Decadal change of dissolved inorganic carbon in the subarctic western North Pacific Ocean

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ABSTRACT

Dissolved inorganic carbon (DIC) was measured from 1992 to 2008 at two time-series sites in the subarctic western North Pacific; this region is a source of atmospheric CO$_2$ in winter due to vertical mixing of deep waters rich in DIC. To estimate the decadal DIC increase resulting from CO$_2$ uptake from the atmosphere, we corrected DIC for the contribution of biological activity below the temperature minimum ($T_{\text{min}}$) layer ($\sim 100$ m), which is the remnant of the mixed layer from the preceding winter. Decadal DIC increases in the $T_{\text{min}}$ layer and upper intermediate water (1.3–1.5 μmol kg$^{-1}$ yr$^{-1}$; 100–200 m) were higher than those expected from oceanic equilibration with increasing atmospheric CO$_2$ and those previously reported in the open North Pacific. The increase in water column CO$_2$ was estimated to be 0.40 ± 0.08 mol m$^{-2}$ yr$^{-1}$. The decadal DIC change in the $T_{\text{min}}$ layer affects winter CO$_2$ emission. The increase of atmospheric $x$CO$_2$ in winter (2.1 ± 0.0 ppm yr$^{-1}$) is higher than that of oceanic $x$CO$_2$ (0.7 ± 0.5 ppm yr$^{-1}$) that calculated from DIC and total alkalinity in the $T_{\text{min}}$ layer. This difference suggests reduction of CO$_2$ emission in winter is possibly controlled by the increase of total alkalinity.

1. Introduction

Anthropogenic CO$_2$ was taken up by the global ocean at a rate of 2.2 ± 0.4 PgC yr$^{-1}$ during the 1990s (Denman et al., 2007). The most fundamental and reliable approach to the direct detection of the CO$_2$ uptake rate and its variation is to make more accurate and longer time-series observations at fixed stations, for example, station ALOHA in the subtropical North Pacific Ocean (e.g. Dore et al., 2003), BATS in the subtropical western North Atlantic Ocean (e.g. Bates et al., 2002), ESTOC in the subtropical eastern North Atlantic Ocean (e.g. Santana-Casiano et al., 2007), Ocean Station Papa (OSP) in the subarctic eastern North Pacific (e.g. Wong and Chan, 1991) and stations KNOT (44°N, 155°E) (e.g. Tsurushima et al., 2002) and K2 (47°N, 160°E) (e.g. Honda et al., 2006) in the subarctic western North Pacific Ocean (Fig. 1). Time-series observations are essential to understanding temporal variation of dissolved inorganic carbon (DIC) in the ocean.

Increases of DIC in the surface waters from 1997 to 2001 and in the subsurface waters from 1992 to 2001 were detected by time-series observations at KNOT (Tanaka et al., 2003; Wakita et al., 2005). DIC is positively correlated with AOU in the subsurface water due to the decomposition of organic matter. AOU was calculated by subtracting the observed concentration of dissolved oxygen (DO) from the saturated concentration calculated from temperature and salinity with the equation of Weiss (1970). A linear increase in AOU is superimposed on a bidecadal oscillation of AOU in the Oyashio region (near the subarctic western North Pacific) (Ono et al., 2001). Because the previously reported DIC increases in the subarctic western North Pacific are based on time-series data only from the 10-year period 1992 to 2001 (Wakita et al., 2005), more accurate and longer time-series data are required to confirm the increase of DIC.

At least annually since 1997, the Japan Agency for Marine-Earth Science and Technology (JAMSTEC) has conducted hydrographic observations at KNOT or K2 in the subarctic western North Pacific (Fig. 1). Hydrographic studies in this region were also conducted by other organizations between 1992 and 2003 (e.g. Tsurushima et al., 2002; Wakita et al., 2005). In this study,
we compiled data obtained by JAMSTEC and others and investigated the temporal variation of DIC and related properties [AOU and total alkalinity (TA)] below the winter mixed layer of the subarctic western North Pacific for 1992–2008. We also discuss the decadal increase of DIC resulting from the uptake of CO$_2$ from the atmosphere through gas exchange at the air–sea interface.

2. Data sources and analyses

We prepared a set of data to investigate long-term climate change in the subarctic western North Pacific. All data were collected from the observations at the Japanese time-series stations KNOT and K2 from 1992 to 2008, which were made in 61 CTD casts on 44 cruises at KNOT and in 28 CTD casts on 15 cruises at K2; these cruises were made by seven research vessels: T/S Hokusei Maru (Hokkaido University), T/S Oshoro Maru (Hokkaido University), R/V Bosei Maru (Tokai University), R/V Hakuo Maru (Tokyo University), R/V Kaiyo Maru (Japan Fisheries Agency), R/V Hakurei No. 2 (Metal Mining Agency of Japan), R/V Natsumia (JAMSTEC) and R/V Mirai (JAMSTEC) (Tsurushima et al., 2002; Wakita et al., 2005; Watanabe et al., 2007). Approximately, 60% of all observations were done from the R/V Mirai. Data sets from K2 from 1999 to 2006 (Watanabe et al., 2007) are published at the Carbon Dioxide Information and Analysis Center (http://cdiac.ornl.gov/) and the JAMSTEC data web site (http://www.godac.jamstec.go.jp/k2/index.html). DIC and TA were measured by using coulometric and potentiometric techniques, respectively. The precision of both DIC and TA was ±0.1% or better from duplicate determinations.

