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Seasonal Variability of Carbon Chemistry at the SEATS Time-Series Site, Northern South China Sea Between 2002 and 2003

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ABSTRACT

Analyses of total carbon dioxide (TCO_2) and titration alkalinity (TA) in the mixed-layer was performed approximately bimonthly at the SEATS time series site (18°15'N, 115°35'E) in the northern South China Sea (SCS) from March 2002 to April 2003. These measurements and the calculated-fCO_2 were then used to document their seasonal variations and to estimate the seasonal air-sea flux of CO_2 during the observed period at the site. Results show that the normalized TCO_2 ($NTCO_2 = TCO_2 \times 35/S$) fluctuates seasonally between ~1972 and ~1997 μmol kg^{-1}, with the highest value in winter. The decline of $NTCO_2$ in spring-summer mainly results from *in situ* biological utilization, while the resurgence of $NTCO_2$ in fall-winter is due to entrainment of the TCO_2-rich subsurface waters from below. TA varies from ~2190 to ~2220 μmol kg^{-1} in tandem with salinity, suggesting the prime control of physical processes. fCO_2 increases progressively from spring to summer, reaches a maximum in July (~382 μatm), then decreases from fall to winter to a minimum (~347 μatm) in January with an amplitude of ~35 μatm. The seasonal variability of fCO_2 is in phase with temperature changes but is inversely correlated with the fluctuation of $NTCO_2$, suggesting that the fCO_2 seasonality is primarily controlled by temperature changes, though other factors have compensated partially to yield the

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observed low amplitude of its variability. The sea-to-air CO_2 fluxes for spring, summer, fall and winter are estimated to range from 0.00 ± 0.01 to -0.02 ± 0.05, +0.03 ± 0.01 to +0.23 ± 0.06, +0.18 ± 0.10 to +0.45 ± 0.25, and -0.62 ± 0.20 to -1.42 ± 0.46 $molC\ m^{-2}\ year^{-1}$, respectively. Throughout the year, the annual flux is calculated to be -0.11 ± 0.08 ~ -0.23 ± 0.18 $molC\ m^{-2}\ year^{-1}$ during the observed period. Furthermore, although there is a drawdown of $NTCO_2$ of ~25 $\mu mol\ kg^{-1}$ from winter to summer, which implies a net community production of 6.80 ± 0.77 $mmolC\ m^{-2}\ year^{-1}$ in the mixed layer at the SEATS site, there is no corresponding change of nitrate observed, suggesting other sources of nitrogen required to sustain the new production.

(Key words: Carbon dioxide, Air-sea flux, Time-series, South China Sea)

1. INTRODUCTION

In response to growing concern over the role of oceanic uptake of increasing atmospheric CO_2 concentration, enormous research efforts have been made to better quantify the spatial and temporal variability of CO_2 fluxes between the atmosphere and the oceans. Among these, the most direct method to unravel the temporal variations of CO_2 fluxes is to carry out long-term time-series measurements in the world's oceans. For instance, the Bermuda Atlantic Time-series Study (BATS; 31°50'N, 64°10'W) and the Hawaii Ocean Time-series program (HOT; 22°45'N, 158°00'W) were established in the subtropical Atlantic and Pacific during the late 1980s, respectively. Results from these long-term oceanic time-series observations have helped scientists to better understand processes modulating the CO_2 exchange between the atmosphere and the upper oceans (Winn et al. 1994; 1998; Bates et al. 1996; Bates 2001; Gruber et al. 2002; Dore et al. 2003). The general consensus has been that the temperate and polar oceans of both hemispheres act as major sinks for atmospheric CO_2, while the equatorial oceans are major sources for CO_2. Furthermore, in terms of the processes controlling seasonal amplitude of surface-water fCO_2, the biological effect is shown to be dominant in high-latitude and equatorial waters, whereas in the temperate gyre regions it is dominated by the temperature effect (Takahashi et al. 1993; 1997; 2002). However, in order to gain a better air-sea CO_2 flux scenario on a global scale, time-series observations from other key regions in the world's oceans is required (Feely et al. 2001; Karl et al. 2001; Quay, 2002). Thus, the establishment and subsequent study of the South East Asia Time-series Study site (SEATS; 18°15'N, 115°35'E; Fig. 1) in the northern South China Sea (SCS) are timely needed, because it is the only one time-series station situated in a subtropical marginal sea (Shiah et al. 1999; Karl et al. 2003).

The SCS, one of the largest marginal seas in the world ocean, is a semi-enclosed marginal sea off the Asian continent in the West Pacific. Previous studies have revealed intensive upwelling and vertical mixing in the SCS (Chen and Huang 1995; Chao et al. 1996). The upwelling of deep water brings nutrient-laden water closer to the surface, thus nutricline in the

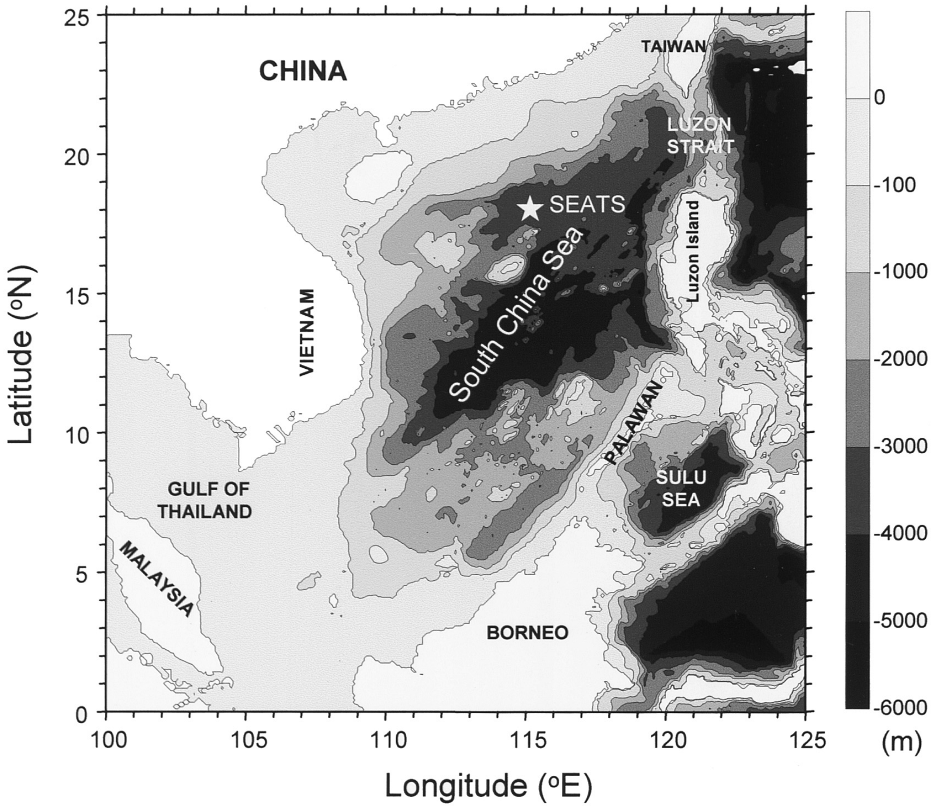

Fig. 1. Bathymetric map showing the location of the SEATS (South East Asia Time-series Study) time-series site (18°15’N, 115°35’E) at a water depth of ~3770 m in the northern South China Sea.

