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Effect of large-scale kelp and bivalve farming on seawater carbonate system variations in the semi-enclosed Sanggou Bay

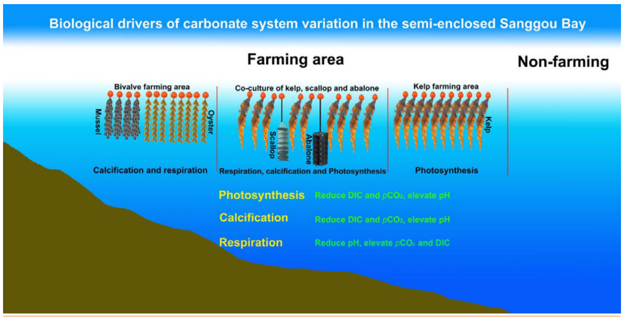
Jiaqi Li, Wenwen Zhang, Jingkun Ding, Suyan Xue, Enze Huo, Zhanfei Ma, Wenhan Yu, Zengjie Jiang, Jianguang Fang, Yuze Mao *

Yellow Sea Fisheries Research Institute, Chinese Academy of Fishery Sciences, Qingdao 266071, China
Laboratory for Marine Ecology and Environmental Science, Piolet National Laboratory for Marine Science and Technology (Qingdao), Qingdao 266071, China

HIGHLIGHTS

- Kelp and bivalve farming are important biological drivers of carbonate chemistry variations within the Sanggou Bay.
- The fluctuation of carbonate systems in farming areas were much larger than those in non-farming area.
- Kelp farming may favour the calcification of farmed bivalves and provide essential refuge for these species.
- Farmed bivalves can fix larger amounts of inorganic carbon by calcification than that released into seawater by respiration.

GRAPHICAL ABSTRACT



Large-scale of kelp and bivalve farming are important biological drivers of carbonate chemistry variations within the semi-encolsed Sanggou Bay.

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ABSTRACT

Although cultured algae and shellfish can be the dominant species in some localized coastal waters, research on the effect of large-scale mariculture on the carbonate system variations in these local waters is still lacking. We conducted five cruises from May to September and studied spatiotemporal variations in the seawater carbonate system in the semi-closed Sanggou Bay, which is famous for its large-scale mariculture. Our results showed that both kelp and bivalve farming induced significant spatiotemporal variations in the carbonate system within the bay. When cultured kelp reached its highest biomass in May, the maximum ΔDIC, ΔpCO$_2$ and ΔpH$_T$ between the seawater from the kelp farming area and the non-farming outer bay area was -156 μmol kg^{-1}, -102 μatm and 0.15 pH units, respectively. However, no significant effect of kelp farming on seawater total alkalinity (TA) was observed. Kelp farming also caused the carbonate system variations of seawater from the bivalve farming area. Assuming no kelp was farmed in May, the average pH and pCO$_2$ would reduce by 0.12 pH units and increase by 179 μatm, respectively, in the bivalve farming area. Bivalve farming significantly reduced seawater TA, indicating that fast deposition of calcium carbonate occurred in the bivalve farming area. Although bivalve respiration released CO$_2$ into seawater and elevated seawater pCO$_2$ level and reduced seawater pH$_T$, surprisingly, seawater dissolved inorganic carbon (DIC) reduced significantly in the bivalve farming area. These results indicated that bivalves fixed a larger amount of inorganic carbon by calcification than that released into seawater by respiration. Overall, large-scale kelp and bivalve farming are important biological drivers of variations in the carbonate system within the semi-enclosed Sanggou Bay. Altered carbonate systems by kelp farming may favour calcification of farmed bivalves and provide an essential refuge for these species during the future ocean acidification.

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* Corresponding author at: Yellow Sea Fisheries Research Institute, Chinese Academy of Fishery Sciences, Qingdao 266071, China.
E-mail address: maoyz@ysfri.ac.cn (Y. Mao).

