reanalysis runs with a 3-hr data assimilation which we averaged over the months of August 2016 and 2018. Calculated fluxes were not scaled by ice cover as August fjord conditions were ice-free.

2.3. Statistical Modeling

To examine the impact of environmental parameters on $p\text{CO}_2$ levels, statistical models were applied to our data, focusing on the upper ocean, where biological, physical, and chemical drivers can influence CO_2 uptake potential. CTD profiles published in Henson et al. (2023) reveal the strongest density differences occur in the upper few meters due to intense freshening, making traditional calculation of the mixed layer depth based on seawater density challenging for examining biogeochemical processes (Figure S2 in Supporting Information S1). Instead, we define a biologically relevant dynamic surface layer (DSL) based on chlorophyll-*a* fluorescence profiles. This upper ocean layer captures dynamic signals from biological and chemical processes. To identify the DSL depth, we first located the deep fluorescence maximum (DFM) and then selected the depth corresponding to the maximum standard deviation in fluorescence values below the DFM and above 50 m, marking the bottom of the fluorescence peak. All data from above this depth at each station were included in the statistical models.

First, general linear models were fit to identify relationships between predictors and $p\text{CO}_2$ in the dynamic surface layer. 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 & 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* = 0.34, p -value < $6\text{e}-5$). Spatial autocorrelation can inflate type 1 errors and potentially influence parameter estimation (Kreft & 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 $p\text{CO}_2$. 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 < $5\text{e}-4$). 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.

3. Results

3.1. Heterogeneity and Drivers of $p\text{CO}_2$

Comparison between the east and west coasts of Greenland revealed differences extending beyond surface waters. Depth profiles of $p\text{CO}_2$ depicted distinct signals in both regions. Western Greenland exhibited more variable $p\text{CO}_2$ 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 m (Figure 2, Figure S3 in Supporting Information S1). Conversely, the eastern coast displayed more uniform $p\text{CO}_2$ levels across the upper water column, with values slightly below atmospheric levels (221–399 μatm at 1 m and 330–414 at 50 m; Figure 2). These differences in surface $p\text{CO}_2$ levels between East and West Greenland are also reflected in the magnitude of the air-sea CO_2 exchange that can be seen in Figure 1.

The concentrations of $p\text{CO}_2$ in the water column are influenced by a combination of biological and physical/chemical factors. The main drivers affecting $p\text{CO}_2$ 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 $p\text{CO}_2$.

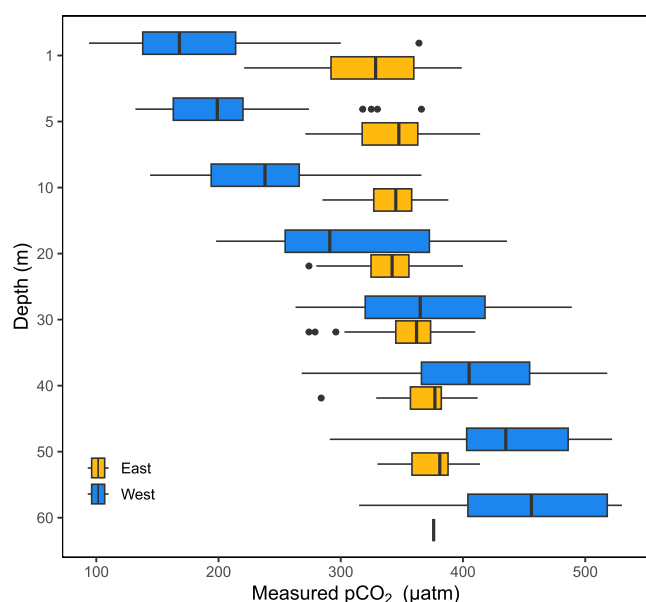


Figure 2. Boxplots of $p\text{CO}_2$ profiles on the East and West coast of Greenland. Outliers are depicted as points outside the quartile lines.

Primary producers' biological incorporation of carbon decreased $p\text{CO}_2$ levels in the surface layer (Figure 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 inferred (Figure 3b), leading to a decoupling between AOU and measured $p\text{CO}_2$. This region, known for its lower productivity (Henson et al., 2023), exhibited less variation in $p\text{CO}_2$ levels.

