Causes of increased dissolved inorganic carbon in the subsurface layers in the western shelfbreak and high latitudes basin in the Arctic Pacific sector

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Abstract
The expansion of dissolved inorganic carbon (DIC)-rich water carried by the Pacific inflow creates a DIC maximum layer and exerts important influences on ocean acidification in the subsurface Arctic Ocean. This study analyzed shifts in the DIC distribution of the subsurface Arctic Ocean during 1998–2015 through hindcast simulation using a three-dimensional ocean-sea ice-biogeochemical model. For this purpose, the study was divided into two time periods (1998–2007 and 2008–2015). The results showed that the lower boundary layer of the Pacific Winter Water, defined as an isopycnal of 27 kg m\(^{-3}\), became deeper by \(\sim 50\) m in the central Canada Basin and expanded northward during 2008–2015 relative to 1998–2007. Accordingly, the subsurface DIC maximum layer deepened and expanded northwards into the Makarov Basin at high latitudes around 85\(^\circ\) N. During 2008–2015, DIC concentrations, averaged over a 50–250 m water column, increased significantly in the Chukchi-East Siberian Shelfbreak and Makarov Basin. The DIC increase over the shelfbreak is mainly attributable to increased local biological degradation and the transportation of DIC-rich water from the Chukchi Shelf through Barrow Canyon. Estimates of the DIC budget indicated that advection controlled the increase in DIC content in the Makarov Basin during 2008–2015. This is attributed to the shift of the ocean circulation pattern, in which the ocean current along the Chukchi-East Siberian Slope to the Makarov Basin became stronger during 2008–2015, promoting the transport of DIC-rich Pacific Water into the Makarov Basin.

1. Introduction
The Pacific inflow transports water rich in dissolved inorganic carbon (DIC) [1, 2] through the Bering Strait to the Arctic deep basins [3, 4], contributing to the subsurface DIC maximum layer in the Arctic Ocean [5, 6]. This layer is generally located in the core Upper Halocline Layer with a salinity of \(\sim 33\) [5, 7]. DIC concentrations within it decrease from the Bering Strait to the Canada Basin in the horizontal orientation [2], and vary interannually with environmental changes. Chen [8] found that DIC concentrations in the subsurface of the Makarov Basin and the western Canada Basin (80\(^\circ\) N, 170\(^\circ\) W) were higher in 2010 than in 1994. They suggested that this variation was associated with the higher fractional contribution of Pacific Water in 2010. High DIC concentrations act as an indicator of ocean acidification, along with low calcium carbonate saturation [2, 9]. With reference to 1997, the calcium carbonate saturation in the 100–200 m subsurface water in the Canada Basin decreased significantly in 2008 [10]. Qi et al [9] analyzed observation-based aragonite saturation along multiple transects (150\(^\circ\) W–170\(^\circ\) W) in the western Arctic Ocean and reported that subsurface acidified waters expanded to higher latitudes from 1994 to 2010. This suggests shifts in the expansion of acidified Pacific Water in the Arctic Ocean.

Under Borealization, the Arctic Ocean has been undergoing rapid environmental changes in recent decades [11]. Satellite records show a downward trend of sea ice in the Arctic Ocean from 1979 to present [12, 13]. However, Lewis et al [14] found that the increasing rate of annual open water area (<50% sea ice cover) has slowed considerably after 2008, while Perovich et al [15] reported a slower
decline of sea ice extent in September after 2007 following an accelerated extent loss during 1993–2006. The Pacific inflow carrying abundant acidified water \cite{10, 16} entered the Arctic Ocean at a mean increasing rate of 0.01 Sv yr\(^{-1}\) from 1990 to 2015 \cite{17, 18} and the interface of Pacific water migrated over and back from the Makarov Basin \cite{19}. The ocean circulation pattern in the Arctic Ocean has also changed. Recently, the center of the Beaufort Gyre (BG) moved towards the southwestern Canada Basin and BG spun up after 2007 \cite{20}. Ocean currents presented a long-term increase in the southwestern BG during 2003–2014, and accelerated in late 2007 with higher speeds sustained until 2011 in the southeastern BG \cite{21, 22}. Accordingly, the transport pathway of Pacific Winter Water (PWW) was altered, shifting to the northwest toward the Chukchi Borderland and thickening and expanding with depth \cite{20}. These findings reflect an obvious change in sea ice and ocean circulation after 2007. Therefore, the responses of DIC distributions and associated ocean acidification in the Arctic Ocean are being widely discussed. Previous studies presented snapshots of biogeochemical characteristics along single or multiple transects through limited cruises \cite{5, 9, 23}. However, the continuous spatiotemporal DIC distribution in the Arctic Ocean and its response to Pacific Water expansion remain unclear. Zhong et al \cite{20} explored the expansion of Pacific Water during 2012–2016 using observations from multiple sources. These samples were mainly collected from the Canada Basin. Questions that remain to be resolved include what other regions will be affected by the expansion of Pacific Water? What are the underlying mechanisms of DIC changes in the vast Arctic Ocean?