TA from July 2000 to July 2001 during T/S Hokusei Maru cruise was determined by the improved single-point titration method, with precision of ±0.2% from duplicate determinations (Wakita et al., 2005). The DIC and TA values were determined with calibration against certified reference material provided by Prof. A. G. Dickson (Scripps Institution of Oceanography). The DO and nutrient concentrations from JAMSTEC observations were measured with an automatic titrator and a continuous flow analyser, respectively (e.g. Watanabe et al., 2007).

We merged the KNOT data obtained by JAMSTEC cruises with those published by Wakita et al. (2005) and Tsurushima et al. (2002) for T/S Hokusei Maru, T/S Oshoro Maru, R/V Bosei Maru and R/V Hakuo Maru cruises. In addition, we added the oceanic physical and chemical data from KNOT and K2 collected as part of WOCE–P1 (1999) (http://whpo.ucsd.edu/data_access/show_cruise?ExpoCode=49KA199905_1) and WESTCOSMIC (1998, 1999) (http://www.kanso.co.jp/ocean/html-doc/english/top1.html).

The values of DIC, TA, DO and nutrients (silicate, phosphate and nitrate) had systematic errors among cruises, because the analytical methods used for these determinations and the precision and standards for analysis varied slightly from cruise to cruise. These systematic errors probably derive from the standardization because of the absence of a common reference material. DIC data for 1992–1997 during T/S Hokusei Maru cruise by Wakita et al. (2005) were not calibrated against certified reference material.

To investigate the temporal variation of chemical properties from the winter mixed layer to intermediate water, we corrected the systematic errors by assuming that ocean chemical
properties in the North Pacific Deep Water (NPDW) were constant in our study area from 1992 to 2008 (Wakita et al., 2005; Watanabe et al., 2007). NPDW was defined as the water mass between 27.69σθ (~2000 db) and 27.77σθ (~3500 db), because chlorofluorocarbons were not detected below 27.69σθ (Watanabe et al., 2001) and the bottom water deeper than the NPDW in the western North Pacific expanding from the Samoan Passage is characterized by potential temperature (θ) lower than 1.2 °C (~27.77σθ) (Johnson et al., 1994). Because the residence time of NPDW is ~500 years (Stuiver et al., 1983), we can assume that the NPDW did not change significantly from 1992 to 2008 for the purpose of investigating the temporal variations in shallower waters.

The values of the systematic errors and factors were –6–19 μmol kg⁻¹ for DIC, –15–8 μmol kg⁻¹ for TA, 0.82–0.97 for DO, 0.95–1.09 for silicate, 0.98–1.12 for phosphate and 0.96–1.04 for nitrate. We corrected the values of these properties at the isopycnal surfaces of NPDW (σθ = 27.69, 27.70, 27.71, 27.72, 27.73, 27.74, 27.75, 27.76 and 27.77) to fit the mean observed values from 2006 to 2008 at the same isopycnal surfaces. The values at each of these isopycnal surfaces were obtained by linear interpolation. The minimum systematic errors were ±0.2% for DIC and TA, ±3% for DO and ±1% for nutrients. The standard deviations of DIC, TA, DO, silicate, phosphate and nitrate values in NPDW after the corrections were estimated to be ±1.9, ±1.3, ±1.5, ±1.0, ±0.01 and ±0.1 μmol kg⁻¹, respectively. These values agree well with the measurement precision for these properties and serve for use in evaluations of the temporal variation of chemical properties.

Because KNOT and K2 are both located in the western subarctic gyre (Fig. 1) and we lacked sufficient decadal time-series data for either station, we combined data from KNOT and K2. The typical water column structure in this region has a minimum temperature (Tmin) layer at about 26.5σθ (~100 m), which is the remnant of the mixed layer from the preceding winter; the maximum temperature layer is at about 27.1σθ (~370 m) (Fig. 2) (e.g. Osafune and Yasuda, 2006). Because KNOT is located just North of the Subarctic Front (Fig. 1), data obtained there on some days (10 and 11 May 1999 (MR99–K02); 5 June 1999 (HO99–1); 22 July 1999 (HO99–3)) include no Tmin layer as a result of the migration of subtropical water (Tsurushima et al., 2002). Data from these days were excluded from the analysis of the Tmin layer in this study.

We here define the Tmin layer as the remnant of the winter mixed layer in early April, when the surface mixed layer was the coldest in the year (Fig. 3a). The depth, salinity and σθ of the surface mixed layer reached annual maxima from middle March to early April (Figs 3b–d). Because the mean values of depth and σθ in the winter mixed layer in early April (~120 m, ~26.5 kg m⁻³) were similar to those of the Tmin layer over all season from 1992 to 2008 (108 ± 18 m, 26.5 ± 0.1 kg m⁻³) (Figs 3b and c), in the Tmin layer there was no influence of the surface mixed layer from spring to fall. We can thus define the Tmax layer as the remnant of the winter mixed layer in early April.

