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Key Points:
- The subsurface dissolved inorganic nitrogen (DIN) reservoir in the Canada Basin increased by ~35% during 1990–2015
- The subsurface DIN concentration in the Canada Basin remained fairly constant during 1990–2015
- The ratio of subsurface delta dissolved inorganic carbon (DIC) to delta DIN (ΔDIC/ΔDIN) increased by ~33% during 1990–2015

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

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Increased DIN Storage and ΔDIC/ΔDIN Ratio in the Subsurface Water of the Canada Basin, 1990–2015

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Abstract The subsurface layer of the Canada Basin contains large reservoirs of dissolved inorganic nitrogen and carbon (DIN and DIC, respectively). Under rapid change of Arctic climate system, these reservoirs may not only serve as potential material bases for primary production, but also for carbon source and sink. However, the long-term trends in reservoirs and the interaction between reservoirs and the upper ocean are not fully understood. This study used two data sets to evaluate the long-term trends in the DIN and DIC reservoirs in the subsurface layer of the Canada Basin during 1990–2015. The results indicate an increase of ~35% in the subsurface DIN reservoir over 25 years, whereas the DIN concentration remained fairly constant. The discrepancy between the DIN reservoir and DIN concentration was primarily owing to the expansion of the subsurface layer, which increased the DIN stock but maintained a relatively stable DIN concentration. Additionally, the ΔDIC/ΔDIN ratio in the subsurface layer increased by ~33% over 25 years, primarily due to denitrification on the pathway of Pacific inflow, such as Chukchi shelf. In the Canada Basin, the enhanced mixing between the subsurface and surface layers may promote primary production and render the subsurface layer a carbon source. Consequently, when studying the biogeochemical cycles in the changing Arctic Ocean, the long-term interactions between the subsurface and surface layers in the Canada Basin should be further considered.

Plain Language Summary Nutrients in the subsurface layer of the Canada Basin are potential material bases for primary production. Rapid climate change and enhanced mixing between the subsurface and surface layers impact the biogeochemical cycles in the Canada Basin. Two data sets were collected to determine the dissolved inorganic nitrogen and carbon (DIN and DIC, respectively) concentrations, DIN reservoir, and ΔDIC/ΔDIN ratio in the subsurface layer from 1990 to 2015. The results revealed that the subsurface DIN concentration remained constant, whereas the DIN and DIC reservoirs and ΔDIC/ΔDIN ratio dramatically increased. The increased Pacific inflow and expansion of subsurface layer may have led to an increase in the subsurface DIN and DIC stocks in the Canada Basin. Simultaneously, denitrification on the Chukchi shelf may have slowed the increase in the DIN reservoir, leading to an increase in the ΔDIC/ΔDIN ratio. Increased regional events such as upwellings, eddies, and storms can enhance the mixing of the upper layers (subsurface and surface layers) and thus may promote primary production. However, the increased ΔDIC/ΔDIN ratio implies that the mixing of the upper layers also supplies DIC upwardly, which might negatively impact the air-sea CO_2 flux in the Canada Basin and reduce carbon uptake from the atmosphere.

1. Introduction

The subsurface layer of the Canada Basin contains large reservoirs of dissolved inorganic nitrogen (DIN) and dissolved inorganic carbon (DIC), which are potential materials for primary production (McLaughlin & Carmack, 2010; Qi et al., 2017; Yamamoto-Kawai et al., 2006). Over the last three decades, DIN and DIC reservoirs have been steadily increasing, primarily due to the enhanced biogeochemical cycles in the Chukchi shelf (Zhuang et al., 2022): Since the 1990s, with drastic sea-ice retreat and prolonged open water duration, increased Pacific inflow water has brought abundant nutrients to the Chukchi shelf, which has led to an increase in the primary production (Grebmeier, 2012; Serreze et al., 2016; Woodgate, 2018). The enhanced biological pump and remineralization have led to increased DIC and DIN in the Chukchi shelf bottom water (Brown et al., 2015; Lewis et al., 2020). With a considerable portion of the Chukchi shelf bottom water entering the subsurface layer of the Canada Basin through Barrow Canyon, the subsurface reservoirs have been increasing (Weingartner et al., 1998).

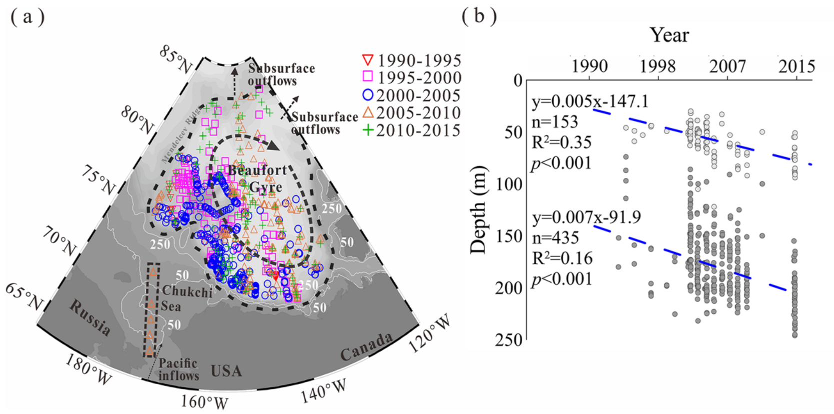

Figure 1. (a) Schematic diagram of topography and circulation in the study area. The figure was created with Ocean Data View version 5.6.3 (Schlitzer, 2021). The black dashed circles indicate the study area in the Canada Basin and the Chukchi shelf, and the white solid lines are the 50 and 250 m isobaths; (b) Upper and lower boundary depths (25.5 kg/m^3, light gray points; 27 kg/m^3, dark gray points) of subsurface water in the Canada Basin between 1990 and 2015, the blue dashed lines are the linear trends of the upper and lower boundary over the study period.

