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# Surface $CO_2$ partial pressure and air–sea $CO_2$ flux on the China side of the South Yellow Sea based on multiple-year underway measurements during 2005–2011 and comparison with results for 2011–2018

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#### ABSTRACT

By resolving spatiotemporal variations in sea surface partial pressure of  $CO_2$  ( $pCO_2$ ) based on multiple-year underway measurements, we quantified basin-scale air–sea  $CO_2$  exchange flux on the China side of the South Yellow Sea between 2005 and 2011, and compared it with the result obtained between 2011 and 2018. Over the three subregions under study, the area-weighted average of the  $CO_2$  influx rate was estimated to be  $1.1 \pm 1.5$  mol m<sup>-2</sup> yr<sup>-1</sup> during 2005–2011, which is only half the annual  $CO_2$  influx rate over the adjacent East China Sea shelf but not remarkably different from the rate ( $0.4 \pm 2.1$  mol m<sup>-2</sup> yr<sup>-1</sup>) estimated for almost the same sea area during 2011–2018. Over central and western parts of the South Yellow Sea, subregion-specific fitting curves of monthly variations in air–sea  $CO_2$ flux during 2005–2011 were consistent with those monthly variations during 2011–2018. However, over the southern part of the South Yellow Sea near the Changjiang Estuary, the fitting curve of monthly variations in air–sea  $CO_2$  fluxes during 2005–2011 located systematically on one side of the monthly variation estimated for 2011–2018, indicating that annual  $CO_2$  uptake had weakened in this subregion. Both cases were different from some other ocean margin areas characterized by enhanced  $CO_2$  uptake following rise in atmospheric  $CO_2$ . To study the potential shift in terms of shelf  $CO_2$  sources and sinks over multiple decades and under environmental changes, further field observations and data-based research are needed on several large continental shelves.

#### 1. Introduction

Despite comprising just 7–8 % of the world's ocean surface area, continental shelf seas play a nonnegligible role in the global oceanic uptake of atmospheric CO<sub>2</sub> (e.g., Wanninkhof et al., 2013; Laruelle et al., 2014; Dai et al., 2022). Recent evidence has shown that the CO<sub>2</sub> influx rate of some continental shelf systems might have increased during the past several decades (Laruelle et al., 2018; Mathis et al., 2024). However, quantifying air–sea CO<sub>2</sub> fluxes on continental shelves and understanding their variations remain challenging (Resplandy et al., 2024) because coastal waters are highly heterogeneous in terms of air–sea CO<sub>2</sub> exchange (Laruelle et al., 2010; Cai, 2011; Chen et al., 2013; Dai et al., 2013; 2022). Moreover, continental shelves are affected by intense human disturbances such as sewage discharges and coastal wetland reclamation (Bauer et al., 2013; Regnier et al., 2013; Li et al., 2023a).

The East China Sea (ECS) and the South Yellow Sea (SYS) together represent a globally important shelf system, characterized by a wide shelf and water exchange between complex coastal currents and the Kuroshio Current system (Fig. 1). The ECS serves as a remarkable annual net sink of atmospheric  $CO_2$  (Zhang et al., 1997; Tsunogai et al., 1999;

Tseng et al., 2014; Guo et al., 2015), whereas the SYS serves as a weak annual sink of atmospheric CO<sub>2</sub> (Choi et al., 2019; Wang and Zhai, 2021; Ko et al., 2022). In 2023, Li et al. (2023b) applied a random-forestalgorithm-based machine learning method to modeling of the surface partial pressure of CO<sub>2</sub> (pCO<sub>2</sub>) in the Yellow Sea, using Moderate Resolution Imaging Spectroradiometer (MODIS) derived sea surface temperature, chlorophyll-a concentration, diffuse attenuation of downwelling irradiance, and salinity. Their modeled pCO2 increased at a rate much slower than the rate of increase in atmospheric CO<sub>2</sub>, implying that the Yellow Sea might have changed from an annual source of atmospheric CO<sub>2</sub> in the 2000 s to an annual sink in the 2010 s (Li et al., 2023b). However, the correctness of machine learning methods is highly subject to the training datasets used, and Li et al. (2023b) trained their model using only a dataset obtained between 2011 and 2019, thereby lacking verification through the 2000 s. By contrast, another satellite retrieval algorithm (combining a semi-mechanistic algorithm and machine learning method) indicated that the rate of increase in Yellow Sea pCO<sub>2</sub> during 2003-2019 should be close to that of the rise in atmospheric CO<sub>2</sub> (Yu et al., 2023). To examine the reliability of those machine learning results, further investigation of additional field data is

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#### needed.

Between 2005 and 2011, we conducted 26 cruises through combined areas of the ECS and the SYS (Supplementary Table S1), obtaining underway measurements of sea surface pCO<sub>2</sub> (Supplementary Fig. S1). Most of those cruises passed through two or three of the three physical-biogeochemical domains in the SYS categorized in the present study (Table 1). Although Zhai and Dai (2009) and Guo et al. (2015) both used some data other than those obtained in September 2008, June 2009, and November/December 2011, they focused on the air-sea CO<sub>2</sub> exchange in the ECS and largely ignored those data obtained in the SYS. In this study, we revisited this dataset and elucidated the seasonal or even monthly variations in air-sea CO2 flux in the SYS between 2005 and 2011 based on underway pCO2 measurements. We then compared our results with the data published by Wang and Zhai (2021), who reported monthly variations in air-sea CO<sub>2</sub> flux over the same shelf region between 2011 and 2018. The objective of this study was to conduct a trend analysis of air-sea CO2 flux in the Yellow Sea, an important continental shelf sea, using the available datasets. To avoid potential confusion induced by zoning overlapping between this study and Guo et al. (2015), we also recalculated results for the ECS shelf (see Supplementary Materials), based on a slightly updated zoning scheme from Guo et al. (2015) over the ECS region (Fig. 1C, Supplementary Table S2).

# 2. Materials and methods

During the cruises, temperature and salinity of the pumped surface seawater were measured continuously using either a YSI 6600 meter (Yellow Springs Instrument Co., U.S.) or a set of Idronaut Multiparameter "Flow Through" sensor modules (IDRONAUT S.r.l., Italy). All salinity data were corrected to the practical salinity scale 1978, while sea surface temperature (SST) and dissolved oxygen (DO) were corrected on the basis of intercomparison experiments between underway pumping measurements and vertical profile measurements at sampling stations. For  $pCO_2$  determination, a Li-Cor® nondispersive infrared spectrometer (Li-7000) was used to measure the dried CO<sub>2</sub> fraction ( $xCO_2$ ) in the equilibrator and in the air. For calibration purposes, CO<sub>2</sub> gas standards with  $xCO_2$  values in the range of 138–967 ppmv (parts per million by volume in dry air) were applied. The  $pCO_2$  data was transformed from corrected  $xCO_2$  based on either barometric pressure along the transect or air pressure in the Li-7000 detector. Intercalibration testing showed that both air pressure data sets were consistent at a relative error level of < 0.3 % (i.e., < 3 hPa). The Weiss and Price (1980) saturated water vapor pressure and the Takahashi et al. (1993) temperature effect coefficient of 4.23 % °C<sup>-1</sup> were used to calculate in situ sea surface  $pCO_2$ . The overall uncertainty of the  $xCO_2$  measurements and the  $pCO_2$  data processing was < 1 % (Zhai and Dai, 2009).

Considering the complexity of the geographical features and the hydrological conditions in coastal seas (Guo et al., 2015; Wang and Zhai, 2021), the study area of the SYS was divided into three subregions: (I) the western SYS, (II) the Chinese side of the central SYS, and (III) the southern part of the SYS (Fig. 1C, Table 1). The western SYS is characterized by the Jiangsu Shallows and the resuspension of local sediments. The boundary between the western and central SYS areas is aligned approximately along the Jiangsu Front and the Shandong Peninsula Front in winter (Chen, 2009). The southern SYS, which is confined to the region south of 34°N, is affected by the Changjiang Diluted Water in summer (Chen, 2009; Guo et al., 2015). In this study, we assumed that the arithmetic mean of the field data was representative of the given subregion during the survey.