SCS is much shallower and its chlorophyll level in surface water is twice as high as those in the adjacent western North Pacific (Gong et al. 1992; Liu et al. 2002). In addition, several studies suggest that the seasonality of biogeochemical cycles is affected profoundly by the alternate monsoon system prevailing in the SCS. For instance, both modeled and SeaWiFS-derived primary production show a winter maximum during the stronger northeast monsoon and a summer minimum during the weaker southwest monsoon (Liu et al. 2002). Wong et al. (2002) reported that the concentrations of nitrate anomaly in the upper nutricline were systematically higher in fall through early spring during the northeast monsoon than in the summer during the southwest monsoon. The authors suggest that this phenomenon could be the result of higher nitrogen fixation during the northeast monsoon period. The objectives of this study are to document the carbon system in the mixed-layer, to calculate the seasonal variations of CO_2 flux, and to delineate the processes controlling its variability at the SEATS site over an annual cycle from March 2002 to April 2003.

2. METHODS

2.1 Sampling and Analysis

During the course of this study, the SEATS site was investigated aboard the *R/V Ocean Research I* and *III* in March, July, September, and November 2002, as well as January, March, and April 2003. On board, temperature and salinity were recorded with a CTD (SeaBird Inc. model 911 plus). All raw data and other pertinent measurements are archived in the SEATS time-series database of the Ocean Data Bank (ODB) at the National Center for Ocean Research (NCOR). Interested readers may contact NCOR-ODB for data requests. Discrete water samples at depths of 5, 10, 20, 40, 60, 80, 100, 125, 150, and 200 m were sampled and transferred into 250-ml BOD bottles from Go-Flo bottles mounted onto a Rosette sampling assembly (General Oceanic Inc.). All water samples are poisoned with 50 μl saturated $HgCl_2$ solution immediately after sampling and stored at 5°C in darkness to prevent biological alteration (DOE 1994).

Determinations of TCO_2 and TA follow the standard operating procedures described in DOE (1994). For TCO_2 analysis, a single operator multiparameter metabolic analyzer (SOMMA) system was used to control the pipetting system and extraction of CO_2 from seawater samples. The CO_2 gas was then measured by a coulometric detector (UIC, Coulometric Inc., model 5011) (Johnson et al. 1993). TA was determined by the potentiometric titration method (Bradshaw et al. 1981; Millero et al. 1993; DOE 1994). The set-up of the titration system is composed of a Radiometer pH meter (pHM-85), a GK 2041C combination electrode, an autoburette (Metrohm 665 Dosimat), an open titration cell, and a temperature-controlled water bath at 25 ± 0.05°C. A specific amount of seawater (~150 g) was first dispensed into the cell, and then titrated with hydrochloric acid, which was prepared in a NaCl solution with an ionic strength similar to that of seawater of 0.68, by passing the carbonic acid endpoint. The concentration of acid titrant (~0.1 N) was calibrated against solutions of Na_2CO_3, and then verified by both internal (i.e., seawater samples at a depth of 2500 m at SEATS) and international (provided by A. G. Dickson of the Scripps Institution of Oceanography) TA reference materials. Titration data passing the carbonic acid endpoint (~4.5 pH) were used to calculate TA, using a proton condition to define TA (Dickson 1981; 1992; Butler 1992). TCO_2 and TA were normalized to a salinity of 35 in order to eliminate the evaporation/precipitation effect, and are denoted as $NTCO_2$ and NTA, respectively.

Seawater references (batch number #51) prepared and provided by A. G. Dickson are used throughout this study for assessing the accuracy of our TCO_2 and TA measurements. Differences between the certified values (2050.28 ± 0.03 and 2269.86 ± 0.78 μmol kg^{-1} for TCO_2 and TA respectively) and our measurements are less than 2 μmol kg^{-1} and 3 μmol kg^{-1} for TCO_2 and TA, respectively. Precisions of the TCO_2 and TA analyses, estimated by repeated measurements of deep-water samples from 2000 - 3000 m on each cruise, are better than ±2 μmol kg^{-1} and ±2.5 μmol kg^{-1}, respectively. Finally, the fugacity of CO_2 (fCO_2) is computed from temperature, salinity, TCO_2, TA, phosphate, and silicate data for samples collected from each cruise using the program of Lewis and Wallace (1998), in which the carbonic dissociation constants are adopted from Mehrbach et al. (1973) and refitted by Dickson and Millero (1987). It is worth mentioning, however, that phosphate and silicate are almost undetectable at the micro-molar level in the mixed-layer at SEATS site. The error of the calculated fCO_2 is less than ±5 μatm.

2.2 Correction for Temperature Effect on fCO_2

In this study, the extent of temperature effect is assessed using the equation of Takahashi et al. (2002):

$$fCO_2 \text{ at } T_{obs} = (\text{Mean annual } fCO_2) \times \exp[0.0423(T_{obs} - T_{mean})], \quad (1)$$

where T is temperature in °C, and the subscripts "mean" and "obs" stand for the annual average and observed values, respectively. In our calculations, we use the averaged values of fCO_2 (364 μatm) and temperature (27°C) measured throughout the period of this study as the mean. The resultant fCO_2, designated as "fCO_{2mean} corrected for ΔT", are the expected fCO_2 values that are only affected by temperature if a parcel of water with a fCO_2 value of 364 μatm are subjected to seasonal temperature changes (the difference between observed and annual mean temperature) under constant TA and TCO_2. Moreover, in order to further discern other factors besides temperature that may affect fCO_2 changes, we normalized the observed fCO_2 to a constant temperature using the following equation (Takahashi et al. 2002):

$$fCO_2 \text{ at } T_{mean} = (fCO_2)_{obs} \times \exp[0.0423(T_{mean} - T_{obs})], \quad (2)$$

where T and the subscripts "mean" and "obs" are defined as the same as those in Eq. (1).