Itoh et al. (2013) and Pickart et al. (2016) suggested that the Chukchi shelf bottom water influx through Barrow Canyon would increase with the increased Pacific inflow water. As a result, the subsurface reservoirs in the Canada Basin are expected to increase. Previous studies have indicated increased subsurface DIC and DIN stocks, while these works lacked quantitative estimations for such increasing.

Under rapid climate change, the enhanced interaction between the subsurface and surface layers impacts the biogeochemical cycle of the Canada Basin (Arrigo et al., 2008; Tremblay et al., 2015). Because of the stratification, the subsurface layer in the Canada Basin was considered to be a stable DIN and DIC reservoir (Coachman & Aagaard, 1974; Proshutinsky et al., 2019). However, in recent years, enhanced mixing between the upper layers (subsurface and surface layers) may have increased the upward supply of nutrients (mainly DIN), thus promoting primary production in the Canada Basin (Pickart et al., 2013; Timmermans & Toole, 2022; Tremblay et al., 2011). In addition, the upward supply of DIC decreases the CO_2 uptake by the upper water layer (Brown et al., 2016; Griffith et al., 2012). The interaction between the upper layers emphasizes the importance of subsurface water in the biogeochemical cycle. It also implies that the long-term trends in the DIC and DIN reservoirs in this layer could be used to evaluate the influence of subsurface water on primary production and CO_2 uptake in the Canada Basin.

In the past decades, researchers have extensively studied the biogeochemical cycles in the changing Arctic Ocean. It should be noted, however, due to the scarcity of high-latitude cruises, the long-term trends of subsurface DIN and DIC reservoirs in the Canada Basin are poorly understood (Arrigo & van Dijken, 2015; Tremblay et al., 2015; Wassmann et al., 2020; Yamamoto-Kawai et al., 2006). In this study, in situ data was collected from the subsurface layer of the Canada Basin from the Chinese Arctic Scientific Expedition (CHINARE; https://datacenter.chinare.org.cn/data-center/dindex) and the Global Ocean Data Analysis Project v2 (GLODAPv2) (Key et al., 2015; Olsen et al., 2016). We calculated the DIN and DIC concentrations, DIN reservoir, and ΔDIC/ΔDIN ratio in the subsurface layer between 1990 and 2015. Our main goal was to understand the long-term variations in subsurface DIN and DIC reservoirs and their effects on primary production and CO_2 uptake in the Canada Basin. This study will enhance our understanding of biogeochemical processes in the Canada Basin under rapid climate change and supplements the investigation of biogeochemical cycles in the western Arctic Ocean.

2. Materials and Methods

2.1. Data Source

All data (DIN and DIC concentrations, density, and water depth) of 1,180 stations in the study area (Figure 1a) were assembled from CHINARE (2010, 2012, and 2014) and GLODAPv2 (1990–2015). Because GLODAPv2 lacked Canada Basin's data from 2010 to 2014, the CHINARE data from 2010 to 2014 were used as suitable supplements. Data for Pacific water inflow from 1990 to 2015 were derived from Woodgate (2018).

2.2. Geographical Setting and Circulation

Figure 1a shows the geographical location of the study area, including the Canada Basin and the Chukchi Shelf. The southern boundary of the Canada Basin comprises the Beaufort and Chukchi shelves. The Canadian Arctic Archipelago marks the eastern border of the Canada Basin. The Alpha Ridge lies at the northern edge of the Canada Basin, while Northwind Ridge is considered as its west boundary (Timmermans & Toole, 2022). In this study, we extended the western boundary of the Canada Basin to the Mendeleev Ridge because the subsurface water considered in our research is generally shallower than 250 m. Under the influence of the Beaufort Gyre (BG), a clockwise surface-intensified circulation system centered over the Canada Basin, the nutrient-rich subsurface water can be observed in the Chukchi Abyssal Plain (Figure S1 in Supporting Information S1; Regan et al., 2019). The total Canada Basin is ∼1.5×10^6 km^2, which is slightly larger than a previous estimation (1.3×10^6 km^2; Grantz et al., 1990).

2.3. Boundaries of the Subsurface Layer

The boundaries of the subsurface layer were defined before calculating the DIN and DIC concentrations and the DIN reservoir. McLaughlin et al. (1996) suggested that the depth between 50 and 200 m in the Canada Basin can be defined as the subsurface layer. However, the spin-up BG strengthens the Ekman convergence of freshwater in the surface water and enhances the stratification between the surface and subsurface layers (Timmermans & Toole, 2022). This leads to the accumulation of freshwater and deepens the upper boundary of the subsurface layer (Kenigson & Timmermans, 2021; Proshutinsky et al., 2019). Additionally, the lower boundary of the subsurface layer is also deepening. This is primarily caused by the lateral advection of subsurface water from the Chukchi Borderland and an increased lateral eddy flux in this layer (Zhong et al., 2019; Zhong & Zhao, 2014). The deepening of the upper and lower boundaries (Figure 1b) renders the definition of the subsurface layer proposed by McLaughlin et al. (1996) no longer suitable. In this study, we integrated the analyses of subsurface water from different researchers and defined a density between 25.5 and 27 kg/m^3 as the subsurface layer (Nishino et al., 2008; Pickart et al., 2016; Zhong et al., 2019).