The values of properties on each isopycnal surface (26.6σθ–27.1σθ) were obtained by linear interpolation of discrete bottle sampling data. For the Tmax layer, after the values of Tmax, depth, salinity and σθ were determined from continuous CTD data (1 db resolution), the contents of chemical parameters (DO, DIC, TA and nutrients) on the Tmax isopycnal surface were obtained by linear interpolation of discrete bottle sampling data. This was done because the values of depth, salinity and σθ in the Tmax layer are not necessarily identical to those from discrete bottle sampling.

3. Results and discussion

3.1. Seasonal variations of DIC and related properties in the surface water

In winter, the western subarctic gyre is a source of atmospheric CO2 because of strong vertical mixing: from spring to fall it is a sink for CO2 because of biological production (e.g. Tsurushima et al., 2002; Kawakami et al., 2007). At KNOT and K2, the maximum values of ρCO2 and net air–sea CO2 flux occurred in early April (late winter); these maximum values were larger than the climatological mean values for this region reported by Takahashi et al., 2009 (Figs 3i and j). The values of ρCO2 in the ocean were calculated from DIC and TA by using the CO2SYS program (Pierrot et al., 2006), by using all data.
Fig. 3. Seasonal variations of (a) \( \theta \), (b) maximum mixed layer depth (MLD), (c) salinity, (d) \( \sigma_\theta \), (e) DIC, (f) AOU, (g) TA and (h) phosphate (PO4) in the surface mixed layer (circles) and minimum temperature (\( T_{\text{min}} \)) layer (triangles) at stations KNOT (open symbols) and K2 (solid symbols). We also show the seasonal variations of (i) oceanic and atmospheric \( p\text{CO}_2 \) and (j) monthly net air–sea \( p\text{CO}_2 \) flux in the surface mixed layer. These figures were plotted by using all data from 1992 to 2008 in order to examine the typical features of seasonal variations at KNOT and K2 and to compare them with the climatological monthly mean from Takahashi et al. (2009). The density criterion in the surface mixed layer was smaller than 0.125 kg m\(^{-3}\) (de Boyer Montégut et al., 2004). Values of oceanic \( p\text{CO}_2 \) (i) were calculated from TA and DIC. The individual \( p\text{CO}_2 \) values calculated in different years are normalized to a reference year 2000 using 1.5 \( \mu \text{atm year}^{-1} \) as the rate of \( p\text{CO}_2 \) increase (Takahashi et al., 2009). Values of monthly net air–sea \( p\text{CO}_2 \) flux (j) were estimated with the sea–air \( p\text{CO}_2 \) difference at KNOT and K2 and the climatological monthly mean value of air–sea gas transfer rate for 48°N, 157.5°E as the center of a 4° (latitude) \( \times \) 5° (longitude) grid cell that is parameterized as a function of (wind speed)\(^2\) with a scaling factor of 0.26 from Takahashi et al. (2009) (T09). Positive values indicate effluxes from the sea and negative values influxes from the air. The thick black lines in (i) and (j) are the monthly mean values averaged for KNOT and K2. The other lines in (i) are \( p\text{CO}_2 \) in the seawater (doublet) and in the atmosphere (thin dash) and the doublet line in (j) is the monthly net air–sea \( p\text{CO}_2 \) flux. These values are the climatological monthly mean values at 48°N, 157.5°E, the center of the designated box from Takahashi et al. (2009).
from 1992 to 2008 to examine the typical features of seasonal variation. In this calculation, we used the carbonate dissociation constants of Mehrbach et al. (1973) as refitted by Dickson and Millero (1987). The other properties also reached annual maxima in late winter (Fig. 3): mixed layer depth (135 m), salinity (33.2), density (26.5 kg m$^{-3}$), DIC (2130 μmol kg$^{-1}$), AOU (8 μmol kg$^{-1}$), TA (240 μmol kg$^{-1}$) and phosphate (1.9 μmol kg$^{-1}$). The amplitude of seasonal variation of DIC in the surface mixed layer at KNOT (160 μmol kg$^{-1}$) was larger than that at K2 (130 μmol kg$^{-1}$) (Fig. 3). These amplitudes in the western subarctic gyre are larger than those at other pelagic ocean time-series sites and are mainly attributed to biological production from spring to fall and strong vertical mixing in winter (e.g., Tsurushima et al., 2002). In summer, DIC and phosphate were lower at KNOT than at K2 (Figs. 3e and h), which suggests that the difference of seasonal amplitude between KNOT and K2 is caused by differences in biological production (Kawakami et al., 2007). The values of DIC, AOU, TA, phosphate, pCO$_2$ and net air–sea CO$_2$ flux in winter (early April) at KNOT were similar to those in winter at K2 (Fig. 3). During winter, there were no differences in biological production and CO$_2$ emission between KNOT and K2. CO$_2$ emission during winter is controlled by strong vertical mixing of subsurface waters rich in DIC, TA and nutrients because of decomposition of organic matter and dissolution of CaCO$_3$ (e.g., Takahashi et al., 2006). These results indicate that the decadal DIC increase in the $T_{\text{min}}$ layer is affected by not only the increase of anthropogenic CO$_2$, but also the temporal variation of CO$_2$ emission in winter due to strong vertical mixing.