Figures 3c and 3d highlight the primary physical and chemical drivers of $p\text{CO}_2$ in this region: freshwater influx. Glacial meltwater input has been documented to reduce seawater $p\text{CO}_2$ through both thermodynamic effects and dilution (Meire et al., 2015). The relationship between freshwater input and $p\text{CO}_2$ is apparent in both eastern and western Greenland, where lower $p\text{CO}_2$ levels correspond with lower salinities. This relationship, however, was somewhat obscured in western Greenland where high levels of primary production reduce $p\text{CO}_2$ values even at more elevated salinities. Meanwhile, in Eastern Greenland, where biological production is weak, a strong linear relationship between salinity and $p\text{CO}_2$ can be observed.

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 $p\text{CO}_2$ in relation to environmental predictors within the dynamic surface layer. 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

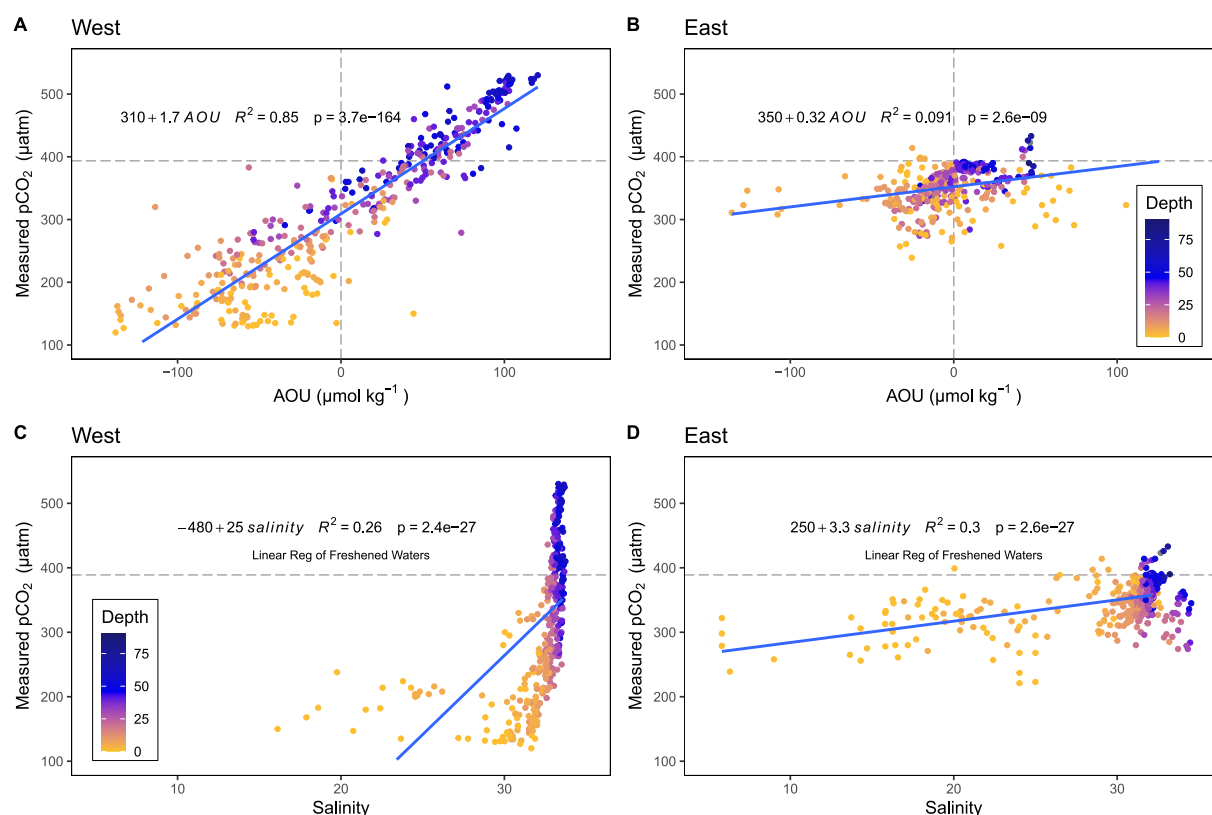


Figure 3. Relationships between apparent oxygen utilization (AOU) and $p\text{CO}_2$ (a, b) and salinity and $p\text{CO}_2$ (c, d) for East and West coasts. Linear regression fits use all data in (a, b) while (c, d) fit regression for all data below salinity of coastal seawater endmembers (Henson et al., 2023). Atmospheric $p\text{CO}_2$ concentrations and the equilibrium between net auto- and heterotrophy are depicted with horizontal and vertical gray dashed lines respectively.