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1. Introduction

Marine organisms rely on seawater carbonate systems to process biological activities such as photosynthesis (Koch et al., 2013; Kroeker et al., 2010) and calcification (Riebesell et al., 2000; Weiss et al., 2002). Accordingly, they are also capable of changing the seawater carbonate chemistry that surrounds them (Cyronak et al., 2018; Hendriks et al., 2014; Jiang et al., 2015). In open environments, seawater carbonate systems usually remain stable and seldom show strong variation. However, it is much easier to observe strong variations in the seawater carbonate systems of macroalgae or calcifying organisms that dominate shallow water ecosystems. Marked seasonal variations in the carbonate systems produced by seagrass meadows (Bouillon et al., 2007; Challener et al., 2016) and macroalgal beds (Delille et al., 2000) have been reported. In kelp dominated seabeds, kelp can elevate the pH, oxygen, and aragonite saturation state of local seawater, but lower dissolved inorganic carbon (DIC) content and total alkalinity (TA) (Pfister et al., 2019). Coral dominated areas always have a lower mean DIC and TA and also exhibit greater variability than outer reef areas (Page et al., 2019). There are also a large amounts of seagrass and macroalgae that co-exist in some coral reefs; thus, both strong temporal and spatial variations in seawater pH and pCO_2 in shallow coral reefs have also been observed (DeCarlo et al., 2017). Even larger variations in seawater carbonate systems have been observed in tide pools, which are essentially closed during low tides and restore relatively small amounts of seawater. Metabolism of tidal pool communities induces marked pH variability within and between tide pools, in which the pH varied up to 1.0 pH unit during the daytime (Jellison and Gaylord, 2019; Silbiger and Sorte, 2018). By conducting photosynthetic activity, macrophytes drive the decrease of DIC and pCO_2, and the increase of pH in local seawaters (Buapet et al., 2013; Delille et al., 2000), playing an essential role in regulating carbonate system fluctuations in surrounding seawater (Howland et al., 2000), especially in shallow coastal waters where the water exchange rate is low (Hendriks et al., 2014; McGrath et al., 2016). Corals also use dissolved inorganic carbon to build their calcium carbonate frameworks that is produced through calcification. Calcification by reef-building organisms is the main factor that controls the variations in seawater carbonate systems inside the coral reef (Watanabe et al., 2006). The calcification rate in shallow coral reef flats can be so rapid that it can drive seawater that is highly saturated with calcium carbonate ($\Omega_{Arag} \gg 1$) towards unsaturation ($\Omega_{Arag} < 1$) (DeCarlo et al., 2017). Community metabolism manipulates strong variations in tide pools, and as the communities vary substantially among tide pools; producer-dominated pools always have the highest and most variable pH values, whereas consumer-dominated pools had lower pH values and minimal diel variation (Jellison and Gaylord, 2019). Strong biological influence can also extend to the ecosystem-scale; Lowe et al. (2019) quantified seawater pH variations across ~7500 km^2 of coastal ecosystems in Washington State and the relationship to abiotic and biotic ecological factors based on monthly observations for over 25 years. Their study provided evidence that seasonal changes in local ecosystem metabolism dominate other sources of variability of carbonate system.

Variations in the carbonate systems in these coastal waters may have remarkable impacts on local organisms. It has been hypothesized that highly productive seagrass meadows could elevate the pH and aragonite saturation state of seawater and establish ocean acidification (OA) refugia. The high photosynthetic rates of seagrass contributed to increased calcification rate of calcifying organisms (Hendriks et al., 2015; Hendriks et al., 2014; Hurd, 2015; Krause-Jensen et al., 2015). Besides calcification, Hoshijima and Hofmann (2019) also found that adult sea urchins that completed sexual maturation inside the kelp forest produced more protein-rich eggs that developed into more pH-resilient embryos than those matured outside the kelp forest. These studies suggested that seagrass meadow could provide refuge against OA effects and increase the resilience of species susceptible to OA (Bergstrom et al., 2019; Hendriks et al., 2014).

Although cultured algae and shellfish can be the dominant species in some localized coastal waters, the effect of large-scale algae and shellfish aquaculture on carbonate system variations the in semi-enclosed bays still need to be studied. Sanggou Bay is a shallow semi-enclosed bay, and has been used for mariculture for over 30 years. It is also one of the most representative bays for the integrated multi-trophic aquaculture (IMTA) of macroalgae and shellfish (Fang et al., 2016). The annual production of seafood in the Sanggou Bay is approximately 240 000 t fresh weight (Fang et al., 2016). In this study, we reported the seawater carbonate variations in the semi-enclosed Sanggou Bay produced by the large-scale and high-density kelp and bivalve farming.

2. Material and methods

2.1. Studying area and sampling stations

Sanggou Bay is situated on the eastern tip of Shandong Peninsula to the northwest of the Yellow Sea (37° 01′ to 37° 09′ N, 122° 24′ to 122° 35′ E), with a total area of approximately 144 km^2 and a mean depth of 7.5 m (Zhang et al., 2009). The bay mostly produces kelp (*Saccharina japonica*), the Pacific oyster (*Crassostrea gigas*), blue mussel (*Mytilus galloprovincialis*), the Pacific abalone (*Haliotis discus hannai*) and *zhikong* scallop (*Chlamys farreri*).