3.2. Temporal variations of DIC and related properties in the subsurface water

We detected distinct trends in $\theta$, salinity and isopycnal depths in the $T_{\text{max}}$ and 26.6–26.8σ$_\theta$ layers (Fig. 4, Table 1). There was no apparent DIC increase observed in the subsurface water and AOU significantly decreased in the 26.6–26.8σ$_\theta$ layer ($P < 0.05$) (Fig. 5, Table 1), even though DIC and AOU reportedly increased from 1992 to 2001 at KNOT (Wakita et al., 2005). These linear regressions model was fitted using the least squares approach.

Long-term trends and bidecadal oscillations of values of $\theta$, salinity, isopycnal depth, AOU and nutrients in the subsurface water in the northwestern subarctic Pacific Ocean are known (Ono et al., 2001; Osafune and Yasuda, 2006; Watanabe et al., 2008). Recently, the bidecadal oscillations were linked to the nodal tidal cycle with an 18.6-year period (e.g., Osafune and Yasuda, 2006). Oxidation and denitrification of organic matter includes oxidation and denitrification of organic matter in the subsurface seawater (e.g., Sabine et al., 2002). The decomposition of organic matter includes oxidation and denitrification of organic matter in the aragonite saturation horizons of the subarctic region in the North Pacific.

3.3. Estimate of DIC increase due to air–sea CO$_2$ gas exchange

The change in DIC in the subsurface water is controlled by the uptake of atmospheric CO$_2$ through gas exchange at the air–sea interface, the decomposition of organic matter and the dissolution of CaCO$_3$ (e.g., Gruber et al., 1996; Sabine et al., 2002). The decomposition of organic matter includes oxidation and denitrification of organic matter in the subsurface seawater (e.g., Sabine et al., 2002). Oxidation and denitrification of organic matter are related to AOU and $N^* [=\text{nitrate} + \text{nitrite}] - 16 \times \text{phosphate} + 2.90$ as an index of nitrogen fixation–denitrification (Deutsch et al., 2001), respectively.
Fig. 4. Temporal variations of depth (a), $\theta$ (b) and salinity (c) from 1992 to 2008 in the temperature minimum ($T_{\text{min}}$, circles) and 26.8$\sigma_{\theta}$ (triangles) layers at stations KNOT (open symbols) and K2 (solid symbols); note the shifted scales for $T_{\text{min}}$ (left) and 26.8$\sigma_{\theta}$ (right) in panel (b). The depths in the $T_{\text{min}}$ layer are those determined from continuous CTD data (1 db resolution). Where significant ($P < 0.05$), linear regressions for 1997 to 2008 are shown for the $T_{\text{min}}$ (solid lines) and 26.8$\sigma_{\theta}$ (dashed lines) layers.

Table 1. Rates of increase for DIC and related properties in the western subarctic gyre from 1997 to 2008

| Layer | Depth (m)$^a$ | $\theta$ (°C yr$^{-1}$) | Salinity (yr$^{-1}$) | AOU ($\mu$mol kg$^{-1}$ yr$^{-1}$) | TA ($\mu$mol kg$^{-1}$ yr$^{-1}$) | $\Delta C^* + C_{280}$ ($\mu$mol kg$^{-1}$ yr$^{-1}$) |
|-------|--------------|--------------------------|----------------------|-----------------------------------|---------------------------------|-----------------------------------------------|
| $T_{\text{min}}$ | 108 ± 17 | $1.3 \pm 0.6$ | $0.039 \pm 0.021$ | $0.009 \pm 0.003$ | $-2.1 \pm 0.9$ | $1.6 \pm 0.2$ | $1.3 \pm 0.3$ |
| 26.6$\sigma_{\theta}$ | 121 ± 13 | $-1.3 \pm 0.4$ | $-0.020 \pm 0.024$ | $-0.002 \pm 0.002$ | $-2.8 \pm 0.8$ | $0.9 \pm 0.1$ | $1.5 \pm 0.3$ |
| 26.7$\sigma_{\theta}$ | 146 ± 16 | $2.0 \pm 0.5$ | $-0.028 \pm 0.016$ | $-0.003 \pm 0.002$ | $-3.0 \pm 0.6$ | $0.8 \pm 0.1$ | $1.4 \pm 0.3$ |
| 26.8$\sigma_{\theta}$ | 175 ± 23 | $-2.4 \pm 0.8$ | $-0.025 \pm 0.011$ | $-0.003 \pm 0.001$ | $-1.8 \pm 0.5$ | $0.5 \pm 0.1$ | $1.3 \pm 0.2$ |
| 26.9$\sigma_{\theta}$ | 222 ± 32 | $-2.2 \pm 1.2$ | $0.012 \pm 0.010$ | $-0.001 \pm 0.001$ | $-0.4 \pm 0.6$ | $0.5 \pm 0.1$ | $0.8 \pm 0.2$ |
| 27.0$\sigma_{\theta}$ | 287 ± 37 | $-1.3 \pm 1.4$ | $-0.004 \pm 0.007$ | $0.000 \pm 0.001$ | $-0.3 \pm 0.6$ | $0.5 \pm 0.1$ | $0.7 \pm 0.2$ |
| 27.1$\sigma_{\theta}$ | 375 ± 38 | $-0.3 \pm 1.4$ | $-0.004 \pm 0.007$ | $0.000 \pm 0.001$ | $-0.2 \pm 0.5$ | $0.5 \pm 0.1$ | $0.4 \pm 0.2$ |

Note: Error values are the SE of the slope of the linear regression. Bold numbers indicate significant rates derived from linear regressions ($P < 0.05$).