In this study, the characteristics of DIC concentration in various Arctic water masses were first analyzed through a decadal hindcast simulation (1998–2015) using an ocean-sea ice-biogeochemical model and field measurements. To this end, data from 12 scientific investigation projects encompassing 27 cruises from 1994 to 2015 were collected (table 1), and the locations of stations were shown in figure 1(a). The shift of DIC-rich water distribution in the Arctic Pacific Sector was explored by dividing the 1998–2015 hindcast simulation into two time periods, 1998–2007 (10 years) and 2008–2015 (8 years), considering shifts in sea ice and ocean circulation in recent decades. Finally, the main controls on the increased DIC during 2008–2015 relative to 1998–2007 in the Chukchi-East Siberian Shelfbreak and the Makarov Basin were identified.

2. Model description and \textit{in situ} measurements

The three-dimensional ocean-sea ice-biogeochemical model (NAPA-BGC) includes the Nucleus for European Modelling of the Ocean (NEMO3.6 \cite{24}), the Louvain-la-Neuve Ice Model (LIM3 \cite{25, 26}), and a modified biogeochemical model of the Pelagic Interactions Scheme for Carbon and Ecosystem Studies (PISCES-v2 \cite{27}). There are 19 prognostic variables in the biogeochemical component, containing the carbonate system.

NAPA represents an abbreviated notation of model domain covering the North Atlantic Ocean (north of 26° N), the North Pacific Ocean (north of 45° N), and the entire Arctic Ocean. The nominal horizontal resolution of NAPA-BGC is 1/4° in longitude and latitude. The vertical space is discretized into 75 layers. The hindcast simulation of NAPA-BGC covers a period from 1 October 1993 to 31 December 2015. In this study, 5 \& averaged model fields were analyzed. The detailed configuration and evaluation of the coupled ocean and sea ice model are provided in Luo et al \cite{28} and Zhang et al \cite{29}. The description and assessment of the biogeochemical model are documented in Wei et al \cite{30} and Zheng et al \cite{31, 32}.

