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Key Points:

- An extensive data set of western Arctic coastal ocean seawater pCO₂ was analyzed
- Sea ice-adjusted annual carbon uptake was 5% of global coastal ocean exchange
- Areas of uncertainty were used to pinpoint next steps for future research

Supporting Information:

Figures S1–S3

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Sea-air CO₂ exchange in the western Arctic coastal ocean

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Abstract The biogeochemical seascape of the western Arctic coastal ocean is in rapid transition. Changes in sea ice cover will be accompanied by alterations in sea-air carbon dioxide (CO₂) exchange, of which the latter has been difficult to constrain owing to sparse temporal and spatial data sets. Previous assessments of sea-air CO₂ flux have targeted specific subregional areas of the western Arctic coastal ocean. Here a holistic approach is taken to determine the net sea-air CO₂ flux over this broad region. We compiled and analyzed an extensive data set of nearly 600,000 surface seawater CO₂ partial pressure (*p*CO₂) measurements spanning 2003 through 2014. Using space-time colocated, reconstructed atmospheric *p*CO₂ values coupled with the seawater *p*CO₂ data set, monthly climatologies of sea-air *p*CO₂ differences (Δp CO₂) were created on a 0.2° latitude × 0.5° longitude grid. Sea-air CO₂ fluxes were computed using the Δp CO₂ grid and gas transfer rates calculated from climatology of wind speed second moments. Fluxes were calculated with and without the presence of sea ice, treating sea ice as an imperfect barrier to gas exchange. This allowed for carbon uptake by the western Arctic coastal ocean to be assessed under existing and reduced sea ice cover conditions, in which carbon uptake increased 30% over the current 10.9 ± 5.7 Tg C (1 Tg = 10¹² g) yr⁻¹ of sea ice-adjusted exchange in the region. This assessment extends beyond previous subregional estimates in the region in an all-inclusive manner and points to key unresolved aspects that must be targeted by future research.

1. Introduction

Relative to the vast open ocean, coastal oceans exhibit large carbon dioxide (CO₂) disequilibria with the atmosphere [Borges and Frankignoulle, 1999; Evans et al., 2011; Hales et al., 2005; Thomas et al., 2004], indicating that these settings can be large sources or sinks of atmospheric CO2. Collectively, existing data have implied that the global coastal oceans are an important net sink for atmospheric CO₂ ranging between 10 and 20% of the contemporary open ocean uptake [Borges et al., 2005; Cai et al., 2006; Dai et al., 2013; Laruelle et al., 2010, 2014; Takahashi et al., 2009; Wanninkhof et al., 2013]. This unified view of the coastal ocean is in general a weighted average of a mosaic of atmospheric CO₂ source and sink areas that vary widely in the quantity of data collected within each region and across seasons [e.g., Hales et al., 2008]. The overall scarcity of measurements in both time and space presents a significant challenge for accurately constraining net exchange in coastal ocean settings that inherently contain large carbonate system variability [e.g., Evans et al., 2011]. This problem is also complicated by the fact that these systems are being impacted by a changing climate that is altering the frequency distribution of both internal and external sources of variability [Bader et al., 2011; Bauer et al., 2013; Doney et al., 2012; Overland et al., 2014; Sydeman et al., 2014; Wassmann, 2011]. Even as the number of surface seawater CO₂ partial pressure (pCO₂) measurements has increased during the past decade, there are still large expanses of unsampled coastline, especially in the rapidly changing Arctic Ocean [Bakker et al., 2014; Overland et al., 2014]. Expanding data collections in coastal

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Figure 1. Map of the western Arctic coastal ocean, defined here by ocean area within the blue polygon. Red dots within the blue polygon mark the grid used for the construction of monthly sea-air CO₂ flux climatologies, with the offshore boundary set by the Surface Ocean CO₂ Atlas (SOCAT) 400 km continental margin mask (http://www.socat.info). Grid cells are 0.2° latitude $\times 0.5^{\circ}$ longitude (~420 km²). The surface area of the western Arctic coastal ocean is 1.2×10^{12} m². The blue polygon is split between the Chukchi Sea (green polygon; 2.9×10^{11} m²) to the west and the Beaufort Sea (black polygon; 9.2×10^{11} m²) to the east. Upper Barrow Canyon (marked with a black arrow) is the divide between these coastal seas.

settings is the key to refining estimates of net sea-air CO_2 exchange from early assessments of large influx nearly equivalent to open ocean uptake based on spatially and temporally limited data from single settings [*Thomas et al.*, 2004; *Tsunogai et al.*, 1999] to more recent evaluations that drastically downsize the coastal ocean sink magnitude using analyses of broad data compilations built from either many independent studies or extensive pCO_2 data sets [*Borges et al.*, 2005; *Cai*, 2011; *Cai et al.*, 2006; *Chen and Borges*, 2009; *Chen et al.*, 2013; *Dai et al.*, 2013; *Laruelle et al.*, 2010, 2014]. The western Arctic coastal ocean is one such region believed to support large atmospheric CO_2 uptake based on a small number of studies that generally report over short time and space scales (e.g., seasonal and subregional). However, a more holistic approach is necessary to better constrain surface seawater CO_2 exchange with the atmosphere in this rapidly changing polar seascape.

The western Arctic coastal ocean is defined here as the combination of Chukchi and Beaufort seas extending 400 km in the offshore direction into Canada Basin (Figure 1). The 400 km from shore boundary is the coastal ocean definition used by the Surface Ocean CO₂ Atlas (SOCAT) community [Bakker et al., 2014; Pfeil et al., 2013], and we employ this definition here in order to be consistent with the SOCAT community as well as for the following operational and functional reasons: (1) the 400 km from shore distance is roughly the size of nearshore pixels excluded in open ocean syntheses of sea-air CO₂ exchange [Takahashi et al., 2002, 2009], and (2) coastally influenced biogeochemical signals are in general attenuated within this distance from shore [Chavez et al., 2007; Hales et al., 2012]. The areal definition of the western Arctic coastal ocean used here encompasses two contrasting subregional seas: the Chukchi Sea with a broad and shallow shelf (<50 m), and the Beaufort Sea that consists of three narrower shelf regions in addition to portions of Amundsen Gulf and M'Clure Strait (Figure 1). These subregions are similar in that they experience periods of reduced sea ice cover and greater open water beginning in approximately June and ending in November [Bader et al., 2011], as well as an extreme solar cycle with nonlimiting irradiances from April to September [Hill et al., 2013; Wassmann, 2011]. They differ, however, with regard to select internal physical forcings and estimated rates of biological processes. The Chukchi Sea is an extremely productive inflow shelf ecosystem [Carmack and Wassmann, 2006; Carmack et al., 2006], where Pacific water entering the pan-Arctic region from Bering Strait is modified by some of the highest rates of primary production in Arctic surface waters [Hill et al., 2013; Mathis et al., 2009; Popova et al., 2010] and extensive benthic organic matter remineralization [Grebmeier et al., 2006] prior to advecting into the adjacent Beaufort Sea and Canada Basin [Anderson et al., 2013, 2010; Danielson et al., 2014]. In contrast to the Chukchi, the Beaufort Sea contains narrow interior shelves [Carmack and Wassmann, 2006; Carmack et al., 2006] that experience episodic wind-driven upwelling [Pickart et al., 2011; *Pickart et al.*, 2013] and a significant freshwater source from the Mackenzie River (Figure 1), which is the paramount contributor of sediment [*Holmes et al.*, 2002] and a major supplier of terrestrially derived dissolved inorganic carbon [*Tank et al.*, 2012] to the Arctic Basin. Rates of primary production are reduced in the Beaufort Sea relative to the Chukchi [*Carmack and Wassmann*, 2006; *Codispoti et al.*, 2013; *Popova et al.*, 2010], although phytoplankton blooms under sea ice are likely important for the ecosystems in both subregions [*Arrigo et al.*, 2012; *Shadwick et al.*, 2011] and their contribution to annual estimates of primary productivity has been difficult to constrain. The combination of cold and productive surface water sets the stage for potentially large and persistent CO₂ disequilibria between the western Arctic coastal ocean sea surface and the overlaying atmosphere.