Table 1
Fitted GLS Model With Rational Quadratic Spatial Structure

	Estimate	T Statistic	p-value
Intercept	54.88	2.199	0.0288
AOU	0.178	2.081	0.0384
Salinity	8.953	12.53	<0.0001
Temperature	12.71	9.366	<0.0001
Coast (Categorical—west)	−24.29	−1.454	0.1471
AOU:Coast (west)	1.044	10.25	<0.0001
Temperature:Coast (west)	−17.88	−10.58	<0.0001

on each coast. Both biological and physical factors emerged as robust predictors for $p\text{CO}_2$ levels. The spatial model illustrates that in western Greenland, fluctuations in $p\text{CO}_2$ were primarily driven by AOU (biological processes). Conversely, in eastern Greenland, salinity and temperature exerted the most substantial influence on $p\text{CO}_2$ levels. This spatial linear model duly accounted for the spatial autocorrelation of measured $p\text{CO}_2$ values. The estimated range (distance at which spatial correlation becomes negligible) is approximately 0.84 longitudinal degrees, and the nugget parameter (unaccounted variability) was 0.51.

Model performance underwent rigorous assessment via cross-validation, utilizing subsets of the data set to train and validate the model. Our spatial linear model (SLM) accurately predicted 79% of the variance in $p\text{CO}_2$ within the test data set, indicating that a relatively simple model can accurately estimate

coastal $p\text{CO}_2$ levels within the dynamic surface layer. 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 36.4 and 27.3 μatm , respectively, between the predicted and measured $p\text{CO}_2$ values. Furthermore, the prediction error rate, calculated as the RMSE divided by the mean of the observed values, was 12.7%. This suggests that the model's error is small relative to the overall $p\text{CO}_2$ range, further validating its predictive accuracy. 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 $p\text{CO}_2$ levels around coastal Greenland.

Spatial variability in $p\text{CO}_2$ (Figure S3 in Supporting Information S1) 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 ($p\text{-value} < 5\text{e-}5$) where subglacial discharge drives upwelling of nutrient-rich bottom water. Meanwhile, AOU did not vary spatially on the east coast (Figure 4a). Similar to AOU, $p\text{CO}_2$ values in western Greenland were elevated near LTGs compared to near MTGs, with even higher values in shelf waters (Figure 4b). Eastern Greenland did not demonstrate a difference in $p\text{CO}_2$ values between stations near LTGs and MTGs ($p\text{-value} = 0.997$). However, the stations close to glacial output exhibited lower $p\text{CO}_2$ values than mid-fjord or shelf waters (Figure 4b).

3.2. CO_2 Fluxes

The air-sea exchange of CO_2 is a product of wind speed driven turbulence and $\Delta p\text{CO}_2$. 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 the monthly and 3-hour wind speed averages available from the Schyberg et al. (2020). The monthly wind speeds in August ranged between 1.7 and 6.5 m s^{-1} across our stations within Greenland fjords while 3-hour averages ranged from 0 to 16.6 m s^{-1} . Notably, we found a strong correlation ($R^2 = 0.77$; $P = 1.1\text{e-}36$) between these two flux calculations. However, the air-sea CO_2 flux derived from 3-hour wind averages exceeded the estimates using monthly wind speeds by an average of 26% (median of 8%). Disparities in these calculations are attributed to the non-linear relationship between gas transfer velocity and wind speed (Equation 2), leading to significant exchanges during periods of high wind speed events. As a result, if 1 hr or 10 min average wind speeds were available, one could expect an even larger difference to monthly averages, as was observed in Sejr et al. (2011).