$^a$Averages and standard deviations at the isopycnal surfaces.
The change of DIC from the pre-industrial period caused by the oceanic uptake of CO2 from the atmosphere through gas exchange (\(\Delta C^*\)) is defined as follows:

\[
\Delta C^* = \text{DIC}_m - 117/170\text{AOU}_m
- 0.5(\text{TA}_m - \text{TA}^* + 16/170\text{AOU}_m)
+ 106/104N^* - C_{\text{eq}280},
\]

where DIC\(_m\), AOU\(_m\) and TA\(_m\) are the measured values. TA\(^*\) is the preformed TA estimated using the equation of Sabine et al. (2002). The effect of decomposition from \(N^*\) would be small, but not trivial; Watanabe et al. (2008) demonstrated a linear increase of \(N^*\) on 26.8\(\sigma_\theta\) surface superimposed on a biodecadal oscillation. 

\(C_{\text{eq}280}\) is the theoretical DIC content of waters in equilibrium with the pre-industrial atmospheric CO2 (280 \(\mu\)atm). Because \(C_{\text{eq}280}\) over time remains constant and its trend can be cancelled out, we calculated \(\Delta C^* + C_{\text{eq}280}\) and its trend represents the DIC increase caused by the uptake of CO2 from the atmosphere through the gas exchange at the air–sea interface.

\(\Delta C^* + C_{\text{eq}280}\) has significantly increased at 0.7–1.5 (average 1.2 ± 0.3) \(\mu\)mol kg\(^{-1}\) yr\(^{-1}\) \((P < 0.05)\) (Fig. 6, Table 1). These estimated increases in the winter mixed layer and upper intermediate water (\(T_{\text{min}}\) and 26.6\(\sigma_\theta\)–26.8\(\sigma_\theta\) layers), which are between 100 and 200 m thick, are greater than those in deeper water (26.9\(\sigma_\theta\)–27.0\(\sigma_\theta\)); \(\Delta C^* + C_{\text{eq}280}\) has remained unchanged below 27.1\(\sigma_\theta\). We estimated the increase of DIC accumulated in the water column to be 0.15 ± 0.04 mol m\(^{-2}\) yr\(^{-1}\) in the surface mixing layer and 0.25 ± 0.07 mol m\(^{-2}\) yr\(^{-1}\) in the intermediate

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**Fig. 5.** Temporal variations of AOU (a), DIC (b) and TA (c) for 1992 to 2008 in the temperature minimum (\(T_{\text{min}}\), circles) and 26.8\(\sigma_\theta\) (triangles) layers at stations KNOT (open symbols) and K2 (solid symbols). Where significant \((P < 0.05)\), linear regressions for 1997 to 2008 are shown for the \(T_{\text{min}}\) (solid lines) and 26.8\(\sigma_\theta\) (dashed lines) layers.
water (100–400 m), using the $\Delta C^* + C_{eq280}$ increase and the thickness of the average depth on each isopycnal surface from $T_{min}$ to 27.1$\sigma_\theta$ at increments of 0.1$\sigma_\theta$ (Table 1). The total DIC inventory change in the surface and intermediate waters (0.40 ± 0.08 mol m$^{-2}$ yr$^{-1}$; maximum depth ~400 m) is related to the CO$_2$ uptake rate; this value is similar to that previously reported in the subarctic western North Pacific [0.66 ± 0.22 mol m$^{-2}$ yr$^{-1}$; maximum depth ~900 m; 1973–1993 (Ono et al., 2000)] and in the global ocean [0.51 ± 0.09 mol m$^{-2}$ yr$^{-1}$; 1990s (Denman et al., 2007)].

This estimated inventory increase in the western subarctic gyre is higher than the anthropogenic CO$_2$ inventory increase in the Alaskan gyre (>45°N along 152°W) (<0.2 mol m$^{-2}$ yr$^{-1}$; maximum depth <~400 m; 1991/1992–2005/2006) estimated using an extended multiple linear regression technique (Sabine et al., 2008). This different value is the reason why the decadal DIC increase caused by the uptake of CO$_2$ from the atmosphere in the western subarctic gyre is larger than that in the Alaskan gyre despite the same penetration depth in the two gyres.

3.4. Factors controlling the DIC increase due to air–sea CO$_2$ gas exchange

The $\Delta C^*$ increase reflects the increase in anthropogenic CO$_2$ and the temporal variation in the CO$_2$ difference between atmosphere and ocean in the surface mixed layer at the time of water mass formation (e.g. Gruber et al., 1996; Sabine et al., 2002).