The flux calculation was based on the formula  $F = k \times K_H \times \Delta pCO_2$ , where *k* is the gas transfer velocity of CO<sub>2</sub>, *K*<sub>H</sub> is the solubility of CO<sub>2</sub> in seawater (Weiss, 1974), and  $\Delta pCO_2$  is the mean sea-to-air  $pCO_2$  difference. A positive flux value represented net CO<sub>2</sub> exchange from the sea to the atmosphere, whereas a negative flux value represented net CO<sub>2</sub> exchange from the atmosphere to the sea. Because in situ measurements of atmospheric CO<sub>2</sub> concentration were not always available during our surveys, we calculated the air-equilibrated  $pCO_2$  levels based on flask analysis data of the atmospheric CO<sub>2</sub> mole fraction at the adjacent Taeahn Peninsula site (36°44′N, 126°08′E), which is located to the west of South Korea (data from the U.S. NOAA/ESRL's Global Monitoring Division, https://www.esrl.noaa.gov/gmd/), and corrected them to survey-based barometric pressure at 10 m above the sea surface and 100 % humidity at SST and sea surface salinity. Usually, the flask analysis data were satisfactorily comparable to our shipboard measurements of



**Fig. 1.** Area maps. (a) The Bohai Sea, North Yellow Sea (NYS), South Yellow Sea (SYS), East China Sea (ECS), and circulation in winter; (b) circulation in summer (Chen, 2009; Zhai, 2018); and (c) the physical-biogeochemical domains categorized in this study, as detailed in Table 1 (for the SYS) and Table S2 (for the ECS). In panel (a), YSCC = Yellow Sea Coastal Current, and YSWC = Yellow Sea Warm Current. In panel (b), YSCWM = Yellow Sea Cold Water Mass. Depth contours of 25, 50, 100, 200 and 1000 m are shown as thin gray lines; boundaries of sea areas are shown as red dashed lines. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Phy	sical-biogeochemical	domains in the So	outh Yellow S	Sea (SYS)	categorized in the	present study

Subregion #	Subregion name / description	Characteristics	Latitude (°N)	Longitude (°E)	Area (10 <sup>3</sup> km <sup>2</sup> )	Surveying months
Ι	Western part of the SYS	Shallow waters (<50 m), sediment resuspension	32 - 37	120 - 122.3	58.138	Apr. 2005, May 2005, Sep. 2006, Oct. 2006, Jul. 2007, Nov. 2007, Mar. 2008, Apr. 2008, Aug. 2008, Sep. 2008, Dec. 2008, Jan. 2009, Mar. 2009, Apr. 2009, May 2009, Jun. 2009, Jul. 2009, Aug. 2009, Feb. 2010, Dec. 2010, Apr. 2011, Jun. 2011, Nov. 2011, Dec. 2011
Π	Chinese side of the central SYS	Highly stratified in summer, warm current in winter	34 – 37	122.3 – 124	51.330	May 2005, Sep. 2006, Aug. 2008, Mar. 2009, Apr. 2009, May 2009, Jun. 2009, Aug. 2009, Feb. 2010, Jun. 2011, Nov. 2011, Dec. 2011
III	Southern part of the SYS	Highly stratified in summer, with river plume signals visible in flood seasons	32 - 34	122.3 – 125	55.958	Apr. 2005, May 2005, Sep. 2006, Oct. 2006, Nov. 2006, Jul. 2007, Mar. 2008, Apr. 2008, Aug. 2008, Sep. 2008, Mar. 2009, Apr. 2009, May 2009, Jul. 2009, Aug. 2009, Feb. 2010, Dec. 2010, Apr. 2011, Jun. 2011, Dec. 2011

atmospheric xCO<sub>2</sub> (Supplementary Fig. S2).

In situ measurements of the k value in the SYS were unavailable, and therefore we calculated the k value using the Sweeney et al. (2007) empirical function of wind speed at 10-m height  $(u_{10})$ , which is a modification from Wanninkhof (1992) empirical functions. Notably, the Sweeney et al. (2007) formula has been verified by Takahashi et al. (2009), Ho et al. (2011) and Yang et al. (2022) using their own reanalysis and/or field-measured data. Wanninkhof (2014) also proposed an empirical formula that produces results only  $\sim 10$  % lower than those obtained using the Sweeney et al. (2007) formula. According to Yang et al. (2022) synthesis of air-sea CO2 transfer velocity estimates from ship-based eddy covariance (EC) measurements, estimates of CO2 fluxes obtained using a  $u_{10}^2$  dependence with zero intercept are likely biased when the  $u_{10}$  values are  $< 7 \text{ m s}^{-1}$ . They developed an empirical function that produces k values similar to those derived using the Sweeney et al. (2007) parameterization at moderate-high wind speed, but that substantially exceed the latter at low wind speed, possibly attributable in part to chemical enhancement in air-sea CO<sub>2</sub> exchange (Wanninkhof and Knox, 1996). In this study, we also calculated the k value based on the Yang et al. (2022) empirical function to compare our results with those considering the effect of chemical enhancement at low wind speed.

Wind speed data with  $0.25^{\circ} \times 0.25^{\circ}$  spatial resolution and daily temporal resolution were obtained from the Cross-Calibrated Multi-Platform (CCMP) Ocean Surface Wind dataset developed by the U.S. Jet Propulsion Laboratory. The data were derived through cross-calibration and assimilation of data from several sensors (i.e., SSM/I, TMI, AMSR-E, SeaWinds on QuikSCAT and SeaWinds on ADEOS-II), which were also combined with conventional observations and a starting estimate of the wind field derived using a variational analysis method. The monthly average was adopted in the calculation of the air–sea CO<sub>2</sub> flux.

Following Wanninkhof (1992), the effect of short-term variability in wind speed over a month on the gas transfer velocity was determined using  $C2 = (u_j^2)_{\text{mean}}/(u_{\text{mean}})^2$ , where  $u_j$  is all of the available satellite-derived near surface wind speeds (units: m s<sup>-1</sup>) in a month, the subscript "mean" indicates the average, and  $u_{\text{mean}}$  is the monthly mean wind speed. During our study period, the satellite-derived *C*2 varied between 1.14 and 1.53 (Tables 2–4), similar to the range reported earlier (1.17–1.65) as derived from field data recorded by onboard weather stations (Wang and Zhai, 2021).

The Sweeney et al. (2007) equation (S07 for short) is expressed as follows:

$$k(/\text{cm h}^{-1}) = C2 \times 0.27 \ u_{10}^2 \times (\text{Sc}/660)^{-0.5},$$
 (1)

while the EC-derived (EC22 for short) equation developed by Yang et al. (2022) is expressed as follows:

$$k(/\text{cm h}^{-1}) = C2 \times (0.36 + 1.203 u_{10} + 0.167 u_{10}^2) \times (\text{Sc/660})^{-0.5}, (2)$$

where  $u_{10}$  (unit: m s<sup>-1</sup>) is the satellite-derived monthly mean nearsurface wind speed at 10-m height; *C*2 is the nonlinearity coefficient, assuming that the long-term wind follows a Raleigh (Weibull) distribution (Wanninkhof, 1992); Sc is the Schmidt number of CO<sub>2</sub> in seawater; and 660 is the Sc value in seawater at salinity of 35 and temperature of 20°C (Wanninkhof, 1992).

## 3. Results

#### 3.1. Data overview

Over the SYS, survey-averaged SST varied between 4.2  $\pm$  1.0  $^\circ C$  in the western part (Subregion I) in February 2010 and 27.6  $\pm$  1.0  $^\circ\text{C}$  in the southern part (Subregion III) in August 2008 (Fig. 2). In contrast to the substantial seasonal variation in SST, survey-averaged surface salinity usually showed limited variation, ranging between 30.9  $\pm$  0.5 (in November 2007) and 32.1  $\pm$  0.5 (in March 2009) in Subregion I, and from 30.9  $\pm$  0.5 (in August 2008) to 33.2  $\pm$  0.6 (in April 2009) in Subregion II (i.e., the central SYS). In Subregion III, survey-averaged surface salinity exhibited the lowest value of 29.6  $\pm$  1.6 (in July 2009) recorded in this study, and the relatively high value of 33.0  $\pm$  0.7 was recorded in November 2006. The relatively low values of salinity of < 30 and substantial standard deviation of salinity of > 1.0 were observed in late spring and summer (Fig. 2F), indicating the Changjiang Diluted Water in this subregion (Fig. 1B). It is also worth noting that in Subregions I and II, relatively high survey-averaged surface salinity values of 32–33 were usually associated with winter/spring cold waters, while relatively low survey-averaged surface salinity values of 30-31 were usually associated with summertime/autumnal warm waters (Fig. 2).

During the study period, sea surface  $pCO_2$  exhibited dramatic heterogeneity, with huge variations in both temporal and spatial spans. In the SYS, the two lowest  $pCO_2$  values of ~ 100 µatm (120 µatm at 32°17′N 122°47′E in May 2009 and 88 µatm at 32°01′N 122°40′E in June 2011) and the two highest  $pCO_2$  values of ~ 810 µatm (804 µatm at 32°05′N 122°12′E in August 2008 and 818 µatm at 32°29′N 122°42′E in early September 2008) were observed near the Changjiang Estuary. In Subregion I, survey-averaged surface  $pCO_2$  varied between 285 ± 16 µatm in February 2010 and 458 ± 55 µatm in June 2011 (Fig. 2G), while survey-averaged surface  $pCO_2$  in Subregion II ranged from 298 ± 26 µatm in September 2006 to 374 ± 43 µatm in August 2008 (Fig. 2H). In Subregion III, survey-averaged surface  $pCO_2$  usually ranged between 287 ± 66 µatm in July 2007 and 449 ± 83 µatm in October 2006 (Fig. 2I), while the highest survey-averaged value of surface  $pCO_2$  of 642 ± 79 µatm was detected in early September 2008.