Based on either measurements of sea-air pCO₂ difference combined with established wind speed-dependent parameterizations of the gas transfer rate or directly measured sea-air CO₂ fluxes using a micrometeorological technique known as the eddy correlation method [McGillis et al., 2001; Wanninkhof and McGillis, 1999], all studies conducted to date within the western Arctic coastal ocean that compute mean annual fluxes report net uptake of atmospheric CO₂, albeit with substantial variability both within and across subregions. In general, the Chukchi Sea is thought to be a massive sink for atmospheric CO₂ with mean annual flux estimates ranging between -10 and -40 mmol CO₂ m⁻² d⁻¹ (negative fluxes = atmospheric CO₂ uptake; positive fluxes = CO₂ outgassing to the atmosphere) [Bates, 2006; Gao et al., 2012; Semiletov et al., 2007], while estimates for the Beaufort Sea are more moderate with values between -0.3 and -10 mmol CO₂ m⁻² d⁻¹ [Else et al., 2013a; Mucci et al., 2010; Shadwick et al., 2011]. Annual mean fluxes from carbon mass balance assessments are basin-scale analyses; however, these also point to large atmospheric CO₂ uptake in the region [Anderson and Kaltin, 2001; MacGilchrist et al., 2014]. Previous studies have not assessed mean annual sea-air CO2 flux over the entire western Arctic coastal ocean. Combining published and unpublished data sets, which have grown considerably in recent years, provides the opportunity to assess net exchange over this broad region. The pertinent questions related to western Arctic coastal ocean sea-air CO₂ exchange then become (1) what is the annual mean flux for the entire region, (2) what is the contribution of this exchange to global estimates of coastal ocean CO_2 uptake, and (3) how may we expect this contribution to change under conditions of shrinking sea ice cover in this rapidly warming region [Overland and Wang, 2013]? The general assumption has been that reduced sea ice cover equates to enhanced exchange of CO₂ with the overlaying atmosphere owing to the larger areas of open water [Parmentier et al., 2013]. Intrinsic to this view is the assumption that sea ice acts as a barrier to CO₂ exchange between the sea surface and atmosphere [Bates et al., 2011; Bates et al., 2006; Stephens and Keeling, 2000]. There are well-established arguments both for enhanced atmospheric CO₂ uptake due to sea ice loss [Arrigo et al., 2010; Bates, 2006; Bates and Mathis, 2009; Bates et al., 2006] and against any increase in atmospheric CO₂ exchange [Cai et al., 2010; Else et al., 2013b], as well as arguments for a more active role from sea ice in the transfer of CO_2 between the sea surface and the atmosphere [Else et al., 2011; Loose et al., 2011, 2014; Miller et al., 2011; Moreau et al., 2015; Semiletov et al., 2004]. In this analysis, we treat sea ice as an imperfect barrier for sea-air CO₂ flux using a simple approach prescribed by Takahashi et al. [2009] and compute mean annual exchanges over the broad western Arctic coastal ocean using an extensive compilation of published and unpublished surface seawater pCO_2 data collected from 2003 to 2014 (Table 1). We provide answers for the three questions listed above with careful consideration of the caveats involved.

2. Data Sets and Calculations

Climatologies of western Arctic coastal ocean sea-air CO_2 flux with and without the presence of sea ice were computed following steps outlined in the schematic shown in Figure 2. To conduct this analysis and build flux climatologies, measurements made from 2003 through 2014 were acquired from publicly available repositories (e.g., SOCAT; http://www.socat.info) and from a number of contributors spanning more than five nations (Table 1). A total of nearly 600,000 surface seawater pCO_2 data points were compiled. These data were either directly measured with nondispersive infrared analyses of CO_2 content in equilibrated headspace gas by underway measurement systems or calculated from discrete measurements of dissolved inorganic carbon (DIC) and total alkalinity (TA). The temporal and spatial distribution of the compiled data from each contributor is shown in the supporting information Figure S1. It is important to note that within this extensive data compilation, only one contribution contained measurements spanning the course of a full year

as Defined in Figure 1		
Contributor	Country of Origin	# of Measurements
1. T. Takahashi (underway)	USA	92013
2. SOCAT V2 (underway)	Multiple	3206
3. B. Else and T. Papakyriakou (underway)	Canada	473509
4. M. DeGrandpre and F. Islam (underway)	USA	6092
5. WJ. Cai and B. Chen (underway)	USA/China	5729
6. M. Yamamoto-Kawai (underway)	Japan	3889
7. N. Bates (discrete)	Bermuda	380
8. L. A. Miller (discrete)	Canada	887
9. M. Yamamoto-Kawai (discrete)	Japan/Canada	70
TOTAL		585775

 Table 1.
 The Number of Measurements From Each Contributor That Was Made Within the Western Arctic Coastal Ocean as Defined in Figure 1

[*Else et al.*, 2012]. The underway and discrete data and their associated calculations are described in detail below. All data sets contained sea surface temperature (SST) and salinity measurements, on the Practical Salinity Scale (PSS-78, dimensionless), in addition to CO₂ system parameters.

2.1. Underway Seawater pCO₂ Data

Directly measured pCO_2 data were provided by each contributor with the exception of the SOCAT data set that distributes CO_2 fugacity (fCO_2). Owing to most of the data contributions being pCO_2 and not fCO_2, and that fCO_2 is derived using the virial coefficients for CO_2 [*Dickson et al.*, 2007; *Weiss*, 1974], we chose to conduct our analysis using pCO_2 . Note that the difference between partial pressure and fugacity is small for CO_2 and on the order of 0.3% (http://cdiac.ornl.gov/oceans/ndp_047/backdisc047.html). Furthermore, the difference mostly cancels when the sea-air pCO_2 difference is computed. The SOCAT data were converted back to pCO_2 by first reverting fCO_2 at SST to fCO_2 at equilibrator temperature, which was included in the provided data set, using SST and equilibrator temperatures with the relationship for pCO_2 temperature dependence from *Takahashi et al.* [1993], then correcting by the virial coefficients to obtain pCO_2 at equilibrator temperature using equation (5) in *Sutton et al.* [2014], and finally calculating pCO_2 at SST using the temperature dependence. The protocols for these calculations are described in detail elsewhere [*Pierrot et al.,* 2009; *Sutton et al.,* 2014]. The compilation of directly measured pCO_2 was next combined with contributions of pCO_2 calculated from surface measurements of DIC and TA.



Figure 2. Analysis scheme used to build climatologies of sea-air CO₂ flux for the western Arctic coastal ocean.

Table 2. RMSEs (μ atm) Between Directly Measured pCO_2 and pCO_2 Calculated From Discrete Measurements of TA and DIC Using Six Common Sets of Equilibrium Constants With a Customized MathWorks MATLAB Version of CO2SYS^a

Millero et al. [2002]	9.3
Millero et al. [2006]	9.0
Millero [2010]	9.2
Mehrbach refit by Dickson and Millero	
Lueker et al. [2000]	9.9
Roy et al. [1993]	12.6

^aA lower RMSE value indicates closer agreement between directly measured and calculated pCO_2 . Time-matched measured and calculated pCO_2 data used in this comparison were collected during the U.S. Coast Guard Cutter Healy expedition in October 2012 (n = 50). The equilibrium constants of *Millero et al.* [2006] produced the lowest-RMSE value and so were applied to all discrete data acquired for this analysis to calculate seawater pCO_2 values. RMSEs, root-mean-square errors; DIC, dissolved inorganic carbon; TA, total alkalinity.

