Key Points:

- In a regional Arctic Ocean model, carbon uptake increases by 20% and mean sea surface pH decreases from 8.1 to 8.0 from 1980 to 2015
- Tests show that sea ice algae impact multidecadal runs and the ice carbon pump impacts the seasonal sea surface carbonate saturation
- There is large regional and seasonal variability, highlighting problems with extrapolating Arctic observations in both space and time

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Modeled Impacts of Sea Ice Exchange Processes on Arctic Ocean Carbon Uptake and Acidification (1980–2015)

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Abstract A regional Arctic ice ocean model incorporating biogeochemical processes occurring inside the sea ice and water column is used to assess changes to the Arctic Ocean's carbon system, including oceanic carbon uptake and ocean acidification, over the recent period of Arctic sea ice decline (1980–2015). Two novel modifications are the following: (1) incorporation of carbon uptake by sea ice algae and (2) modification of the sea ice carbon pump to allow for vertical transport by brine plumes with high concentrations of dissolved inorganic carbon (DIC) and total alkalinity (TA) to the bottom of the mixed layer. The simulated carbon uptake north of 66.5°N increases by 20%, from 110 to 135 Tg C yr⁻¹ from 1980–2015, and the mean pan-Arctic sea surface pH decreases from 8.1 to 8.0. There is substantial regional and seasonal variability, highlighting potential problems with interpolating sparse measurements. Two sensitivity studies assess the effects of modifications to the near-surface carbonate system. First, excluding the sea ice carbon pump results in a marked decrease in seasonal variability of pan-Arctic-mean sea surface DIC (~25% less than the standard run) and TA (~10% less), suggesting that neglecting the ice carbon pump results in models overestimating the saturation state in summer. Second, neglecting the sea ice algae results in an underestimation of the Arctic Ocean's annual carbon uptake (~3% per year), indicating that the accumulating year-on-year underestimation of annual carbon uptake, as a result of neglecting sea ice algae, would lead to increasing error over multidecadal runs of polar ocean models.

1. Introduction

In the last half century, the state of the Arctic Ocean has undergone dramatic shifts, particularly at the sea surface and along coastal shelves. In addition to the loss of sea ice immediately apparent from the satellite record (e.g., Barber et al., 2015; Stroeve et al., 2012), the Arctic Ocean has also experienced accelerated warming and freshening (Carmack et al., 2016). The Arctic has also undergone biogeochemical changes. In particular, it has become lower in pH and the calcium carbonate saturation state of the biologically relevant forms aragonite and calcite, Ω_{arag} and Ω_{calc} , have decreased faster than the global ocean-averaged trends (Bellerby et al., 2018; Miller et al., 2014; Steiner et al., 2015; Wallhead et al., 2017).

In the polar oceans, sea ice provides a unique interface that both directly and indirectly affects the air-sea exchange of carbon. Many of the carbon-related processes associated with sea ice are often neglected or oversimplified in current ocean and climate models. In particular, observations demonstrate a marked difference between the dissolved inorganic carbon (DIC) concentration of sea ice and that of the seawater from which it formed (and similarly for total alkalinity, TA). From a budgetary view, this difference necessitates a flux of carbon out of the ice as it forms as well as the dilution of near-surface seawater as ice melts. This two-part process is called the sea ice carbon pump (Rysgaard et al., 2011). Only very recently have model studies quantitatively examined the effects of sea ice on the ocean carbon system (Grimm et al., 2016; Moreau et al., 2016). Both of those studies include an (inorganic) sea ice carbon pump parameterization that is restricted to the surface modeled layer. They found that the contribution of the sea ice carbon pump could have a substantial regional effect on air-sea exchange, as well as on carbon distribution in the polar oceans. A novel treatment in the present study is the nonlocal transport of the DIC and TA during ice growth, associated with dense brine rejection, which is implemented by depositing both DIC and TA at the bottom of the mixed layer rather than at the surface. The development and analysis of this parameterized ice carbon pump in a 1-D

model study are described in detail in Mortenson et al. (2018). Here, we apply the parameterization into a regional coupled sea ice and ocean biogeochemical model of the Arctic Ocean and evaluate its regional and pan-Arctic impact. The depth of deposition of brine-rejected DIC and TA in the present study and Mortenson et al. (2018) is based on results from Jin et al. (2015), in which subgrid scale brine-rejected salt in a simulation best represents observed salinity profiles when deposited at the bottom of the mixed layer. The scenario with brine-rejected tracers at the mixed layer depth is an idealization of comparable simplicity to the surface release assumed in Grimm et al. (2016) and Moreau et al. (2016) or an intermediate scenario of mixing throughout the mixed layer as described in DeGrandpre et al. (2019).

In addition to the sea ice carbon pump, representations of both the pelagic and sympagic (sea ice associated) ecosystems are also implemented in the model (Hayashida et al., 2019; Mortenson, 2019; Mortenson et al., 2017). The ice algae, along with the simulated pelagic ecosystem, act as a sink of DIC associated with primary production, and a source of DIC associated with remineralization of detritus. Likewise, the sympagic and pelagic ecosystems affect the ocean TA at the surface and throughout the water column (although the effect on TA is not entirely analogous to that on DIC Mortenson et al., 2018). While the effect of ice algae on the carbon system is considered to be small compared to that of the pelagic ecosystem (Else et al., 2019), the ice algae bloom begins before that of the pelagic phytoplankton and thus extends the time period for biological impacts on the carbon system. In addition to the direct effect of ice algae on the carbon system, they are suggested to contribute to seeding the pelagic bloom when they get released into the water column (Michel et al., 1993). To evaluate the impact of these processes, we have included the sympagic ecosystem into the model.

This model study is novel in its inclusion of both pelagic and sympagic ecosystems into the assessment of the Arctic Ocean's carbon uptake as well as a representation of the ice carbon pump as a nonlocal transport mechanism for DIC and TA. The standard run simulation is driven by an atmospheric reanalysis product over the satellite record (1980–2015, including a spin-up period from 1969–1979) which spans the recent retreat of Arctic sea ice. After a description of the model setup in section 2, the results of the standard run are presented in section 3 including a description of regions that deviate from the pan-Arctic mean in the standard run. In addition, the importance of representing the sea ice carbon pump and sympagic ecosystem are independently assessed through two sensitivity studies in section 3. Results of the model study, implications, and potential future research directions are presented in section 4.

2. Methods

The study uses the Nucleus for European Modelling of the Oceans, version 3.4 (NEMO Madec & the NEMO team, 2008) in a domain encompassing the Arctic Ocean. The model system includes physical and biogeochemical models for the ocean and the ice as well as coupling between ocean and sea ice biogeochemistry. The period of the standard run is 1980–2015, with a spin-up period from 1969–1979. Detailed sensitivity analyses regarding various model settings related to the Arctic sea ice and forcing were carried out in Hayashida et al. (2019).

2.1. Ocean and Sea Ice Physics Models

The ice ocean physics in NEMO is simulated using Océan Parallélisé (OPA), coupled to the Louvain-la-Neuve sea Ice Model, version 2 (LIM2). The physical ocean model, OPA, represents ocean dynamics and thermodynamics through the primitive equations along with the nonlinear seawater equation of state and a free surface formulation for sea surface height (Madec & the NEMO team, 2008). The ice model, LIM2, is composed of two ice layers and one snow layer, and represents dynamics and thermodynamics within the ice (Bouillon et al., 2009). The default setting for LIM2 does not allow any light penetration when there is snow cover. In order to provide adequate light penetration at the time of observed ice algae blooms, the default setting for LIM2 was modified to allow some light penetration through a thin snow cover, as described in Hayashida et al. (2019). Documentation and source files for both sea ice and ocean physical models (OPA and LIM2) can be found online (at <https://www.nemo-ocean.eu>).