On the basis of monthly averaged values of the flask analysis data of atmospheric CO<sub>2</sub> mole fraction at the Tae-ahn Peninsula site, the air-equilibrated levels of sea surface  $pCO_2$  varied between 363 µatm (in Subregion I in August 2009) and 401 µatm (in all of the three subregions

#### Table 2

Summary of pCO<sub>2</sub>, salinity and SST along cruise tracks, and monthly satellite-derived wind speed, and air–sea CO<sub>2</sub> flux estimation in Subregion I, with positive values indicating that the sea area release CO<sub>2</sub> to the atmosphere. The annual air–sea CO<sub>2</sub> flux amounts to  $-0.9 \pm 1.5$  mol CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup> using S07 equation or  $-1.0 \pm 2.1$  mol CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup> using EC22 equation (Yang et al., 2022). Note that seasonal average CO<sub>2</sub> fluxes were calculated from the monthly averages of the three calendar months in the given season. Errors in air–sea CO<sub>2</sub> flux estimation are the temporal variability of CO<sub>2</sub> fluxes based on those cruises/surveys.

Observation	Sea surface pCO <sub>2</sub> (µatm)	Sea surface	Air	Air Salinity	SST Wind	Wind	C2	Air-sea $CO_2$ fluxes (mmol $CO_2 m^{-2} d^{-1}$ )			
time		$pCO_2$ ( $\mu$ atm)	(°C)	(°C)	(m s <sup>-1</sup> ))))))))))))))))))))))))))))))))))))		Survey average (EC22)	Seasonal average (EC22)	Survey average (S07)	Seasonal average (S07)	
Dec 2008	$336 \pm 8$ (320–358)	390.2	$31.25 \pm 0.56$ (29.74–32.08)	$9.5 \pm 1.2$ (7.78–11.66)	$\begin{array}{c} 8.2 \pm \\ 3.2 \end{array}$	1.14	-11.28	$-14.07 \pm 7.72$ (in winter)	-9.56	$-11.11 \pm 5.96$ (in winter)	
Dec 2010	$355 \pm 18$ (319–396)	390.6	$31.55 \pm 0.13$ (31.22–31.79)	$12.9 \pm 0.5$ (11.90–14.09)	8.5 ± 4.2	1.24	-8.19		-7.04		
Dec 2011	$400 \pm 20$ (361-441)	398.7	$31.22 \pm 0.23$ (30.65–31.70)	$14.3 \pm 0.7$ (12.43–15.25)	$6.7 \pm 3.1$	1.21	0.20		0.15		
Jan 2009	$313 \pm 10$ (292–331)	396.3	$\begin{array}{c} 31.68 \pm 0.44 \\ (30.07  32.15) \end{array}$	7.3 ± 1.2 (5.29–8.72)	6.9 ± 4.0	1.32	-15.72		-12.21		
Feb 2010	285 ± 16 (266–320)	398.1	31.88 ± 0.04 (31.73–31.94)	4.2 ± 1.0 (1.84–5.31)	7.0 ± 3.3	1.22	-20.08		-15.63		
Mar 2008	295 ± 24 (225–319)	395.6	$\begin{array}{c} 32.02 \pm 0.43 \\ (31.57  32.88) \end{array}$	7.6 ± 1.1 (6.51–9.46)	$\begin{array}{c} \textbf{4.9} \pm \\ \textbf{2.2} \end{array}$	1.19	-10.46	$-4.21\pm5.57$ (in spring)	-6.62	$-2.95\pm3.77$ (in spring)	
Mar 2009	322 ± 39 (269–398)	391.4	$\begin{array}{c} 32.07 \pm 0.44 \\ (30.90  33.26) \end{array}$	7.4 ± 0.9 (6.05–9.71)	$\begin{array}{c} \textbf{6.7} \pm \\ \textbf{3.0} \end{array}$	1.20	-11.30		-8.63		
Apr 2005	347 ± 19 (313–392)	384.4	$\begin{array}{c} 31.86 \pm 0.22 \\ (31.4232.47) \end{array}$	$\begin{array}{c} \textbf{7.9} \pm \textbf{0.8} \\ \textbf{(6.21-9.10)} \end{array}$	$\begin{array}{c} 4.3 \pm \\ 2.5 \end{array}$	1.33	-3.66		-2.14		
Apr 2008	371 ± 38 (326–437)	393.3	$\begin{array}{c} 31.98 \pm 0.31 \\ (31.51  32.73) \end{array}$	$\begin{array}{c} 11.6 \pm 1.2 \\ \textbf{(9.58-13.43)} \end{array}$	$\begin{array}{c} 4.8 \pm \\ 3.0 \end{array}$	1.37	-2.54		-1.60		
Apr 2009	364 ± 44 (314–444)	393.9	$\begin{array}{c} 32.07 \pm 0.20 \\ (31.57  32.53) \end{array}$	$11.1 \pm 1.1$ (9.69–12.88)	$\begin{array}{c} \textbf{4.8} \pm \\ \textbf{2.7} \end{array}$	1.30	-3.29		-2.06		
Apr 2011	320 ± 17 (270–349)	401.1	$\begin{array}{c} 32.00 \pm 0.19 \\ (31.71  32.45) \end{array}$	8.2 ± 0.5 (7.44–9.20)	$\begin{array}{c} 5.0 \pm \\ 2.6 \end{array}$	1.26	-8.99		-5.73		
May 2005	407 ± 29 (363–471)	384.3	$\begin{array}{c} 31.47 \pm 0.60 \\ \textbf{(28.08-31.82)} \end{array}$	$13.7 \pm 1.7$ (11.15–16.57)	$\begin{array}{c} \textbf{4.8} \pm \\ \textbf{2.8} \end{array}$	1.32	2.49		1.56		
May 2009	425 ± 65 (343–619)	386.7	$\begin{array}{c} 31.16 \pm 0.86 \\ \textbf{(28.62-32.32)} \end{array}$	$15.1 \pm 1.1$ (12.61–17.63)	3.9 ± 2.4	1.35	3.24		1.74		
Jun 2009	435 ± 40 (375–642)	375.9	$31.57 \pm 0.46$ (29.75–32.17)	$19.4 \pm 1.2$ (16.31–22.30)	$\begin{array}{c} \textbf{4.4} \pm \\ \textbf{2.0} \end{array}$	1.19	5.09	$5.00 \pm 4.01$ (in summer)	3.00	$2.96 \pm 2.53$ (in summer)	
Jun 2011	458 ± 55 (317–609)	386.2	$\begin{array}{c} 31.47 \pm 0.47 \\ \textbf{(29.98-32.17)} \end{array}$	$\begin{array}{c} 18.6 \pm 1.5 \\ (14.88 – 21.23) \end{array}$	4.9 ± 2.4	1.23	7.28		4.57		
Jul 2007	433 ± 54 (311–564)	370.6	$\begin{array}{c} 31.11 \pm 0.49 \\ (30.50  31.93) \end{array}$	$\begin{array}{c} 22.7 \pm 0.9 \\ (20.14  23.66) \end{array}$	4.0 ± 2.0	1.25	4.86		2.67		
Jul 2009	449 ± 56 (314–507)	367.7	$\begin{array}{c} 31.13 \pm 0.47 \\ (29.66  31.83) \end{array}$	$21.9 \pm 1.1$ (18.54–23.13)	4.6 ± 2.4	1.27	7.79		4.72		
Aug 2008	$447 \pm 124$ (232–804)	374.0	$\begin{array}{l} 30.30 \pm 0.76 \\ (27.36  31.54) \end{array}$	$\begin{array}{c} 25.7 \pm 1.2 \\ (22.4027.90) \end{array}$	$4.5 \pm 3.1$	1.44	7.74		4.65		
Aug 2009	343 ± 32 (265–381)	366.4	$\begin{array}{c} 31.10 \pm 0.08 \\ (30.93  31.20) \end{array}$	$\begin{array}{l} 24.7 \pm 0.5 \\ (23.85  25.30) \end{array}$	$5.6 \pm 2.3$	1.17	-2.73		-1.87		
Sep 2006	333 ± 14 (313–363)	370.1	$\begin{array}{c} 31.32 \pm 0.11 \\ (31.10  31.63) \end{array}$	$\begin{array}{c} 22.9 \pm 0.2 \\ (22.56 – 23.43) \end{array}$	$\begin{array}{c} 5.0 \ \pm \\ 2.9 \end{array}$	1.33	-4.11	$2.28\pm5.47$ (in autumn)	$2.28 \pm 5.47$ (in $-2.61$ autumn)	$1.77 \pm 3.82$ (in autumn)	
Sep 2008	448 ± 65 (370–573)	382.7	$\begin{array}{l} 30.42 \pm 0.26 \\ (29.87  30.76) \end{array}$	$\begin{array}{c} 24.1 \pm 0.7 \\ (22.53  25.61) \end{array}$	$\begin{array}{c} 5.7 \ \pm \\ 3.1 \end{array}$	1.29	8.50	5.91			
Oct 2006	365 ± 28 (311–459)	378.3	$\begin{array}{c} 31.04 \pm 0.20 \\ (30.55  31.31) \end{array}$	$\begin{array}{c} 22.6 \pm 0.4 \\ (21.93  23.07) \end{array}$	4.9 ± 2.7	1.28	-1.35		-0.85		
Nov 2007	419 ± 20 (376–467)	384.1	$\begin{array}{c} 30.91 \pm 0.49 \\ (29.83  31.44) \end{array}$	$\begin{array}{c} 18.7 \pm 0.3 \\ \textbf{(18.20-19.77)} \end{array}$	$\begin{array}{c} \textbf{6.2} \pm \\ \textbf{3.0} \end{array}$	1.22	5.02		3.68		
Nov 2011	$445 \pm 22$ (421–495)	400.6	$\begin{array}{c} 31.00 \pm 0.12 \\ (30.69  31.12) \end{array}$	$\begin{array}{c} 16.6 \pm 0.5 \\ (15.42  16.92) \end{array}$	6.7 ± 3.2	1.21	6.98		5.32		

in 2011). Through comparison with the air-equilibrated levels of sea surface  $pCO_2$ , it was determined that the three subregions of the SYS tended to always absorb  $CO_2$  from the atmosphere in winter, but that the status varied in summer, showing  $CO_2$ -degassing in Subregion I, air-sea  $CO_2$  equilibrium in Subregion II, and  $CO_2$ -absorbing from the atmosphere in Subregion III (Fig. 2G–I). In spring and autumn, Subregion II was the only area in this study that always served as a  $CO_2$ -absorbing region during both transitional seasons, while Subregions I and III tended to release  $CO_2$  to the atmosphere in autumn.