2.2. Discrete Seawater pCO₂ Data

A total of 1337 discrete measurements of DIC and TA collected from depths <6 m were ingested into this analysis, including measurements from a recently published data set for the Canadian Arctic [*Giesbrecht et al.*, 2014]. All discrete measurements were collected using standard carbonate system sampling protocols [*Dickson et al.*, 2007]. Values of pCO_2 were calculated from these measurements using a modified MathWorks MATLAB version of CO2SYS [*van Heuven et al.*, 2011] with the "ideal" carbonate dissociation constants for

this setting selected based on the approach described below. Following the selection of ideal constants, calculated and directly measured pCO_2 data sets were combined and trimmed to remove measurements made outside of the defined western Arctic coastal ocean domain (Figure 1).

2.2.1. Selection of Ideal Equilibrium Constants for Western Arctic Coastal Ocean

During an October 2012 cruise aboard the U.S. Coast Guard Cutter (USCGC) Healy operating in the western Arctic coastal ocean, discrete surface water DIC, TA, SST, and salinity measurements were made while in transit along with directly measured pCO_2 , SST, and salinity data collected by the Lamont-Doherty Earth Observatory (LDEO) Carbon Dioxide Research Group underway pCO2 system (http://www.ldeo.columbia. edu/res/pi/CO2/). The pCO₂ data from the LDEO system were time matched with the DIC and TA measurements, and pCO₂ was then calculated from the discrete DIC, TA, SST, and salinity data, excluding nutrients, using a variety of equilibrium constants for the dissociation of carbonic acid [Dickson and Millero, 1987; Lueker et al., 2000; Millero, 2010; Millero et al., 2006, 2002; Roy et al., 1993]. The calculated pCO₂ values from each set of equilibrium constants were then compared with the directly measured underway LDEO pCO_2 data. In general, calculated pCO₂ from all pairs of equilibrium constants tested tracked directly measured pCO_2 to within approximately ±15 µatm. However, at times calculated pCO_2 deviated by as much as 70 µatm from the directly measured values, similar to what was seen during comparisons made along ocean basin transects [Wanninkhof et al., 1999]. To determine the ideal set of equilibrium dissociation constants, rootmean-square errors (RMSEs) were computed for only periods when calculated pCO_2 from all sets of constants closely tracked directly measured pCO₂ (Table 2). The ideal set of equilibrium constants were selected based on the lowest-RMSE value.

2.3. ΔpCO_2 and Solubility Grids

Sea-air pCO_2 difference (ΔpCO_2), an integral quantity for the calculation of sea-air CO_2 flux, was calculated in the same manner as in *Evans and Mathis* [2013] and *Cross et al.* [2014]. As was the case in those studies, we do not correct the compiled pCO_2 data to a reference year as is typically done for open ocean climatologies [*Takahashi et al.*, 2002, 2009]. Instead, we reconstructed atmospheric pCO_2 with the National Oceanic and Atmospheric Administration (NOAA) Earth System Research Laboratory (ESRL) Greenhouse Gas Marine Boundary Layer (MBL) Reference (http://www.esrl.noaa.gov/gmd/ccgg/mbl/) and coupled this reconstruction with the surface seawater pCO_2 data. This approach intrinsically captures secular increases in both atmospheric and surface seawater pCO_2 records [*Evans and Mathis*, 2013] but does not account for interannual variability associated with the El Niño–Southern Oscillation or other similar perturbations that may be present in the data. The MBL reference is a global, zonally averaged record of weekly CO_2 mole fractions in dry air (xCO_2). The MBL data were extracted for the latitudinal range of the western Arctic coastal ocean (Figure 1) and averaged into monthly values. Monthly atmospheric xCO_2 data were corrected to pCO_2 using the following equation:

 $pCO_{2(Monthly)} = xCO_{2(Monthly)} \times (P_{sl(NCEPMonthly)} - P_{w(NCEPMonthly)}),$

where $P_{sl(NCEP monthly)}$ and $P_{w(NCEP monthly)}$ are the sea level and water vapor pressures, respectively. Sea level pressure was provided directly from the National Center for Environmental Prediction (NCEP)-Department of Energy (DOE) Reanalysis 2 product (http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis2.html). The sea level pressure field is a monthly product with 2.5° latitude × 2.5° longitude spatial resolution. Water vapor pressures are assumed to be saturated and were computed from NCEP-DOE Reanalysis 2 1000 hPa (surface) air temperatures using the relationship described by *Buck* [1981]. Values of ΔpCO_2 were then calculated by subtracting the time-matched atmospheric pCO_2 data from the surface seawater pCO_2 measurements collected within each 2.5° latitude × 2.5° longitude pixel. Following the calculation of ΔpCO_2 , CO₂ solubility was computed using the SST and salinity data with the relationship described by *Weiss* [1974]. Averages and standard deviations of monthly ΔpCO_2 , CO₂ solubility, SST, and salinity data within 0.2° latitude × 0.5° longitude pixels were then calculated to construct gridded monthly data for the western Arctic coastal ocean.

2.4. Wind Speed Second Moments

Using u and v 10 m wind velocities from 2003 through 2014 extracted from the NCEP North American Regional Reanalysis product (NARR; http://www.esrl.noaa.gov/psd/data/gridded/data.narr.monolevel.html), we calculated monthly averages of the daily second moment of the wind speed (<wind speed²>). This time period tightly brackets the compiled surface seawater pCO_2 data set. NARR is a North American extension of the global NCEP products that is based on both actual measurements and model projections [Mesinger et al., 2006]. The NARR data are provided by ESRL on a Lambert Conformal grid with approximately 32 km resolution, which was interpolated here to a uniform 0.25° latitude × 0.25° longitude grid. From the near decade of daily wind speed observations, we calculated the second moment of the wind speed for each day. Daily second moments were then regridded to 0.2° latitude \times 0.5° longitude resolution to match the ΔpCO_2 grid and then averaged into monthly values. Second moments were calculated as opposed to daily averages because short-term (daily) variability in the winds is retained leading to higher gas transfer rates during periods of greater wind speed variability [Wanninkhof et al., 2004]. Sea-air CO₂ fluxes calculated with gas exchanges estimated from simple time-averaged wind speeds typically utilize enhancement factors to account for short-term variability [Ho et al., 2011; Jiang et al., 2008; Wanninkhof et al., 2004], which, if not employed would underestimate the magnitude of the flux. Employing second moments of the wind speed removes the need for enhancement factors.

2.5. Sea Ice Data

Measurements of sea ice concentration (% cover) made by the Special Sensor Microwave Imager (SSM/I) and processed by the National Snow and Ice Data Center were provided by the French Research Institute for Exploration of the Sea (IFREMER) at daily and 12 km resolution (http://cersat.ifremer.fr/oceanography-from-space/our-domains-of-research/sea-ice). These data were obtained for the period from 2003 through 2014 to remain consistent with the surface seawater pCO_2 data set. Three representations of the sea ice concentration were used for the western Arctic coastal ocean: (1) a monthly climatology of sea ice concentrations built from 2003 to 2014 satellite observations, (2) sea ice concentrations from 2003 alone, and (3) sea ice concentrations from 2014 alone. All representations of sea ice concentration were regrided to 0.2° latitude $\times 0.5^{\circ}$ longitude resolution to match the ΔpCO_2 grid. The climatology of sea ice concentration was built by averaging the daily ice data for each calendar day of the year and then averaging those daily means into monthly values. The 2003 and 2014 daily data were also averaged into monthly values.