2.3 Calculation of ΔfCO_2 and Air-sea CO_2 Exchange Flux

Since there are no direct measurements of atmospheric fCO_2 at SEATS, we adopted 361.7 μatm as the atmospheric fCO_2 in our calculations. The value is modified from the average value of 370.89 ppmv reported at Mauna Loa Observatory for 2001 (Keeling and Whorf 2002) after corrections for the equilibrium water vapor pressure at a temperature of 27°C and a salinity of 33.5 (i.e., the annual mean temperature and salinity of the surface water at SEATS). Values of ΔfCO_2 in different seasons are derived as follows: spring value is the mean ΔfCO_2 of the observed data in March 2002 and 2003 as well as April 2003; summer value is the observed ΔfCO_2 in July 2002; fall value is the average of ΔfCO_2 in September and November 2002; winter value is the observed ΔfCO_2 in January 2003. The net sea-to-air CO_2 flux, F, is then computed using the following equation:

$$F = K \times (\Delta fCO_2), \quad (3)$$

where K is the CO_2 gas exchange coefficient, and ΔfCO_2 is $fCO_{2(seawater)}$ - $fCO_{2(atmosphere)}$. The seasonal wind-speed data at a 10-m height are computed from monthly average values for

1985-1999 from the ECMWF database (Liang et al. 2000). The potential errors in this calculation stem mainly from the uncertainties of the relationship between K and wind speed, as well as the calculated $fCO_{2(seawater)}$, which includes both analytical errors in TA and TCO_2, and the uncertainties of thermodynamic data applied in the calculation of $fCO_{2(seawater)}$. Therefore, we have used three formulations of K (i.e., Liss and Merlivat 1986; Tans et al. 1990; and Wannikohf 1992), and given an error of ±5 μatm to the calculated $fCO_{2(seawater)}$ in order to evaluate uncertainties of the estimated CO_2 flux.

3. RESULTS AND DISCUSSION

3.1 The Seasonal Hydrography at the SEATS Site

The relationships between potential temperature and salinity from different cruises at the SEATS site are depicted in Fig. 2. They are consistent with the general features reported previously for South China Sea waters (Gong et al. 1992; Chen and Huang 1996; Shaw et al. 1996; Lin et al. 1999; Chen et al. 2001; Wong et al. 2002), and have been used frequently to distinguish them from other water masses (e.g., the Western Philippine Sea water) upon mixing. As shown in Fig. 2, the SCS waters are characteristic of both a distinct shallow salinity maximum (S > 34.6; σ_θ = ~24.5) at approximately 150 m and a pronounced minimum (S = ~34.4; σ_θ = ~26.8) at about 350 m. The former is indicative of the North Pacific Tropical Water, while the later fingerprints the core of the North Pacific Intermediate Water (Nitani 1972). Below 15°C (which corresponds to a depth of about 200 m), data from the seven cruises are indistinguishable from each other throughout the year. On a closer examination, Fig. 2 further shows that sea surface temperature was moderately high (~26°C) in March 2002, increased gradually to a maximum of ~30°C in July 2002, then decreased to a minimum of ~24°C in January 2003, and rebounded back to ~28°C in April 2003, giving a total range of ~6°C over the annual cycle. In the meantime, salinities fluctuated between 33.4 and 33.7 with higher values in summer and winter and lower values in spring and fall. Thus, the seasonal variability of temperature and salinity at the SEATS site appears to be confined within the upper 200 m of the water column and characterized by the summer heating and winter cooling.

3.2 The Seasonal Patterns of TCO_2/ $NTCO_2$, TA/NTA and fCO_2 in the Mixed-layer

In this study, depths of mixed-layer and the data of TCO_2/ $NTCO_2$, TA/NTA, and the calculated-fCO_2 collected within the mixed-layer from each cruise are averaged to depict their overall seasonality in the mixed-layer at the SEATS site (Fig. 3). As seen in Fig. 3a, the mixed-layer depth, defined as "(σ_θ at the bottom of mixed-layer - σ_θ at 10 m) = 0.1 kg m^{-3}", remains relatively constant between 20 - 30 m until late fall (November), then reaches the maximal depth of ~90 m in January. The deepening of mixed-layer from late fall to winter is caused by the vigorous vertical mixing induced by winter cooling and increased wind stress as a much stronger northeast (winter) monsoon prevails in the SCS during this season (Liu et al. 2002; Tseng et al. 2005). A similar monsoon-induced temperature forcing and convective

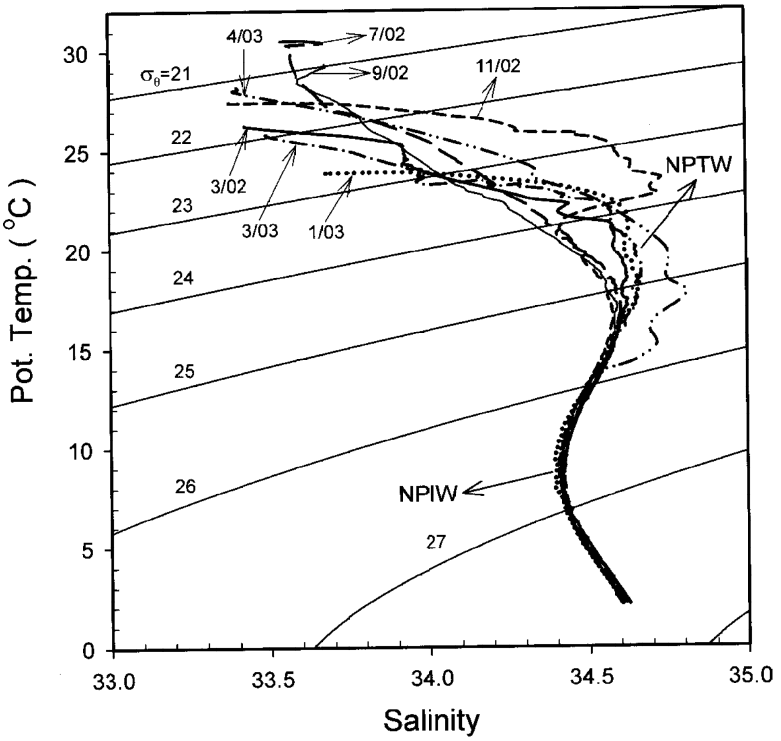

Fig. 2. T-S diagram showing the distinct shallow salinity maximum (S > 34.6; σ_θ = ~24.5) at approximately 150 m and the pronounced salinity minimum (S > 34.4; σ_θ = ~26.8) at about 350 m, a typical feature of the South China Sea waters. (NPTW: North Pacific Tropical Water; NPIW: North Pacific Intermediate Water)

mixing during northeast monsoon and inter-monsoon seasons are also observed in the central Arabian Sea (Goyet et al. 1998; Sarma et al. 1998; 2000).