Kelp cultivation occurred mainly in the middle parts and around the mouth of the Sanggou Bay (Fig. 1) from late November to early July of the next year, while bivalves were cultivated in the bottom of the bay (Fig. 1) all year round. The highest biomass of kelp appeared in May, while the biomass of bivalves remained relatively stable all year round. In May, we launched the first cruise, in which 54 sampling stations were surveyed and the cultivated species around these sampling stations were also recorded (Fig. 1). The sampling stations were carried out in May and June to September (Fig. 1). In May, we used a patrolling speedboat to complete the whole bay survey, while for the rest of the surveys we used the local fishing boat 'Lu Rong Yu Yang 65579'.

2.2. Sampling strategy

As the locations of the sampling stations were distributed from the bottom of the Sanggou Bay to the outer bay area, the semidiurnal tidal current in this bay could have caused temporal variations in seawater chemistry within the same station. To minimize the effects of tidal currents on sampling, we carried out the survey on the same lunar day of each month. During each cruise, sample collection started around 8:00 AM and finished around 2:00 PM.

At each sampling location, in situ temperature and salinity were measured using a multi-electrode analyzer (Multi 3630, WTW; Germany) approximately 0.50 m below the sea surface. A Ruttner sampler (HYDROBIOS; Germany) was used for collecting seawater from the sea surface (0.5 m depth). The collected seawater was transferred into 2000 mL NIKKO polypropylene bottles and immediately poisoned with 0.5 mL of saturated solution of mercuric chloride ($HgCl_2$, 10 g of mercuric chloride was added into 100 mL of distilled water) to halt any biological activity within the seawater. These samples were shipped to a nearby laboratory in the National Engineering Center of Marine Shellfish where they were analyzed for pH and total alkalinity (TA).

2.3. Analytical methods

The pH_T was measured using spectrophotometry in 10 cm pathlength quartz cuvettes with a purified *m-cresol* dye following the standard protocols for seawater CO_2 measurements (Dickson et al., 2007). Seawater samples were incubated in a 25 °C thermostat bath for 2 h before the

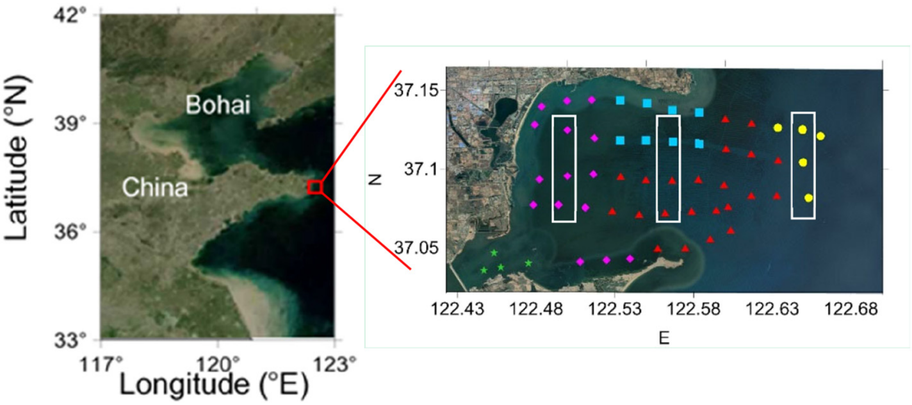

Fig. 1. Study site and the sampling stations in the Sanggou Bay. Five sampling surveys were carried out from May to September 2019. Coloured symbols represent the sampling stations in the survey carried out in May, yellow round dots represent non-farming areas, red triangles represent kelp farming areas, blue squares represent IMTA areas, pink diamond represent bivalve farming areas and green stars represent fish cage areas. Stations in the three rectangles represent the sampling stations in the surveys carried out from June to September.

pH_T measurements were taken using a double-beam spectrophotometer (UV-9000S, METASH) at room temperature (25 °C). Before being used for the carbonate system calculation, the measured pH_T was transformed into in situ pH_T using CO2SYS (Pierrot et al., 2006). TA was measured using open-cell automatic potentiometric titration (848 Titrino plus; Metrohm, Switzerland) (Perez et al., 2000). The accuracy and precision of the TA measurements were determined using Certified Reference Material (BATCH #178) from the laboratory of A. Dickson (Scripps Institution of Oceanography) that yielded TA values within ~5 $\mu mol\ kg^{-1}$ of the nominal value. The carbonate system parameters of the sampled seawater were determined with the CO2SYS (Pierrot et al., 2006) using in situ pH_T and the measured values of TA, temperature, and salinity. For the calculation we used constants provided by Mehrbach et al. (1973) that have been refitted by Dickson and Millero, 1987and KSO4 constants from Dickson (1990).