2.6. Sea-Air CO₂ Flux Calculations

The following equation was used to calculate the monthly sea-air CO_2 fluxes for each 0.2° latitude $\times 0.5^{\circ}$ longitude pixel:

Sea – AirCO₂Flux =
$$k_{SST} \times K_{CO2} \times \Delta pCO_2$$
,

where k_{SST} is the gas transfer rate at SST (m d⁻¹) and K_{CO2} is the CO₂ solubility (mmol m⁻³ atm⁻¹). k_{SST} is calculated from k_{660} , the gas transfer rate at a Schmidt number of 660 computed here using the wind speeddependent parameterization of *Wanninkhof* [2014] with the monthly wind speed second moments, adjusted to in situ SST conditions with the Schmidt number correction. We did not correct k_{SST} for salinity, because the difference between k_{SST} in freshwater and at a salinity of 35 is on the order of a few percent [*Evans et al.*, 2011]. Monthly sea-air CO₂ fluxes for each pixel were then adjusted according to the sea ice concentration

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Figure 3. Monthly climatology of ΔpCO_2 (µatm) computed using the grid shown in Figure 1 and the seawater data depicted in supporting information Figure S1 with reconstructions of atmospheric pCO_2 following *Evans and Mathis* [2013] and *Cross et al.* [2014].

present during the particular month and within the corresponding pixel. For this analysis, we consider sea ice to be an imperfect barrier for turbulent-driven gas exchange, such that ice-adjusted fluxes were calculated by

$$\mathsf{Sea} - \mathsf{AirCO}_2\mathsf{Flux}_{(i)} = \mathsf{Sea} - \mathsf{AirCO}_2\mathsf{Flux}_{(i)} \times \frac{100 - \mathsf{Sea} \quad \mathsf{lce} \quad \mathsf{Concentration}}{100}.$$

Sea ice concentrations used to adjust the fluxes were the three representations of the ice cover discussed above. The satellite data cannot resolve fine-scale cracks and lead in the ice that can allow for rapid gas exchange [*Else et al.*, 2013a]; therefore, we employ the technique used by *Takahashi et al.* [2009] for Antarctic sea ice where a total open water area of 10% is assumed for all cases where satellite observations show > 90% ice cover. In this way, 10% of the open water flux persists for > 90% ice cover conditions where SSM/I satellite data fail to resolve fine-scale structure in the dynamic winter icescape. Monthly area-weighted fluxes were then computed for the entire western Arctic coastal ocean domain and the subregional seas, the Chukchi Sea and the Beaufort Sea, using the calculated fluxes with and without the presence of sea ice and the equation:

$$\mathsf{Sea} - \mathsf{AirCO}_2\mathsf{Flux}_{\mathsf{area-weighted}} = \sum_{i=1}^n \bigl(\mathsf{Sea} - \mathsf{AirCO}_2\mathsf{Flux}_{(1)} \times A_{(i)}\bigr) / \sum_{i=1}^n A_{(i)},$$

where *i* represents the individual pixel and *A* is the area of the corresponding pixel. Pixel area varies with latitude, ranging from 509 km² at 65°N to 316 km² at 75°N. Annual mean sea-air CO₂ fluxes with and without the presence of sea ice were calculated by averaging monthly area-weighted fluxes. Uncertainty around the annual mean fluxes was estimated as the sum of uncertainties associated with the monthly mean Δp CO₂ and the gas transfer rate. Monthly Δp CO₂ uncertainty was calculated using the standard deviations of Δp CO₂ in each pixel. The difference in sea-air CO₂ fluxes recomputed using the Δp CO₂ standard deviation both added

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Figure 4. Climatology of monthly average second moment of the wind speed (<Wind Speed²>; m² s⁻²) computed from daily NCEP North American Regional Reanalysis (NARR) 1000 hPa level meridional and zonal wind components from 2003 to 2014.

to and subtracted from the monthly ΔpCO_2 provided a measure of uncertainty associated with the mean ΔpCO_2 . This term was added to a 20% uncertainty in the wind speed-dependent parameterization of the gas transfer rate reported by *Wanninkhof* [2014]. The annual mean sea-air CO₂ fluxes and their respective uncertainties were used to estimate annual mean carbon uptake for the western Arctic coastal ocean.

3. Results

Sea-air CO₂ fluxes were computed here using an extensive data set of compiled surface seawater pCO₂ that was either directly measured using shipboard underway systems or calculated from discrete DIC and TA measurements. An important point revealed by the comparison of measured and calculated pCO₂ data was that the tested equilibrium constants all produced values not too disparate from directly measured pCO₂ in most instances (not shown); however, the equilibrium constants from *Millero et al.* [2006] produced the lowest-RMSE value (Table 2) suggesting that these constants are ideal for the western Arctic coastal ocean. While this ideal set of equilibrium constants did not produce a RMSE value largely different from the value derived from data calculated with the *Lueker et al.* [2000] constants recommended for use as best practice [*Dickson et al.*, 2007], other equilibrium constants produced a notably larger error (Table 2). The results of combining directly measured values and calculated pCO₂ using the ideal constants produced a near 600,000-measurement data set of western Arctic coastal ocean pCO₂ (Table 1).

3.1. ∆*p*CO₂

Average monthly ΔpCO_2 for December through April was based on data collected over a limited number of pixels during the 2007–2008 International Polar Year Circumpolar Flaw Lead System Study in the Beaufort

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Figure 5. Climatology of monthly sea ice concentration (% Cover) computed from daily 12 km Special Sensor Microwave Imager (SSM/I) data provided by the French Research Institute for Exploration of the Sea (IFREMER) (http://cersat.ifremer.fr/oceanography-from-space/our-domains-of-researc2h/sea-ice) collected from 2003 to 2014.

Sea [*Else et al.*, 2012]. Pixels in these months all had ΔpCO_2 ranging from $-60 \mu atm$ to near 0 (Figure 3). During May, more pixels were occupied in the Beaufort Sea as well as some in the Chukchi Sea by an early 2014 USCGC Healy expedition in the region. Instances of positive and negative $\Delta p CO_2$ were evident in both seas during this month, but 60% of pixels contained undersaturated seawater pCO_2 with values averaging $-80 \,\mu$ atm. At this time the largest area of oversaturation, with levels up to +220 µatm, was in the Chukchi Sea beneath sea ice cover (supporting information Figure S2). Only one instance of positive ΔpCO_2 was seen in June near the Mackenzie Shelf (Figures 1 and 3), and average ΔpCO_2 in our study was $-108 \,\mu$ atm. July had select areas of positive ΔpCO_2 in the southern Chukchi Sea and in portions of the Beaufort Sea but predominantly negative ΔpCO_2 with values reaching $-277 \,\mu$ atm near the divide between the two subregions (the head of Barrow Canyon; Figure 1) and extending broadly over the northern Chukchi Sea. This area of greatest undersaturation with respect to atmospheric CO₂ persisted through August, with the Beaufort Sea containing more moderate levels of undersaturation and instances of $\Delta p CO_2$ near and above zero. The region of greatest negative Δp CO₂ was still strongly evident in September and October; however, the spatial extent of this area had abated during these 2 months from the apparent August peak in coverage. Moderate undersaturation with levels averaging -60 µatm was widely distributed over the western Arctic coastal ocean during these 2 months, with instances of positive Δp CO₂ in select areas of the Chukchi and Beaufort Seas. The extent of November data coverage was limited and similar to that of May, but in comparison to other months, a higher percentage of pixels was near or above saturation with respect to atmospheric CO₂. Sixty percent of the 263 November pixels had negative ΔpCO_2 averaging -45 µatm, and 25% of pixels contained ΔpCO_2 above +5 µatm. Areas in the western Arctic coastal ocean where positive November ΔpCO_2 pixels occurred were the Mackenzie Shelf and Amundsen Gulf areas of the Beaufort Sea and in the southern portion of the Chukchi Sea (Figures 1 and 3).