TCO_2 remains fairly constant from ~1887 to ~1898 μmol kg^{-1} throughout the period of this study except for the highest value of ~1925 μmol kg^{-1} in winter (Fig. 3b). The TCO_2 maximum thus coincides with the deepening of the mixed-layer (Fig. 3a) and the winter cooling. Such a close association suggests that the strong northeast monsoon not only causes vigorous downward mixing to deepen the mixed-layer but also brings in more cold, TCO_2-rich subsurface waters from the deep. The drawdown of $NTCO_2$ in spring-summer (Fig. 3c) manifests the biological uptake in the mixed-layer. Similar observations were also reported previously at time-series stations of BATS (Bates et al. 1996) and HOT (Winn et al. 1994).

Figure 3d shows the seasonal variability of TA measured in the mixed-layer at SEATS site. TA values vary from ~2190 to ~2220 μmol kg^{-1} with higher values in January, July and September and lower values in March, April and November. The observed seasonal oscilla-

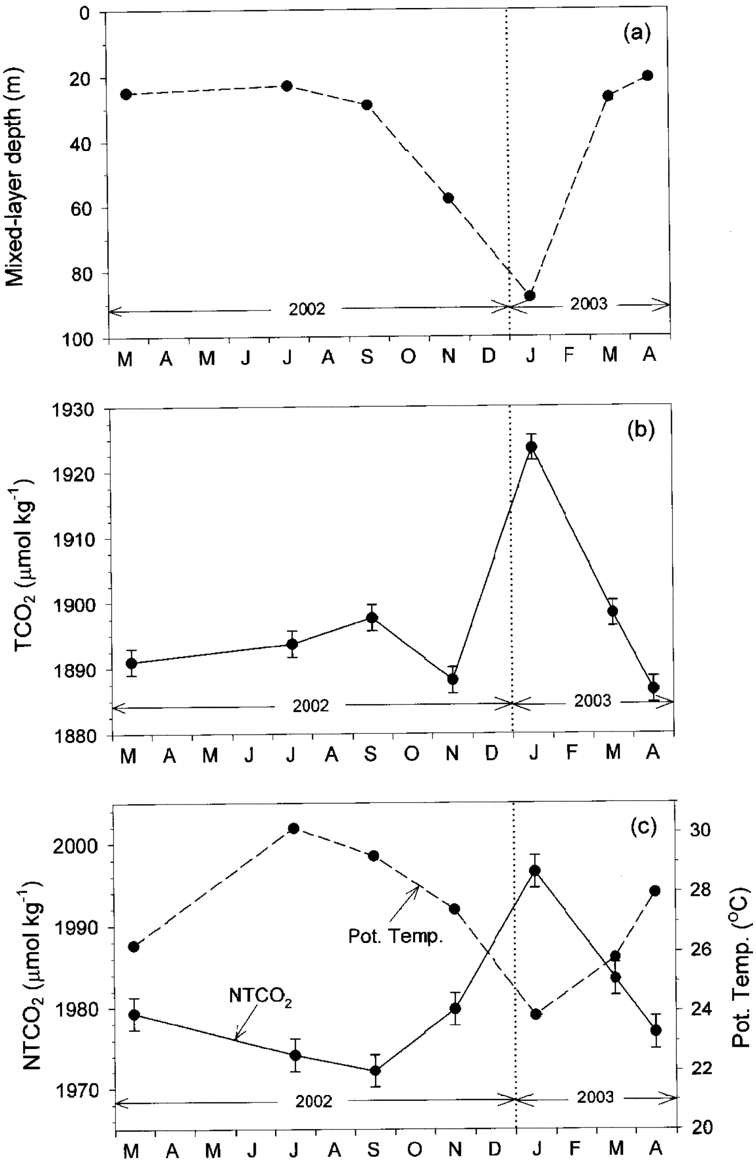

Fig. 3. Seasonal fluctuations of (a) the mixed-layer depth, (b) TCO_2, (c) $NTCO_2$ (solid line) and potential temperature (dash line), (d) TA (solid line) and salinity (dash line), (e) NTA and (f) fCO_2 (solid line) and potential temperature (dash line) in the mixed-layer at the SEATS site from March 2002 to April 2003. The data points represent the averaged values of all samples measured in the mixed-layer from each cruise. The vertical bars indicate the analytical errors ($\pm 1\sigma$).

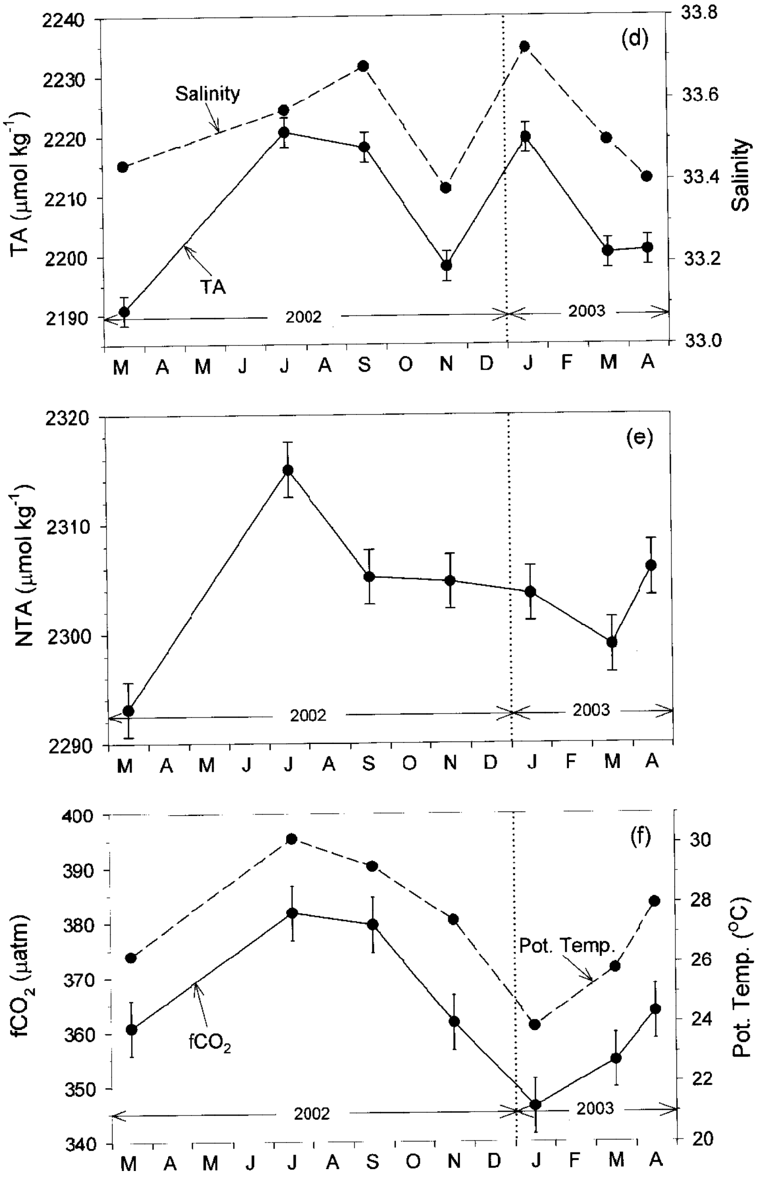

Fig. 3. Continued.

tion of TA is thus controlled primarily by the same factors affecting salinity. A similar relationship between TA and salinity is also documented at BATS and HOT sites (Winn et al. 1994; Bates et al. 1996). However, as revealed in Fig. 3e, NTA remains variable throughout the year, suggesting the potential contribution of the formation and/or dissolution of $CaCO_3$ and the consumption and/or remineralization of nitrate to the observed NTA variability.