In August, both the East and West coasts of Greenland showed $p\text{CO}_2$ values in surface water below atmospheric levels, indicating this region acts as a CO_2 sink. However, considerable geographic variability existed in $\Delta p\text{CO}_2$ values, leading to varied flux magnitudes. Fluxes in West Greenland ranged from -64.23 to $-2.62 \text{ mmol m}^{-2} \text{ day}^{-1}$ while those in East Greenland ranged from -21.27 to $0.23 \text{ mmol m}^{-2} \text{ day}^{-1}$, 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

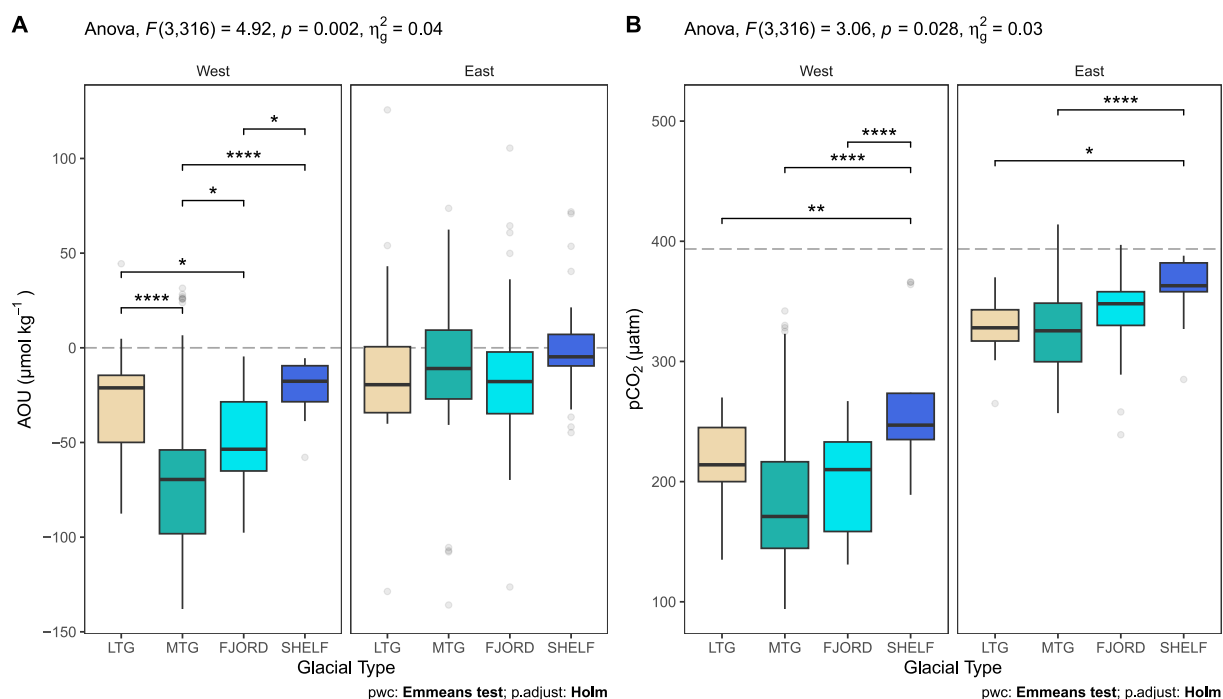


Figure 4. Boxplots and two-way ANOVA analysis of AOU with fjord location (a) and $p\text{CO}_2$ (b) with fjord location for surface layer samples (above 20 m depth). Significant interactions between coast and fjord location on AOU and on $p\text{CO}_2$ are described above each plot. Significance codes: “*” 0.05 “***” 0.01 “****” 0.001 “*****” 0.0001. 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 $p\text{CO}_2$ concentrations (b) are depicted as gray dashed lines. Meanwhile outliers are depicted as gray points outside the quartile lines.

smaller $\Delta p\text{CO}_2$, was recorded in shelf waters (Figure 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.

However, estimating flux magnitudes continues to be affected by significant uncertainties. The calculation of gas transfer velocities, needed to estimate air-sea exchange, relies on empirically derived relationships. To date, existing studies have primarily quantified k_{660} values in regions that are rather dissimilar to Arctic coastal ecosystems, making the choice of parameterization for coastal Greenland waters difficult. It is not known which of these parameterizations best represents gas exchange in this cold, freshened ecosystem. By calculating fluxes using a range of k -parameterizations from open-ocean, coastal, and estuarine systems, we see dramatic differences (Figure 5a). This choice changes the maximum measured air-sea exchange from -42.4 to -100.3 $\text{mmol m}^{-2} \text{ day}^{-1}$ and alters median values from -4.06 to -25.13 $\text{mmol m}^{-2} \text{ day}^{-1}$. Comparing Greenland coastal fluxes using Borges et al. (2004) and Wanninkhof and McGillis (1999) parameterizations, for example, results in a median flux difference of 672% or 21.06 $\text{mmol m}^{-2} \text{ day}^{-1}$. These extreme differences highlight just one of the major uncertainties that remain with estimating carbon sequestration in Arctic Coastal environments.