# 3.2. Air-sea CO<sub>2</sub> fluxes and comparison with those in 2011-2018

The calculated air–sea  $CO_2$  fluxes along the cruise tracks in the three subregions are summarized in Tables 2–4. On the basis of the assumption that these cruise track fluxes were representative of the entire subregions, we obtained an overview of the seasonal variation in air–sea  $CO_2$  exchange over the SYS (Fig. 3). In this study, we used the CCMP wind speed data and the S07 equation (Eq. 1) to calculate the gas transfer velocities, from which we then determined the air–sea  $CO_2$  fluxes. We also calculated the air–sea  $CO_2$  fluxes based on the EC22 equation (Eq. 2) to better quantify the gas transfer velocities and fluxes at relatively low wind speeds. Generally, the two equations produced similar temporal variations, although the EC22 equation sometimes produced relatively large values (Fig. 3).

In the western SYS (i.e., Subregion I), a total of 24 surveys were conducted, spanning 12 calendar months. The survey-averaged air–sea CO<sub>2</sub> fluxes ranged from - 15.6 mmol CO<sub>2</sub>  $m^{-2}$   $d^{-1}$  in February 2010 to 5.9 mmol CO<sub>2</sub>  $m^{-2}$   $d^{-1}$  in early September 2008 (Fig. 3A). In terms of the seasonal average, the air–sea CO<sub>2</sub> fluxes varied from - 11.1  $\pm$  6.0 mmol CO<sub>2</sub>  $m^{-2}$   $d^{-1}$  in winter and - 3.0  $\pm$  3.8 mmol CO<sub>2</sub>  $m^{-2}$   $d^{-1}$  in

#### Table 3

Summary of pCO<sub>2</sub>, salinity and SST along cruise tracks, and monthly satellite-derived wind speed, and air–sea CO<sub>2</sub> flux estimation in Subregion II, with positive values indicating that the sea area release CO<sub>2</sub> to the atmosphere. The annual air–sea CO<sub>2</sub> flux amounts to  $-1.6 \pm 0.5$  mol CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup> using S07 equation or  $-2.3 \pm 0.7$  mol CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup> using EC22 equation (Yang et al., 2022). Note that seasonal average CO<sub>2</sub> fluxes were calculated from the monthly averages of the three calendar months in the given season. Errors in air–sea CO<sub>2</sub> flux estimation are the temporal variability of CO<sub>2</sub> fluxes based on those cruises/surveys.

Observation	Sea surface	Sea surface	Air	Salinity	SST	Wind	C2	Air–sea $CO_2$ fluxes (mmol $CO_2 m^{-2} d^{-1}$ )			
time	pCO <sub>2</sub> (µatm)	pCO <sub>2</sub> (µatm)		(°C)	speed (m s <sup>-1</sup> )	speed (m s <sup>-1</sup> )		Survey average (EC22)	Seasonal average (EC22)	Survey average (S07)	Seasonal average (S07)
Dec 2011	$363\pm15$	398.6	$31.65\pm0.32$	$14.1\pm0.8$	7.4 $\pm$	1.20	-6.50	$-7.64\pm1.62$ (in	-5.22	$-6.14\pm1.31$ (in	
	(345–405)		(30.71–32.18)	(12.10–15.01)	3.3			winter)		winter)	
Feb 2010	$350\pm20$	396.5	$\textbf{32.48} \pm \textbf{0.46}$	$\textbf{8.2}\pm\textbf{1.7}$	7.4 $\pm$	1.20	-8.78		-7.07		
	(310–388)		(31.81–33.80)	(4.39–11.67)	3.4						
Mar 2009	$347 \pm 15$	391.0	$33.20\pm0.43$	$\textbf{9.6} \pm \textbf{0.8}$	$7.2 \pm$	1.22	-7.92	$-9.03\pm3.02$ (in	-6.29	$-5.97\pm2.17$ (in	
	(307–369)		(32.07–33.80)	(7.24–10.60)	3.4			spring)		spring)	
Apr 2009	$289 \pm 23$	395.5	$33.22\pm0.56$	$11.4\pm0.7$	4.8 $\pm$	1.43	-12.73		-7.98		
	(255–335)		(32.20–33.78)	(10.21 - 12.23)	3.2						
May 2005	$319\pm15$	383.3	$32.91\pm0.33$	$13.6\pm0.7$	4.3 $\pm$	1.53	-6.97		-4.04		
	(293–363)		(31.83–33.39)	(12.27–14.94)	3.2						
May 2009	$314\pm39$	385.7	$32.16\pm0.56$	$13.6\pm0.8$	$4.0 \pm$	1.27	-5.89		-3.23		
	(277–414)		(31.31–32.94)	(10.98–14.92)	2.2						
Jun 2009	$366 \pm 37$	374.9	$32.23\pm0.43$	$19.5\pm1.4$	4.7 ±	1.22	-0.80	$-1.10 \pm 1.04$ (in	-0.49	$-0.70 \pm 0.67$ (in	
	(281–464)		(31.22–33.09)	(16.84–22.17)	2.2			summer)		summer)	
Jun 2011	$373\pm24$	386.3	$32.03\pm0.47$	$18.6 \pm 1.8$	4.7 ±	1.35	-1.37		-0.84		
	(331–442)		(30.85–32.84)	(12.57–21.47)	2.8						
Aug 2008	$374 \pm 43$	373.0	$30.92\pm0.44$	$\textbf{27.4} \pm \textbf{0.5}$	$4.5 \pm$	1.39	0.12		0.07		
	(292–436)		(30.25–31.71)	(25.92–28.33)	2.9						
Aug 2009	$344\pm45$	365.2	$31.10\pm0.57$	$\textbf{25.8} \pm \textbf{0.6}$	$5.1 \pm$	1.26	-2.34		-1.52		
	(262–392)		(29.97–31.72)	(24.27–26.58)	2.7						
Sep 2006	$298\pm26$	370.3	$31.07\pm0.30$	$23.5\pm0.3$	$5.2 \pm$	1.30	-8.43	$-7.28 \pm 1.62$ (in	-5.54	$-5.10\pm0.63$ (in	
	(246–325)		(30.49–31.39)	(23.08–24.21)	2.9			autumn)		autumn)	
Nov 2011	$363\pm28$	401.1	$31.30\pm0.36$	$15.1\pm0.5$	$6.7 \pm$	1.22	-6.14		-4.66		
	(334–432)		(30.74–31.92)	(14.15–16.18)	3.2						

spring to  $3.0 \pm 2.6 \text{ mmol CO}_2 \text{ m}^{-2} \text{ d}^{-1}$  in summer and  $1.8 \pm 3.9 \text{ mmol}$ CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in autumn (Table 2). The annual air–sea CO<sub>2</sub> flux estimation ( $-0.9 \pm 1.5 \text{ mol m}^{-2} \text{ yr}^{-1}$  using the S07 equation and  $-1.0 \pm 2.1 \text{ mol CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$  using the EC22 equation) indicated that this subregion tended to be a weak sink area of atmospheric CO<sub>2</sub>, consistent with the earlier estimate over the western SYS (with an annual air–sea CO<sub>2</sub> flux of  $-0.2 \pm 2.2 \text{ mol m}^{-2} \text{ yr}^{-1}$  using the S07 equation) based on data obtained during 2011–2018 and the same atmospheric CO<sub>2</sub> data set at the Tae-ahn Peninsula site (Wang and Zhai, 2021).

In the central SYS (Subregion II), a total of 12 surveys were conducted, spanning 9 out of 12 calendar months, i.e., excluding January, July and October. The survey-averaged air-sea CO<sub>2</sub> fluxes ranged from - 8.0 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in late April 2009 to 0.1 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in August 2008 (Fig. 3B). In terms of the seasonal average, the air-sea CO<sub>2</sub> fluxes varied from - 6.1  $\pm$  1.4 mmol CO $_2$  m $^{-2}$  d $^{-1}$  in winter and - 6.0  $\pm$ 2.2 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in spring to - 0.7  $\pm$  0.7 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in summer and  $-5.1 \pm 0.7$  mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in autumn (Table 3). The annual air-sea CO<sub>2</sub> flux estimation ( $-1.6 \pm 0.5$  mol m<sup>-2</sup> yr<sup>-1</sup> using the S07 equation and - 2.3  $\pm$  0.7 mol CO\_2  $m^{-2}\,yr^{-1}$  using the EC22 equation) indicated that this subregion used to be a stronger CO<sub>2</sub>-absorbing area during 2005–2011 compared with the estimate derived by Wang and Zhai (2021) for annual air-sea CO2 flux over the central SYS (i.e.,  $-0.8\pm1.8$  mol  $m^{-2}\,yr^{-1}$  using the S07 equation based on data obtained during 2011-2018 and the same atmospheric CO2 data set at the Taeahn Peninsula site).