NAPA-BGC captured interannual variations in Arctic sea ice coverage, including the extremely low ice condition in the summers of 2007 and 2012, through comparisons with the climate data record of passive microwave sea ice concentration \cite{28}. For the ice thickness, the model provided consistent variations with drill-hole observations in the Chukchi and Beaufort Seas during 2002–2004 \cite{33} with bias = 0.1 m, and the two time series presented a significant correlation of \( r = 0.80 (p < 0.01) \) \cite{29}. The simulated interannual velocity anomalies through the Bering Strait during 1998–2015 matched well with the mooring observation by Woodgate \cite{18}, yielding a significant correlation coefficient of 0.88 (\( p < 0.01 \)) \cite{34}. The main ocean circulation pattern \cite{28} and structure of water masses were reproduced \cite{35}. NAPA-BGC provided a reasonable vertical distribution of water masses, including melt-water (MW) in the upper thin layer, Pacific Summer Water within 50–100 m, PWW centered at 150 m, and Atlantic Water (AW) below 200 m in the Canada Basin \cite{35}. In this study, we further evaluated the model representation of temperature, salinity, density, and DIC concentration based on \textit{in situ} measurements from the 27 cruises during 1994–2015 covering the entire Arctic Ocean. These measurements were obtained from various sources, namely, GEOTRACES, Western Arctic Shelf Basin Interactions Project (SBI), Arctic Ocean Section cruise (AOS), Chinese Arctic Research Expedition (CHINARE), Impacts of Climate on the Ecosystems and Chemistry of the Arctic Pacific Environment (ICECAPE), Study of Under-ice Blooms In the Chukchi Ecosystem (SUBICE), International Siberian Shelf Study (ISSS), Joint Ocean Ice System Study (JOIS), Surface Heat Budget of the Arctic Ocean Project (SHEBA), Swedish Icebreaker Oden cruises, Polarstern cruises, and AKADEMIEK FEDOROV cruise (table 1).
| Project          | Research vessel | Cruise | Sampling date               | Source                                                                 |
|------------------|-----------------|--------|-----------------------------|------------------------------------------------------------------------|
| AKADEMIK FEDOROV | TUNDRA94        | 06 July 1994–07 August 1994 | Anderson L. 2015. Bottle data from Cruise 90AQ19940706, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/90AQ19940706. Access date 23 April 2015 |
| AOS              | Louis St Laurent| AOS94  | 26 July 1994–01 September 1994 | Swift J, Carmack E and Aagaard K 2015. Bottle data from Cruise 18SN19940724, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/18SN19940724. Access date 23 April 2015 |
| Oden             | AOS05           | 21 August 2005–23 September 2005 | Anderson L. 2015. Bottle data from Cruise 77DN20050819, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/77DN20050819. Access date 23 April 2015 |
| Polarstern cruise| Polarstern       | ARK XII| 24 July 1996–04 September 1996 | Anderson L. 2015. Bottle data from Cruise 06AQ19960712, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/06AQ19960712. Access date 23 April 2015 |
| Polarstern cruise|                 | ARK XXII/2| 29 July 2007–24 September 2007 | Ober S, van Heuven S 2015. Bottle data from Cruise 06AQ20070728, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/06AQ20070728. Access date 23 April 2015 |
| Polarstern cruise|                 | ARK XXVI/3| 29 August 2011–26 September 2011 | Anderson L. 2018. Bottle data from Cruise 06AQ20110805, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/06AQ20110805. Access date 09 October 2018. |
| Polarstern cruise|                 | ARK XXIX/3| 21 August 2015–23 September 2015 | Jones E and Ulfsbo A 2018. Bottle data from Cruise 06AQ20150817, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/06AQ20150817. Access date 24 January 2018. |
| GEOTRACES        | Healy           | HLY1502| 12 August 2015–07 October 2015 | Swift J, Carmack E and Aagaard K 2015. Bottle data from Cruise 18SN19940724, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/18SN19940724. Access date 23 April 2015. |
| CHINARE          | Xuelong         | CHINARE03| 28 July 2003–18 August 2003 | Zhongyong Gao. DIC data sets in Arctic during CHINARE-2nd (2003), National Arctic and Antarctic Data Center, 2019. |
| CHINARE          |                 | CHINARE08| 01 August 2008–08 September 2008 | Zhongyong Gao. DIC data sets in Arctic during CHINARE-3rd (2008), National Arctic and Antarctic Data Center, 2019. |