A strong seasonality with an amplitude of ~35 μatm during the annual cycle is found in the mixed-layer fCO_2 at the SEATS site (Fig. 3f). The fCO_2 increases progressively in spring to summer with the maximum in July (~382 μatm) followed by a decrease in fall to winter with the minimum (~347 μatm) in January. The seasonal changes of fCO_2 are closely in phase with temperature (Fig. 3f) but are inversely correlated with $NTCO_2$ (Fig. 3c). These relationships suggest that the variation in fCO_2 at SEATS is controlled ultimately by seawater temperature change, and will be examined in detail in the following sections.

3.3 Processes Controlling the Mixed-layer fCO_2 at the SEATS Site

Figure 4 depicts graphically the seasonal variability of "fCO_{2mean} corrected for ΔT", the observed fCO_2, and "fCO_2 at 27°C" at the SEATS site. As can be seen, the magnitude of seasonal variation of the "fCO_{2mean} corrected for ΔT" is greater than the observed seasonal fCO_2 fluctuation, and the "fCO_2 at 27°C" and the "fCO_{2mean} corrected for ΔT" are inversely correlated. These relationships imply that the temperature effect on the seasonal variability of fCO_2 must have been compensated in part by other factors to yield the observed smaller amplitude.

These factors other than temperature include biological processes, air-sea exchanges of CO_2 and vertical transport of subsurface waters. Although it is difficult to discern their respective effects on the seasonal fCO_2 oscillations, the decrease in "fCO_2 at 27°C" from winter to summer (the warming period) can be attributed chiefly to biological effects because both the upward transport of subsurface waters and the air-sea CO_2 exchange are minimal during this period of time. The shoaling of the mixed-layer during the warming period (Fig. 3a) results in a stronger stratification that, in turn, prevents subsurface waters underneath the mixed-layer from moving upward. To further estimate the extent of biological effects on the mixed-layer fCO_2, we will use the "fCO_2 at 27°C" in January 2003 and July 2002 as winter and summer values, respectively, in the following discussion. Accordingly, the decline of "fCO_2 at 27°C" from winter to summer is ~60 μatm (Fig. 4), yet the drawdown of TCO_2 is only about one-half of that figure (~30 μmol kg^{-1}; Fig. 3c). Using a Revelle factor of 10 (Takahashi et al. 1993), the observed drop in TCO_2 is calculated to be equivalent to a decrease of fCO_2 of ~55 μatm. The close agreement between the measured difference and the hypothetical value (60 vs. 55 μatm) thus confirms the aforementioned biological utilization responsible for the decrease of "fCO_2 at 27°C" during the warming period.

On the other hand, the increase of "fCO_2 at 27°C" from summer to winter (the cooling period) mainly results from the entrainment of subsurface water from the deep. Assuming the biological effect is the same as that of the warming period (i.e., ~ -60 μatm) and the effect of air-sea exchange is negligible on the fCO_2 variation during the cooling period, a simple mass-balance calculation shows that the entrainment of the subsurface water will lead to an increase of fCO_2 of +120 μatm in the mixed-layer during this period, providing that the temperature effect on the fCO_2 change is about -90 μatm (i.e., the difference of "fCO_{2mean} corrected for ΔT" between July 2002 and January 2003; Fig. 4) and the observed fCO_2 change is about -30 μatm.

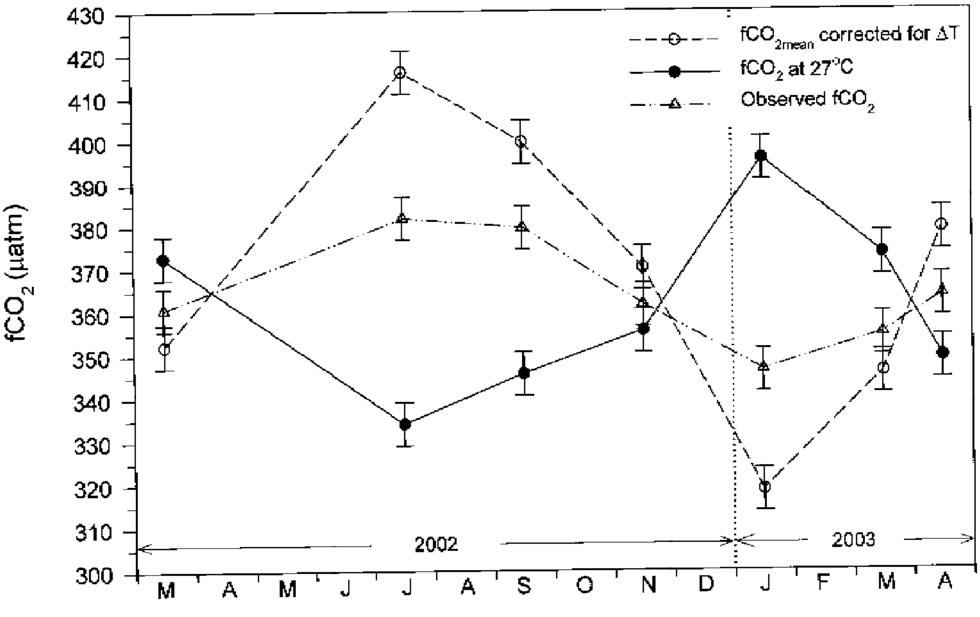

Fig. 4. Seasonal variability of "fCO$_{2mean}$ corrected for ΔT" (open circle), "fCO$_2$ at 27°C" (filled circle), and observed fCO$_2$ (open triangle) in the mixed layer at the SEATS site from March 2002 to April 2003. The vertical bars represent the range of potential errors. (see text for details in the definitions and calculations of "fCO$_{2mean}$ corrected for ΔT" and "fCO$_2$ at 27°C")

3.4 Assessing the Relative Importance of the Biological and Temperature Effects on Seasonal fCO$_2$ Variability at the SEATS Site