In the southern SYS (i.e., Subregion III), a total of 20 surveys were conducted, spanning 11 out of 12 calendar months, i.e., excluding January. The survey-averaged air–sea CO<sub>2</sub> fluxes ranged from - 8.4 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in February 2010 to 5.5 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in October 2006 and 28.0 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in early September 2008 (Fig. 3C). In terms of the seasonal average, the air–sea CO<sub>2</sub> fluxes varied from - 3.8 ± 5.8 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in winter and - 3.5 ± 2.7 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in spring to - 3.9 ± 2.5 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in summer and 0.8 ± 5.0 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> in autumn (Table 4), ignoring the extremely high value (~28 mmol CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup>) obtained in September

2008. The annual air–sea CO<sub>2</sub> flux estimation  $(-0.9 \pm 2.3 \text{ mol m}^{-2} \text{ yr}^{-1}$  using the S07 equation or  $-1.3 \pm 3.1 \text{ mol CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$  using the EC22 equation) indicated that this subregion also tended to be a CO<sub>2</sub> sink area, comparable to the estimate derived by Wang and Zhai (2021) over the southern SYS (i.e.,  $-0.1 \pm 2.0 \text{ mol m}^{-2} \text{ yr}^{-1}$  using the S07 equation based on data obtained during 2011–2018 and the same atmospheric CO<sub>2</sub> data set at the Tae-ahn Peninsula site).

To further compare the seasonal variations in air-sea CO<sub>2</sub> fluxes during the two periods, we manually fitted the monthly variations in air-sea CO<sub>2</sub> fluxes in 2005-2011 based on the data investigated in this study (Fig. 3A-C), and copied the fitting curves to those plots based on data obtained during 2011–2018 (Fig. 3D–F). Although neither the zoning details nor the air-sea CO<sub>2</sub> flux estimates of the two periods were exactly the same, the fitting curves of the monthly variations in Subregions I and II in 2005-2011 were consistent with those of the monthly variations during 2011-2018 (Fig. 3D-E), showing that similar air-sea CO<sub>2</sub> exchanges and seasonal variations occurred during the two periods. In Subregion III, the fitting curve (showing the monthly variation in the air-sea CO2 flux during 2005-2011) located systematically to one side of those estimated for 2011-2018, indicating that annual CO2 uptake had weakened in this subregion (Fig. 3F). Even if we could properly integrate the episodic CO2-release event observed in early September 2008 into the seasonal air-sea CO<sub>2</sub> flux estimation of this subregion, the systematic change between the two periods would not be reversed.

Despite the systematic change revealed in Subregion III, our results suggest that the SYS did not experience a change from being an annually CO<sub>2</sub>-degassing area to becoming a CO<sub>2</sub>-absorbing area during the two periods. By contrast, it served as a sink for atmospheric CO<sub>2</sub> during both periods. The area-weighted average of the CO<sub>2</sub> influx rate was estimated to be  $1.1 \pm 1.5$  mol m<sup>-2</sup> yr<sup>-1</sup> (using the S07 equation) or  $1.5 \pm 2.0$  mol m<sup>-2</sup> yr<sup>-1</sup> (using the EC22 equation) over the three subregions during 2005–2011. Given the dynamic and heterogeneous nature of the air–sea CO<sub>2</sub> flux in shelf seas, the new estimate for 2005–2011 was broadly consistent with the value of  $0.4 \pm 2.1$  mol m<sup>-2</sup> yr<sup>-1</sup> reported for 2011–2018 (using the S07 equation) based on another SYS dataset

#### Table 4

Summary of pCO<sub>2</sub>, salinity and SST along cruise tracks, and monthly satellite-derived wind speed, and air–sea CO<sub>2</sub> flux estimation in Subregion III, with positive values indicating that the sea area release CO<sub>2</sub> to the atmosphere. The annual air–sea CO<sub>2</sub> flux amounts to  $-0.9 \pm 2.3$  mol CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup> using S07 equation or  $-1.3 \pm 3.1$  mol CO<sub>2</sub> m<sup>-2</sup> yr<sup>-1</sup> using EC22 equation (Yang et al., 2022). Note that seasonal average CO<sub>2</sub> fluxes were calculated from the monthly averages of the three calendar months in the given season. Errors in air–sea CO<sub>2</sub> flux estimation are the temporal variability of CO<sub>2</sub> fluxes based on those cruises/surveys. The extremely high value obtained in September 2008 has been removed from the seasonal average and the annual mean flux estimation, while it was involved in the assessment of the temporal variability.

Observation	Sea surface pCO <sub>2</sub> (µatm)	Air	Air Salinity pCO <sub>2</sub> (μatm)	SST (°C)	Wind speed (m s <sup>-1</sup> )	C2	Air–sea $CO_2$ fluxes (mmol $CO_2 m^{-2} d^{-1}$ )			
time		pCO <sub>2</sub> (µatm)					Survey average (EC22)	Seasonal average (EC22)	Survey average (S07)	Seasonal average (S07)
Dec 2010	$408\pm21$	393.2	$\textbf{32.29} \pm \textbf{0.88}$	$15.6\pm1.1$	8.7 $\pm$	1.26	3.57	$-4.88\pm7.12$ (in	3.11	$-3.81\pm5.77$ (in
	(376–451)		(31.08–33.52)	(13.79–16.81)	4.5			winter)		winter)
Dec 2011	$388 \pm 19$	398.4	$31.66\pm0.18$	$15.3\pm0.7$	$7.9 \pm$	1.18	-1.98		-1.64	
	(349–417)		(31.32–31.98)	(14.57–16.64)	3.4					
Feb 2010	$337\pm14$	395.6	$32.80\pm0.92$	$10.0\pm1.6$	$7.2 \pm$	1.22	-10.55		-8.36	
	(312–355)		(31.46–34.06)	(7.58–12.15)	3.5					
Mar 2008	$354\pm13$	392.5	$32.51\pm0.20$	$\textbf{9.3}\pm\textbf{0.4}$	$5.9 \pm$	1.24	-5.35	$-5.20\pm4.07$ (in	-3.78	$-3.48\pm2.64$ (in
	(324–371)		(32.19–33.06)	(8.75–10.35)	2.9			spring)		spring)
Mar 2009	$359\pm13$	390.4	$32.31\pm0.79$	$\textbf{9.2}\pm\textbf{0.9}$	$7.2 \pm$	1.18	-5.39		-4.26	
	(330–379)		(31.21–33.86)	(7.70–10.74)	3.1					
Apr 2005	$391 \pm 15$	385.8	$31.74 \pm 1.24$	$\textbf{9.2}\pm\textbf{0.8}$	$4.5 \pm$	1.35	0.51		0.30	
	(354–419)		(27.65–32.74)	(8.53–12.12)	2.7					
Apr 2008	$394\pm52$	394.0	$31.67 \pm 1.36$	$13.2\pm0.7$	4.8 $\pm$	1.25	0.04		0.02	
	(282–443)		(26.99–32.55)	(12.29–15.02)	2.4					
Apr 2009	$305\pm69$	395.2	$32.00 \pm 1.20$	$12.3\pm1.0$	$5.2 \pm$	1.36	-11.42		-7.52	
	(231–440)		(27.92–33.45)	(11.33–15.46)	3.2					
Apr 2011	$364\pm60$	400.9	$\textbf{32.43} \pm \textbf{0.29}$	$\textbf{9.7}\pm\textbf{0.9}$	5.6 $\pm$	1.22	-4.63		-3.17	
	(189–423)		(31.56–33.07)	(8.72–11.96)	2.7					
May 2005	$351\pm59$	383.5	$31.39 \pm 2.30$	$15.0\pm0.8$	$4.7 \pm$	1.39	-3.61		-2.22	
	(214–453)		(26.31–33.47)	(13.33–16.39)	3.0					
May 2009	$292\pm81$	387.6	$30.58 \pm 1.32$	$16.1\pm1.2$	$4.5 \pm$	1.26	-9.08		-5.42	
	(120–431)		(27.10–32.83)	(13.05–19.05)	2.3					
Jun 2011	$323\pm59$	386.0	$31.71\pm0.87$	$18.4\pm0.8$	$5.2 \pm$	1.24	-6.97	$-5.93\pm3.63$ (in	-4.56	$-3.89\pm2.44$ (in
	(88–501)		(27.82–32.85)	(16.53–19.97)	2.6			summer)		summer)
Jul 2007	$287\pm 66$	370.5	$31.33\pm0.80$	$23.0\pm1.1$	$4.4 \pm$	1.25	-7.45		-4.38	
	(165–417)		(28.81–32.32)	(21.83–25.05)	2.3					
Jul 2009	$374\pm78$	368.2	$29.59 \pm 1.60$	$21.9 \pm 0.9$	$5.4 \pm$	1.22	0.72		0.48	
	(272–544)		(26.67–31.53)	(20.47–23.43)	2.6					
Aug 2008	$308\pm65$	371.7	$30.94\pm0.69$	$\textbf{27.6} \pm \textbf{1.0}$	$5.1 \pm$	1.26	-6.97		-4.51	
	(181–401)		(29.27–32.15)	(24.60–29.29)	2.6					
Aug 2009	$302\pm72$	363.2	$30.25 \pm 1.78$	$\textbf{26.7} \pm \textbf{0.8}$	$6.2 \pm$	1.15	-7.96		-5.81	
	(182–463)		(24.92–32.34)	(25.22–28.05)	2.5					
Sep 2006	$333\pm57$	370.6	$31.58\pm0.72$	$23.0\pm0.9$	$6.7 \pm$	1.21	-5.77	$\begin{array}{cc} -5.77 & 1.33 \pm 19.14 \mbox{ (in autumn)} \end{array}$	-4.39	$0.82 \pm 14.15$ (in autumn)
	(252–414)		(30.48–33.10)	(21.89–24.33)	3.1					
Sep 2008	$642\pm79$	381.8	$31.21\pm0.44$	$\textbf{24.3} \pm \textbf{0.8}$	$6.3 \pm$	1.24	37.87		27.95	
	(538–818)		(30.49–32.10)	(22.23–25.08)	3.2					
Oct 2006	$449\pm83$	377.3	$31.82 \pm 0.83$	$\textbf{23.4} \pm \textbf{0.7}$	5.4 $\pm$	1.22	8.17		5.47	
	(193–580)		(30.84–33.37)	(21.96–24.18)	2.6					
Nov 2006	$389\pm34$	381.7	$33.02\pm0.65$	$19.4 \pm 1.0$	$\textbf{8.8}~\pm$	1.12	1.58		1.39	
	(318–456)	(31.54–33.65)	(17.11–20.22)	3.0						