(Continued.)
| Project | Research vessel | Cruise | Sampling date | Source |
|---------|----------------|--------|---------------|--------|
| CHINARE10 |                | 15 July 2010–29 August 2010 | Zhongyong Gao. DIC data sets in Arctic during CHINARE-4th (2010), National Arctic and Antarctic Data Center, 2019. |
| CHINARE12 |                | 15 July 2012–07 September 2012 | Zhongyong Gao. DIC data sets in Arctic during CHINARE-5th (2012), National Arctic and Antarctic Data Center, 2019. |
| CHINARE14 |                | 22 July 2014–29 August 2014 | Di Qi. DIC data sets in Arctic during CHINARE-6th (2014), National Arctic and Antarctic Data Center, 2019. |
| ICESCAPE | Healy          | HLY1001 | 18 June 2010–16 July 2010 | Ocean Biogeochemistry Lab. 2011. [http://ocean.stanford.edu/icescape/#hly1001](http://ocean.stanford.edu/icescape/#hly1001). |
|          |                | HLY1101 | 28 June 2011–24 July 2011 | Ocean Biogeochemistry Lab. 2011. [http://ocean.stanford.edu/icescape/#hly1101](http://ocean.stanford.edu/icescape/#hly1101). |
| SUBICE   | Healy          | HLY1401 | 15 May 2014–20 June 2014 | Ocean Biogeochemistry Lab. 2014. [http://ocean.stanford.edu/subice](http://ocean.stanford.edu/subice). |
| ISSS     | Yacov Smirnitsky | ISSS08 | 18 August 2008–18 September 2008 | Anderson L. 2015. Bottle data from Cruise 90JS20080815, exchange version. Accessed from CCHDO [https://cchdo.ucsd.edu/cruise/90JS20080815](https://cchdo.ucsd.edu/cruise/90JS20080815). Access date 23 April 2015. |
| JOIS     | Louis St Laurent | JOIS97 Leg IV | 24 September 1997–15 October 1997 | Jones P and Azetsu-Scott K 2015. Bottle data from Cruise 18SN19970924, exchange version. Accessed from CCHDO [https://cchdo.ucsd.edu/cruise/18SN19970924](https://cchdo.ucsd.edu/cruise/18SN19970924). Access date 23 April 2015. |
|          |                | JOIS15  | 21 September 2015–14 October 2015 | Williams B 2019. Bottle data from Cruise 18SN20150920, exchange version. Accessed from CCHDO [https://cchdo.ucsd.edu/cruise/18SN20150920](https://cchdo.ucsd.edu/cruise/18SN20150920). Access date 25 June 2019. |
| Oden cruises | Oden         | ODEN02  | 26 April 2002–30 May 2002 | Anderson L. 2015. Bottle data from Cruise 77DN20020420, exchange version. Accessed from CCHDO [https://cchdo.ucsd.edu/cruise/77DN20020420](https://cchdo.ucsd.edu/cruise/77DN20020420). Access date 23 April 2015. |
|          |                | ODEN07  | 15 August 2007–07 September 2007 | Anderson L. 2018. Bottle data from Cruise 77DN20070812, exchange version. Accessed from CCHDO [https://cchdo.ucsd.edu/cruise/77DN20070812](https://cchdo.ucsd.edu/cruise/77DN20070812). Access date 23 February 2018. |
|          |                | ODEN14  | 15 July 2014–25 September 2014 | Anderson L. 2015. Hydrochemistry measured on water bottle samples in the Arctic Ocean during Oden cruise SWERUS-C3 (77DN20140705). PANGAEA, [https://doi.org/10.1594/PANGAEA.843909](https://doi.org/10.1594/PANGAEA.843909). |
Table 1. (Continued.)

| Project | Research vessel | Cruise   | Sampling date       | Source                                                                 |
|---------|-----------------|----------|---------------------|----------------------------------------------------------------------|
| SBI     | Healy           | HLY0201  | 08 May 2002–12 June 2002 | Bates N 2015. Bottle data from Cruise 32H120020505, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/32H120020505. Access date 23 April 2015. |
|         |                 | HLY0203  | 18 July 2002–21 August 2002 | Bates N 2015. Bottle data from Cruise 32H120020718, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/32H120020718. Access date 23 April 2015. |
|         |                 | HLY0402  | 17 May 2004–21 June 2004 | Bates N 2015. Bottle data from Cruise 32H120040515, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/32H120040515. Access date 23 April 2015. |
|         |                 | HLY0403  | 19 July 2004–24 August 2004 | Bates N 2015. Bottle data from Cruise 32H120040718, exchange version. Accessed from CCHDO https://cchdo.ucsd.edu/cruise/32H120040718. Access date 23 April 2015. |
| SHEBA   | Des Groseilliers| SHEBA98 | 15 November 1997–30 September 1998 | Cota G, Siferd T, Pomeroy L. 2007. Arctic Nutrient Database 1997–1998. Version 1.0. UCAR/NCAR—Earth Observing Laboratory; 10.5065/D6DN43FS. Accessed 20 July 2021. |

3. Extension of DIC-rich water

Taylor diagrams are used to present the quality of model simulations by expressing correlation, normalized centered root-mean-square error, and normalized standard deviations with observations [36]. Figures 2(a)–(c) respectively show the Taylor diagrams of observed and corresponding simulated temperature, salinity, and DIC concentration within the water column. In general, the correlation coefficients concerning the three variables exceeded 0.6, and approximately 60% and 90% of the cruises exhibited higher correlation (coefficients larger than 0.8) for temperature and salinity, respectively. Moreover, the simulated and observed DIC concentrations presented significant correlation with coefficients greater than 0.7 for 75% of the cruises.