2.2. Ocean Biogeochemistry Model

Ocean biogeochemistry and ecosystem dynamics are simulated using the Canadian Ocean Ecosystem model (CanOE). The biogeochemical model CanOE was developed at the Canadian Centre for Climate Modelling

Table 1

List of Studies That Have Analyzed the Arctic Ocean Carbon Uptake, Along With a Brief Description of the Method Used and an Identification of Whether the Range Indicates an Uncertainty or an Increasing Trend Over the Given Time Period

Study	Carbon uptake rate (Tg C yr ⁻¹)	Method	Range
Bates and Mathis (2009)	66–199	literature review	uncertainty
Arrigo et al. (2010)	111–125	model study	trend (1998–2003)
Manizza et al. (2013)	52–64	model study	trend (1996–2007)
Schuster et al. (2013)	60–180	cross-method study	trend (1990–2009)
MacGilchrist et al. (2014)	106–226	obs. and mass balance	uncertainty
Yasunaka et al. (2016)	180 (±210)	obs. sea surf. pCO ₂	uncertainty
Yasunaka et al. (2018)	180 (±130)	obs. sea surf. pCO ₂	uncertainty
Manizza et al. (2019)	136–164	model study	trend (2006–2013)

Note. obs. = observed; surf. = surface.

and Analysis for implementation in the Canadian Earth System Model version 5 (CanESM5). CanOE includes a pelagic ecosystem composed of four functional groups: two phytoplankton groups, representing small dinoflagellates and large diatoms, and small and large zooplankton grazers. Nutrients in CanOE include both nitrogen components (NH₄ and NO₃) as well as an inorganic carbon system with a sink of DIC associated with primary productivity and a source of DIC associated with remineralization of detritus. Within the water column, NH₄ is converted to NO₃ by nitrification. Documentation of CanOE is described by Christian et al. (unpublished) available in the appendix of Mortenson (2019). For the present study, the model is modified to include both a sympagic ecosystem and a sea ice carbon pump.

2.2.1. Sea Ice Ecosystem and Carbon Pump

The ice algae functional group, located in the bottom of the ice (denoted as the skeletal layer, with a prescribed thickness of 3 cm), is coupled to the pelagic ecosystem through the release of ice algae detritus and to water column nutrients via exchange at the ice-water interface. The specific growth rate for ice algae depends on temperature and the availability of light, NO₃, NH₄, and silicate (Si). The ice algae take up nutrients and DIC within the ice skeletal layer during photosynthetic growth, and release both detritus and organic carbon into the water column during senescence. Also, consistent with the 1-D model described in Mortenson et al. (2017, 2018), ice algal detritus contributes to the pelagic large phytoplankton through seeding. The detrital organic carbon is converted to DIC through remineralization as the detritus sinks through the water column.

In addition to inclusion of ice algae, the model's inorganic carbon system is modified to account for DIC and TA fluxes via the seasonal growth and melt cycle. These fluxes are denoted as the sea ice carbon pump. CanOE has been modified to include an ice carbon pump with nonlocal vertical transport by brine plumes (high in DIC and TA relative to in situ sea water concentrations) during ice growth, and the dilution of near-surface waters during ice melt. The depth of deposition of brine-rejected DIC and TA during ice formation in the present study and Mortenson et al. (2018) is based on results from Jin et al. (2015), in which subgrid scale brine-rejected salt in a simulation best represents observed salinity profiles when deposited at the bottom of the mixed layer. The scenario with brine-rejected tracers at the mixed layer depth can be viewed as one extreme scenario among the likely deposition depths, as opposed to surface release as the other extreme assumed in Grimm et al. (2016) and Moreau et al. (2016) and an intermediate scenario of mixing throughout the mixed layer as described in DeGrandpre et al. (2019). The DIC and TA fluxes associated with the sea ice carbon pump (respectively, ϕ_{ice}^{DIC} and ϕ_{ice}^{TA}) are parameterized in terms of a constant ice-ocean concentration difference:

$$\phi_{ice}^{DIC} = P_{SW}(DIC_{ice}^{ref} - DIC_{SW}^{ref})\frac{\Delta h_{ice}}{\Delta t}, \quad (1)$$

where Δh_{ice} is the change in ice thickness over a model time step, Δt , and DIC_{ice}^{ref} and DIC_{SW}^{ref} are the constant reference concentrations of DIC in the ice and seawater (Table 1 in Mortenson et al., 2018). The proportion of DIC released to the water column is denoted P_{SW} and is set at 99% as in the 1-D study described in Mortenson et al. (2018). The remaining proportion $1 - P_{SW}$ is released from the ice directly into the atmosphere. The parameter settings for ice-ocean and ice-air exchange of DIC (and TA below) are based on observations which indicate that large amounts of DIC and TA are expelled into the water column during ice formation, and low concentrations in sea ice lead to dilution of the near-surface seawater during melting (e.g., Brown et al., 2015; Geilfus et al., 2015; Rysgaard et al., 2011).

Similarly, for TA,

$$\phi_{ice}^{TA} = (TA_{ice}^{ref} - TA_{SW}^{ref})\frac{\Delta h_{ice}}{\Delta t}, \quad (2)$$

where TA_{ice}^{ref} and TA_{SW}^{ref} are the reference concentrations of total alkalinity in the ice and seawater. Unlike

carbon, there is no flux of TA into the atmosphere. The inclusion of ikaite precipitation in the ice and dilution in the water column, discussed in Mortenson et al. (2018), was not treated explicitly in the DIC and TA budgets of the 3-D formulation but are implicit in the reference concentration differences for each between ice and sea water. In Mortenson et al. (2018), explicit representation of ikaite was shown to have little effect on air-sea carbon exchange.

One of the novel features of this model study is the nonlocal treatment of the sea ice carbon pump. In general, models release the brine-associated TA and DIC fluxes into the surface layer of the model, while in the present study they are released at the bottom of the mixed layer as a representation of the nonlocal transport by brine plumes as in the 1-D study described in Mortenson et al. (2018), analogous to the brine-associated release of salt in Jin et al. (2015).

The carbon system properties of pH, $p\text{CO}_2$, Ω_{arag} , and Ω_{calc} presented in this study were calculated offline from model output of DIC, TA, temperature and salinity, following Dickson et al. (2007), with equilibrium constants of Lueker et al. (2000). The saturation states were based on Mucci (1983) with the pressure correction of Millero (1995). Spatially or temporally averaged carbon system quantities are first computed at each point in time and space and then averaged.

2.3. Model Domain

The model domain consists of the North Atlantic and Arctic (NAA) grid developed at the University of Alberta (https://knossos.eas.ualberta.ca/xianmin/anha/model_configuration.psp1#naa). The NAA grid is rotated and has three major boundaries: at approximately 60°N (about 100 km south of Bering Strait) in the North Pacific, across the North Atlantic at about 50°N, and at the mouth of Hudson Bay. In this study, area averages and totals for most variables (with the exception of sea ice area) are over the Arctic Ocean region north of 66.5°N, as shown in Figure 1a. The horizontal grid resolution ranges from 11 km near the North American coastline to 15 km along the northern Eurasian coastline. The vertical resolution is modified from the original NAA configuration to have higher vertical resolution near the ocean surface (e.g., from 6 m for the upper surface layer to 1 m), as described in Hayashida et al. (2019).