The relative importance of the temperature and biological effects on surface-water fCO$_2$ can be evaluated in terms of (T/B) and/or (T-B), in which (T/B) = $[(\Delta fCO_2)_{temp}/(\Delta fCO_2)_{bio}]$ and/or (T-B) = $[(\Delta fCO_2)_{temp} - (\Delta fCO_2)_{bio}]$ (Takahashi et al. 2002). According to the authors, the biological effect, $(\Delta fCO_2)_{bio}$, on the surface-water fCO$_2$ can be represented by the seasonal amplitude of "fCO$_2$ at T$_{mean}$", i.e., $(\Delta fCO_2)_{bio} = (fCO_{2mean}\ \text{at}\ T_{mean})_{max} - (fCO_{2mean}\ \text{at}\ T_{mean})_{min}$, while the temperature effect, $(\Delta fCO_2)_{temp}$, can be estimated from the seasonal magnitude of the "fCO$_{2mean}$ corrected for ΔT", i.e., $(\Delta fCO_2)_{temp} = (fCO_{2mean}\ \text{corrected for}\ \Delta T)_{max} - (fCO_{2mean}\ \text{corrected for}\ \Delta T)_{min}$. In any given oceanic regime, if the effect of temperature exceeds the biological effect, the (T/B) ratio is greater than 1 and the (T-B) is positive, whereas in areas where the biological effect appears to surpass the temperature effect, the (T/B) ratio varies from 0 to 1, and the (T-B) is negative. In areas where the two effects are equally important and cancel each other out, the (T/B) ratio is 1 and (T-B) is 0.

Figure 4 shows that the maximum and minimum of "fCO_2 at 27°C" at the SEATS site are ~395 and ~333 μatm, respectively, while the maximum and minimum of the "fCO_{2mean} corrected for ΔT" are ~415 and ~318 μatm. The maximal changes of $(\Delta fCO_2)_{bio}$ and $(\Delta fCO_2)_{temp}$, therefore, are ~62 μatm and ~97 μatm, respectively. Accordingly, the (T/B) ratio and the (T-B) difference are calculated to be 1.6 and 35 μatm, respectively. The results thus indicate that temperature overwhelms the biological effect on regulating the mixed-layer fCO_2 at the SEATS site.

A comparison of our results with those from other time-series stations (e.g., HOT, BATS, KNOT and OSP) is shown in Table 1. It is evident that the effect of temperature surpasses the biological effect at all of the subtropical time-series stations (SEATS, HOT and BATS), while the opposite is evident for stations located at high latitudes (KNOT and OSP). The contrast is attributed to the fact that the nutrient-rich surface waters in high latitudes usually are high in biological production and tend to reinforce the biological effect on regulating the fCO_2 in the mixed-layer. It should be noted that both temperature (97 vs. 59 μatm) and biological (62 vs. 23 μatm) effects on the fCO_2 in the mixed-layer at the SEATS site are greater than those at

Table 1. Comparison of the relative importance of temperature and biological effects on fCO_2 variations at the SEATS site with those at HOT, BATS, KNOT and OSP time-series stations.

	Location and Oceanic regime represented	Temperature effect / Biological effect	Temperature effect - Biological effect	References
SEATS	18°15'N , 115°35'E South China Sea, the largest subtropical marginal sea	1.6 (97/62 μatm)	+35 μtam	This study
HOT	22°45'N, 158°W North Pacific subtropical gyre	2.6 (59/23 μatm)	+36 μtam	Calculated based on Winn et al. (1994)
BATS	31°50'N, 64°10'W Western North Atlantic subtropical gyre	2.7 (150/55 μatm)	+95 μtam	Bates et al. (2001) Takahashi et al. (2002)
KNOT	44°N, 155°E Northwestern subarctic Pacific Ocean	0.8 (174/228 μatm)	-54 μtam	Calculated based on Tsurushima et al. (2002)
OSP	50°N, 145°W Northeastern subarctic Pacific Ocean	0.9 (100/115 μatm)	-10 μtam	Wong and Chan (1991) Takahashi et al. (2002)

HOT, despite the fact that both sites are situated at similar latitudes in the oligotrophic oceanic regime and have a similar seasonal change in mixed-layer depth (Neuer et al. 2002). The discrepancy thus can be attributed to the shallower thermocline, nutricline and TCO_2-cline at SEATS than those at HOT (Fig. 5), because the cold, nutrient-laden, TCO_2-rich subsurface waters can entrain easily from the deep to result in a greater amplitude of temperature and biological effects on the surface water fCO_2.

3.5 The Seasonal Pattern of ΔfCO_2 and Air-sea Exchange of CO_2 at the SEATS Site

Figure 6 shows the seasonal variability of ΔfCO_2 at the SEATS site, in which the maximum positive and negative ΔfCO_2 occur in summer (~ +20 μatm; July 2002) and in winter (~ -15 μatm; January 2003), respectively. In other words, there has been an efflux of CO_2 from the SCS in summer and fall but an influx of CO_2 into the surface SCS in winter. The seasonal variability also reveals that the ΔfCO_2 increases gradually from spring to summer, and then decreases progressively from fall to winter. Values of the calculated sea-to-air CO_2 flux at the SEATS site using different formulations (see Methods) are listed in Table 2. As seen, they vary remarkably in different seasons and depend on which formula is used in calculations, ranging from 0.00 ± 0.01 to -0.02 ± 0.05 (spring), +0.03 ± 0.01 to +0.23 ± 0.06

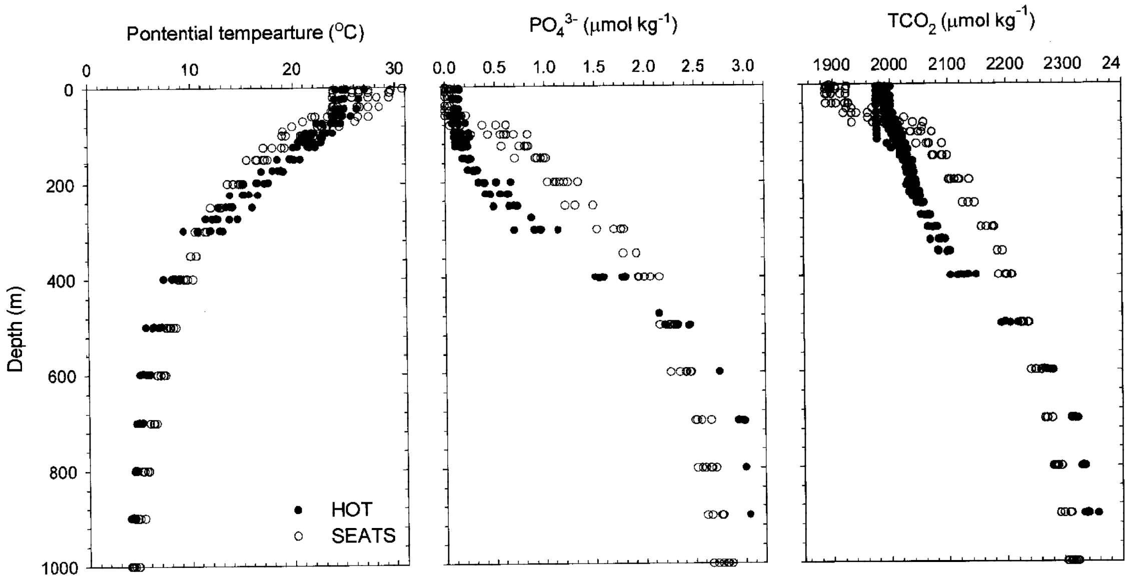

Fig. 5. Comparison of depth distributions of potential temperature, PO_4^{-3}, and TCO_2 from surface to 1000 m between the SEATS and HOT sites. The profiles of HOT are plotted from data collected in June, July, August, and December 1998, as well as January and February 1999 (R. Lukas and D. Karl, HOT project, 1999).