In addition, the widely used cost function (CF) proposed by OSPAR Commission [37] was applied for the uncertainty assessments:

$$\text{CF} = \frac{1}{N} \sum \frac{|\text{obs} - \text{mod}|}{\text{std(obs)}}$$

where \(\text{obs}\) and \(\text{mod}\) are values of observation and model, respectively, \(\text{std(obs)}\) denotes the standard deviation of observation, and \(N\) is the number of observations. This quantifies the goodness of fit between model and observation. The criteria for CF are as follows: \(<1 = \text{very good}, 1–2 = \text{good}, 2–3 = \text{reasonable}, >3 = \text{poor}\) [38]. The results of CF for DIC ranged from 0.41–0.89 depending on cruise with the mean value being 0.45. For temperature, CFs ranged within 0.17–0.70. For salinity, CFs were less than 0.93 for all cruises except for the SUBICE cruise in 2014 (CF = 1.04). These indicate that NAPA-BGC captured the main spatial distribution of temperature, salinity, and DIC concentration in the Arctic Ocean.

Following Itoh et al [39] and Pisareva et al [40], five water masses in the Arctic Ocean were recognized, including Alaskan Coastal Water (ACW: \(S < 32\) and \(T > 3\) °C), Bering Summer Water (BSW: \(S < 32\) and \(0 < T < 3\) °C or \(32 < S < 33.64\) and \(T > 0\) °C), Sea-ice MW (\(S < 31.5\) and \(T < 0\) °C), PWW (\(31.5 < S < 33.64\) and \(T < 0\) °C or \(S > 33.64\) and \(T < -1.26\) °C), and AW (\(S > 33.64\) and \(T > -1.26\) °C). Similar to the observed features, the model simulated temperature-salinity-DIC diagram indicates that water masses with high DIC concentrations were mainly distributed in PWW, but a certain high-DIC mass was observed in AW, which could be attributed to mixing between PWW and AW (figures 2(d) and (e)). This was identified in previous research [2, 6, 41]. Density is also applied to distinguish water masses.
PWW is usually characterized as water bounded by isopycnals of 26 and 27 kg m\(^{-3}\) [20]. Diagrams of observed and corresponding simulated density versus DIC revealed the occurrence of high DIC masses mainly in PWW, although the simulated DIC was lower than the observed DIC (figures 2(f) and (g)).
We also found that the simulated density of surface water was higher than the corresponding observation, but the bias rapidly decreased with increasing depth. Considering this bias, the density of 27 kg m\(^{-3}\) was used to determine the extension of the lower boundary of PWW.

Variations in the vertical distribution of DIC were presented along the meridional transects in 1994, 2008, and 2015 (figures 3(a)–(i)). The positions of these sampling transects in the 3 years were similarly designed along 170° W, extending from the Bering Strait to the North Pole, crossing the Chukchi Shelf, Canada Basin and Makarov Basin. Both observation and corresponding simulation provided consistent vertical DIC distributions in each year. From the sea surface to the deep, DIC concentration presented a
Figure 3. (a) Observed and (b) simulated DIC distributions along the transect marked with red dots in the subfigure of (a) in 1994. (c) Scatter plot of observed and simulated DIC in the top 500 m of the transect in 1994. Color shading represents sampling depth. X and Y axes represent observed and simulated DIC, respectively. Panels (d)–(f) and (g)–(i) show the corresponding information in 2008 and 2015, respectively.