2.4. Atmospheric Forcing

The simulation is forced by meteorological data for the Arctic Ocean from 1969 to 2015, from the Drakkar Forcing Set (DFS), version 5.2 (<https://www.drakkar-ocean.eu/>). The DFS data set provides atmospheric information on a 0.7° resolution grid, with temporal resolution of 3 hr for winds at 10 m above the surface, air temperature and humidity at 2 m, and incoming shortwave and longwave radiation, as well as daily resolution for snowfall and total precipitation (rain plus snow). The period 1969–1979 is used as a spin-up period and not included in the model evaluation.

The atmospheric $p\text{CO}_2$ is set to monthly mean values from observations at Mauna Loa (<https://scrippsco2.ucsd.edu>), which increase from $\sim 335 \mu\text{atm}$ in 1980 to $400 \mu\text{atm}$ by 2015. It should be noted that the actual seasonal variability in atmospheric $p\text{CO}_2$ is more pronounced in the Arctic than in the lower latitudes as measured at Mauna Loa. A more accurate representation of atmospheric $p\text{CO}_2$ over the Arctic Ocean would be data from stations located at Point Barrow, Alaska, or Alert, NWT, Canada. Although the trends and annual averages for Mauna Loa and the Arctic stations largely overlap, the seasonal variability is $\sim 15 \mu\text{atm}$ in the Arctic locations, while the range at Mauna Loa is about half of that. Despite the difference in seasonal variability, Mauna Loa was chosen due to the continuity of measurements (there are sporadic gaps in both the Barrow and Alert data sets, and both Arctic time series begin later than the Mauna Loa time series).

2.5. Initial and Boundary Conditions

The model is initialized at rest (velocity fields of 0) with thermodynamic variables (temperature and salinity) representative of January 1969 from the Ocean Reanalysis System 4 (ORAS4) (Balmaseda & Weaver, 2013). For grid cells with initial sea surface temperature (SST) within 2°C of freezing, sea ice thickness and concentration were respectively set to 2.5 m and 95%, with 10 cm of snow cover; otherwise these quantities were set to 0. The boundary conditions for physical variables at open-ocean boundaries (temperature, salinity and velocity) were based on climatological monthly mean fields of ORAS4, and interpolated to the NAA grid's

Figure 1. (a) Arctic Ocean model bathymetry, (b) subregions described in section 3.2 (subregions are, clockwise from left: Canadian Polar Shelf, Beaufort Sea shelf and basin, Laptev Sea, Barents Sea, and Norwegian Sea), and comparison of (c) the simulated mean sea surface salinity (SSS) of this study over the period of the standard run and (d) SSS based on observations compiled for the Polar Science Center Hydrographic Climatology.

open-ocean boundaries. Relaxation time scales at boundaries were set to 1 day for inflow and 15 days for outflow, consistent with Dupont et al. (2015).

Initial conditions for biogeochemical variables (NO_3 , DIC, and TA) were based on the GLObal Ocean Data Analysis Project version 2 (GLODAPv2) (Lauvset et al., 2016). For biogeochemical variables, the open-ocean boundary conditions were held constant at the initial values. The constant value for biogeochemical variables imposed at the open-ocean boundaries does not account for interannual variability or trends, in particular of sea surface DIC. While a positive trend in the surface concentration of DIC at the boundaries, associated with the positive trend in atmospheric $p\text{CO}_2$, would be more accurate, it should be noted that the model domain at the largest boundary (in the North Atlantic at 45°N) is relatively far from the domain analyzed and defined as the pan-Arctic (north of 66.5°N). The distance between the boundary and the region of analysis could mitigate the inaccuracy due to the constant DIC boundary condition, because any surface water mass reaching the pan-Arctic from the boundary would have time to equilibrate with the atmosphere during the transit.

The ocean surface is subject to air-sea gas exchange as well as dilution (concentration) due to river runoff and precipitation (evaporation). Air-sea exchange of CO_2 is calculated based on the formulation of Wanninkhof (1992), multiplied by the percent of open water exposed to the atmosphere (uncovered by

sea ice) within each grid cell, as described in Aumont et al. (2015): $k_{CO_2} = k_{CO_2}^{Wanninkhof} \frac{A_{total} - A_{seaice}}{A_{total}}$. The river flows at the six major Arctic rivers (Mackenzie and Yukon Rivers in North America, and Kolyma, Lena, Yenisey, and Ob in Eurasia) were prescribed based on the monthly mean, annually varying estimates of Dai et al. (2009) from 1969–2007. From 2007–2015, river flow from the Year 2007 was repeated annually. Each of the rivers was assigned a fixed concentration for DIC, TA, and NO_3 based on the studies of Cooper et al. (2008), Tank et al. (2012), and McClelland et al. (2012). Concentrations of dissolved organic carbon (DOC) and dissolved organic nitrate (DON) for each river were based on the studies of Cooper et al. (2008), Holmes et al. (2012), and McClelland et al. (2012). In the model, the riverine inputs of DOC and DON are added directly to the respective inorganic pools of DIC and nitrate.

2.6. Experimental Setup

To investigate changes in the Pan-Arctic carbon system and the impacts of specific parameterizations, three simulations were performed, a standard run and two sensitivity runs, described below:

1. The standard run (1980–2015, with a spin-up period from 1969–1979) includes both the sea ice algae and the sea ice carbon pump implementations. In addition to the standard run, we perform two sensitivity experiments to estimate the contribution of ice algae and the sea ice carbon pump to carbon fluxes in the Arctic.
2. In the first sensitivity run, denoted the noICP run, the sea ice algae implementation is included, but the sea ice carbon pump is suppressed. This experiment branches off the standard run in Year 1990 and are integrated until 2010. In the noICP run, DIC and TA fluxes associated with ice growth and melt are set to 0 by setting the reference seawater and ice concentrations in Equations 1 and 2 to be equal (i.e., $DIC_{ice}^{ref} = DIC_{SW}^{ref}$ and $TA_{ice}^{ref} = TA_{SW}^{ref}$).
3. In the second sensitivity run, denoted the noIA run, the sea ice carbon pump implementation is included, but the sea ice algae are suppressed. This experiment is initialized the same as the standard run in Year 1969 and are presented (as in the standard run, post-spin-up) over the period 1980–2015.

3. Results and Discussion

The standard run encompasses the recent epoch in which the Arctic Ocean has transitioned from a largely sea ice covered ocean (in both area and time) to a seasonally open-water dominated ocean. A particularly important aspect of this transitional period is the potential effect the loss of sea ice may have on the annual uptake of atmospheric carbon by the Arctic Ocean. In addition to the ice-ocean exchange of DIC and TA described above, sea ice also serves as a barrier to the exchange of carbon between the ocean and atmosphere.