The $\Delta C^* + C_{eq280}$ increase in the $T_{min}$ layer in the subarctic western North Pacific would be affected by not only the increase of anthropogenic CO$_2$ but also the temporal variation of a strong CO$_2$ source during winter, because the $T_{min}$ layer is a remnant of the preceding winter mixed layer (e.g. Osafune and Yasuda, 2006). The western subarctic areas along the Kuril and western Aleutian arcs are strong CO$_2$ sources during winter owing to convective mixing of deep waters rich in respired CO$_2$ (e.g. Takahashi et al., 2006). This seasonal variation of pCO$_2$ has an opposite pattern to that of other open ocean time-series sites (ALOHA, BATS and OSP) (e.g. Tsurushima et al., 2002), because the primary causes of seasonal change in pCO$_2$ in the subarctic region (K2 and KNOT) and these other time-series sites are seasonal changes in DIC and temperature, respectively (e.g. Takahashi et al., 2006). The temporal variation of CO$_2$ emission in winter must affect that of DIC in the $T_{min}$ layer. If the winter increase rate of atmospheric CO$_2$ were higher than that of oceanic CO$_2$ from year-to-year, then the oceanic CO$_2$ uptake over time in this region would increase as a result of the reduction of CO$_2$ emission in winter due to decrease in the CO$_2$ difference between atmosphere and ocean.

The decadal increases of $\Delta C^* + C_{eq280}$ we observed in the $T_{min}$ layer (1.3 ± 0.3 μmol kg$^{-1}$ yr$^{-1}$) were higher than expected from oceanic equilibration with increased anthropogenic CO$_2$ in the atmosphere (0.7 μmol kg$^{-1}$ yr$^{-1}$), when calculated using the increase of atmospheric CO$_2$ (1.9 ppm yr$^{-1}$) (Foster et al., 2007) and the Revelle factor in the $T_{min}$ layer (15.6 ± 0.4) at constant TA. This Revelle factor is consistent with previous results in winter (Takahashi et al., 2006). This $\Delta C^* + C_{eq280}$ increase was larger than the anthropogenic DIC increase since the 1990s at other open North Pacific stations [<1.1 μmol kg$^{-1}$ yr$^{-1}$; <30°N along 149°E (WHP P10) (Murata et al., 2009), <1.0 μmol kg$^{-1}$ yr$^{-1}$; 145°E–140°W along 30°N (WHP P02), 0–55°N along 152°W (WHP P16) (Sabine et al., 2008)], and is comparable to higher anthropogenic CO$_2$ increases associated with the Kuroshio and California Currents on both edges of the basin (<1.5 μmol kg$^{-1}$ yr$^{-1}$; WHP P02) (Sabine et al., 2008).

The CO$_2$ emission in winter is estimated from the difference between atmospheric and oceanic xCO$_2$. We calculated oceanic xCO$_2$ by using the values of DIC and TA in the mixing layer (early April) (DIC$_{win}$·TA$_{win}$). Phosphate and nitrate in the winter mixing layer also were calculated (PO$_{4win}$, NO$_3win$). DIC, TA,
where $S_{aln}$ is salinity in the $T_{min}$ layer. The values of DIC, AOU, PO$_4$ and NO$_3$ in the $T_{min}$ layer (DIC$_{T_{min}}$, AOU$_{T_{min}}$, PO$_4{T_{min}}$ and NO$_3{T_{min}}$) were annual minima in winter (Figs 3e, f and h), whereas TA$_{T_{min}}$ had no a distinct seasonal variation (Fig. 3g). This increase of observed DIC$_{T_{min}}$, PO$_4{T_{min}}$ and NO$_3{T_{min}}$ are caused by the decomposition of organic matter after the previous winter (Figs 3e, f and h). We assumed that DO in the winter mixed layer is homogeneously saturated (i.e. AOU = 0) (e.g. Tsurushima et al., 2002), because the degree of saturation of DO in early April is ~100% (98 ± 2%) because of the strong vertical mixing and air–sea exchange. DIC$_{win}$, PO$_4{win}$ and NO$_3{win}$ were obtained from the observed DIC$_{T_{min}}$, PO$_4{T_{min}}$ and NO$_3{T_{min}}$ and decomposition with the following equations:

$$\text{DIC}_{\text{win}} = \text{DIC}_{T_{\text{win}}} - C/ - O_2(=117/170) \times \text{AOU}_{T_{\text{win}}},$$
$$\text{PO}_4{\text{win}} = \text{PO}_4{T_{\text{win}}} - P/ - O_2(=1/170) \times \text{AOU}_{T_{\text{win}}},$$
$$\text{NO}_3{\text{win}} = \text{NO}_3{T_{\text{win}}} - N/ - O_2(=16/170) \times \text{AOU}_{T_{\text{win}}},$$

where C/–O$_2$, P/–O$_2$ and N/–O$_2$ are the stoichiometric ratios of carbon, phosphorus and nitrogen to oxygen during the decomposition of organic matter (Anderson and Sarmentino, 1994). We estimated the xCO$_2$ of surface seawater in winter from $n\text{DIC}_{\text{win}}$ and $n\text{TA}_{\text{win}}$ (Figs 7a and b). The contents of atmospheric xCO$_2$ in late winter (at the beginning of April) are cited by GLOBALVIEW-CO$_2$ (2009) from 1997 to 2008 at 44.4° N.