2.4. Calculation of DIN Reservoir and Subsurface ΔDIC/ΔDIN Ratio

The integrated DIN (I_{DIN}) and DIN reservoir (R_{DIN}) of the surface layer ($\sigma < 25.5$ kg/m^3) and subsurface layer (25.5 kg/m^3 $< \sigma <$ 27 kg/m^3) were estimated in the Canada Basin from 1990 to 2015 for each year as follows:

$$\mathrm{I_{DIN}} = \sum_{n}^{m-1} (N_{i+1} + N_i) \times (D_{i+1} - D_i)/2 \tag{1}$$

$$\mathrm{R_{DIN}} = \frac{\sum I_{DIN}}{Z} \times S \tag{2}$$

where n and m represent the depth range for each station when calculating the I_{DIN} of the surface or subsurface layer. When calculating the I_{DIN} of the surface layer, n and m represent depth of surface water (0 m) and depth at which the potential density is 25.5 kg/m^3, respectively. When calculating the $\mathrm{I_{DIN}}$ of the subsurface layer, n and m represent depths at which the potential density is 25.5 and 27 kg/m^3, respectively. N_i is the DIN concentration at each sampling depth (D_i) from n to m. S and Z represent the total area of the study region (∼1.5×10^6 km^2) and the number of stations for each year, respectively. $\sum I_{DIN}$ represents the sum of $\mathrm{I_{DIN}}$ of all stations in each year.

There was a significant linear relationship between the subsurface DIC and DIN concentrations for each year ($y_{DIC} = kx_{DIN} + b$; Figure S2 in Supporting Information S1). The slope k (i.e., ΔDIC/ΔDIN ratio) during the study period was used to reflect the changes in DIC relative to DIN in the subsurface water.

3. Results and Discussion

3.1. Long-Term Trends in DIN and DIC Stocks in the Subsurface Layer

In the subsurface layer, the average DIN and DIC concentrations exhibited minimal difference between the years 1990 (11.78 ± 3.81 μmol/kg and 2176 ± 37 μmol/kg, respectively) and 2015 (12.13 ± 3.15 μmol/kg and 2202 ± 44 μmol/kg, respectively; Figures 2a and 2b). The DIN and DIC concentrations in the subsurface layer

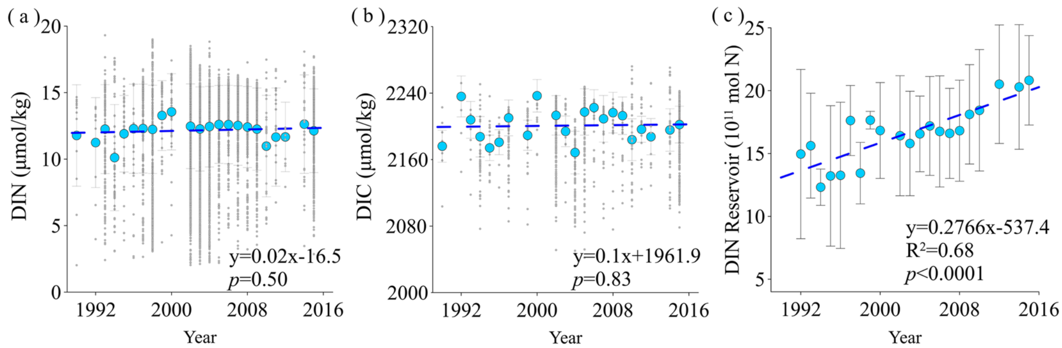

Figure 2. Annual variations of (a) dissolved inorganic nitrogen (DIN) concentrations, (b) dissolved inorganic carbon (DIC) concentrations and (c) DIN reservoirs in the subsurface layer of the Canada Basin between 1990 and 2015. The gray dots indicate the original data, the blue dots indicate the averaged data over the years, and the blue dashed lines indicate the linear trends of the averaged data over the years.

were stable during the study period ($p = 0.50$ and $p = 0.83$, respectively), suggesting that the average DIN and DIC concentrations in the subsurface layer of the Canada Basin have remained constant over the last 25 years.

During 1990–2015, the subsurface DIN reservoir increased from $15.0 \pm 7.5 \times 10^{11}$ mol N to $20.8 \pm 4.0 \times 10^{11}$ mol N (Figure 2c) and exhibited a notable increasing trend. According to Tremblay et al. (2015), the annual nitrate input to the surface of the Arctic Ocean contributed $\sim 22 \times 10^{11}$ mol C to net community production. By assuming a C:N ratio of 6.7 (Redfield, 1934), the annual surface nitrate input was estimated to be $\sim 3.3 \times 10^{11}$ mol N ($22 \times 10^{11} \div 6.7 = 3.3 \times 10^{11}$ mol N), which accounted for approximately one-seventh to one-sixth of the subsurface DIN reservoir. This suggests that the subsurface layer has a vast reservoir of DIN in the Arctic Ocean. The subsurface DIN reservoir was significantly correlated with time ($R^2 = 0.64$, $p < 0.0001$; Figure 2c), increasing at a rate of 0.3×10^{11} mol N yr^{-1} (~2% per year).