(summer), +0.18 ± 0.10 to +0.45 ± 0.25 (fall), and -0.62 ± 0.20 to -1.42 ± 0.46 (winter) molC m^{-2} $year^{-1}$. It further shows that the annual sea-to-air CO_2 flux is dominated by the influx of CO_2 from the atmosphere onto the surface waters in winter. It is worth pointing out that despite the bigger positive ΔfCO_2 in summer, the modest wind speed results in a rela-

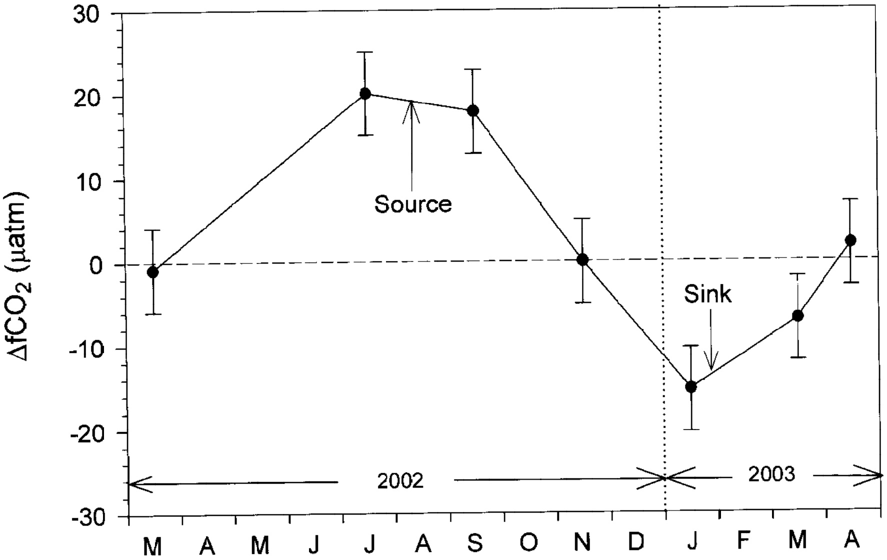

Fig. 6. Seasonal variations of ΔfCO_2 (surface seawater fCO_2 - atmospheric fCO_2) in the mixed layer at SEATS site from March 2002 to April 2003. The vertical bars represent the range of potential errors.

Table 2. Estimates of seasonal CO_2 flux at the SEATS site using formulations of Liss and Merlivat (1986), Tans et al. (1990), and Wanninkhof (1992). The seasonal wind speed data are calculated from monthly averaged values for 1985-1999 from the ECMWF database (Liang et al. 2000).

	ΔfCO_2	Wind speed	CO_2 flux (F, mol C m^{-2} yr^{-1}) and exchange coefficient (K, mol C m^{-2} $year^{-1}$ μatm^{-1})					
	(µatm)	(ms^{-1})	Liss and Merlivat (1986)		Tans et al. (1990)		Wanninkhof (1992)	
			K	F	K	F	K	F
Spring	-1.9 ± 5	3.1	0.001	0.00 ± 0.01	0.002	0.00 ± 0.01	0.011	-0.02 ± 0.05
Summer	20.1 ± 5	3.2	0.001	0.03 ± 0.01	0.003	0.06 ± 0.02	0.012	0.23 ± 0.06
Fall	9.0 ± 5	6.1	0.018	0.18 ± 0.10	0.050	0.45 ± 0.25	0.042	0.38 ± 0.21
Winter	-15.3 ± 5	8.8	0.041	-0.62 ± 0.20	0.093	-1.42 ± 0.46	0.087	-1.33 ± 0.43
Yearly Average				-0.11 ± 0.08		-0.23 ± 0.18		-0.19 ± 0.19

tively small CO_2 efflux. In contrast, the high wind speed in winter gives rise to higher CO_2 influx regardless of the smaller ΔfCO_2. The annual sea-to-air CO_2 flux at the SEATS site is estimated to be around -0.11 ± 0.08 to -0.23 ± 0.18 molC m^{-2} $year^{-1}$ during the observed period (Table 2), and is in good agreement with that reported previously by Takahashi et al. (2002) in the SCS region (~ 0 to -0.4 molC m^{-2} $year^{-1}$).

Similar seasonal patterns have also been documented at BATS and HOT (Winn et al. 1994; Bates et al. 1996). Our estimates, however, are less than -0.7 molC m^{-2} $year^{-1}$ found at HOT (Winn et al. 1994) or -0.3 to -0.6 molC m^{-2} $year^{-1}$ found at BATS (Bates et al. 1998). The lower values at SEATS can be attributed to the higher sea surface temperature at SEATS, which increases fCO_2 in the surface water and depresses the capacity for CO_2 uptake, and the general upwelling circulation in the SCS (Chao et al. 1996; Chen et al. 2001). Finally, if the estimated annual sea-to-air CO_2 flux at SEATS are extrapolated to the entire SCS (3.5×10^6 km^2), it could take up 4.5 ± 3.3 ~ 9.6 ± 7.7 TgC yr^{-1}, i.e., only about 0.20 ± 0.15 to 0.44 ± 0.35% of the total CO_2 uptake by the global oceans (~2.2 PgC yr^{-1}; Takahashi et al. 2002). These values are small, considering the SCS's share that occupies 0.97% of the total ocean area. The small fraction of CO_2 uptake estimated for the SCS in this study, however, is consistent with the small percentage of CO_2 inventory reported by Chen et al. (2005), in which the SCS contains 0.6 ± 0.1 PgC of anthropogenic CO_2, while the global ocean inventory is about 100 PgC.