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Figure 6. Monthly climatology of sea-air CO₂ fluxes in the presence of sea ice (mmol m⁻² d⁻¹; Sea-Air CO₂ Flux_{sea ice clim}) using the 2003–2014 sea ice climatology.

3.2. <Wind Speed²> and Sea Ice

Monthly averages of the daily second moments of the wind speed (<wind speed²>) showed a seasonal amplification from broadly lower <wind speed²> values from January through June to higher values during the second half of the year (Figure 4). During the months of lower <wind speed²>, select areas had values up to $25 \text{ m}^2 \text{ s}^{-2}$. <wind speed²> intensified in the Chukchi Sea in July, with higher levels becoming more wide-spread over the western Arctic coastal ocean by August. September and October had broad areas of <wind speed²> above 40 m² s⁻². High <wind speed²> values persisted in the Chukchi Sea in November and then declined by December (Figure 4).

The climatology of sea ice concentration showed above 90% cover over the entire region until May when select areas of open water (sea ice concentration < 10%) began to develop (Figure 5). During June, the open water regions grew to cover larger areas in both the Chukchi and Beaufort seas. The area of open water increased to a maximum in September. A rapid decrease in the expanse of open water occurred from October to November, at which time only the central and southern Chukchi Sea had low sea ice cover. By December, the southern Chukchi Sea had the lowest sea ice cover with levels near 60% in the climatology. It is important to note that the general seasonal timing of sea ice recession and expansion was similar between the 2003 and 2014 sea ice data sets, albeit with differences in spatial patterns for most months (supporting information Figures 2 and 3).

3.3. Sea-Air CO₂ Fluxes

Sea-air CO₂ fluxes adjusted for sea ice concentrations maintained near zero levels from January until May and into June when areas of open water began to develop (Figures 5 and 6). Fluxes during these months averaged $-3 \text{ mmol m}^{-2} \text{ d}^{-1}$. With expanded measurements (supporting information Figure S1) and larger regions of

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Figure 7. Monthly climatology of sea-air CO₂ fluxes without the presence of sea ice (mmol $m^{-2} d^{-1}$).

open water in July (Figure 5), widespread areas of atmospheric CO₂ uptake were apparent with an average flux also near $-3 \text{ mmol m}^{-2} \text{ d}^{-1}$. Rates of atmospheric CO₂ uptake strengthened in August, with maximal influx reaching $-20 \text{ mmol m}^{-2} \text{ d}^{-1}$. Many instances of near-zero sea-air CO₂ exchange (dark grey points; Figure 6) were also apparent during this month as were a few pixels showing CO₂ efflux to the atmosphere in the Beaufort Sea. September and October both contained the greatest spatial extent of negative fluxes, as these were the months with the highest degree of open water (Figure 5). Maximal influxes were near $-25 \text{ mmol m}^{-2} \text{ d}^{-1}$ with averages near $-7 \text{ mmol m}^{-2} \text{ d}^{-1}$ during both months. The large uptake during September and October (Figure 6) was due to the trifecta of strong and widespread undersaturation of surface seawater *p*CO₂ (Figure 3), strong winds (Figure 4) resulting large gas transfer rates, and the peak in open water coverage (Figure 5). Instances of CO₂ efflux were limited to the southern Chukchi Sea in September and October, and in few Beaufort Sea pixels in October. Average sea ice adjusted November sea-air CO₂ fluxes were slightly above zero (+0.2 mmol m⁻² d⁻¹), with many instances of atmospheric CO₂ uptake still occurring. Some CO₂ efflux regions were evident in the southern Chukchi Sea and to a lesser extent in the Beaufort Sea. By December, sea ice-adjusted fluxes had returned to zero.

Neglecting the sea ice concentration adjustment of the sea-air CO₂ fluxes provides a sense of two possibilities: (1) what the "unrealized" atmospheric CO₂ exchange may be in the future under more limited sea ice cover scenarios, and (2) what the sea-air CO₂ fluxes may be if our treatment of sea ice cover as an imperfect barrier for gas exchange is inaccurate. Differences were evident between fluxes with and without the presence of sea ice for every month sea ice was present in the western Arctic coastal ocean (Figure 7). Near-zero winter month fluxes become average CO₂ influxes without the presence of sea ice. Largely, wintertime no sea ice fluxes are low and negative because of the combination of marginally negative ΔpCO_2 (Figure 3) and low <wind speed²> values (Figure 4); however these low-flux values will impact area-weighted fluxes and carbon



Figure 8. Monthly area-weighted sea-air CO_2 fluxes (mmol m⁻² d⁻¹) for the western Arctic coastal ocean calculated from flux climatologies with no sea ice (black), 2003 sea ice concentrations (green), 2014 sea ice concentrations (red), and the 2003–2014 sea ice climatology (blue).

uptake estimates. The flux adjustment for sea ice concentration may be most important during the months of rapid seasonal shifts in sea ice cover (i.e., May and November, Figure 5). During these months, potentially large CO_2 efflux to the atmosphere is being suppressed in May, whereas in November atmospheric CO_2 uptake would still occur in northern portions of the Chukchi Sea if not for our assumption regarding the role of sea ice cover.

Area-weighted mean sea-air CO₂ fluxes clearly reveal the degree of sensitivity for the exchanges calculated with and without the presence of sea ice (Figures 6 and 7). Maximal winter month, area-weighted, no sea ice fluxes for the western Arctic coastal ocean would be nearly half of the values seen during peak uptake months, if not for our treatment of sea ice as an imperfect barrier to gas exchange (Figure 8). Monthly area-weighted fluxes with and without the presence sea ice were within 1 mmol m⁻² d⁻¹ of each other from June through November. Note that the calculation of sea ice-adjusted area-weighted fluxes was fairly insensitive to which sea ice cover representation was used (i.e., 2003, 2014, or the sea ice climatology). The sea ice adjustment reversed the sign of the fluxes during May and November, although in different directions. Area-weighted May fluxes would favor outgassing without sea ice, whereas the slight efflux in November would turn to uptake under more sea ice-free conditions. In general, the broad western Arctic coastal ocean acts as a persistent atmospheric CO₂ sink throughout the year with maximal uptake from August through October (Figure 8).

Total carbon exchanges were calculated by scaling the annual mean sea-air CO₂ fluxes, estimated by averaging the monthly area-weighted fluxes (Figure 8), by the days of the year, the molar mass of carbon, and the western Arctic coastal ocean surface area $(1.2 \times 10^{12} \text{ m}^2; \text{ Table 3})$. Sea ice-adjusted carbon uptake based on this analysis of nearly 600,000 data points was $10.9 \text{ Tg C yr}^{-1}$ ($1 \text{ Tg} = 10^{12} \text{ g}$). Neglecting the sea ice adjustment of the fluxes increased the carbon uptake by ~42% to $15.4 \text{ Tg C yr}^{-1}$. Very little difference was seen between the sea ice-adjusted annual mean fluxes and the carbon uptake estimates using sea ice cover data from 2003, 2014, or the sea ice climatology (Table 3). Uncertainty in the annual mean sea-air CO₂ fluxes and the carbon uptake estimates was nearly 50% in all cases (Table 3).