3.1. Large-Scale Characteristics of the Baseline Arctic Ocean Simulation (1980–2015)

Simulated physical sea surface and sea ice variables, averaged over the domain north of 66.5°N, are presented in Figure 2. Simulated Arctic SST shows a seasonal range of around 2°C (Figure 2a) with an approximate increase in the annual mean of 0.3°C over the period of the standard run (from −0.6°C in the decade 1980–1990 to −0.3°C in the decade 2005–2015). Simulated Arctic sea surface salinity shows a seasonal range between minima of around 2 PSU below the annual mean (of 30.75 PSU) to 1 PSU above the annual mean. There is some interannual variability but no substantial trend around the long-term average (annual averages are largely within a range of ± 0.25 PSU; Figure 2b). The simulated sea ice area decreases from an annual mean of $\sim 10 \times 10^6$ to $\sim 9 \times 10^6$ km² over the standard run period, with a larger decrease in annual minima from $\sim 6 \times 10^6$ to $\sim 4 \times 10^6$ km² (Figure 2c). The simulated sea ice area generally has slightly lower maxima relative to observed sea ice area of the National Snow and Ice Data Center (NSIDC Fetterer et al., 2017). A contributing factor to the difference in maxima is the larger domain used for NSIDC sea ice area calculations. The simulated sea ice volume shows generally good agreement when compared to the sea ice volume of the Pan-Arctic Ice Ocean Modeling and Assimilation System (PIOMAS Zhang & Rothrock, 2003; Figure 2d). One exception is the slight offset in ice volume extrema between our simulation and PIOMAS for a ~ 5 -yr period centered around the Year 2000 and over 2011–2012. When interpreting these differences, it is important to note that both quantities being compared are model products, and the maximum deviation between extrema is less than 4,000 km³ (<20% of the mean annual ice volume). In

Figure 2. Annual (solid lines) and monthly mean (dashed lines) of simulated Pan-Arctic (a) sea surface temperature, (b) sea surface salinity, (c) sea ice area, and (d) sea ice volume. The dotted lines in panels (c) and (d) indicate, respectively, the maximum and minimum monthly values of sea ice area provided by the National Snow and Ice Data Center (NSIDC) and of sea ice volume provided by the Pan-Arctic Ice Ocean Modeling and Assimilation System. The domain of the modeled sea ice area calculations is the entire model domain and north of 31°N for NSIDC ice area. All other variables are analyzed over the region north of 66.5°N .

general, both PIOMAS and the present study show similar ranges around the annual mean and a consistent decline in simulated ice volume with time.

A comparison of sea surface salinity between the model mean over the standard run period (1980–2015) and a long-term mean based on observations from the Polar Science Center Hydrographic Climatology (PHC Steele et al., 2001) is presented in Figures 1c and 1d. Both the modeled and observed sea surface salinities are highest in the Atlantic sector of the Arctic, and low salinity values are found in both data sets along the coasts of Eurasia and Alaska. The most prominent difference between the two is a lower average model salinity through the Chukchi and Beaufort Sea and in the Canadian Polar Shelf. It is important to note, however, that the PHC data product is interpolated from observations which are particularly sparse in the northern parts of both the Chukchi and Beaufort Sea and throughout the Canadian Polar Shelf (see Steele et al., 2001, Figure 1). Therefore, the low salinity simulated in these regions is not necessarily incorrect. Given the importance of sea surface salinity to carbon system properties, the deviation between the model output and interpolated observations in the Canadian Polar Shelf, the Beaufort Sea basin, and northern Chukchi Seas indicate that these are key regions of uncertainty in which there is a particular need for observations.

The annual carbon uptake of the Arctic Ocean north of 66.5°N (Figure 3a) starts at around 110 Tg C yr^{-1} at the beginning of the standard run and increases to 135 Tg C yr^{-1} by the end of the standard run, an

Figure 3. (a) Net annual CO_2 uptake over the Arctic Ocean (solid black line); net seasonal CO_2 uptake (solid color lines) over summer (red), fall (orange), winter (blue), and spring (green); and net monthly CO_2 uptake (dashed line). (b) Annual (solid black line) and monthly mean (dashed line) sea surface $p\text{CO}_2$ and atmospheric $p\text{CO}_2$ (red line). Annual (solid lines) and monthly mean (dashed lines) sea surface (c) DIC, (d) TA, and (e) ratio of DIC/TA. All variables are analyzed over the Arctic north of 66.5°N .

increase of $\sim 20\%$ over that period of time (1980–2015). The trend in carbon uptake is attributed to a secular increase in atmospheric $p\text{CO}_2$. The simulated pan-Arctic uptake of carbon in this study is within the range of several recent studies employing various distinct methods, including Bates and Mathis (2009), Arrigo et al. (2010), Schuster et al. (2013), MacGilchrist et al. (2014), and Yasunaka et al. (2016) (see Table 1 for details of these studies). Our results fall outside of the range estimated in Manizza et al. (2013) and are at the lower bound of the range estimated by Manizza et al. (2019). The modeling study by Manizza et al. (2013) over the period 1996–2007 finds a smaller uptake for the pan-Arctic of ~ 50 to 65 Tg C yr^{-1} from beginning to end of the model run. A contributing factor to their relatively small pan-Arctic uptake is their overestimation of sea ice extent by $\sim 10\%$ over the entire run when compared to satellite data, as discussed in Manizza et al. (2019).

Figure 3a also includes values of seasonal uptake for summer (red; July, August, September), fall (orange; October, November, December), winter (blue; January, February, March), and spring (cyan; April, May, June). The seasonal analysis shows that the largest uptake rate occurs in summer and the smallest in winter, as expected. The summer carbon uptake rate is ~ 2.5 times the annual average, and the winter

Figure 4. Arctic Ocean annual (solid lines) and monthly (dashed lines) mean sea surface (a) pH, (b) aragonite (Ω_{arag} , black) and calcite (Ω_{calc} , red) saturation states. Arctic Ocean total biomass of (c) phytoplankton (black) in the upper 50 m of the water column and sea ice algae (red), both in Tg C.

carbon uptake rate is ~ 0.5 times the annual average. Over the standard run period, the increases in the spring and summer uptake rates (of $\sim 40\%$ and $\sim 35\%$) are slightly higher than the trend in the annually averaged carbon uptake rate over the standard run period. The fall exhibits the smallest increase in carbon uptake rate of $\sim 10\%$ over the standard run period.

The increase in carbon uptake rate from 1980 to 2015 is coincident with increases in the (prescribed) atmospheric and (simulated) sea surface $p\text{CO}_2$ (Figure 3b). For the pan-Arctic domain, the average sea surface $p\text{CO}_2$ increases from $300 \mu\text{atm}$ at the beginning of the standard run to $340 \mu\text{atm}$ by the end (Figure 3b). As mentioned above, the increase in Arctic Ocean carbon uptake rate during this period is also coincident with an increase in SST and a decrease in sea ice area. The increased Arctic Ocean uptake of carbon can be mostly attributed to increases over the model run of both atmospheric CO_2 and open water (which leads to increased direct exposure to the atmosphere).

Two other factors influence the changes in ocean carbon uptake over the period considered. Part of the increase of $p\text{CO}_2$ in surface waters is due to a temperature-based reduction of solubility: Following Takahashi et al. (1993) the modeled increase in pan-Arctic SST of 0.3°C implies that $\sim 10\%$ of the total simulated increase in sea surface $p\text{CO}_2$ can be explained by the temperature-based solubility change. This reduction in solubility reduces the ocean uptake of carbon relative to what it would be for fixed SST. In contrast, warmer temperatures and more light exposure can also lead to increased primary production and a strengthened biological pump, and a larger change in ocean carbon uptake than that associated only with abiotic processes. In fact, the annually averaged phytoplankton biomass does show an increase of just over 10% over the standard run period (see below and Figure 4).