3.6 Estimate of Net Community Production and the "Mysterious Carbon Drawdown"

In our previous discussion, we attributed the decrease of $NTCO_2$ and the drawdown of "fCO_2 at 27°C" from winter to summer to the biological uptake. The net amount of $NTCO_2$ consumed biologically in the mixed-layer during this warming period can be regarded as the net community production (NCP), i.e., the difference between primary production and net community respiration. The observed change of $NTCO_2$ between the overturned period in winter (Tseng et al. 2005) and the stratified period in summer is about 25 μmol kg^{-1} (Fig. 3c). Assuming a mean mixed-layer depth of 50 m, the NCP in the mixed-layer can be calculated to be about 1.25 ± 0.14 molC m^{-2}. Since this value only represents the phytoplankton growth over the first six months in a year, the annual net community production would be about 6.8 ± 0.77 mmolC m^{-2} $year^{-1}$, if the NCP during the cooling period, i.e., from summer to winter, were the same as that during the warming period. The source of TCO_2 as well as nutrients in the later six months in a year is assumed to be supplied by less dramatic but significant diffusive fluxes, including typhoon mixing (Lin et al. 2003). The uncertainty in this calculation was assumed to derive only from the analytical error of TCO_2, which was reported as ±2 μmol kg^{-1}. Accordingly, a corresponding error of ±2.8 μmol kg^{-1} in $\Delta NTCO_2$ (the square root of two times of TCO_2's error) was applied to estimate the uncertainty of calculated NCP.

In a thorough investigation of the roles of nitrogen in modulating phytoplankton growth in the northern SCS basin, Chen et al. (2004) noted a great variability of primary production between 15.0 and 27.5 mmolC m^{-2} $year^{-1}$ in March 2000 and 2001, respectively, while the new production varied from 5.0 to 10.0 mmolC m^{-2} $year^{-1}$. In addition, using a modeling result and the data from SeaWiFS ocean color images, Liu et al. (2002) reported an annual mean primary productivity of ~23.3 - 29.5 mmolC m^{-2} $year^{-1}$, and predicted an e-ratio of 0.12 in the SCS. In other words, the averaged export production in the SCS would range from 2.80 to 3.54 mmolC m^{-2} $year^{-1}$. Our estimated NCP using carbon data in this study is thus in reasonable agreement with the reported new and export productions, and supports the importance of biological utilization in controlling the observed seasonal drawdown of "fCO_2 at 27°C" and $NTCO_2$ from winter to summer at the SEATS site in the South China Sea.

Nonetheless, nitrate concentrations in the mixed-layer at the SEATS site are undetectable except a trace of 0.5 μmol kg^{-1} found in January during the course of this study. The observation of no measurable nitrate in the mixed-layer during spring and summer is also reported by Wong et al. (2002) and Chen et al. (2004). The continuous decrease of $NTCO_2$ in the absence of measurable nitrate, the so-called "mysterious carbon drawdown", observed in this study has also been observed at BATS and HOT time-series stations (Michaels et al. 1994; Bates et al. 1996; Karl et al. 1997). A recent global ocean analysis further shows that such a phenomenon is a common feature for all tropical and subtropical marine habitats (Lee et al. 2001). Moreover, it has been shown that in seawaters of low-nitrate concentrations the C : N ratio utilized by marine organisms is very different from the conventional Redfield (Copin-Montegut 2000; Michaels et al. 2001). Assuming a C : N ratio of 16 (ref. Copin-Montégut 2000), the utilization of 0.5 μmol kg^{-1} nitrate can only sustain 8 μmol kg^{-1} of $NTCO_2$ drawdown, i.e., only 32% of the overall drawdown of $NTCO_2$ (25 μmol kg^{-1}; Fig. 3c) measured between winter and summer.

It thus appears that other sources of nitrogen from processes such as nitrogen fixation, riverin inputs, vertical migration of zooplankton, utilization of dissolved organic nitrogen, vertical diffusion of nutrients from the deep, horizontal advection and atmospheric deposition are required to compensate for the deficit. For instance, nitrogen fixation has been the most frequently mentioned process in the literature and shown to be a new production pathway in nutrient-depleted waters (Karl et al. 1997; Michaels et al. 2000). In fact, Wong et al. (2002) report high values of nitrate anomaly up to 2.5 μmol kg^{-1} at the SEATS site from the fall through early spring during the northeast monsoon, indicating that the remineralization of nitrogen-rich organic matter formed by nitrogen fixation have played an important role in nutrient dynamics in the northern SCS. However, as shown by Chen et al. (2004), the nitrogen-fixing cynobacteria, *Trichodesmium* sp. and *R. intracellularis*, are both sparsely distributed in the mixed layer during spring, and constitute less than 3% of the new production. The dichotomy between rates of nitrogen fixation measured directly by biological techniques and inferred from geochemical data in the North Atlantic has been thoroughly investigated by Bates and Hansell (2004) and Hansell et al. (2004). Further studies, therefore, are necessary to elucidate the sources of nitrogen in the mixed layer at SEATS site in the northern SCS.

4. CONCLUSIONS

A better assessment of seasonal and inter-annual variability of CO_2 fluxes in different oceanic regimes is a premise for adequately quantifying the global oceanic CO_2 uptake. However, our present knowledge from time-series studies has been largely achieved by studies in the open ocean (e.g., BATS and HOT), with scant attention to the marginal seas. In this study, we report, for the first time, the seasonal variations of carbon system at the SEATS time-series site in the northern South China Sea, based on the measurements from March 2002 to April 2003. Results show that the seasonal pattern of $NTCO_2$ in the mixed-layer is characterized by a progressive decline from spring to early fall and by a subsequent increase in fall and winter. This seasonal variability of $NTCO_2$ is mainly due to biological utilization in spring-summer, and to strong vertical mixing in fall-winter, respectively. The seasonal changes of fCO_2, however, are nearly opposite to that of $NTCO_2$. The observed fCO_2 augmentation in spring-summer appears to be primarily the result of temperature increases, despite the fact that about 60% of such a temperature effect is compensated by biological utilization. In the fall and winter, the temperature effect dominates over the combined effects of subsurface water entrainment and biological production, giving rise to diminishing fCO_2 during the cooling period.

By comparison, our data show that the shallower thermalcline, nutricline and TCO_2-cline at SEATS have resulted in a greater extent of biological and temperature effects on fCO_2 variations than at HOT. According to the seasonal fCO_2 distributions, surface waters at the SEATS site act as an atmospheric CO_2 sink in winter, yet a source in summer and fall during the observed period. Over the annual cycle, however, a net sea-to-air CO_2 flux is estimated to be -0.11 ± 0.08 to -0.23 ± 0.18 $\mathrm{molC\ m^{-2}\ year^{-1}}$. In response to the observed $NTCO_2$ drawdown, an estimated net community production of 6.8 ± 0.77 $\mathrm{mmolC\ m^{-2}\ year^{-1}}$ in the mixed-layer is obtained. The continuous spring-summer drawdown of $NTCO_2$ with the absence of nitrate, however, suggests other sources of nitrogen are required to support the observed new production.

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