4. Discussion

4.1. The Western Arctic Coastal Ocean Contribution to Atmospheric Carbon Uptake

Areas of the western Arctic coastal ocean have been marked as globally some of the strongest coastal ocean sink regions for atmospheric CO₂ [*Bates*, 2006; *Laruelle et al.*, 2014] due to increased CO₂ solubility in

Table 3. Annual Mean Sea-Air CO₂ Flux (mmol $m^{-2} d^{-1}$) and Carbon Uptake (Tg C yr⁻¹) for the Western Arctic Coastal Ocean Calculated With No Sea Ice, 2003 and 2014 Sea Ice Concentrations (Supporting Information Figures S2 and S3), and the 2003–2014 Sea Ice Climatology (Figure 5)^a

	Annual Mean Sea-Air CO ₂ Flux (mmol CO ₂ m ^{-2} d ^{-1})	Annual Mean Carbon Uptake (Tg C yr ⁻¹)
No ice	-2.9 ± 1.5	15.4 ± 8.1
2003 sea ice	-2.1 ± 1.1	11.1 ± 5.8
2014 sea ice	-2.0 ± 1.1	10.8 ± 5.7
Sea ice climatology	-2.0 ± 1.1	10.9 ± 5.7

^aUncertainties in the fluxes and carbon uptake estimates are shown and calculated based on combining uncertainty around the mean monthly Δ*p*CO₂ and the gas transfer parameterization [*Wanninkhof*, 2014]. Note the minimal difference in sea-air CO₂ flux and carbon uptake using any of the three ice products. cold waters in combination with high rates of primary production that draw down surface seawater pCO_2 to low levels. Historically, limited data coverage has made constraining this exchange difficult and reliant on measurements from few cruises or single research programs. Now, in this contribution, nearly 600,000 data points collected over an ~11 year time frame have been analyzed with the aim to better constrain the flux in this key coastal region. Constraining atmospheric CO_2 exchange is an important step toward the deconvolution of **Table 4.** Monthly Area-Weighted Sea-Air CO_2 Flux (mmol m⁻² d⁻¹) Calculated With and Without the 2003–2014 Sea Ice Climatology for the Chukchi Sea and Beaufort Sea Subregions of the Western Arctic Coastal Ocean Shown in Figure 1^a

	Chukcki Sea Sea Ice	Beaufort Sea Sea Ice	Chukcki Sea No Sea Ice	Beaufort Sea No Sea Ice
JAN	NaN	-0.3	NaN	-2.7
FEB	NaN	-0.1	NaN	-1.3
MAR	NaN	-0.1	NaN	-0.7
APR	NaN	-0.1	NaN	-0.8
MAY	0.3	-0.3	3.2	-2.4
JUN	-4.8	-0.3	-6.1	-2.4
JUL	-4.3	-1.0	-6.0	-1.6
AUG	-11.5	-1.5	-11.8	-1.7
SEP	-10.5	-3.9	-10.6	-4.2
OCT	-10.4	-4.0	-10.5	-4.6
NOV	0.5	-0.1	0.0	-0.6
DEC	NaN	-0.2	NaN	-2.2
Annual mean sea-air CO ₂ flux (mmol CO ₂ m ^{-2} d ^{-1})	-3.5	-1.0	-4.1	-2.1
Annual mean carbon uptake (Tg C yr $^{-1}$)	4.4	4.0	5.3	8.5

^aAnnual mean sea-air CO₂ fluxes are shown for each region. To estimate the annual mean sea-air CO₂ flux for the Chukchi Sea, it was assumed that fluxes there are similar to those in the Beaufort Sea during the ice-covered months of the year (December–April). Annual mean sea-air CO₂ fluxes were scaled up to carbon uptake estimates using the surface areas for each region described in Figure 1.

natural and anthropogenic components of net sea-air CO₂ flux, and then subsequently detailing the implications of the anthropogenic component for the shifting biogeochemical seascape [Robbins et al., 2013; Yamamoto-Kawai et al., 2011]. The analysis we conducted clearly addressed the first question posed above regarding the magnitude of annual mean CO₂ exchange and carbon uptake in this region (Table 3). In the absence of a replicate global-scale analysis using the same coastal ocean definition and approach as utilized here, defining the contribution of western Arctic coastal ocean carbon uptake to global coastal ocean CO_2 exchange then becomes an accounting exercise using previously published estimates. The most recent analysis estimated carbon uptake at 185 ± 46 Tg C yr⁻¹ for coastal oceans globally [*Laruelle et al.*, 2014], with Arctic shelf seas contributing 40% or 71 Tq C yr⁻¹. These values from Laruelle et al. [2014] are noteworthy for a number of reasons: (1) the analysis differs from previous global syntheses that rely on compilations of local estimates [Cai et al., 2006; Dai et al., 2013; Laruelle et al., 2010] but rather uses the SOCAT data set also employed here albeit with a very different, globally oriented integration strategy; and (2) as is the case with recent global estimates of the coastal ocean sink term, the Arctic coastal ocean contribution as a whole decreased in magnitude from 100 to 71 Tg Cyr⁻¹ [*Cai*, 2011; *Laruelle et al.*, 2014]. Given the 10.9 Tg Cyr⁻¹ of sea ice-adjusted carbon uptake reported here (Table 3), the reduction in estimates of polar shelf sea CO_2 exchange as a whole implies a greater contribution, increasing from 11 to 15%, from the western Arctic coastal ocean. On a global scale, this region is nearly equivalent in its contribution to coastal ocean surface area and carbon uptake, making up nearly 5% of both using the estimates from Laruelle et al. [2014]. This inherently implies that sea ice corrected annual mean sea-air CO₂ flux for the western Arctic coastal ocean is close to the global average coastal ocean flux.

The analysis presented here is the first to attempt to integrate over the entire western Arctic coastal ocean, but by reanalyzing the compiled data set parsed into the two subregional seas (Figure 1), some large differences become apparent that also suggest significant reduction in sink terms at the subregional scale. In order to compute annual mean carbon uptake for each subregional sea, we assumed that winter fluxes were similar between the two areas, such that December through April fluxes from the temporally well-resolved Beaufort Sea could be applied to the Chukchi Sea (Figure 6). In this way, annual mean sea-air CO₂ fluxes and carbon uptake estimates could be calculated for both regions (Table 4). The landmass-corrected surface areas of the polygons defining each subregional sea were 2.9×10^{11} m² for the Chukchi and 9.2×10^{11} m² for the Beaufort (Figure 1). These areas scaled the mean annual fluxes to carbon uptake values near 4 Tg C yr⁻¹ for both regions (Table 4). Note that the sum of the uptake values from each sea does not exactly equal the carbon uptake for the overall western Arctic coastal ocean (Table 3). Area-weighted fluxes calculated using the different data distributions between the subregional areas and the entire study region resulted in mismatched estimates of net exchange. This discrepancy is within the uncertainty of the flux reported here

 $(5.7 \text{ Tg C yr}^{-1}; \text{ Table 3})$ and arises because no interpolation or extrapolation was used to fill empty pixels for the calculation of annual carbon uptake for the entire study area or the subregions. This is an important point because interpolation/extrapolation in dynamic coastal settings may resolve this mismatch but at the risk of inaccurately populating vacant pixels. The estimates of carbon uptake reported here are based solely on the data set we compiled and not interpolated/extrapolated values.