Sea surface DIC, TA, and their difference, averaged over the Arctic Ocean, are shown in Figures 3c–3e. Comparable magnitudes of variability are found in Arctic-wide sea surface DIC and TA, ranging from $\sim 100 \text{ mmol m}^{-3}$ above to 150 mmol m^{-3} below the respective annual means of $1,875$ and $1,975 \text{ mmol m}^{-3}$. Neither DIC nor TA show strong trends such as that apparent in the time series of $p\text{CO}_2$. The differences

in the trends of $p\text{CO}_2$ and DIC is a consequence of the fact that the relationship between sea surface DIC and TA, rather than DIC alone, determines the values of other quantities in the marine carbon system. Analogous to the study of Xue and Cai (2020) regarding pH, the difference of DIC and TA can be treated as a proxy for $p\text{CO}_2$ (outside of a range where DIC and TA are within 50 mmol m^{-3} of one another.) To first order, DIC and TA have opposing effects on $p\text{CO}_2$, and the difference of annually averaged pan-Arctic DIC minus TA increases, from -110 to -90 , over the period 1980–2015 in a manner comparable to the trend in $p\text{CO}_2$ (Figure 3e).

Additional sea surface carbon variables averaged over the Arctic Ocean north of 66.5°N (pH, Ω_{arag} , and Ω_{calc}) are shown in Figures 4a and 4b. The annually averaged pan-Arctic sea surface pH shows a steady decline from 8.1 to just over 8.0 in the standard run, with seasonal summer maxima approximately 0.1 above the annual mean and winter minima that fall below 8 by the end of the simulated period (2010–2015). The pan-Arctic annual average of Ω_{arag} shows a trend from just over 1.2 to just under 1.1, with seasonal minima in winter that approach the threshold of aragonite undersaturation by the end of the standard run. The annual averaged Ω_{calc} shows a general decline over the standard run from ~ 2 to 1.7, but neither the annual nor monthly averaged pan-Arctic means approach aragonite undersaturation at any time in the standard run.

The pan-Arctic simulated phytoplankton biomass integrated over the upper 50 m of the water column and the ice algae biomass, both in units of carbon content, are presented in Figure 4c. Over the standard run period, the annually averaged phytoplankton biomass increases by just over 10% (from 400 to 450 Tg C). The phytoplankton biomass ranges from near 0 in winter to a maximum of 1,000–1,200 Tg C, which is roughly twice the biomass presented in Arrigo and van Dijken (2015) and in Kahru et al. (2016). The estimates $400\text{--}600 \text{ Tg C yr}^{-1}$ presented in these studies are based on satellite observations of open-water regions of the Arctic and show much more pronounced positive trends. Our result suggests that pelagic primary production is not as strongly coupled to the ice cover as is implied in Arrigo and van Dijken (2015) and Kahru et al. (2016), and that both under-ice and subsurface blooms are not negligible, as indeed noted in Arrigo et al. (2012). Ice algae biomass peaks in the spring, generally between 20 and 60 Tg C. This value is 2 orders of magnitude less than the maximum phytoplankton biomass. This large ratio between phytoplankton and ice algae biomass is consistent with Else et al. (2019), which finds the under-ice phytoplankton bloom to be the main contributor to water column DIC and $p\text{CO}_2$ changes in the spring. There is no evident reduction in pan-Arctic sea ice algae biomass despite the loss in sea ice area (Figure 2c). The stability of the pan-Arctic sea ice algae bloom throughout the standard run, despite the loss of habitat (sea ice area), can be attributed to the fact that the remaining sea ice is thinner, allowing for more light at the bottom of the ice and a larger ice algae bloom (Hayashida, 2018).

3.2. A Regional Perspective on Carbon Uptake in Arctic Seas

While the Arctic Ocean averages presented above illustrate a general pan-Arctic picture of the variability in physical and biogeochemical variables, all these quantities exhibit substantial regional variability. The regions that exhibit the largest deviations from the pan-Arctic mean, with regards to the oceanic uptake of carbon and the oceanic carbon system, are the Laptev, Norwegian, and Barents Seas in the Atlantic sector; the Canadian Polar Shelf (CPS, a region also known by its terrestrial designation as the Canadian Arctic Archipelago) and the Beaufort Sea basin and shelf in the sector adjacent to the Pacific (Figure 1). Figure 5 displays the annual mean carbon flux, sea surface $p\text{CO}_2$, pH, and sea ice cover for each of these regions. The corresponding pan-Arctic annual averages are indicated in black for comparison. The changes in the properties displayed in Figure 5 are also tabulated in Appendix B.

Regional differences are particularly pronounced in the annually averaged mean carbon flux, with high annual uptake rates in the Barents and Norwegian Seas relative to the pan-Arctic mean, and low uptake rates in both the Beaufort Sea basin and shelf and the Laptev Sea. Increases in carbon uptake rates in both the Norwegian and Barents Seas over the standard run time period (of approximately $5 \text{ g C m}^{-2} \text{ yr}^{-1}$ for each of these regions) account for a large proportion of the pan-Arctic increase in net annual oceanic carbon uptake described in section 3.1 (together the Barents and Norwegian Seas comprise roughly 20% of the entire Arctic Ocean surface area).

Figure 5. (a) Annual air-to-sea carbon flux (positive equals into the ocean) for the entire Arctic Ocean (black line) as well as regions in panel (b) of Figure 1, colored similarly. The regions in all panels are the Barents (dashed orange), Norwegian (red), Canadian Polar Shelf (dotted blue), Beaufort Sea shelf (dashed blue) and basin (cyan), and Laptev (olive) Seas. Regional and pan-Arctic (north of 66.5°N) annual mean (b) sea surface and atmospheric (black dashed line) $p\text{CO}_2$, (c) sea surface pH, (d) aragonite saturation state, and (e) ice cover.

The trend in pH is negative in all regions displayed in Figure 5. Most regions have similar trends in pH as the pan-Arctic average (a drop of ~ 0.1 in pH over the period of the standard run), with the exception of the CPS region which exhibits a more rapid decrease in pH (of 0.15 drop in pH over the standard run period). Regions with low pH (Laptev Sea and Beaufort Sea basin and shelf, Figure 5) relative to the pan-Arctic mean are similarly low in both Ω_{arag} and Ω_{calc} .

The timing and location of undersaturation events have a potentially negative

Potentially harmful impacts on marine species (e.g., calcifying species such as pteropods) are influenced by the timing and location of undersaturation events (Bednaršek et al., 2014; Comeau et al., 2012). The Laptev Sea and Beaufort Sea basin and shelf all exhibit annually averaged sea surface aragonite undersaturation over most of the standard run period. The low aragonite saturation state and pH in the Laptev Sea are consistent with observations (Semiletov et al., 2016). In the Beaufort Sea, however, observations indicate that aragonite became undersaturated year-round only from around the Year 2000, and that undersaturation was sporadic before that (Bellerby et al., 2018; Qi et al., 2017).