4. Discussion

4.1. Greenland Coastal $p\text{CO}_2$ Is Highly Heterogeneous

Our observation of low surface water $p\text{CO}_2$ 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 $\Delta p\text{CO}_2$ 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 Kangerittivaq. 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 and -134 μatm in Young Sound, Rysgaard et al. (2012) observed $\Delta p\text{CO}_2$ 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

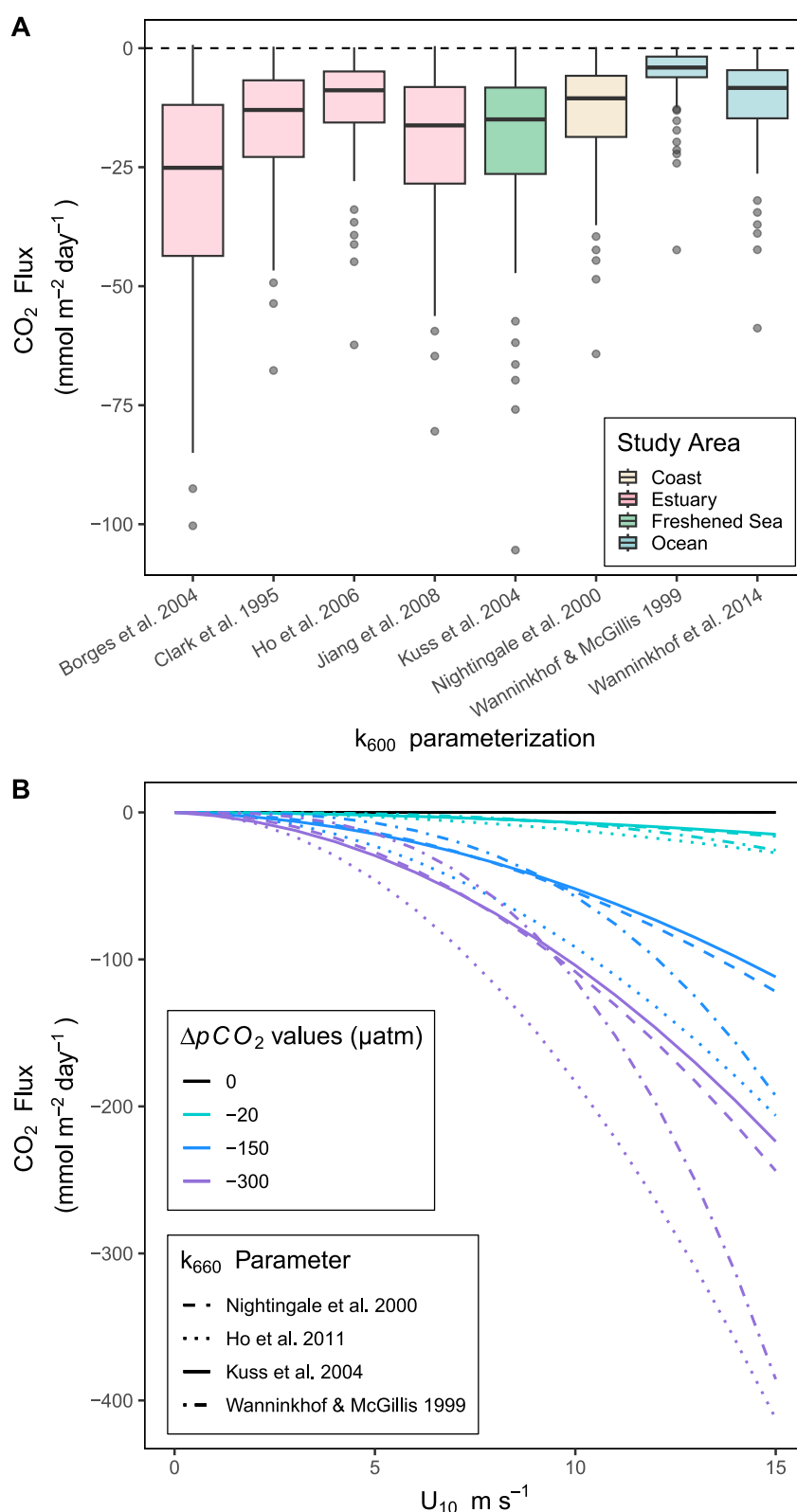


Figure 5. Calculated flux from coastal Greenland waters using different gas transfer velocity parameterizations (Borges et al., 2004; Clark et al., 1995; Ho et al., 2011; Jiang et al., 2015; Kuss et al., 2004; Nightingale et al., 2000; Wanninkhof & McGillis, 1999; Wanninkhof, 2014) (a). Variability in CO₂ uptake explained by wind speed, ΔpCO₂ magnitude, and choice of gas transfer parameterization.

variability and levels of $\Delta p\text{CO}_2$ that we observed on the east and west coast. Additionally, when comparing to syntheses of global shelf seas, our measured ranges of $\Delta p\text{CO}_2$ fall within low-end of measurements at the same latitude (Dai et al., 2022).

Although $p\text{CO}_2$ values have been reported before, this is the first study to examine CO_2 uptake potential in more than one fjord system. We measured $p\text{CO}_2$ 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 $p\text{CO}_2$ 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.

4.2. Autotrophy/Heterotrophy Balance Dictates $p\text{CO}_2$ in West Greenland

Both biological and physical parameters were found to be significantly related to surface water $p\text{CO}_2$ within SLM models (Table 1). As a result, oxygen saturation in combination with salinity and temperature can be used to predict $p\text{CO}_2$ 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 $p\text{CO}_2$ levels and AOU in western Greenland fjords (Figure 3a). Similarly, limited biological productivity in eastern Greenland resulted in less variability in $p\text{CO}_2$ profiles with depth (Figure 2). Western Greenland is known to be a more productive ecosystem compared to eastern Greenland due to advection of surrounding water masses with higher nutrient concentrations (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 $p\text{CO}_2$ levels remains uncertain.

4.3. Glacial Meltwater Lowers $p\text{CO}_2$ Levels Along the Coasts of Greenland