The largest difference between these uptake estimates and those previously reported within the western Arctic coastal ocean were for the Chukchi Sea. Bates [2006] estimated a massive 40 Tg carbon uptake for the Chukchi Sea per year, vastly different from that reported here (Table 4). There are two key reasons for this striking difference: (1) contrasting maximal exchanges based on single-cruise measurements versus synthesized observations from multiple cruises with greater spatial coverage over 11 years and (2) different surface areal definitions. By rescaling the monthly carbon uptakes from Table 1 in Bates [2006] using the surface area he applied for the Chukchi Sea, maximal fluxes are more than double what we report here for the period of annual peak uptake (Figure 6). The difference in maximal fluxes from what we report is the result of using single-cruise observations of seawater pCO_2 combined with high gas transfer rates from instantaneous wind speeds, which coupled with a sparse data set, create biases that lead to gross overestimations of monthly net exchange. In contrast, our more modest maximal monthly fluxes are based on surface seawater pCO_2 data collected over many cruises (Table 1, supporting information Figure S1) combined with gas transfer rates computed using monthly <wind speed²> values, in addition to resolving more months of the year (7 months here as opposed to 3 in Bates [2006]) before relying on extrapolation. These strategies better enable the variability to be represented by not creating large biases, which then produces a more representative estimate of annual exchanges. Contrary to the Chukchi Sea analysis, our assessment of the Beaufort Sea mean flux was in closer agreement with previous estimates because that region is much better resolved relative to the Chukchi [Else et al., 2013a; Mucci et al., 2010; Shadwick et al., 2011]. The second factor that is responsible for at least half the discrepancy between the Chukchi Sea estimates is the areal definition. By applying the annual average sea-air CO₂ flux calculated using the rescaled monthly uptake estimates from Table 1 of Bates [2006] to the surface area we define for the Chukchi Sea, the high uptake reported by Bates [2006] becomes reduced by half. Areal definitions of coastal ocean settings have been a source of debate [Evans and Mathis, 2013; Liu et al., 2010], and the point stressed here is that uptake estimates for specific coastal areas will never align if differing coastal ocean areal definitions are continually used. Here we rely on the SOCAT coastal ocean definition [Bakker et al., 2014; Pfeil et al., 2013] that has been agreed upon by the international community, has been used in past regional analyses of sea-air CO₂ exchange [Evans and Mathis, 2013; Hales et al., 2008; Hales et al., 2012], and makes sense given the relationship to open ocean syntheses. The assessment presented here accounts for these two factors and reduces the Chukchi Sea sink term by nearly 70%.

4.2. Sea-Air CO₂ Flux Uncertainties

Through the analysis of calculated mean annual fluxes and addressing the western Arctic coastal ocean contribution to global coastal sea-air CO₂ exchange, we quantified sources of uncertainty associated with the monthly mean $\Delta p CO_2$ and the gas transfer rate, but there are two additional key and likely larger sources of uncertainty that are difficult to quantify: (1) undersampling in time and space across the western Arctic coastal ocean domain and (2) the multifaceted role of sea ice in sea-air CO₂ transfer. Takahashi et al. [2009] eloquently estimated the error due to undersampling in their global synthesis using differences between their SST data set and an extensive climatological record of global SST coupled with the relationship for pCO₂ temperature dependence [Takahashi et al., 1993]. By this approach, Takahashi et al. [2009] estimated a 20% uncertainty from undersampling in this global context. We made a similar calculation by comparing our monthly gridded SST data with objectively analyzed monthly SST climatologies from the World Ocean Atlas 2013 (WOA13) computed for the 2005 through 2012 time period and extracted for the western Arctic coastal ocean. The WOA13 data were 0.25° with nearly 5000 pixels per month in this domain. The mean bias in SST between our data set and the WOA13 climatologies was +0.53°C. Assuming the warm bias in our data reflects the undersampling bias in surface seawater pCO_2 , using the pCO_2 temperature dependence [Takahashi et al., 1993] we estimate that the mean surface water pCO_2 in our study (332 µatm) may be biased by +7.1 μ atm. Using a mean gas transfer coefficient ($k_{SST} \times K_{CO2}$) of 0.05 mmol m⁻² μ atm⁻¹ d⁻¹ calculated from our k_{SST} and K_{CO2} data sets, we estimate that the annual mean sea-air CO₂ flux for the western Arctic coastal ocean may be underestimated by 13% (-3.4 mmol m⁻² d⁻¹) due to undersampling. This corresponds



Figure 9. Standard deviation of November and December sea ice concentration (% Cover) using SSM/I data from 2003 through 2014.

in an underestimate in carbon uptake for the region of 0.7 Tg yr^{-1} . This estimate of undersampling bias is < 10% of the annual mean carbon uptake reported here, suggesting our analysis is fairly robust. However, the entire Arctic Ocean domain suffers from undersampling that affects even the extensive WOA13 climatologies. Below we discuss the time periods when undersampling potentially has the biggest impact on net CO₂ exchange as a way to target future measurement programs within these areas. The rapidly evolving science of sea ice gas exchange can then be factored into this discussion as it relates to time periods when data coverage suffers most, with a final hypothesis regarding how the contribution of western Arctic coastal ocean carbon uptake will change under conditions of reduced sea ice cover.

4.2.1. Late Season Efflux

The high rates of primary production in western Arctic coastal ocean surface waters, which are partly responsible for the persistent undersaturated sea surface pCO_2 conditions, are coincident with high organic matter sedimentation to continental shelf bottom waters, where this organic matter is respired back into CO₂ by an active microbial loop [Grebmeier et al., 2006]. In addition to this biological mediated process, brine rejection during periods of sea ice growth can also enrich bottom water CO₂ by forming dense near-surface water that sinks and carries with it solutes that include DIC rejected during sea ice formation [Rysgaard et al., 2007]. The seasonal increase in bottom water CO₂ content has implications for patterns in calcium carbonate corrosivity [Mathis and Questel, 2013], with the possibility of atmospheric ventilation during prefreezeup late season storm events [Bates and Mathis, 2009; Hauri et al., 2013]. The current perception is that CO2-enriched shelf bottom waters are advected off-shelf laterally into the adjacent Canada Basin and not mixed vertically to outgas at the sea surface [Anderson et al., 2013, 2010; Mathis et al., 2007]. The largest window for CO₂ ventilation to the atmosphere would be during stormy autumn conditions [Vavrus, 2013] in the absence of photon fluxes to support late season phytoplankton blooms [Ardyna et al., 2014] and prior to winter sea ice freezeup [Markus et al., 2009]. Hauri et al. [2013] speculate on the importance of autumn CO₂ efflux using a limited number of observations, in a manner similar to Bates [2006], that show sea surface pCO₂ oversaturations mostly in the southern Chukchi Sea region. We note that pixels showing CO₂ efflux were apparent in our analysis during the late season in the same area; the same data were used here albeit combined with a vast quantity of additional data points covering the same time period (Table 1 and supporting information Figure S1). Results from our analysis indicate a dominance of undersaturated pixels broadly over the western Arctic coastal ocean from September through November (Figure 3) when wind intensities were greatest (Figure 4). These conditions led to large net CO₂ influxes until November (Table 4), when open water was mostly limited to the southern Chukchi Sea (Figure 5). It is clear from the results described here and by Hauri et al. [2013] that this area of the Chukchi is susceptible to outgassing favorable conditions during this time of year, but how important that is for the overall net exchange is strongly dependent on sea ice conditions. Shown in Figure 9 is the standard deviation of November and December sea ice concentration for the 11 year data set used to build the sea ice climatologies (Figure 5). The areas of large standard deviation illustrate the significant degree of variability in the extent of open water during these months. Instances of CO₂ efflux in the southern Chukchi Sea region will be less important for net exchange during years with greater November open water owing to the prevalence of undersaturated conditions in the northern region (Figure 3). However, years of greater November open water also imply open water conditions persisting into December, a month when no seawater pCO_2 data exist for this area. The increasing delay in freezeup [Markus et al., 2009] and the growing prevalence of storm conditions during autumn months [Ardyna et al., 2014; Hakkinen et al., 2008; Vavrus, 2013] point to these late season effluxes as becoming increasingly important in the future. These potentially ephemeral source regions, which also include some select areas of the Beaufort Sea that experience seasonal upwelling-induced CO₂ outgassing [*Else et al.*, 2013a; *Mathis et al.*, 2012], should be targeted as regions and time periods that require a drastic increase in data coverage.