The low uptake rate in the Laptev Sea can be attributed to the high sea surface $p\text{CO}_2$ in the region, which in our simulation is largely at or above atmospheric $p\text{CO}_2$ throughout the model run. The high sea surface $p\text{CO}_2$ in the Laptev Sea is likely due to the influence of the Lena River, which reduces the sea surface TA (raising $p\text{CO}_2$) through dilution (the river's TA concentration is prescribed at 788 mmol m^{-3}) but has a less pronounced effect on sea surface DIC because the difference in concentration between the Lena River (prescribed at $1.658 \text{ mmol C m}^{-3}$) and the Laptev Sea is smaller.

The Beaufort Sea basin has the highest annual ice coverage ($\sim 90\%$), and the ice cover of the Beaufort Sea shelf drops from 80% in the first half of the standard run to 60% by the decade 2005–2015 (Figure 5e). The low carbon uptake in the Beaufort Sea basin is consistent with the generally high ice coverage, despite the sea surface $p\text{CO}_2$ in the region being below atmospheric $p\text{CO}_2$ (Figures 5a and 5b). The Beaufort Sea shelf exhibits one of the highest increases in average sea surface $p\text{CO}_2$ throughout the standard run (from ~ 320 to almost $400 \mu\text{atm}$, Figure 5b), despite the very low ocean carbon uptake (as shown in Figure 5a) in the region over the period of the standard run. The result that $p\text{CO}_2$ is increasing in the Beaufort Sea shelf, despite minimal local atmospheric carbon uptake, indicates that the trend in sea surface $p\text{CO}_2$ is not primarily due to local air-sea exchange. This result is at least partly consistent with Cai et al. (2010), who describe, through shipboard measurements taken in 1999 and 2008, a weak oceanic carbon uptake in the Beaufort Sea (both basin and shelf) along with an increase in sea surface $p\text{CO}_2$ with time. The increase in simulated $p\text{CO}_2$ in the Beaufort Sea shelf could be due to changes in one or more of the following: vertical mixing, horizontal advection from other water masses (e.g., low-pH Pacific water inflow), or the outflow of the Mackenzie River, with a high prescribed DIC concentration and low TA concentration (Table A1).

The simulated progression of annually averaged DIC in the upper 300 m of the water column for both the Beaufort Sea basin and shelf over the period of the standard run (Mortenson, 2019) is similar to that observed by Miller et al. (2014). Although the simulated surface Ω_{arag} is lower than observed, the decrease in subsurface Ω_{arag} over time (Figures 6a and 6b) is consistent with observations of both Qi et al. (2017) and Bellerby et al. (2018). As many of the observations described in Miller et al. (2014), Qi et al. (2017), and Bellerby et al. (2018) were taken in the summer months, we repeated the analysis of simulated Ω_{arag} for the summer period (July–September; Figures 6c and 6d). Simulated summer-averaged Ω_{arag} values are closer to observations than in the simulated annual averages, with higher values at the surface in both the Beaufort Sea basin and shelf and generally smaller values in the subsurface (particularly for the Beaufort Sea shelf; Figures 6e and 6f). The seasonal variability in Ω_{arag} shown in this study, along with the sparsity of observations in winter due to inaccessibility of the region, illustrates both the value of using model studies to fill in gaps in observable data sets and the limitations of extrapolating observations taken at particular times and places.

3.3. Sensitivity Analyses

Two simulations were carried out to determine the sensitivity of the modeled output to the implementations of an ice carbon pump and to sea ice algae. The first sensitivity run has the ice carbon pump suppressed (denoted the “noICP run”), with initial conditions from Year 1990 of the standard run and spanning a 20-yr time period from 1990–2010. The second sensitivity run has the ice algal production suppressed (denoted the “noIA run”), with the same initial conditions as the standard run (starting from Year 1969 with an independent spin-up period from 1969–1979) and analyzed over the same period as the standard run, from 1980–2015. The pan-Arctic total carbon uptake rate (seasonal and annual), and monthly and annual mean sea surface DIC and TA, and aragonite saturation state in the upper 50 m of the water column for the sensitivity and standard runs are presented in Figure 7.

3.3.1. No Ice Carbon Pump (noICP) Run

In the initial year of the noICP run, pan-Arctic net annual carbon uptake has a relatively large deviation from the standard run. After the first year, the pan-Arctic net annual carbon uptake rate adjusts so that there is very little difference between the noICP run and the standard run (Figure 7a). In the noICP run, both sea surface DIC and TA have a much diminished seasonal variability relative to the standard run (Figures 7b and 7c). The magnitude of the seasonal range in pan-Arctic sea surface DIC (TA) is reduced to $\sim 25\%$ ($\sim 10\%$) of the seasonal range in the standard run. The fact that the pan-Arctic annual ocean carbon uptake rate remains relatively unchanged ($\sim 1\%$ difference from the standard run) despite these changes can be explained first by the fact that the decrease in summer uptake in the noICP run is largely offset by the

Figure 6. Hovmueller plots of the vertical distribution of the annual averaged Ω_{arag} in the upper water column in the Beaufort Sea basin (a) and shelf (b) over the period of the standard run, averaged over only the summer months (July, August, and September) for each year (c and d), and the difference between the annual and summer averages (e and f). These plots are meant as a comparison to observations presented in Miller et al. (2014), Qi et al. (2017), and Bellerby et al. (2018).

increased uptake in winter and fall (Figure 7a) and second by the fact that the synchronous addition (or removal) of both DIC and TA inherent in the ice carbon pump have counteracting effects on seawater pCO_2 (Mortenson et al., 2018).

The annual mean pan-Arctic saturation state Ω_{arag} exhibits a similar trend in the noICP run as in the standard run, with an offset of ~ 0.02 . Over the period of the sensitivity run, none of the changes in sea surface carbon properties between the noICP and standard runs appear to be cumulative. Despite the similarity of annual, pan-Arctic averaged Ω_{arag} in the standard and noICP runs, there is a substantial difference in the seasonal cycles. The summertime maxima in the noICP run are ~ 0.075 higher than in the standard run. This indicates that models not representing the ice carbon pump may overestimate the annual summer maximum Ω_{arag} at the sea surface and hence underestimate the time at which species will be exposed to year-round undersaturation. In summary, the ice carbon pump does not appear to be of substantial importance for the annual mean oceanic carbon uptake but does substantially influence the seasonality of carbon system quantities.

Figure 7. (a) Total annual (black) and seasonal (summer = red, fall = orange, winter = blue, and spring = green) ocean carbon uptake of the Arctic Ocean north of 66.5°N (Tg C). The solid lines indicate the standard run (same as in Figure 3a). The dashed lines indicate the noICP run, and the dotted lines indicate the noIA run. Monthly (dashed) and annual (solid lines) pan-Arctic mean sea surface (b) DIC, (c) TA, and (d) aragonite saturation state Ω_{arag} .

3.3.2. No Ice Algae (noIA) Run

The suppression of ice algal production only has a small effect on the pan-Arctic monthly mean sea surface DIC and TA relative to the standard run (Figures 7b and 7c). However, the pan-Arctic annual net carbon uptake rate does show a difference between the standard and noIA runs, with the noIA run ranging between 7% and 15% less carbon uptake in the summer (Figure 7a). The mean reduction in carbon uptake in the noIA run relative to the standard run is 3.25% of the net annual pan-Arctic carbon uptake. This deficit, due to neglecting the role of ice algae, results in a net reduction in carbon uptake of 136 Tg carbon over the period 1980–2015; this value is on the order of 1 yr of pan-Arctic carbon uptake. As the deficit in carbon uptake between the noIA and standard runs accumulates over time, inclusion of ice algae is an important consideration in long-term simulations focused on Arctic Ocean carbon uptake.