released by Wang and Zhai (2021). At most, the CO<sub>2</sub> influx rate over the SYS might have experienced slight weakening during the two periods.

## 3.3. Comparing air-sea CO<sub>2</sub> fluxes with the adjacent East China sea

Over the adjacent ECS shelf, using the S07 equation, annual air–sea  $CO_2$  influx rates ranged from  $1.7 \pm 2.0 \text{ mol m}^{-2} \text{ yr}^{-1}$  in Subregion V to  $2.3 \pm 1.8 \text{ mol m}^{-2} \text{ yr}^{-1}$  in Subregions IV and VI (Supplementary Tables S3–S7), broadly consistent with the earlier data-based estimates (Zhai and Dai, 2009; Tseng et al., 2014; Guo et al., 2015) and a satellite-based estimate  $(1.5 \pm 1.4 \text{ mol m}^{-2} \text{ yr}^{-1}$  for the entire ECS) released by Yu et al. (2023). In the SYS, the only substantial CO<sub>2</sub>-absorbing value was observed in Subregion II (i.e., the central SYS), where the annual air–sea  $CO_2$  influx rate of  $1.6 \pm 0.5 \text{ mol m}^{-2} \text{ yr}^{-1}$ , estimated using the S07 equation (Table 3), reached the lower limit of the range of annual air–sea  $CO_2$  influx rates in the ECS. The area-weighted average of the CO<sub>2</sub> influx rate over the surveyed areas of the SYS was only half that of the ECS shelf value, indicating again that the Yellow Sea serves as a weak  $CO_2$  absorber among various  $CO_2$ -absorbing continental shelf areas (Wang and Zhai, 2021).

#### 4. Discussion

#### 4.1. Factors influencing seasonal variations in sea surface $pCO_2$

As reported by Guo et al. (2015), surface  $pCO_2$  was highly heterogenous over the SYS and ECS, especially near the Changjiang Estuary. It remains a challenge to better understand the complex influences of spring algal blooms, the summer river plume from Changjiang, the autumnal collapse of coastal zone hypoxia, and the cold coastal current in winter on the seasonal variation in surface  $pCO_2$  over the SYS and ECS (e.g., Zhai and Dai, 2009; Wang and Zhai, 2021). To consider certain evidence, survey-averaged  $pCO_2$  was plotted against SST and salinity (Fig. 4). Except for the cold seasons in Subregion I, the relationship between  $pCO_2$  and SST exhibited no clear pattern, while  $pCO_2$  versus salinity showed a mixture of water mixing (between relatively high  $pCO_2$  at low salinity and relatively low and/or nearly air-equilibrated  $pCO_2$  at high salinity) and likely biological CO<sub>2</sub> drawdown/addition at intermediate salinity.

Subregion-specific seasonal variations in cruise-averaged SST were broadly the same between the two periods (Supplementary Fig. S3), and



**Fig. 2.** Seasonal variations in sea surface temperature (SST), salinity and partial pressure of  $CO_2$  (*p*CO<sub>2</sub>) in the three subregions defined in Fig. 1C, and based on data presented in Tables 2–4. Field data are shown as mean  $\pm$  standard deviation, seasonal breaks are indicated by pink dashed lines, and subregion-specific fitting curves of the monthly variations are manually plotted with dashed curves. In panels G–I, the gray band indicates the range of the air-equilibrated level (open circles) of 360–400 µatm. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

therefore the potential temperature effect on seasonal variations in surface *p*CO<sub>2</sub> should have behaved similarly during the two periods. The only difference was the relatively high air-equilibrated level of *p*CO<sub>2</sub> in 2011–2018 (380–420 µatm) compared with that in 2005–2011 (360–400 µatm). To examine this issue, the SST-driven *p*CO<sub>2</sub> values (*p*CO<sub>2</sub><sup>Temp</sup>, unit: µatm) during 2005–2011 were plotted with *p*CO<sub>2</sub><sup>Temp</sup> = (380 ± 20) × e<sup>0.0423×(SST-mSST)</sup>, where the value of 380 ± 20 is comparable to the range of the air-equilibrated level of *p*CO<sub>2</sub> (unit: µatm) at 10-m (Fig. 2G–I), 0.0423°C<sup>-1</sup> is the coefficient for the temperature effect of seawater *p*CO<sub>2</sub> (Takahashi et al., 1993), and *m*SST is the annual average SST, i.e., 15.46°C in the offshore area of the western SYS (Fig. 4A), 16.20°C in the central SYS (Fig. 4B), and 17.32°C in the southern SYS (Fig. 4C). However, only a few surveys followed the ideal SST-driven lines, implying again that non-thermodynamic regulations of *p*CO<sub>2</sub> dynamics occurs in this marginal sea (Wang and Zhai, 2021).

To clarify this non-thermodynamic regulation issue, we further calculated the difference between  $pCO_2$  and  $pCO_2^{\text{Temp}}$ , and plotted it with salinity (Fig. 4G–I). Highly negative values of  $pCO_2 - pCO_2^{\text{Temp}}$  (with a survey-averaged value of below – 100 µatm) were associated with relatively low salinity of < 31.6 and usually warm waters in the SYS (Fig. 2). Although we have no reliable chlorophyll and/or sufficient DO data to characterize the in situ primary production in this study, the SYS has been widely characterized as having high productivity in the warm seasons (e.g., Guo et al., 2024). Therefore, we contend that these highly negative values of  $pCO_2 - pCO_2^{\text{Temp}}$  were induced mostly by biological drawdown, especially for those  $CO_2$  uptake cases in the three subregions in the warm seasons (Fig. 4A–C). By contrast, many very high positive

values of  $pCO_2 - pCO_2^{Temp}$  (with survey-averaged values of usually 40 – 90 µatm) were associated with relatively high survey-averaged salinity of > 31.6 (Fig. 4G–I), with the highest salinity of 33.20 ± 0.43 in Subregion II in March 2009. These very high positive values of  $pCO_2 - pCO_2^{Temp}$  were usually recorded in winter and spring (Fig. 5), implying basin-scale  $pCO_2$  addition of 40–90 µatm in the cold seasons, which might be attributable to vertical mixing with CO<sub>2</sub>-rich bottom water in the SYS (Zhai, 2018; Wang and Zhai, 2021).