The slope of the linear regression of oceanic xCO$_2$ on time was 0.7 ± 0.5 ppm yr$^{-1}$ ($n = 68$), which was significant at the 90% confidence level ($P = 0.1$) (Fig. 7c). This linear regression model was fitted using the weighted least squares approach, because calculated xCO$_2$ in the ocean was dispersing from year-to-year. This annual dispersion was not significantly uniform by the analysis of variance (Figs 4a and c and 7d and e), while $\sigma_p$ remained constant (Fig. 7f). The increasing trend of $n\text{TA}_{\text{win}}$, $n\text{PO}_4{\text{win}}$ and $n\text{NO}_3{\text{win}}$ may be caused by the enhanced winter convective mixing of deep waters rich in DIC, TA and nutrients. This increasing trend of $n\text{PO}_4{\text{win}}$ (Fig. 7d) will follow the increasing trend of phosphate during the 1989–1993 period in the winter mixed layer of the subarctic western North Pacific (Ono et al., 2002). In contrast, multi-decadal decreasing trends of salinity, density and phosphate in the winter mixed layer from the 1970s to the 1990s in the Oyashio region and near the subarctic western North Pacific were thought to have been caused by the

The increase of atmospheric xCO$_2$ (2.1 ± 0.0 ppm yr$^{-1}$, $n = 12$) in winter is significantly higher than that of oceanic xCO$_2$ by the comparison of the two linear regressions ($F < 0.001$) (Fig. 7c). This significance was tested by using the F-distribution of the maximum likelihood estimate of variance, because the regression of oceanic xCO$_2$ was fitted using the weighted least squares approach. The difference of xCO$_2$ between atmosphere and ocean was calculated to be 1.4 ± 0.5 ppm yr$^{-1}$. These results suggest that the declining CO$_2$ emissions in winter were due to the declining xCO$_2$ gradient between atmosphere and ocean over time.

The temporal change in the CO$_2$ source in winter could be caused by increase of TA in the winter mixed layer. In the CO$_2$ system calculation from the DIC change at constant TA in the seawater, the oceanic xCO$_2$ increase in the seawater is proportional to the DIC increase. If TA in the seawater increases over time, the oceanic xCO$_2$-increasing trend is smaller than that calculated by the increase of DIC under constant temperature and TA conditions. Applying this theoretical case to KNOT and K2, we estimated the oceanic xCO$_2$ increase to be 0.8 ppm yr$^{-1}$ by using the increasing values computed from the regression lines of $n\text{DIC}_{\text{win}}$ (Fig. 7a, DIC = 1.2 × year$^{-1}$) and $n\text{TA}_{\text{win}}$ (Fig. 7b, TA = 1.1 × year$^{-1}$) at constant $T_{\text{win}}$ (1.63 °C, mean from 1997 to 2008) (Fig. 8). This estimated increase is similar to that by observed $n\text{DIC}_{\text{win}}$ and $n\text{TA}_{\text{win}}$ (0.7 ± 0.5 ppm yr$^{-1}$, Fig. 7c) and is considerably smaller than that using computed $n\text{DIC}_{\text{win}}$ increasing values and a constant $n\text{TA}_{\text{win}}$ value (2230 μmol kg$^{-1}$ in 1997; 3.9 ppm yr$^{-1}$) (Fig. 8). Thus, the increasing trend of $n\text{TA}_{\text{win}}$ could inhibit the decadal increase of oceanic xCO$_2$ against the $n\text{DIC}_{\text{win}}$ increase, which suggests a reduction of CO$_2$ emissions in winter. This indicates that TA increase favours the uptake of CO$_2$ in the ocean.

If increases of atmospheric and oceanic xCO$_2$ in winter continue in the future at the same rates as those from 1997 to 2008, the gradient between the atmosphere and ocean in winter might gradually decrease with time. Additional time-series data are required to confirm the factors controlling the temporal change of CO$_2$ emission in winter and to evaluate this speculation.

In addition to decadal increase of $n\text{DIC}_{\text{win}}$ and $n\text{TA}_{\text{win}}$, the salinity, depth, $n\text{PO}_4{\text{win}}$ and $n\text{NO}_3{\text{win}}$ in the winter mixed layer ($T_{\text{min}}$ layer) also have significantly increased during the years 1997–2008 ($P < 0.05$) (Figs 4a and c and 7d and e), while $\sigma_p$ remained constant (Fig. 7f). The increasing trend of $n\text{TA}_{\text{win}}$, $n\text{PO}_4{\text{win}}$ and $n\text{NO}_3{\text{win}}$ may be caused by the enhanced winter convective mixing of deep waters rich in DIC, TA and nutrients. This increasing trend of $n\text{PO}_4{\text{win}}$ (Fig. 7d) will follow the increasing trend of phosphate during the 1989–1993 period in the winter mixed layer of the subarctic western North Pacific (Ono et al., 2002). In contrast, multi-decadal decreasing trends of salinity, density and phosphate in the winter mixed layer from the 1970s to the 1990s in the Oyashio region and near the subarctic western North Pacific were thought to have been caused by the
occurrence of surface stratification (Ono et al., 2001, 2002). Moreover, the temporal variability of depth and CO$_2$ emissions from the winter mixed layer must be relevant to atmospheric forcing (wind speed, etc.). The temporal variations of AOU on the isopycnal surface 26.7–27.2$\sigma$$_T$ in the Oyashio region and wintertime wind stress curl anomaly around this region are negatively and positively correlated with the bidecadal component of NPI, respectively (Ono et al., 2001; Ishi and Hanawa, 2005). Because DIC is positively correlated with AOU in the subsurface water ($r = 0.99$) due to the decomposition of organic matter, temporal variation of DIC also possibly has a bidecadal oscillation. Further investigation of the atmospheric effect is required to detect whether there is enhanced winter convective mixing of deep waters or not. The relationships among temporal variability of winter CO$_2$ emissions and atmospheric forcing or climate index must be investigated in the near future. In the future, more accurate and longer time-series data will be required to evaluate these speculations.