The multi-year averaged depth of the lower boundary of PWW (isopycnal of 27 kg m$^{-3}$) and DIC concentration at this depth during 1998–2007 and 2008–2015 were analyzed. The model results showed that the lower boundary layer of PWW was generally deeper in the Canada Basin and gradually became shallower towards the surroundings in both time periods (figures 4(a) and (b)). Compared with the situation during 1998–2007, the lower boundary layer deepened to $\sim$250 m from $\sim$200 m in the central Canada Basin and expanded horizontally to the Makarov Basin, represented by the extensions of isolines of 100 and 150 m during 2008–2015. Variations in PWW explained some changes in the halocline in the Arctic Ocean. Previous studies revealed the deepening halocline in the Arctic Pacific Sector...
recently, especially in the BG region [11, 43]. Zhong et al [21] reported that the 33.1 isohaline (core salinity of PWW) in the Canada Basin deepened by ∼40 m in 2011 relative to 2003. A long time series of mooring data in the Canada Basin (77° N, 150° W) revealed that the isopycnal of 27 kg m$^{-3}$ deepened by ∼70 m from 2003 to 2011, reaching a depth of 250 m in 2011, attributable to the lateral flux convergence of PWW and Ekman pumping [20]. In addition, the deepened PWW in the Canada Basin is also consistent with the increased freshwater content under the persistent anticyclonic atmospheric wind forcing [44, 45], as well as the deepened nutricline [43]. Chen [8] detected PWW in the Makarov Basin in 2010 but in a retrospective analysis of previously collected data from 1994 did not. These observations are consistent with Anderson et al [46] who reported the occurrence of the maximum layer of nutrients in the Makarov Basin in 2014 but not in 1996. The observed findings were reasonably reproduced by NAPA-BGC and provided evidence for the simulated expansion of PWW towards Makarov Basin.
At the lower boundary of PWW, the multi-year averaged simulated DIC concentration generally increased during 2008–2015 (figures 4(c) and (d)). A DIC isopleth of 2180 µmol kg\(^{-1}\) extended to the Makarov Basin along the Chukchi-East Siberian Shelfbreak, where DIC exceeded 2220 µmol kg\(^{-1}\), which was higher than that during 1998–2007 by approximately 40–60 µmol kg\(^{-1}\) on average. Meanwhile, the DIC concentration in the central Canada Basin presented a relatively slight increase of ~20 µmol kg\(^{-1}\) between the two time periods studied.

4. Causes of increased DIC

The difference in DIC concentration averaged within the 50–250 m water column between 2008–2015 and 1998–2007 (figure 5(a)) showed that the DIC concentration generally increased in the subsurface Arctic Pacific Sector, especially in the Chukchi-East Siberian Shelfbreak and Makarov Basin. A slight change was simulated in the central Canada Basin. The DIC budget was estimated to quantitatively evaluate the relative contribution of physical and biological processes to the increased DIC concentration. The governing equation is as follows:

\[
\frac{\partial \text{DIC}}{\partial t} + \text{adv}(\text{DIC}) = \text{diff}(\text{DIC}) + \text{bio}(\text{DIC}) + \text{atm}(\text{DIC}) + \text{riv}(\text{DIC})
\]

where \(\text{adv}\) and \(\text{diff}\) represent advection and diffusion, \(\text{bio}\) represents biological processes mainly including photosynthesis and degradation, and \(\text{atm}\) and \(\text{riv}\) are air-sea carbon dioxide (\(\text{CO}_2\)) exchange and river load, respectively. The final two terms play minor roles in DIC budget within the 50–250 m water column. The diffusion term was divided into vertical and lateral diffusion. Thus, the DIC budget is mainly controlled by four factors: advection (\(\text{adv}\)), net biological effect (\(\text{bio}\)), and vertical (\(\text{zdf}\)) and lateral (\(\text{ldf}\)) diffusion. The relative contribution was calculated according to equation (3), taking the net biological effect as an example:

\[
\text{Relative contribution} = \frac{\text{bio}}{|\text{bio}| + |\text{adv}| + |\text{ldf}| + |\text{zdf}|}
\]

where the denominator is the sum of the absolute values of all terms.