Complex biogeochemical processes dominate the variations in the DIN reservoir, and Pacific inflow is considered a primary factor (Tremblay et al., 2015). Time-lag correlation analyses were performed to further understand the influence of the Pacific inflow on variations in the DIN stock. A lag time of 6–18 months was selected for a series of correlation analyses. The results demonstrated that a lag time of 12 months gave the best linear correlation with the subsurface DIN reservoir ($R^2 = 0.60$, $p < 0.05$; Figure 3a), followed by that with 9 months ($R^2 = 0.51$, $p < 0.05$; Figure S3 in Supporting Information S1). Our results were consistent with those of previous studies on the travel time of the Pacific inflow from the Bering Strait to the Canada Basin: Studies on the Chukchi shelf showed that the Pacific inflow water takes 3–4 months to reach the central Chukchi shelf from the Bering Strait

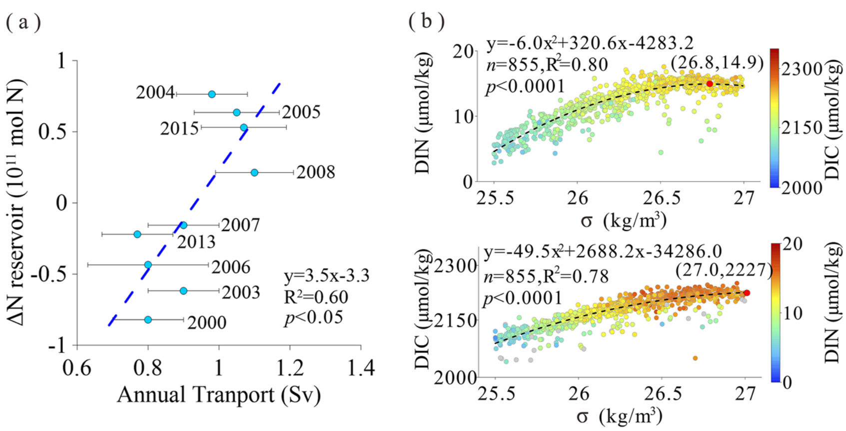

Figure 3. (a) Variation of the subsurface dissolved inorganic nitrogen (DIN) reservoir in the Canada Basin (ΔN reservoir = this year's reservoir − last year's reservoir) versus last year's Pacific Ocean inflow; (b) Distribution of DIN (upper panel) and dissolved inorganic carbon (DIC; lower panel) concentrations in the subsurface layer. The red dots represent the DIN and DIC maxima in the subsurface layer.

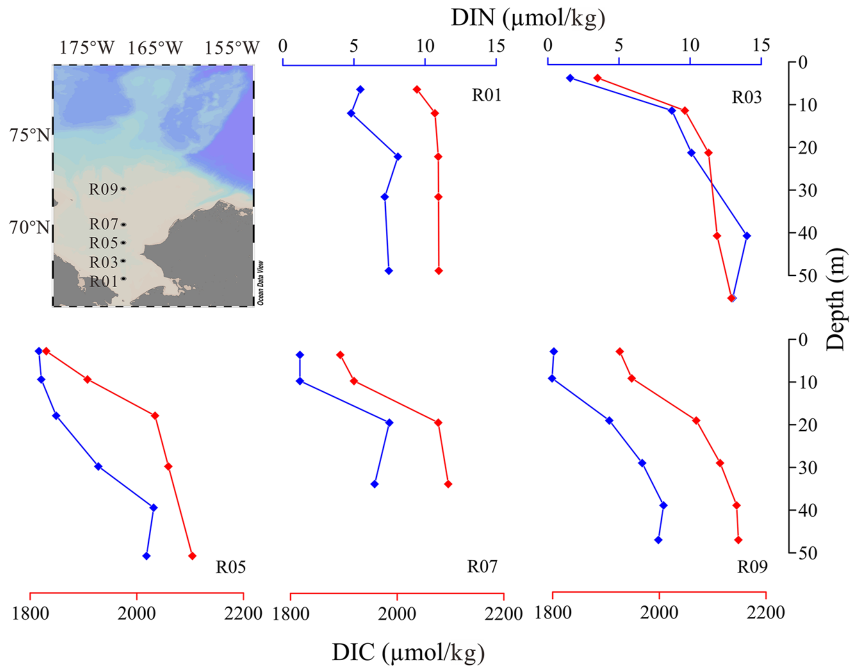

Figure 4. Vertical distribution of dissolved inorganic carbon (DIC) and dissolved inorganic nitrogen (DIN) at R01 to R09 stations on the Chukchi Shelf. The red and blue diamonds represent the DIC and DIN concentrations, respectively. All stations are from the 2010 Chinese Arctic Scientific Expedition.

(Spall, 2007; Winsor & Chapman, 2004; Woodgate et al., 2005) and another 3–6 months to flow eastward from the central Chukchi shelf to Barrow Canyon (Spall, 2007). Woodgate et al. (2005) estimated the renewal time of Pacific Winter Water (PWW) near Barrow Canyon to be approximately 1 year, whereas Spall (2007) determined the residence time of water mass on the Chukchi shelf to be 6–12 months. The results demonstrated a significant correlation between the Pacific inflow and subsurface DIN reservoir, implying that the increased Pacific inflow may be a key factor contributing to the increase in the DIN reservoir.

Direct estimation of the DIC reservoir was not feasible due to the limited availability of the subsurface DIC data; therefore, we inferred the variation in the DIC reservoir from the subsurface DIN reservoir because they both originate from the Chukchi shelf bottom water and undergo comparable biogeochemical processes (Qi et al., 2017; Zhuang et al., 2022). The DIC and DIN concentrations in the subsurface layer were positively correlated during the study period ($R^2 = 0.42$, $p < 0.05$; Figure S4 in Supporting Information S1), which implies that the variation in the DIC reservoir was consistent with that of the DIN reservoir, and the subsurface DIC reservoir increased over the study period.