The summertime saturation state Ω_{arag} is slightly lower in the noIA run than in the standard run, with a difference ranging between 0.3 and 0.8 over the period of comparison (Figure 7d). As discussed in Mortenson et al. (2018), ice algae have a twofold impact on the inorganic carbon system of the water column. First, the growth of ice algae is associated with a reduction of inorganic carbon. Second, and partially offsetting the first effect, the ice algae reduce the size of the pelagic bloom through competition for nutrients.

In summary, the inclusion of ice algae in a biogeochemical model for the Arctic Ocean has a relatively small, but cumulative, effect on the simulated ocean carbon system. Although the effect may be small over subdecadal time scales, over longer time scales, the accumulated deviation in net carbon uptake of the Arctic Ocean between the standard and noIA runs is more pronounced. This sensitivity analysis indicates that the sea ice ecosystem is an important component in the simulation of the Arctic Ocean inorganic carbon

system on time scales longer than a few decades, and especially on the order of centuries. In particular, CMIP5/CMIP6 models that do not include a representation of ice algae may underestimate changes to the polar oceans' sea surface carbonate system, especially for historical and 21st century experiments.

4. Conclusions

A sea ice ecosystem and a subsurface ice carbon pump were implemented in a biogeochemical model representing the Arctic Ocean in order to evaluate their effects on the inorganic carbon system. Carbon uptake and metrics of ocean acidification, including the saturation states Ω_{arag} and Ω_{calc} , were evaluated for the pan-Arctic domain and selected subregions. Simulations were conducted for the recent period of observed sea ice loss (from 1980–2015, with a spin-up period from 1969–1979). The simulated Arctic Ocean sea ice properties are in good agreement with observations of sea ice area and with a previous estimate of simulated sea ice volume, and the net pan-Arctic ocean carbon uptake is within the range of multiple previous studies. This study indicates that there is a $\sim 20\%$ increase in the pan-Arctic ocean uptake of carbon, from 110 to 135 Tg C yr^{-1} , during the period of the standard run. During the period 1980–2015, carbon uptake, open-ocean area, sea surface $p\text{CO}_2$, and temperature all increase coincident with decreases in saturation state (both Ω_{arag} and Ω_{calc}) and annually averaged pH (from 8.1 to 8) over the pan-Arctic surface ocean. The increase in pan-Arctic SST accounts for 10% of the pan-Arctic increase in sea surface $p\text{CO}_2$. The simulated pan-Arctic sea surface salinity shows a fresh bias relative to an interpolated observational data set (PHC) centered in the Canadian Polar Shelf and the Beaufort shelf and northern Chukchi Seas. Minima in Ω_{arag} and Ω_{calc} occur in winter, and regional and seasonal undersaturation of aragonite occurs in increasing frequency from the beginning to the end of the standard run.

Regional analyses indicate that the uptake of carbon in the seas adjacent to the Atlantic (Barents and Norwegian Seas) accounts for a large proportion of the increase in net annual Arctic Ocean carbon uptake over the period of sea ice loss. The Laptev Sea has particularly low pH (below 8.0) and high (sometimes supersaturated) $p\text{CO}_2$ (above 350 μatm), relative to the pan-Arctic mean, in line with observations. The Beaufort Sea shelf and basin exhibit increasing trends in mean annual $p\text{CO}_2$ (respectively 80 and 50 μatm) over the period 1980–2015 but low total annual carbon uptake (at or below 5 $\text{g C yr}^{-1} \text{ m}^{-2}$.) For the Beaufort Sea basin, the low carbon uptake is attributed to the relatively high ice cover. Lastly, the Canadian Polar Shelf does not exhibit a strong trend in ice cover or net annual carbon uptake but does show an accelerated increase in $p\text{CO}_2$ and decrease in pH relative to the pan-Arctic mean.

In the Arctic Ocean, due to prohibitively difficult conditions and costs during other seasons, many measurement campaigns focus on the summer months when the region is relatively accessible. Furthermore, proximity to research stations and ports can often influence decisions on where to make measurements. The strong seasonal and regional variability in the simulated inorganic carbon system properties, as described in this study, indicates that summertime-biased or region-biased observations have the potential for missing important information. These seasonal and regional sampling biases emphasize the importance of well-calibrated model studies which can be compared to available observations and fill in the gaps when assessing the state of the inorganic carbon system and carbon uptake of the Arctic Ocean.

Two sensitivity studies were performed to address the effect of two parameterizations implemented in this study. The first study tests the sea ice carbon pump of DIC and TA. Suppression of both DIC and TA fluxes associated with ice growth/melt shows little effect on the total uptake of carbon by the Arctic Ocean (North of 66.5°N) but a substantial decrease in the seasonal variability in DIC and TA, as well as an increase in the summertime saturation state at the sea surface. Neglecting the ice carbon pump results in an overestimate of the annual maximum simulated Ω_{arag} , which therefore underestimates the stress on aragonite-forming organisms in polar oceans. Differences between the standard and no ice carbon pump runs do not appear to increase with time. Neglect of ice algae results in a cumulative decrease in the pan-Arctic uptake of carbon of $\sim 3\%$ per year relative to the standard run. This analysis indicates that the simulated ice algae have a small effect on the simulated Arctic Ocean on subdecadal time scales, but the cumulative effect on carbon uptake of the Arctic Ocean indicates the sympagic ecosystem should be included in longer-term model runs.

The Arctic Ocean biogeochemical sea ice and ocean 3-D model, as modified in this study, can be used to address many other research questions. Other possible applications of the model could look at how the

inorganic carbon system is affected by changes to river runoff, precipitation, or the representation of the pelagic ecosystem. Another future research direction could be further analysis of the simulated ice carbon pump through differentially varying the flux of DIC relative to TA (difference in ice and seawater reference concentrations) associated with ice growth and melt.

This model study of the Arctic Ocean quantifies the (a priori predictable) increase in carbon uptake over the recent era of sea ice loss. Care must be taken extrapolating this result to conclude that sea ice loss implies that the Arctic will continue to increase in atmospheric carbon uptake into the coming decades. The Arctic Ocean's sensitivity in terms of the inorganic carbon system suggests feedbacks that would impede further increases in the uptake of carbon, both by stress on ecosystems through ocean acidification and by weakening the chemical buffering capacity of seawater. The model can be used to perform a detailed evaluation of the budget contributions which affect differences in regional, seasonal and interannual uptake and what that means for the Arctic as a carbon sink in the future. Therefore, an important next step is to utilize the model described in this study to examine near-future changes to predict how continued sea ice loss will affect the Arctic Ocean in terms of acidification and anthropogenic carbon uptake.