The Chukchi-East Siberian Shelfbreak is mainly influenced by two branches of Pacific Water. One emanates from the Barrow Canyon with a certain fraction continually flowing westward to the East Siberian Slope after separating at the Chukchi Borderland. The other flows towards the Chukchi-East Siberian Shelfbreak after passing through Herald Canyon on the Chukchi Shelf [40, 47]. The transects of Barrow Canyon and Herald Canyon were selected to calculate the simulated annual mean volume flux, DIC concentration, and DIC flux of PWW (figure 5(c)). The volume flux of all water masses through Barrow Canyon averaged over 1998–2015 was 0.48 Sv (1 Sv = 10\(^{6}\) m\(^3\) s\(^{-1}\)), which was six times more than that through Herald Canyon. This annual mean volume transport is similar to the Barrow Canyon flux estimated by Itoh et al (0.44 ± 0.07 Sv) [48] based on mooring data (2000–2008) and Corlett and Pickart (0.53 ± 0.11 Sv) [47] derived from shipboard hydrographic and velocity data (2002–2014). PWW accounted for 43%–86% and 58%–100% of the total volume fluxes passing through Barrow Canyon and Herald Canyon, respectively, from 1998 to 2015. In both transects (figure 5(c)), the DIC flux of PWW presented an increasing trend during 1998–2015. The annual DIC concentration of PWW was relatively higher after 2007. Relative to 1998–2007, DIC concentration increased by 29 and 36 µmol kg\(^{-1}\) in Barrow Canyon and Herald Canyon, respectively, during 2008–2015. A previous study attributed this recent increased DIC concentration to the enhanced remineralization of organic carbon fixed by the increased primary production, which released \(\text{CO}_2\) into PWW when flowing over the Chukchi Shelf bottom [9]. Arrigo and van Dijken [49] suggested that annual net primary production in the Chukchi Sea, estimated from satellites, was higher during 2007–2012 than during 1998–2006. NAPA-BGC also captured the characteristic of the overall higher annual primary production occurring after the extremely low ice year of 2007 in the Chukchi Shelf [31], and increased biological organic carbon in the shelfbreak fueled by nutrients from the Pacific. In contrast, Yun et al [50] reported a recent decrease in summertime primary production from 1974 to 2012 indicated by \textit{in situ} measurements with inconsistent spatial and temporal coverage in the Chukchi Sea. However, this comparison is weak and limited because it is unclear whether any differences are simply the result of differences in the sampling time and location, in methodology, and most importantly, whether the changes are due to the lack of comprehensive spatial and temporal coverage [51]. Samplings from the Distributed Biological Observatory occurring at the same time (mid-July) and using consistent methodology indicated strong interannual variabilities of total chlorophyll-a concentration and primary productivity in the northern Bering Sea and Chukchi Sea, with no clear increasing or decreasing trends from 2006 to 2016 [51]. But it is still unknown whether this long-term trend exists in each phytoplankton growing month depending on \textit{in situ} measurements. Satellites provide a more comprehensive dataset of continuous spatial-temporal scale. The long-term increasing trend of annual satellite-based primary production and its underlying explanation are widely acceptable at least.
for now [14, 49, 52–56]. Other coupled ice-ocean-ecosystem models participating in the Forum for Arctic Modeling and Observational Synthesis project also suggested increases in annual primary production corresponding to decreases in sea ice in the Arctic Ocean including the Chukchi-East Siberian Shelfbreak during 1980–2009 [57]. Based on NAPA-BGC simulations, the DIC budget over the shelfbreak (outlined in black in figure 5(a)) within 50–250 m suggests that during 2008–2015, the net biological effect controlled the DIC source with a relative contribution of 40%, followed by advection (9%). Biological degradation accounted for >93% of the net biological effect, and was highly correlated with the annual primary production in the whole water column ($r = 0.99$, $p < 0.01$), with an increasing trend from 1998 to 2015.

Therefore, the additional DIC along the Chukchi-East Siberian Shelfbreak was mainly driven by the increased local biological degradation and the transportation of DIC-richer water from the Chukchi Shelf through Barrow Canyon.

The Makarov Basin, located west of the Mendeleev-Alpha Ridge (80–88° N), is a region heavily covered by ice (>85%) [8]. DIC concentration in this region also presented a much higher level after 2007. For a representative area of the Makarov Basin (outlined in red in figure 5(a)), the DIC budget within 50–250 m during the two time periods was estimated. During 1998–2007 (figure 5(b)), the net annual DIC content was increased with an amount of $4.14 \times 10^{13}$ mmol. This was mainly attributable to net biological effect (19%), lateral diffusion (20%),