The vertical variations in DIC and DIN concentrations in the subsurface layer were inconsistent (Figure 3b). In the subsurface layer, the DIN increased, followed by a slight decrease with increasing depth. Maximum DIN was observed at 26.8 kg/m^3. The DIC continued to increase with increasing depth, and the maximum DIC was observed at 27 kg/m^3, indicating that the maximum DIC was ~20 m deeper than the maximum DIN. The observed discrepancy could not be attributed to the diffusion from subsurface water because the DIC and DIN concentrations in the Atlantic water (>27 kg/m^3) and in the surface water (<25.5 kg/m^3) were lower than those in the subsurface water (data not shown). Instead, it was deduced that the discrepancy between the DIN and DIC maxima could potentially be caused by denitrification in the Chukchi shelf. Denitrification near the sediment–seawater interface can consume DIN in the bottom water over the Chukchi shelf. The flow of the bottom water into the subsurface layer in the Canada Basi leads to a deeper depth of DIC maxima than DIN maxima (Chang & Devol, 2009; Mills et al., 2015). Figure 4 shows the vertical distribution of DIC and DIN on the Chukchi Shelf. DIN concentration increased with depth, followed by a decrease near the bottom, with the maximum DIN at an average depth of 35.2 ± 10.9 m. However, DIC concentration increased with depth and it maximum concentration was determined at an average depth of 47.5 ± 7.7 m. The vertical distribution of DIC and DIN concentrations over the Chukchi shelf were consistent with those found in previous studies, suggesting that the inconsistent vertical

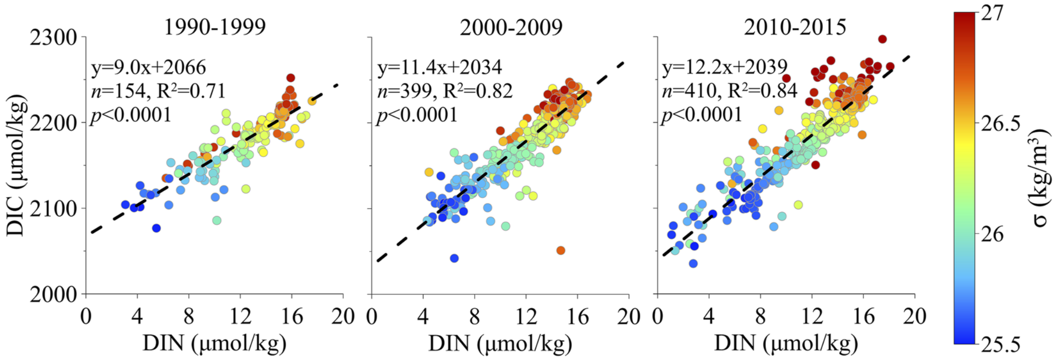

Figure 5. Relationship between subsurface dissolved inorganic carbon (DIC) and dissolved inorganic nitrogen (DIN) in the Canada Basin from 1990 to 1999, 2000 to 2009, and 2010 to 2015. The black dashed line indicates the linear relationship between the DIC and DIN concentrations.

distributions of DIC and DIN concentrations in the subsurface layer of the Canada Basin can be attributed to denitrification over the Chukchi Shelf (Chang & Devol, 2009; Reeve et al., 2019; Zhuang et al., 2022). In view of the enhanced denitrification rate on the Chukchi shelf in recent years (Payne & Arrigo, 2022), we hypothesize that the subsurface DIC reservoir in the Canada Basin should increase faster than the subsurface DIN reservoir.

3.2. Increasing DIN Reservoir Versus Constant DIN Concentration in the Subsurface Layer

Previous studies have reported that the subsurface DIN maximum in the Canada Basin remained constant between 1990 and 2015 (Table S1 in Supporting Information S1), which is consistent with the results of this study (Figure 2a). However, unlike the constant DIN concentration, the subsurface DIN reservoir increased from 1990 (Figure 2c). The variations between the DIN concentration and reservoir were mainly caused by the thickening of the subsurface layer. Zhong et al. (2019) suggested that the total volume of PWW (26–27 kg/m^3, which could be roughly subsurface water) in the Canada Basin increased by ~1.18 times between 2011 and 2016 when compared to that of 2002 and 2006. Considering the relatively stable subsurface DIN concentration and the linear relationship between the years and DIN reservoirs (Figure 2c), we estimated that the total volume of subsurface water in 2015 was ~1.5 times greater than that in 1992 ([2015 × 0.2766–534.7] ÷ [1992 × 0.2766–534.7] = 1.5). During the same study period as that of Zhong et al. (2019) (2011–2015 vs. 2002–2006), the subsurface DIN reservoir increased by ~1.15 times, which is consistent with their results. Although the enhanced biological pump and remineralization in the Chukchi shelf ultimately increased DIN stock and DIN concentration in the subsurface layer in the Canada Basin (Tremblay et al., 2015), the thickening of the subsurface layer maintained a relatively stable DIN concentration. The DIN concentrations and reservoirs in the subsurface layer in 1992 and 2015 were used to illustrate the effect of increased subsurface layer volume on DIN concentrations. The DIN reservoirs in 1992 and 2015 were 15.0 × 10^{11} mol N and 21.0 × 10^{11} mol N, respectively (Figure 2c), while the average DIN concentrations in 1992 and 2015 was 11.8 ± 3.1 μmol/kg and 12.1 ± 3.1 μmol/kg, respectively (Figure 2a). If the volume of the subsurface layer remained constant, the average subsurface DIN concentration in 2015 should have been 16.5 ± 4.3 μmol/kg, which is much higher than the measured value (12.1 ± 3.1 μmol/kg; Figure 2a). If the increased reservoir volume (approximately 1.5 times) is considered, then the estimated subsurface DIN concentration in 2015 should be 11.0 ± 2.9 μmol/kg, which is consistent with the measured value.