4. Summary

DIC and related chemical species have been measured from 1992 to 2008 at Stations KNOT (44ºN, 155ºE) and K2 (47ºN, 160ºE) in the subarctic western North Pacific in the western subarctic gyre. The surface mixed layer there in winter is a source of atmospheric CO$_2$ owing to strong vertical mixing (∼100 m) and from spring to fall it is a sink for CO$_2$ because of biological production (e.g. Tsurushima et al., 2002) (Fig. 3i). This seasonal variation of $p$CO$_2$ has an opposite pattern to that of other time-series sites (ALOHA, BATS and OSP) (e.g. Tsurushima et al., 2002), because the primary cause for seasonal change in the subarctic region is seasonal DIC change (e.g. Takahashi et al.,

![Graphs showing temporal variations of DIC, TA, CO$_2$, and phosphate.](image-url)
The temporal change of CO$_2$ emission in winter must affect that of DIC in the $T_{min}$ layer. Decadal DIC increase below the $T_{min}$ layer is affected not only by the increase of anthropogenic CO$_2$ but also by temporal change of a strong CO$_2$ source during winter, because the typical water column structure here has the $T_{min}$ layer at $\sim 26.5 \sigma_T$ ($\sim 100$ m) (Figs 2 and 3d), which is the remnant of the mixed layer from the preceding winter.

To estimate the decadal increase of DIC resulting from the uptake of CO$_2$ from gas exchange with the atmosphere, we corrected DIC for the contribution of the biological activity by calculating $\Delta C^*$ (e.g., Gruber et al., 1996; Sabine et al., 2002). Decadal changes of corrected DIC have significantly increased at 0.7–1.5 (avg. 1.2 ± 0.2) $\mu$mol kg$^{-1}$ yr$^{-1}$ in the subsurface water from 1997 to 2008 (Fig. 6, Table 1). These estimated increases in the winter mixed layer and upper intermediate water ($T_{min}$ layer and 26.6–26.8$\sigma_T$; 100–200 m) are higher than those in the next deeper water layer (26.9–27.0$\sigma_T$; 200–300 m) and have remained unchanged below 27.1$\sigma_T$ (•400 m). We estimated the water column inventory of CO$_2$ increase to be 0.40 ± 0.08 mol m$^{-2}$ yr$^{-1}$, which is similar to those previously reported for the same region (Ono et al., 2000) and for the global ocean (Bindoff et al., 2007).

The decadal DIC increase by the uptake of CO$_2$ from the atmosphere into the winter mixed layer ($T_{min}$ layer) would be affected not only by the increase of anthropogenic CO$_2$ but also the reduction of CO$_2$ emission in winter. The estimated DIC increase in the $T_{min}$ layer and in the 26.6–26.8$\sigma_T$ layer (1.3–1.5 $\mu$mol kg$^{-1}$ yr$^{-1}$) was higher than that expected from oceanic equilibration with increased anthropogenic CO$_2$ in the atmosphere (0.7 $\mu$mol kg$^{-1}$ yr$^{-1}$) and anthropogenic DIC increase since the 1990s in the other open North Pacific stations ($<$1.1 $\mu$mol kg$^{-1}$ yr$^{-1}$) (Sabine et al., 2008; Murata et al., 2009). The increase of atmospheric xCO$_2$ (2.1 ± 0.0 ppm yr$^{-1}$) in winter (early April) is markedly higher than that of oceanic xCO$_2$ in winter calculated from observed DIC$_{min}$ and TA$_{min}$ (0.7 ± 0.5 ppm yr$^{-1}$) (Fig. 7c), which suggests that the reduction of CO$_2$ emission in winter is due to a decreasing xCO$_2$ gradient between the atmosphere and ocean.

This temporal change of the CO$_2$ source in winter is possibly caused by an increase of nTA$_{win}$ in the winter mixed layer. The oceanic xCO$_2$ increase calculated from observed DIC$_{ta}$ and TA$_{ta}$ was considerably smaller than that calculated using computed nDIC$_{min}$ increasing values and constant $T_{min}$ (1.63 °C, mean from 1997 to 2008) and nTA$_{win}$ (2230 $\mu$mol kg$^{-1}$ in 1997; 3.9 ppm yr$^{-1}$) (Fig. 8). Thus, the increasing trend of nTA$_{win}$ would inhibit decadal increase of oceanic xCO$_2$ against nDIC$_{win}$, which suggests the reduction of CO$_2$ emission in winter. This indicates that TA increase favours the uptake of CO$_2$ in the ocean. Additional time-series data are required to confirm the factors controlling the temporal change of CO$_2$ emission in winter. Moreover, because CO$_2$ efflux from the winter mixed layer must be relevant to atmospheric forcing, the relationships between temporal variability of winter CO$_2$ emissions and atmospheric forcing must be investigated in the near future.

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