According to Yu et al. (2022), the SYS cold water mass persists in accumulating apparent oxygen utilization and supersaturated CO<sub>2</sub> from spring to late autumn (until December). Even in January, the SYS might still contain some supersaturated CO<sub>2</sub> in the bottom waters (Zhai, 2018). The supersaturated CO<sub>2</sub> can be mixed to surface waters during the ventilation period in late autumn and winter, and it tended to be released into the atmosphere over a temporal scale of 3-4 months (e.g., Zhai et al., 2014). In our dataset, taking Subregion II as an example, the wintertime  $pCO_2 - pCO_2^{Temp}$  values (Fig. 4H) increased from 15  $\pm$  15  $\mu$ atm in December (in 2011) to 79  $\pm$  20  $\mu$ atm in February (in 2010), and then declined to 60  $\pm$  15 µatm in March (in 2009) and to - 21  $\pm$  23 µatm in April (in 2009). Therefore, the upwelled supersaturated CO<sub>2</sub> counteracted the cooling-induced pCO<sub>2</sub> decline throughout winter, leading to relatively high pCO2 values in winter when the SST is very low (Fig. 4A–C). Considering horizontal mixing (HM) with the high-salinity Yellow Sea Warm Current in the cold seasons and with the relatively low-salinity Changjiang Diluted Water in the warm seasons (Fig. 1), we proposed a schematic of the non-thermodynamic regulation processes in the SYS (Fig. 4H), involving springtime biological drawdown (BD),



**Fig. 3.** Seasonal variations in air–sea  $CO_2$  fluxes during 2005–2011 and the comparison with those of 2011–2018. Upper two, middle two, and lower two panels represent the western, central and southern parts of the South Yellow Sea, respectively. In panels (A)–(C), filled circles with error bars denote the flux estimation based on the S07 equation (Eq. 1) for calculating the gas transfer velocity, while open circles without error bars show those flux estimation results based on the EC22 equation (Eq. 2). Errors in air–sea  $CO_2$  flux estimation were based on the spatial variability of surface  $pCO_2$  along those cruise tracks in the given subregion. In panels (A)–(C), dashed curves are manually plotted to show subregion-specific monthly variations in air-sea  $CO_2$  fluxes based on data obtained during 2005–2011. In panels (D)–(F), the dashed curves are exact copies of those in the left-hand column.

autumnal vertical mixing (VM), wintertime VM + HM and summertime BD + HM.

Similar non-thermodynamic regulation of pCO<sub>2</sub> was also identified with survey-averaged  $pCO_2 - pCO_2^{Temp}$  values during 2011–2018 (Supplementary Fig. S4). Relevant processes have been discussed using discrete data (Wang and Zhai, 2021). Interestingly, seasonal variations in the survey-averaged  $pCO_2 - pCO_2^{Temp}$  values usually exhibited a consistent pattern among the three subregions and during the two periods (Fig. 5), including wintertime  $CO_2$  addition of usually 40 - 90µatm (through basin-scale vertical mixing during stratification decay in every autumn) and warm-season biological CO<sub>2</sub> drawdown that usually accumulated to a peak of 200–300  $\mu$ atm in every August (i.e., 279  $\pm$  65  $\mu atm$  in August 2008, 262  $\pm$  72  $\mu atm$  in August 2009, and 225  $\pm$  111 µatm in August 2015). In comparison with 2011–2018, the southern SYS (i.e., Subregion III) experienced stronger signals of non-thermodynamic CO2 drawdown of 35-55 µatm (as indicated by the survey-averaged  $pCO_2 - pCO_2^{Temp}$  values) during 2005–2011 (Fig. 5C versus 5F). This might explain the decadal weakening of the air-sea CO<sub>2</sub> flux revealed in Subregion III (Section 3.2), which is discussed further in Section 4.3.

4.2. Exploring the episodic high sea surface  $pCO_2$  as recorded in early September 2008

In early September 2008, the highest  $pCO_2 - pCO_2^{Temp}$  value of 132  $\pm$  79 µatm of this study was recorded in Subregion III (Fig. 4I), associated with relatively low salinity of  $31.21 \pm 0.44$  and relatively high SST of 24.3  $\pm$  0.8 °C, together with extremely high survey-averaged pCO<sub>2</sub> of  $642 \pm 79$  µatm (Table 4). The high pCO<sub>2</sub> even peaked at 818 µatm, where many unsaturated DO data of  $\sim$  80 % were recorded synchronously (Supplementary Fig. S5). In summer 2008, the SYS attracted public attention owing to a massive green algae bloom (e.g., Liu et al., 2009), which increased the concentration of surface dissolved inorganic carbon (DIC) by 80–100  $\mu$ mol kg<sup>-1</sup> during the post-bloom period (Hu et al., 2015). Following Wang and Zhai (2021), this biological addition of DIC could induce an increase in pCO<sub>2</sub> of 228 µatm, i.e., [ $\delta$ DIC/DIC<sup>avg</sup>]  $\times$  RF<sup>avg</sup>  $\times$  pCO<sub>2</sub><sup>avg</sup> = (100/2100)  $\times$  12  $\times$  400 = 228 µatm, where DIC<sup>avg</sup>, RF<sup>avg</sup>, and  $p\mathrm{CO}_2^{\mathrm{avg}}$  are regional average values of DIC (~2100 µmol kg<sup>-1</sup>), the Revelle factor (~12), and  $pCO_2$  (~400 µatm), respectively. Therefore, the post-bloom remineralization could have induced both the high  $pCO_2$  of > 600 µatm and the high  $pCO_2 - pCO_2^{Temp}$  value of ~ 200



**Fig. 4.** Cruise-averaged partial pressure of  $CO_2$  ( $pCO_2$ ) versus sea surface temperature (SST) and salinity (A–F), and the difference in  $pCO_2$  from the SST-driven  $pCO_2$  values ( $pCO_2 - pCO_2^{Temp}$ ) versus salinity (G–I). Upper three, middle three, and lower three panels represent the western, central and southern parts of the South Yellow Sea, respectively. Field data are shown as mean  $\pm$  standard deviation. Open circles indicate cruise-specific air-equilibrated levels of  $pCO_2$ . In panels A-C, red dashed lines show the range of SST-driven  $pCO_2$  values. In panel H, BD = biological drawdown, VM = vertical mixing, and HM = horizontal mixing (with high-salinity Yellow Sea Warm Current in cold seasons and with relatively low-salinity Changjiang Diluted Water in warm seasons). See details in text of Section 4.1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

 $\mu$ atm over the sea surface. Despite no further observation, this assumption has no contradiction with the field data. By contrast, another potential mechanism (but inapplicable in this case) to explain the unexpected high *p*CO<sub>2</sub> is the upwelling of subsurface CO<sub>2</sub>-rich cold water to the sea surface, which must occur together with salinity increase and water temperature decline that were not evidenced by our data.

Given that summertime green algae blooms have occurred in the SYS annually since 2007 (Xiong et al., 2023), it is puzzling why extremely high pCO<sub>2</sub> was observed only once in our surveys, even including this study (Tables 2-4) and the mapping cruises conducted during 2011-2018 (Wang and Zhai, 2021). To investigate this issue, we considered the air-sea equilibration time  $\tau(CO_2)$ . Following Zeebe and Wolf-Gladrow (2001) and Zhai et al. (2014),  $\tau$ (CO<sub>2</sub>) =  $d_{\rm ML}/k \times$  (DIC/  $[CO_2^*]/RF$ ), where  $d_{ML}$  is the mixed layer depth (~10 m in summer), k refers to Eqs. (1)–(2) and varies between 2 and 4 m  $d^{-1}$ , DIC/[CO<sub>2</sub>\*] is the concentration ratio between DIC and free CO<sub>2</sub> (CO<sub>2</sub>\*), and RF is the Revelle factor. During calculation, water temperature and salinity were set to 25 °C and 31, respectively (Supplementary Fig. S3), and total alkalinity (TAlk) was set at 2250 µmol kg<sup>-1</sup> (Zhai, 2018). The calculation program used was CO2SYS.xls (Pelletier et al., 2015), which is an updated version of the original CO2SYS.EXE (Lewis and Wallace, 1998). The dissociation constants for carbonic acid were those determined by Millero et al. (2006), and the dissociation constant for the  $HSO_{\overline{4}}$  ion was determined as per Dickson (1990). The phosphate and silicate concentrations required by the program were replaced by zero.

For the extremely high  $pCO_2$  of 800 µatm, the DIC:[ $CO_2^*$ ] ratio and the RF were calculated to be 91 and 13.6, respectively, and  $\tau(CO_2)$  was

estimated at 16–33 d. This  $\tau$ (CO<sub>2</sub>) value meant that the air–sea disequilibrium of *p*CO<sub>2</sub> (i.e., 800 – 380 = 420 µatm) declined over 0.5–1.0 months to 37 % (~1/*e*) of its initial value, and that the observed *p*CO<sub>2</sub> should be only 380 + 420 × 0.37 = 535 µatm. For the usually high *p*CO<sub>2</sub> of 450 µatm (Fig. 4), the DIC:[CO<sub>2</sub>\*] ratio and the RF were calculated at 154 and 10.7, respectively, and  $\tau$ (CO<sub>2</sub>) under the same conditions should be 36–72 d. For the usually low *p*CO<sub>2</sub> of 300 µatm (Fig. 4), the DIC:[CO<sub>2</sub>\*] ratio and the RF were calculated to be 222 and 9.2, respectively, and  $\tau$ (CO<sub>2</sub>) should be 60–120 d. This comparison explains why marine CO<sub>2</sub> uptake events should be observed more often than CO<sub>2</sub> release events. The episodic CO<sub>2</sub> addition reduces the DIC: [CO<sub>2</sub>\*] ratio and increases the Revelle factor, thereby diminishing the air–sea equilibration time  $\tau$ (CO<sub>2</sub>). If the value of  $\tau$ (CO<sub>2</sub>) was less than 1 month, it would be challenging for researchers to conduct ship-based observation.