3.3. Increasing ΔDIC/ΔDIN Ratio in the Subsurface Layer

The subsurface ΔDIC/ΔDIN ratio increased between 1990 and 2015. Since the residence time of subsurface water is approximately 10 years (Ekwurzel et al., 2001; Macdonald et al., 2005), the study period was thus divided into three intervals (1990–1999, 2000–2009, and 2010–2015) to determine the interdecadal variation in the ΔDIC/ΔDIN ratio. The results revealed a good linear relationship between the subsurface DIC and DIN concentrations for each interval (Figure 5). The subsurface ΔDIC/ΔDIN ratio increased from 9.03 (1990–1999) to 11.41 (2000–2009) and then to 12.16 (2010–2015).

The subsurface ΔDIC/ΔDIN ratio exhibited a positive relationship with time ($R^2 = 0.30$, $p < 0.01$; Figure 6a), which increased by ~33% over 25 years (9.41 in 1990 vs. 12.58 in 2015), and the difference between the subsurface

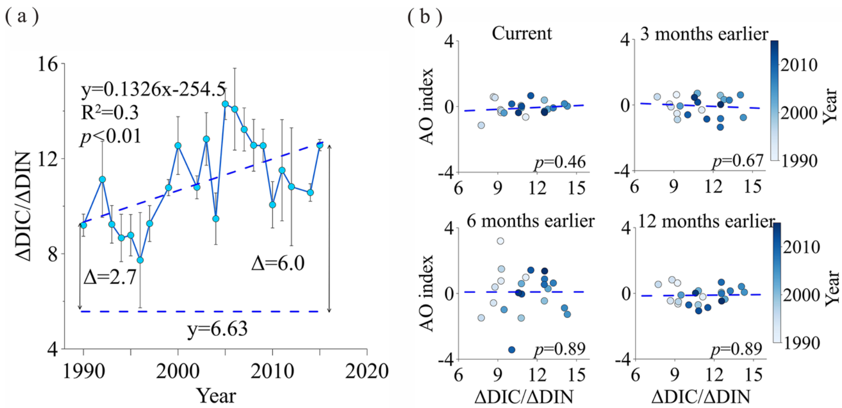

Figure 6. (a) Linear relationship between the subsurface delta dissolved inorganic carbon to delta dissolved inorganic nitrogen (ΔDIC/ΔDIN) and time in the Canada Basin; the flat blue dashed line indicates the C : N uptake ratio by phytoplankton. (b) Linear relationship between the subsurface ΔDIC/ΔDIN ratio and the Arctic Oscillation (AO) index, with the AO index taken from the sampling month, 3 months earlier, 6 months earlier, and the average value over the year. AO index data source: https://www.ncei.noaa.gov/access/monitoring/ao/.

ΔDIC/ΔDIN ratio and the Redfield ratio increased from 2.70 to 6.02 at a rate of ~0.13 yr^{-1} (Redfield, 1934). However, this ratio exhibited considerable fluctuation during the study period, which might be attributed to the changing Arctic Oscillation (AO; Proshutinsky et al., 2015; Timmermans & Toole, 2022). However, the correlation between the subsurface ΔDIC/ΔDIN ratio and the AO index was not significant (Figure 6b). These results indicate that although AO may influence the accumulation of water masses in the Canada Basin, it's impact on the subsurface ΔDIC/ΔDIN was not substantial. Therefore, we suggest that the subsurface ΔDIC/ΔDIN ratio was primarily influenced by Pacific inflow water, biological pumps, denitrification, and physical processes on the Chukchi Shelf.

As most of the subsurface DIC and DIN originated from the Chukchi shelf bottom water (Qi et al., 2017), the significant linear relationship between them (Figure 5) indicates that they underwent the same processes over the Chukchi shelf. According to Tremblay et al. (2015) and Brown et al. (2016), these processes include remineralization (increased DIN and DIC), denitrification (decreased DIN), air–sea CO_2 exchange (increased DIC) and sea–ice brine export (increased DIC). These ultimately influenced the subsurface ΔDIC/ΔDIN ratio as most of this water flowed into the subsurface layer. The Pacific inflow water increased from 0.8 Sv in 1990 to 1.1 Sv in 2015 (Woodgate, 2018), which brought abundant nutrients to the Chukchi shelf and led to an increase in primary production (Grebmeier, 2012; Serreze et al., 2016). With the prolonged open water periods, the enhanced biological pump and remineralization have led to increased DIC and DIN in the Chukchi shelf bottom water (Brown et al., 2015). Simultaneously, denitrification on the Chukchi shelf may have increased the subsurface ΔDIC/ΔDIN ratio. Payne and Arrigo (2022) reported that the interannual denitrification rate on the Chukchi shelf increased at 1.3 ± 0.5 mmol/m^2 yr between 1988 and 2018. Although the biological pump on the Chukchi shelf has been steadily increasing over the years (Arrigo et al., 2008; Grebmeier, 2012), the enhanced denitrification may have decelerated the increase of DIN in the bottom water. As denitrification does not substantially impact DIC in the water column (Brown et al., 2015; Granger et al., 2011), this process increases the ΔDIC/ΔDIN ratio. Furthermore, Zhuang et al. (2022) showed that denitrification on the Chukchi shelf in the 2010s had a 10% greater impact on the subsurface DIN reservoir in the Canada Basin than it did in the 1990s. They calculated the nitrate deficit (N*) and found a northward expansion of the low N* signal (<−10) on the Chukchi shelf from 1994 to 2018. Because denitrification in the Canada Basin is practically negligible (Reeve et al., 2019), the increased denitrification and expansion of the low N* signal on the Chukchi shelf may be the leading cause of the increasing ΔDIC/ΔDIN ratio in the subsurface layer in the Canada Basin.