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**Figure 5.** (a) Difference in DIC concentration averaged within 50–250 m water column between 2008–2015 and 1998–2007. The representative areas of the Makarov Basin and Chukchi-East Siberian Shelfbreak are outlined in the red and black. The DIC budgets (b) during 1998–2007 (blue font color) and 2008–2015 (red font color) were calculated in the Makarov Basin. Inward/outward arrows represent source/sink terms. The black and blue transects in (a) represent Barrow Canyon and Herald Canyon, respectively. (c) Annual mean PWW volume flux, DIC flux, and DIC concentration of Barrow Canyon (marked by star) and Herald Canyon (marked by circle) from 1998 to 2015.
and advection (15%), while vertical diffusion (46%) decreased DIC, acting as a DIC sink. During 2008–2015, the roles of source and sink terms were invariable. The increase in net annual DIC content was up to $1.80 \times 10^{14}$ mmol. This was mainly attributable to enhanced advection with the relative contribution increasing to 31%. The advected DIC in the Makarov Basin was 2.6 times higher than that during 1998–2007. Relative to 1998–2007, the contributions of net biological effect and lateral diffusion were almost constant, and the corresponding DIC amounts were 1.1 times higher. The enhancement of advection closely followed the changed ocean circulation pattern (figures 1(b) and (d)). During 2008–2015, there is a stronger simulated westward current along the Chukchi-East Siberian Slope to the Makarov Basin, favoring DIC transport to the Makarov Basin. A previous study suggested that the volume transport of Pacific water in the Chukchi Slope Current increased from the early regime of 2002–2004 to the recent regime covering 2009–2014 [47]. In addition, NAPA-BGC simulated a northward shift of a portion of the Chukchi Slope Current at the Chukchi Borderland that was ultimately entrained into the Canada Basin, as well as the part of the Chukchi Slope current over the upper continental slope flowing westward into the East Siberian Sea. These agreed with the circulation pattern revealed by Leng et al [58] based on recent mooring/shipboard measurements and a regional ocean–sea ice model. Armitage et al [22] and Zhong et al [21, 59] also found that currents in the southwestern BG intensified recently. These phenomena were also reproduced in NAPA-BGC. Combined with the increased DIC flux from Barrow Canyon and the increased DIC due to biological degradation along the Chukchi-East Siberian Shelfbreak (figure 5(c)), the recent ocean circulation played a key role in DIC increase in the subsurface Makarov Basin.

Previous research suggested the uptake of anthropogenic carbon in AW occupying around 200–500 m of the Makarov Basin increased from 1987 to 2015 at a mean rate of 0.64 $\mu$mol kg$^{-1}$ yr$^{-1}$, which was larger than that in the Canada Basin (0.17–0.50 $\mu$mol kg$^{-1}$ yr$^{-1}$) [60]. Ericson et al [42] pointed out a similar increasing anthropogenic carbon uptake (0.61 $\mu$mol kg$^{-1}$ yr) in AW within the Makarov Basin from 1991 to 2011. Both indicated that the intermediate layer of the Makarov Basin was acidified during recent decades. Coupled with the increased DIC concentration in the subsurface layer, the ecosystem in the Makarov Basin may become more vulnerable to future acidification of the Arctic Ocean.

5. Conclusion

This study investigated shifts in DIC distribution in the subsurface Arctic Ocean during 1998–2015 through hindcast simulation using the NAPA-BGC. Model results suggest that DIC maximum layer deepened by ~50 m in the central Canada Basin and expanded to higher latitudes following the extension of PWW during 2008–2015 relative to 1998–2007. DIC concentrations increased significantly over 50–250 m in the Chukchi-East Siberian Shelfbreak and Makarov Basin during 2008–2015.

The DIC increase over the shelfbreak is attributable to increased local degradation and transport of DIC richer water from the Chukchi Shelf. Estimations of the DIC budget in the subsurface Makarov Basin indicated that advection controlled the increase in DIC content during 2008–2015, contributing 2.6 times more DIC amount than that during 1998–2007. The enhancement of the advection effect corresponded to a shift in the ocean circulation pattern, in which currents along the Chukchi-East Siberian Slope to the Makarov Basin were accelerated recently, promoting the transport of DIC-rich Pacific Water to the subsurface layer of the Makarov Basin. Coupled with the acidifying intermediate water [42, 60], this enhancement of DIC implies a more vulnerable and threatened marine environment in the future Makarov Basin. In addition to acidic water, the Pacific inflow supplies abundant nutrients into the Arctic Ocean [61]. Similar to DIC, the significant increase in nitrate was also simulated in the subsurface Makarov Basin and Chukchi-East Siberian during the past decades. The changes in nutrients structure and their impact on the community composition requires further investigation.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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Conflict of interest

The authors declare that they have no conflict of interest.

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