In summary, the decomposition of summer green algae blooms might result in temporary sudden increases in seawater  $pCO_2$ , leading to rapid  $CO_2$  outgassing that might not be detected through observations. If this interpretation is accurate, the data reported in this study might overestimate the  $CO_2$  absorption capacity of the SYS because the annual decay of green algae blooms is likely to have gone undetected. To fill this gap, volunteer-ship-based and/or mooring-based high-frequency observations might help (e.g., Curbelo-Hernández et al., 2021; Wu et al., 2021).

# 4.3. On the decadal change in shelf air-sea CO<sub>2</sub> fluxes

It has been speculated that increase in marine pCO<sub>2</sub> in shelf waters



**Fig. 5.** Seasonal variations in the difference of  $pCO_2$  from SST-driven  $pCO_2$  values and their comparison with those of 2011–2018. Upper two, middle two, and lower two panels show the western, central, and southern parts of the South Yellow Sea, respectively. To calculate the SST-driven  $pCO_2$  values, the annual average of the air-equilibrated level of  $pCO_2$  was fixed at 380 µatm during 2005–2011 and at 400 µatm during 2011–2018.

should follow rise in atmospheric CO<sub>2</sub> with a temporal lag, leading to enhanced CO<sub>2</sub> uptake over some continental shelves (e.g., Laruelle et al., 2018). This hypothesis has been partially proven in the northern South China Sea based on wintertime data of an 18-year dataset (Li et al., 2020). Because the northern South China Sea shelf lies close to slowlyventilated deep waters, exchange between the surface and deep waters might buffer the effect of atmospheric CO<sub>2</sub> intrusion to a certain extent.

In the SYS, however, water exchange with the adjacent deep ocean is reasonably slow, likely on a scale of several years (Nozaki et al., 1991; Li et al., 2022). Given that the Yellow Sea deep waters are fully ventilated every winter (Zhai, 2018; Yu et al., 2022), annually averaged marine  $pCO_2$  in the Yellow Sea should quickly follow the trend of atmospheric CO<sub>2</sub>. Although many other ocean margins (Dai et al., 2022; Mathis et al., 2024) might exhibit different characteristics to those of the Yellow Sea case, the results of this study appear representative for semi-enclosed shallow seas such as the adjacent Bohai Sea (Zhang et al., 2024). Generally, shallow water restricts the potential sequestration and burial of anthropogenic CO<sub>2</sub> on the annual time scale (e.g., Wei et al., 2022), while the limited circulation places restrictions on the horizontal export of anthropogenic CO<sub>2</sub> out of the SYS (Wang and Zhai, 2021).

Considering the counterflow of the wintertime coastal current and the Yellow Sea Warm Current (Fig. 1A) with different TAlk:salinity and DIC:TAlk ratios, this circulation-induced net carbonate export is equivalent to an annual net CO<sub>2</sub> influx rate of approximately 0.7 mol  $m^{-2}$  yr<sup>-1</sup> over the Yellow Sea (Wang and Zhai, 2021). On the basis of sediment

samples collected in the 2000 s, the Yellow Sea mean burial flux of total organic carbon has been estimated to be approximately 1.3 mol m<sup>-2</sup> yr<sup>-1</sup> (Hu et al., 2016). Both processes are equivalent to a small annual net CO<sub>2</sub> influx rate with magnitude similar to that of our estimates for annual net CO<sub>2</sub> influx in the SYS, suggesting that both circulation-induced net carbonate export and sedimentation of biogenic organic carbon might play a role in maintaining this shelf carbon sink.

For the southern SYS (i.e., Subregion III), the adjacent outer Changjiang Estuary is subject to regional environmental changes. According to Wang et al. (2024), its annual net primary production (NPP) has declined from 280–315 mgC  $m^{-2} d^{-1}$  in the 2000 s to 240–260 mgC  $m^{-2}\;d^{-1}$  in the 2010 s, based on a synthesis study combining field incubation for NPP estimation, correction of satellite-derived chlorophylla and machine learning. Although this decadal change is consistent with declines in nutrient discharges from the Changjiang River since the early 2010 s (Wang et al., 2023), it has also been attributed primarily to the effect of climate-driven events such as the Pacific Decadal Oscillation and/or the El Niño - Southern Oscillation (Wang et al., 2024). Nevertheless, the decline in the NPP of the outer Changjiang Estuary might also affect the adjacent Subregion III, further inducing potential declines in circulation-induced net carbonate export and sedimentation of biogenic organic carbon. If so, weakening of the annual net air-sea CO<sub>2</sub> influx rate in this subregion (i.e., the slight decline from 0.9  $\pm$  2.3 mol  $m^{-2}~{\rm yr}^{-1}$  during 2005–2011 to 0.1  $\pm$  2.0 mol  $m^{-2}~{\rm yr}^{-1}$  during 2011–2018) might be related to this regional environmental change.

Considering the reduction in excess nitrogen in the Bohai and Yellow seas in the 2010 s caused by both air pollution control and reduced inflow through rivers (Liang et al., 2023; Wang et al., 2023; Zheng and Zhai, 2023), the slight weakening of the basin-scale annual net air-sea CO\_2 influx rate over the SYS in the 2010 s (0.4  $\pm$  2.1 mol  $m^{-2}~vr^{-1}$ during 2011–2018) compared with that in the 2000 s (1.1  $\pm$  1.5 mol  $m^{-2} vr^{-1}$  during 2005–2011) might also be meaningful. According to a modeling study, the present enhancement of coastal ocean CO<sub>2</sub> uptake globally is driven largely by the globally increasing biological carbon fixation (Mathis et al., 2024), as evidenced by satellite-derived phytoplankton bloom dynamics over global coasts during 2003-2020 (e.g., Dai et al., 2023). However, satellite-derived chlorophyll-a over the Bohai and Yellow seas increased before 2011 and declined thereafter (Wang et al., 2021), indicating again that basin-scale eutrophication mitigation occurred in the 2010 s. This is different from many other ocean margins. Therefore, the slight weakening of CO<sub>2</sub>-absorbing intensity over the SYS was likely related to eutrophication mitigation and the relevant basin-scale reduction in primary production in the Bohai and Yellow seas, which shares the same logic as Mathis et al. (2024).

Some other factors such as climate-induced changes in open ocean circulation (e.g., Zhang, et al., 2020; Peng et al., 2022; Wang et al., 2022) have severe impact on changes in coastal biogeochemistry and/or air–sea  $CO_2$  flux (e.g., Mathis et al., 2024; Wang et al., 2024). In a given marine system (i.e., the Yellow Sea), it is unclear exactly what would happen following regional environmental changes. Moreover, the uncertainties remain too large to identify changes in the rate of air–sea  $CO_2$  exchange in the Yellow Sea between the 2000 s and 2010 s. Nevertheless, we believe that additional field data should be collected within a certain temporal frame to potentially clarify the changes in  $CO_2$ -absorbing intensity in the changing shelf system of the Yellow and the East China seas.

#### 5. Summary and concluding remarks

In this study, we quantified basin-scale air–sea CO<sub>2</sub> exchange flux by resolving both the temporal and the spatial variations in sea surface *p*CO<sub>2</sub> based on multiple-year underway measurements obtained in the South Yellow Sea (SYS) during 2005–2011. Using the S07 equation, the annual air–sea CO<sub>2</sub> influx rates ranged from 0.9 ± 1.5 mol m<sup>-2</sup> yr<sup>-1</sup> in the western SYS (Subregion I) and 0.9 ± 2.3 mol m<sup>-2</sup> yr<sup>-1</sup> in the southern SYS (Subregion III) to 1.6 ± 0.5 mol m<sup>-2</sup> yr<sup>-1</sup> in the central SYS (Subregion III). The area-weighted average of the CO<sub>2</sub> influx rate was estimated to be 1.1 ± 1.5 mol m<sup>-2</sup> yr<sup>-1</sup> over the three subregions under survey during 2005–2011, which was only half that of the value for the East China Sea shelf (ranging from 1.7 ± 2.0 mol m<sup>-2</sup> yr<sup>-1</sup> in Subregion V to 2.3 ± 1.8 mol m<sup>-2</sup> yr<sup>-1</sup> in Subregions IV and VI).

Over Subregions I and II, both the flux estimation results and the seasonal variation pattern during 2005–2011 were broadly similar to those based on mapping data of discrete fugacity of  $CO_2$ , as calculated from DIC and TAlk data obtained in a similar area during 2011–2018. Over Subregion III, however, comparison of the monthly variation in air–sea  $CO_2$  flux during the two periods indicated that the annual  $CO_2$  uptake had weakened in this subregion. Both cases were different from many other ocean margins characterized by enhanced  $CO_2$  uptake following rise in atmospheric  $CO_2$ . To study the potential shift from a source to a sink of  $CO_2$  over multiple decades, further field observations and data-based research are needed, especially in relation to large continental shelves that are experiencing environmental changes.

#### CRediT authorship contribution statement

Wei-dong Zhai: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. Xiang-hui Guo: Writing – review & editing, Funding acquisition, Data curation. Yan Bai: Resources. Xianqiang He: Writing  review & editing, Resources. Kai Tang: Writing – review & editing, Funding acquisition. Minhan Dai: Resources.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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# Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.pocean.2025.103466.

# Data availability

Data will be made available on request.

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#### W.-d. Zhai et al.

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