In addition, the air–sea CO_2 exchange can increase the ΔDIC/ΔDIN ratio. The surface water over the Chukchi shelf cools in autumn, increasing its CO_2 solubility and enhancing its capacity to absorb CO_2 from the atmosphere (Bates et al., 2005). As the weakened stratification promotes the diffusion of CO_2 toward the bottom water, this direct uptake of CO_2 via air–sea CO_2 exchange contributes to an increase in the ΔDIC/ΔDIN ratio. Sea–ice brine export also contributes DIC to the bottom water, increasing the ΔDIC/ΔDIN ratio. Studies in the Arctic Ocean

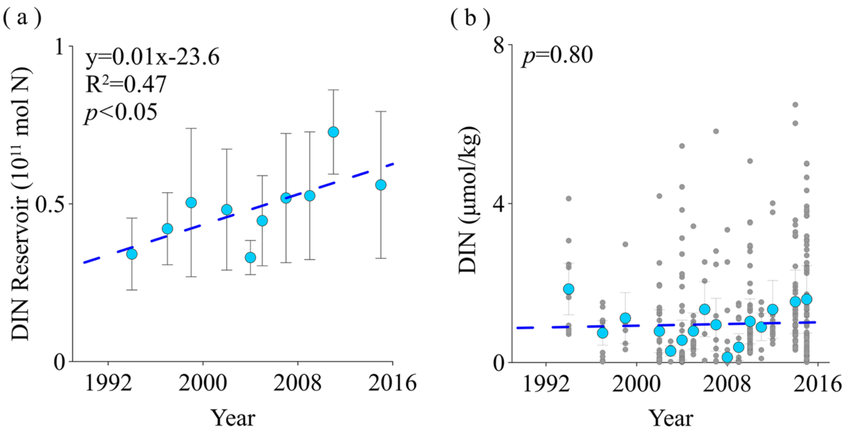

Figure 7. (a) Annual variations of dissolved inorganic nitrogen (DIN) reservoirs in the surface layer of the Canada Basin between 1990 and 2015; the blue dashed lines indicate the linear trends of DIN reservoirs over the years. (b) DIN concentrations in the surface layer of the Canada Basin between 1990 and 2015. The gray dots indicate the original data, the blue dots indicate the averaged data over the years, and the blue dashed lines indicate the linear trends of the averaged data over the years.

suggest that when sea ice forms, CO_2 produced by the inorganic precipitation of $CaCO_3$ is released to the surface water (Brown et al., 2016; Rysgaard et al., 2007). In the Chukchi shelf, with enhanced vertical mixing in the water column during winter, the diffusion of CO_2 toward the bottom water can increase the ΔDIC/ΔDIN ratio. However, according to Brown et al. (2016), the contributions from air–sea CO_2 exchange and sea–ice brine export to the bottom water are not significant. Brown et al. (2016) implied that these two processes are not important contributors to the increased ΔDIC/ΔDIN ratio, and that the impact of remineralization and denitrification on this ratio may be greater than that of air–sea CO_2 exchange and sea–ice brine export over the Chukchi shelf.

3.4. Biogeochemical Implications of Subsurface Water

In the Canada Basin, mixing between the upper layers may increase the surface DIN reservoir. Our results showed that the surface DIN reservoir increased by ~64% over the study period ($3.4 \pm 1.0 \times 10^{11}$ mol N in 1994 vs. $5.6 \pm 1.9 \times 10^{11}$ mol N in 2015; $R^2 = 0.47$; Figure 7a). Meanwhile, the surface DIN concentrations remained constant (1.85 ± 1.08 μmol/kg in 1994 vs. 1.59 ± 1.42 μmol/kg in 2015, Figure 7b), indicating that the increased surface DIN reservoir is mainly attributed to the increased surface layer volume. These results were consistent with the findings of Zhong et al. (2019). They found that the upper bound of PWW, which can be roughly considered as the lower bound of the surface layer, was deepened due to the influence of Ekman convergence. Notably, in recent years, the increased regional events (upwellings, eddies, and storms) have promoted the mixing between the subsurface and surface layers. The enhanced mixing may transport the nutrients upward and thus increase the surface DIN reservoir. Pickart et al. (2013) showed that the frequency and intensity of upwelling events in the southern Canada Basin have increased in recent years. The nutrient-rich subsurface water carried by upwellings can increase the DIN concentration in the surface layer by a factor of two to three, promoting primary production (Tremblay et al., 2011). Several studies have shown that increased storm events and mesoscale eddies in the Canada Basin could also enhance mixing between the upper layers (Nishino et al., 2011; Parkinson & Comiso, 2013; Rinke et al., 2017; Zhao et al., 2016). In addition to regional events, Dosser et al. (2021) showed that the internal wave-driven diffusivity in the Canada Basin increased from $(3 \pm 1) \times 10^{-7}$ m^2 s^{-1} (between 2004 and 2010) to $(7 \pm 1) \times 10^{-7}$ m^2 s^{-1} (between 2011 and 2019), which could have promoted mixing between the upper layers.