Our other primary environmental driver of $p\text{CO}_2$ 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 & 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 $p\text{CO}_2$ 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 $p\text{CO}_2$ values in the upper 20 m (Figure 4, Figure S4 in Supporting Information S1). However, this association seems absent in Eastern Greenland where no apparent difference in biological activity or $p\text{CO}_2$ 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 $p\text{CO}_2$ and salinity was clearly visible (Figures 3c and 3d). Lowest observed $p\text{CO}_2$ values and therefore the strongest carbon

sinks had low salinities and were located near the glacial termini (Figures 3c, 3d and 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 extremely 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 $p\text{CO}_2$ (Meire et al., 2015; Rysgaard et al., 2012). The combination of low temperatures, $p\text{CO}_2$ 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 $\Delta p\text{CO}_2$ 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 $\Delta p\text{CO}_2$ 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).

4.4. Air-Sea Exchange and Sources of Uncertainty

Flux magnitudes are a product of not only $\Delta p\text{CO}_2$ 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 $\Delta p\text{CO}_2$ 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 $\Delta p\text{CO}_2$ 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 $\Delta p\text{CO}_2$ 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 $-25 \text{ mmol m}^{-2} \text{ day}^{-1}$, compared to our range of -6 to $-19 \text{ mmol m}^{-2} \text{ day}^{-1}$ despite relatively similar $\Delta p\text{CO}_2$ measurements between 2006 and 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 local weather station, recording higher wind speeds compared to the more uncertain 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.

Beyond the source of wind data, large uncertainties arise from the choice of gas transfer velocity parameterization. We still do not know if parameterizations derived from coastal, estuarine, or open ocean environments best represent air-sea exchange in the coastal Arctic. Even more worrying is the fact that the equation chosen to derive k_{660} significantly influences the magnitude of the fluxes we calculate (Figure 5a). Uncertainties based on gas parameterization increase with both faster wind speeds and larger air-sea differences in $p\text{CO}_2$ (Figure 5b). While Greenland fjords may not exhibit high wind speeds in the summer season, they certainly demonstrate extreme negative $\Delta p\text{CO}_2$ values, making this choice of gas transfer equation disproportionately important compared to ecosystems at lower latitudes.

These findings underscore the pressing need for better establishment of transfer velocities specific to these Arctic coastal regions, while challenging the level of certainty we can place on historic Arctic flux studies that have not derived a regionally specific k -parameterization.