4.2.2. Sea Ice

There are opposing views for the relationship of sea ice to seawater CO₂ exchange with the overlaying atmosphere, with one view that ice cover acts as a barrier to gas exchange [Bates, 2006; Mucci et al., 2010; Shadwick et al., 2011; Stephens and Keeling, 2000; Takahashi et al., 2009] and another in which, owing to its temperaturedependent permeable nature [Golden et al., 2007], sea ice plays a more dynamic role in the transport of trace gases across the sea surface-atmosphere interface [Else et al., 2011; Loose et al., 2011; Miller et al., 2011; Semiletov et al., 2004, 2007; Vancoppenolle et al., 2013]. In this contribution, we treat sea ice as an imperfect barrier to gas exchange so as to include some transport (10% of open water flux in > 90% ice-covered areas) through cracks and leads in the dynamic winter icescape. However, CO₂ fluxes in areas of a broken, mobile ice cover, based on eddy correlation measurements, have shown large exchanges that suggest active transport of CO₂ at rates an order of magnitude above those in open water [Else et al., 2011; Miller et al., 2011]. If these exchanges are characteristic of the icescape environment, the estimate of carbon uptake presented here that is not adjusted for sea ice cover may be a more accurate assessment of the flux. However, some lines of evidence suggest this may not be the case. The eddy correlation measurements have the potential to be heavily impacted by CO₂-H₂O cross correlation, that if corrected for (the so-called PKT correction), reduce flux magnitudes to values that are more similar to those computed using interfacial pCO₂ gradients and wind speed-dependent parameterizations of gas transfer rates [Landwehr et al., 2014; Lauvset et al., 2011]. In addition to this methodological issue, a recent independent analysis of Radon-222 has shown near-zero interfacial gas transfer in sea ice-covered areas [Rutgers van der Loeff et al., 2014]. Owing to these lines of evidence, we expect limited exchange during most of the winter season except in areas of broken ice cover [Else et al., 2011]. Based on this treatment of the sea ice impact on gas exchange, a simple answer to how the western Arctic coastal ocean contribution to global coastal ocean carbon uptake will change under conditions of shrinking sea ice cover may be provided by the uptake estimates not adjusted for sea ice. Carbon uptake increased by 30% in this setting under ice-free conditions (Table 3), which is supported by a higher-annual mean flux that is nearly double the global average [Laruelle et al., 2014]. This is greatly oversimplified, however, as we expect some degree of seasonal/first-year winter sea ice cover to be maintained in the western Arctic coastal ocean through 2100 even under the highest (Representative Concentration Pathway 8.5) Intergovernmental Panel on Climate Change Fifth Assessment Report CO₂ emissions scenario [Collins et al., 2013]. Moreover, this implicitly assumes no additional adjustments to the western Arctic coastal ocean system as a result of shrinking sea ice cover. As reviewed by others [Grebmeier, 2012; Vancoppenolle et al., 2013], expected changes in physical oceanographic conditions (e.g., changing mixing patterns, upwelling, and stratification) will have cascading effects on ecosystem and biogeochemical patterns, with implications for surface seawater pCO₂ and sea-air CO₂ exchange. However, this oversimplification holds value as a first-order approximation for annual mean ice-free carbon uptake. The recent global estimate by Laruelle et al. [2014] also made the same oversimplification and observed a 46% increase in coastal ocean carbon uptake, signifying the importance of polar shelf sea gas exchange to global coastal ocean CO₂ flux overall.

The consideration of changing sea-air CO₂ flux under conditions of reduced ice cover also needs to factor in the role of melt ponds overlaying the ice surface and the impact sea ice has on gas exchange in adjacent surface water. There appears to be large variability in melt pond CO₂ exchange in both the direction and magnitude of flux [*Bates et al.*, 2014] but with an overall trend toward weak ($-1 \text{ mmol } \text{CO}_2 \text{ m}^{-2} \text{ d}^{-1}$) atmospheric CO₂ uptake [*Geilfus et al.*, 2015] that is supported by meltwater dilution [*Robbins et al.*, 2013]. The meltwater contribution is not included in the analysis conducted here and may be important over short time periods. However, weakly buffered meltwater will equilibrate rapidly making the overall impact short lived. The impact of sea ice on the adjacent surface ocean may be the largest complexity that has the most significant impact on sea-air CO₂ exchange. Wind speed-dependent parameterizations have been shown to be grossly inadequate in sea ice impacted settings where additional sources of turbulence are present beyond that imparted solely by the wind-driven gas exchange [*Loose et al.*, 2011, 2014]. *Loose et al.* [2014] report a near-zero gas transfer rate at 100% ice cover, consistent with our assumption with regard to the role of sea ice in gas exchange, but much higher values than what would be predicted from a wind-dependent parameterization immediately adjacent to the ice pack. This implies a potentially significant underestimation of sea-air CO₂ fluxes in regions with moderate sea ice cover (>30% to < 90%). The effect of underestimating the gas transfer rate in moderate ice cover areas will be greatest during the periods of initial sea ice melt and freezeup in the western Arctic coastal ocean. Higher-gas transfer rates combined with the instances of seawater pCO_2 oversaturation seen in November in some areas with remaining but variable sea ice cover (Figures 5 and 9) will drive larger effluxes, with an accompanying greater impact on net exchange. Clearly, there are key aspects regarding sea-air CO₂ exchange in polar shelf sea settings that require significant additional research, including an increase in the time/space coverage of seawater pCO_2 data in targeted areas and the continued development of sea ice-specific gas transfer parameterizations.

5. Conclusions

Key findings from the analysis of 600,000 surface seawater pCO_2 measurements collected between 2003 and 2014 in the western Arctic coastal ocean are the following.

- 1. Sea ice-adjusted atmospheric CO₂ uptake increased from May to peak in September and October, when strong winds coincided with predominately undersaturated seawater *p*CO₂.
- 2. The average of monthly area-weighted fluxes scaled to the surface area of this region indicated 10.9 ± 5.7 Tg C yr⁻¹ of carbon uptake, representing 5% of the exchange from global coastal oceans.
- 3. Neglecting the sea ice adjustment of the sea-air CO₂ flux increased the uptake by 30% to 15.4 Tg C yr⁻¹.
- 4. Undersampling may result in an underestimate of ocean carbon uptake of about 0.7 Tg C yr^{-1} , which is < 10% of the sea ice-adjusted carbon uptake.
- 5. Carbon uptake in the Chukchi Sea is lower than previously estimated using a sparser data set $(4.4 \text{ Tg C yr}^{-1})$.
- 6. Carbon uptake is similar between the Chukchi and Beaufort Seas, because weaker area-weighted fluxes in the Beaufort Sea operate over a larger surface area. Overall, this analysis has integrated data sets from many sources to constrain CO₂ exchange within this broad and rapidly changing polar coastal ocean.

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