Figure 8 illustrates the differences in the water column and biogeochemical processes between the 1990s and 2010–2015 in the Canada Basin (together with the upstream Chukchi shelf). Rapid sea-ice retreat and increased Pacific inflow accelerated the biological pump on the Chukchi shelf, making great contributions to the subsurface DIN reservoir (Stroeve et al., 2012; Woodgate, 2018). In the Canada Basin, the spin-up BG and the lateral advection of subsurface water from the Chukchi Borderland have led to a deepening subsurface layer during 2010–2015 compared to that in the 1990s (Figure 1b; Proshutinsky et al., 2019; Zhong et al., 2019). Meanwhile,

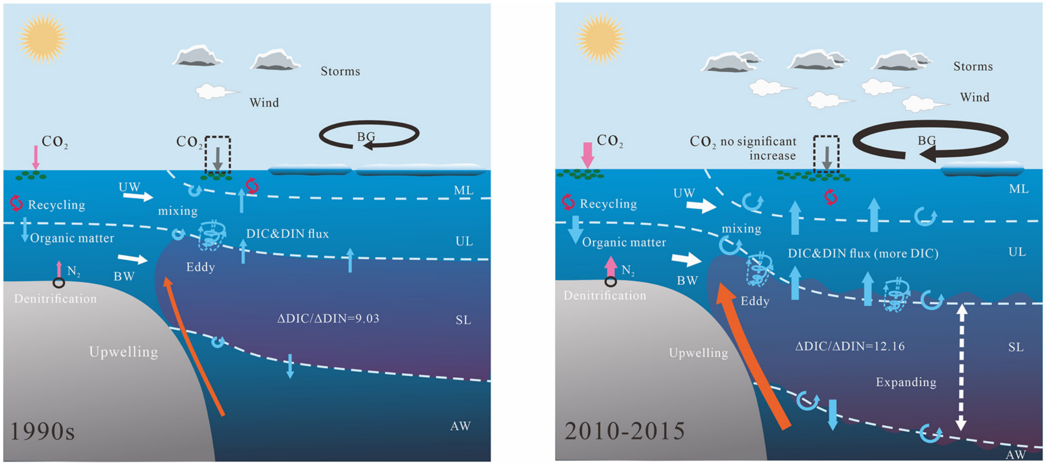

Figure 8. Schematic representation of the biogeochemical processes driven by environmental and climate changes in the western Arctic Ocean (Chukchi shelf and Canada Basin) between the 1990s and 2010–2015. UW = Upper Water, BW = Bottom Water, BG = Beaufort Gyre, ML = Mixed Layer, UL = Upper Layer, SL = Subsurface Layer, AW = Atlantic Water.

the increased upwelling events, mesoscale eddies, and storm events during 2010–2015 promoted mixing between the upper layers, which would increase the upward DIN supply and promote primary production (Arrigo & van Dijken, 2015; Hauri et al., 2013; Mathis et al., 2012; Pickart et al., 2013; Zhao et al., 2016). However, mixing would supply more DIC upward (relative to DIN), rendering the subsurface layer a carbon source, which could have a negative impact on the air–sea CO_2 flux in the Canada Basin (black dashed box in Figure 8; Cai et al., 2010; Ouyang et al., 2022).

4. Conclusions

The results of this study show that the subsurface DIN concentration in the Canada Basin was stable from 1990 to 2015, whereas the DIN reservoir, DIC reservoir, and ΔDIC/ΔDIN ratio in the subsurface layer increased as a result of physical and biogeochemical processes in the Chukchi shelf and Canada Basin. The expansion of subsurface water may be the primary cause of the increasing subsurface DIN reservoirs and relatively stable DIN concentration. Simultaneously, denitrification on the Chukchi shelf may slow the increase in the DIN reservoir, leading to a faster increase in the subsurface DIC reservoir than in the DIN reservoir. With rapid sea ice shrinking, the increased regional events would enhance the mixing of the upper layers, and thus might increase the contribution of subsurface stocks to the upper layers, promoting primary production. However, the increased ΔDIC/ΔDIN ratio implies that the mixing of the upper layers also supplies DIC upward, which might negatively impact the air-sea CO_2 flux in the Canada Basin. Under rapid climate change, it is essential to focus on the long-term trends of the subsurface DIC reservoir, DIN reservoir, and ΔDIC/ΔDIN ratio when investigating biogeochemical processes in the Canada Basin. In the future, sediment traps and biogeochemical models will be essential tools to further assess the impact of subsurface material reservoirs on primary production and the air-sea CO_2 fluxes in the Canada Basin (Tremblay et al., 2015).

Data Availability Statement

All data in the study area during 1990–2009 and 2015 are available through the Global Ocean Data Analysis Project v2, via their website https://www.glodap.info/index.php/merged-and-adjusted-data-product-v2-2022/ (Key et al., 2015; Olsen et al., 2016). All dissolved inorganic nitrogen data and dissolved inorganic carbon data in the Canada Basin and the Chukchi Shelf during 2010–2014 are available online through Figshare (Chen & Qi, 2023).

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