These uncertainties, including accurate wind field data and representative gas transfer velocity parameterizations are only heightened when trying to upscale fluxes to larger areas. Obtaining large-scale estimates of ocean CO_2 uptake is an important task. However, it is often based upon interpolating sparse data in space and time, both of which can be extremely variable in Arctic marine waters. Two previous studies give estimates of coastal Greenland uptake as part of their global budgets of CO_2 exchange, using surface $p\text{CO}_2$ data to calculate fluxes and

then upscale fluxes over ocean shelf areas (Dai et al., 2022; Laruelle et al., 2014). To compare with these assessments, we estimate coastal Greenland uptake using the same approach. By multiplying mean flux rates for fjord and shelf waters with estuarine and shelf surface areas according to Laruelle et al. (2013), we obtain uptake rates of $-0.005 \pm 0.0039 \text{ Tg C day}^{-1}$ in Greenlandic Fjords and $-0.089 \pm 0.074 \text{ Tg C day}^{-1}$ in shelf waters for the summer season. An annual estimate of Greenland coastal uptake can then be estimated 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 $-16.7 \pm 13.7 \text{ Tg C year}^{-1}$. 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 due to changes in primary production and runoff throughout the year (Meire et al., 2015, 2023; Rysgaard et al., 2012). However, for the sake of comparison and context we present this first order estimate. Laruelle et al. (2014) presents a nearly identical estimate while Dai et al. (2022), report significantly higher carbon sequestration in this region (-16.37 and $-26.44 \text{ Tg C year}^{-1}$ respectively). Indeed, this region is beset by wide ranging estimates of air-sea exchange, partly due to the heterogeneity of marine $p\text{CO}_2$ data combined with the choice of gas transfer parameterization. Still, based upon these three available estimates, the Greenland coastal ocean represents between 6.68% and 10.6% of global coastal CO_2 uptake ($0.25 \text{ Pg C year}^{-1}$) and between 12.5% and 19.7% of uptake in the Arctic coastal zone ($-134 \text{ Tg C year}^{-1}$; Dai et al., 2022) despite constituting only 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.

5. Conclusion

The widespread undersaturation of surface water $p\text{CO}_2$ in Greenlandic coastal waters confirms that this region acts as a carbon sink during summer. However, $p\text{CO}_2$ concentrations as well as air-sea exchange rates exhibited considerable geographical variability. Levels of $p\text{CO}_2$ on the West coast of Greenland were controlled by a combination of biological and physical/chemical drivers. Levels of net autotrophy/heterotrophy, indicated by apparent oxygen utilization, combined with freshwater runoff helped to explain spatial variation in West coast $p\text{CO}_2$ levels. In contrast, East Greenland fjords had $p\text{CO}_2$ 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 $p\text{CO}_2$ 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 $-64 \text{ mmol m}^{-2} \text{ day}^{-1}$.

However, quantifying the true flux magnitude is beset by significant uncertainties (up to 672%), stemming from the averaging of wind products, as well as the selection of gas exchange formulations. A rough upscaling of our data across 6° of latitude allows comparison with published literature on CO_2 uptake in the Greenland coastal ocean. Current estimates range from -16 (this study, Laruelle et al., 2014) to $-26 \text{ Tg C year}^{-1}$ (Dai et al., 2022). This equates to between 6.7% and 10.6% of global coastal ocean CO_2 uptake. While these estimates are highly uncertain, they highlight the critical need for accurate quantification of air-sea CO_2 exchange in this disproportionately impactful region. To reduce these uncertainties, we strongly encourage further studies aimed at refining gas transfer velocity parameterizations in the Arctic coastal zone, as this represents the largest source of present uncertainty. Additionally, expanding data collection beyond the summer months is crucial to avoid upscaling from seasonally unrepresentative data. This expanded temporal scope will provide a more holistic view of the year-round dynamics of CO_2 exchange within Greenlandic fjords and shelf waters, shedding light on how these ecosystems sequester carbon throughout the annual cycle.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

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 $p\text{CO}_2$ data used for estimating fluxes are freely available via <https://doi.org/10.5281/zenodo.10679593> (Henson et al., 2024). 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> (Carlson et al., 2020) and <https://doi.org/10.5281/zenodo.5572329> (Holding et al., 2021).

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