Appendix A: Meteorological Forcing and Ocean Boundary Conditions

All model simulations were forced at the surface using a prescribed meteorology based on the Drakkar Forcing Set 5.2 (DFS Dussin et al., 2016). The meteorological variables were precipitation (snow and total), temperature and humidity at 2 m above sea surface, short and longwave radiation, and zonal and meridional velocities at 10m. The DFS data are on a 0.7° grid with a time resolution of 3 hr for all meteorological variables except for precipitation and radiation, which had a daily time resolution. For the spin-up period (1969–1979), precipitation data were not available, so for this period 1979 data were repeated every year over that time period. Physical oceanographic variables (temperature, salinity, and horizontal currents) were initialized in 1969 based on Ocean Reanalysis System 4 (ORAS4 Balmaseda & Weaver, 2013). The physical oceanographic variables were prescribed for the open-ocean boundaries using interannual monthly mean fields, also based on ORAS4. Relaxation time scale for all variables was set to 1 day for inflow and 15 days for outflow following Dupont et al. (2015). The fields for biogeochemical variables (NO_3 , DIC, and TA) at the boundaries, as well as initialization, are based on climatological GLObal Ocean Data Analysis Project version 2 (GLODAP) data (Lauvset et al., 2016). Ecosystem functional groups were initialized at arbitrarily low values, as the time of initialization (January 1969) was at a biologically inert time of year. Lastly, the river flow was prescribed based on the monthly mean annually varying product of Dai et al. (2009) up to 2007. From 2007–2015, river flow from the Year 2007 is repeated annually. Biogeochemical concentrations for river flow (DIC, TA, and NO_3) are based on Cooper et al. (2008), Holmes et al. (2012), McClelland et al. (2012), and Tank et al. (2012) for each of the six respective major Arctic rivers (the Mackenzie and Yukon Rivers in North America, and, from east to west, the Kolima, Lena, Yenisey, and Ob' Rivers out of Eurasia). Organic and inorganic nutrients were combined at the time of outflow. For carbon, the organic and inorganic concentrations for each river were prescribed by the above sources for biogeochemical concentrations. For nitrogen, the organic concentration was assumed based on the DIN:DON ratio prescribed in CanOE. River flow concentrations for all rivers are given in Table A1.

Table A1

Prescribed Annual Mean River Flow Concentrations of Biogeochemical Variables at the Six Major Arctic Rivers in the Model Simulation

Variable	Mackenzie	Yukon	Kolyma	Lena	Yenisey	Ob'
DIC (mmol C m^{-3})	1,717	1,792	642	817	908	1,175
DOC (mmol C m^{-3})	368	544	592	841	611	785
DIC+DOC (mmol C m^{-3})	2,085	2,336	1,234	1,658	1,519	1,955
TA (mmol m^{-3})	1,540	1,707	449	788	845	1,181
DON (mmol N m^{-3})	7.1	14.2	7.2	14.4	14.2	7.1
NO_3 (mmol N m^{-3})	7.1	7.1	3.6	3.6	7.1	7.1

Appendix B: Regional Trends in Carbonate System Surface Properties/Fluxes

This section includes tables (Tables B1–B6) highlighting trends in the pan-Arctic mean and six regions with substantial deviations from the pan-Arctic mean for the following variables relevant to sea surface carbon system properties and to air-sea flux of carbon: pH, sea surface $p\text{CO}_2$, mean carbon flux, Ω_{arag} and Ω_{calc} , and ice coverage.

Table B1
Trends in pH Over the Standard Run

Region	Initial	Final	Difference	% Difference	Difference/decade
pan-Arctic	8.1	8.02	−0.08	−0.99	−0.02
Canadian Polar Shelf	8.18	8.06	−0.11	−1.32	−0.03
Beaufort Shelf	8.05	7.95	−0.1	−1.2	−0.03
Beaufort Basin	8.09	7.98	−0.07	−0.92	−0.02
Laptev	7.96	7.92	−0.06	−0.8	−0.02
Barents	8.15	8.07	−0.08	−1.0	−0.02
Norwegian	8.13	8.06	−0.07	−0.89	−0.02

Note. The initial value is the annual mean for the first year of the standard run (1981), the final value is the annual mean of the last year of the standard run (2015), and the difference, percent difference, and difference/decade are based on trends analyzed over the entire standard run period (1981–2015).

Table B2
Changes to Sea Surface $p\text{CO}_2$ (μatm) Over the Standard Run

Region	Initial	Final	Difference	% Difference	Difference/decade
pan-Arctic	295.4	356.95	60.96	18.8	16.93
Canadian Polar Shelf	258.48	335.05	70.58	23.9	19.61
Beaufort Shelf	318.38	384.6	76.63	22.2	21.29
Beaufort Basin	283.94	355.49	45.28	14.56	12.58
Laptev	377.67	408.74	48.74	12.81	13.54
Barents	278.33	339.18	62.28	20.48	17.3
Norwegian	299.56	353.66	59.25	18.36	16.46

Note. Headings are the same as in Table B1.

Table B3
Changes to Air-Sea Flux of CO_2 ($\text{mg C m}^{-2} \text{yr}^{-1}$, Positive Is Into the Ocean) Over the Standard Run

Region	Initial	Final	Difference	% Difference	Difference/decade
pan-Arctic	8.5	11.79	2.24	22.33	0.62
Canadian Polar Shelf	13.79	13.42	−0.79	−5.93	−0.22
Beaufort Shelf	3.89	5.44	−0.4	−11.59	−0.11
Beaufort Basin	2.62	4.67	2.17	61.0	0.6
Laptev	2.49	2.59	0.58	20.44	0.16
Barents	23.23	31.56	4.43	15.97	1.23
Norwegian	19.19	28.4	5.52	24.07	1.53

Note. Headings are the same as in Table B1.

Table B4
Changes to Sea Surface Ω_{arag} Over the Standard Run

Region	Initial	Final	Difference	% Difference	Difference/decade
pan-Arctic	1.23	1.05	−0.18	−15.83	−0.05
Canadian Polar Shelf	1.55	1.16	−0.32	−24.64	−0.09
Beaufort Shelf	0.97	0.72	−0.14	−17.48	−0.04
Beaufort Basin	1.01	0.77	−0.17	−19.87	−0.05
Laptev	0.73	0.7	−0.1	−13.56	−0.03
Barents	1.67	1.5	−0.22	−13.51	−0.06
Norwegian	1.69	1.5	−0.17	−10.68	−0.05

Note. Headings are the same as in Table B1.

Table B5
Changes to Sea Surface Ω_{calc} Over the Standard Run

Region	Initial	Final	Difference	Fraction Difference	Difference/decade
pan-Arctic	1.98	1.68	−0.29	−15.93	−0.08
Canadian Polar Shelf	2.49	1.87	−0.52	−24.65	−0.14
Beaufort Shelf	1.56	1.17	−0.23	−17.85	−0.06
Beaufort Basin	1.63	1.24	−0.28	−19.81	−0.08
Laptev	1.19	1.14	−0.16	−13.76	−0.05
Barents	2.67	2.38	−0.35	−13.62	−0.1
Norwegian	2.69	2.38	−0.27	−10.84	−0.08

Note. Headings are the same as in Table B1.

Table B6
Changes to Ice Coverage (% of Total Area) Over the Standard Run

Region	Initial	Final	Difference	% Difference	Difference/decade
pan-Arctic	0.71	0.63	−0.09	−13.11	−0.02
Canadian Polar Shelf	0.58	0.58	−0.04	−7.73	−0.01
Beaufort Shelf	0.76	0.67	−0.17	−23.95	−0.05
Beaufort Basin	0.94	0.86	−0.06	−6.8	−0.02
Laptev	0.79	0.7	−0.13	−16.4	−0.04
Barents	0.35	0.21	−0.12	−42.31	−0.03
Norwegian	0.21	0.15	−0.09	−45.86	−0.03

Note. Headings are the same as in Table B1.

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Data Availability Statement

Data supporting this study can be found online (at https://zenodo.org/record/3479936#.XaVP_i1L3UI).

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