CO_2 fluxes at the seawater-air interface were calculated as a function of the air-seawater CO_2 gradient (ΔpCO_2), temperature and salinity dependent solubility (k_0, (Weiss, 1974)) and gas transfer velocity (k) according to Eq. (1).

$$FCO_2 = k * k_0 \Delta pCO_2 \tag{1}$$

where k is the CO_2 gas transfer velocity, k_0 is the solubility of CO_2 and ΔpCO_2 is the difference between sea and air ($pCO_2sea - pCO_2air$). pCO_{2sea} was calculated using CO2SYS, while pCO_{2air} was assumed to be 400 μatm (http://keelingcurve.ucsd.edu/). k was calculated using Eq. (2) following Wanninkhof et al. (2009).

$$k = 0.251u^2(Sc/660)^{1/2} \tag{2}$$

where k is the transfer velocity ($cm\ h^{-1}$), u is the wind speed (ms^{-1}) that was equal to the monthly average of wind speed according to the local meteorological bureau, and *Sc* is the Schmidt number of CO_2 at in situ temperature and salinity.

We compared the statistical differences in seawater inorganic carbon variables among areas and cruises using a one-way ANOVA (SAS 9.0) to detect significant differences. All data were tested for homoscedasticity using Leven's test. A p value <0.05 was considered significant.

3. Results

3.1. Spatial and temporal variability of pH_T and TA

Kelp was cultivated in the first two cruises in May and June, and it was completely harvested in the last three cruises in July, August, and September. Table 1 provides a summary of all the carbonate system variable ranges in each of the three areas. Both strong spatial and temporal variation in seawater pH_T and TA were observed (Fig. 2). In May, the average seawater pH_T values in the kelp farming area, bivalve farming area and outer bay area were 7.98 ± 0.04, 7.94 ± 0.02 and 7.92 ± 0.02, respectively. The seawater pH_T in the kelp farming area was significantly ($F = 9.39$, $df = 2$, $n = 46$) higher than those of the bivalve farming and outer bay areas. In June, the average seawater pH_T in the kelp farming area (7.99 ± 0.01) and the outer bay area (7.98 ± 0.02) were significantly higher than that of bivalve farming area (7.91 ± 0.01). In the following months, the seawater pH_T level increased from the inner bay towards the outer bay area. Meanwhile, the highest and lowest seawater pH_T value appeared in the outer bay and bivalve farming area, respectively. A significant difference ($F = 8.88$, $df = 4$, $n = 9$) in average pH_T was also found between the cruises. In the kelp farming area, the average seawater pH_T decreased from May to September. In the bivalve farming and outer bay area, the lowest average pH_T value for both appeared in September, while the highest average pH_T was found in August and July, respectively.

Seawater TA increased from the inner bay towards the outer bay area and the average seawater TA in bivalve farming area was significantly lower ($F = 29.12$, $df = 2$, $n = 15$) than in the other two areas.

Table 1
Carbonate system variables summary statistics aggregating all nine sampling sites from May to September.

Variable	Area	Minium	Maximum	Mean
DIC ($\mu mol\ kg^{-1}$)	Bivalve	1899	2110	2018
	Kelp	2024	2153	2099
	Non-farming	2030	2185	2116
TA ($\mu mol\ kg^{-1}$)	Bivalve	2114	2326	2218
	Kelp	2243	2391	2340
	Non-farming	2226	2391	2354
pH_T-25	Bivalve	7.73	7.97	7.90
	Kelp	7.89	8.06	7.97
	Non-farming	7.89	8.03	7.96
pCO_2 (μatm)	Bivalve	427	862	555
	Kelp	260	591	417
	Non-farming	349	575	421
HCO_3^- ($\mu mol\ kg^{-1}$)	Bivalve	1724	1934	1855
	Kelp	1882	1963	1908
	Non-farming	1867	2016	1926
CO_3^{2-} ($\mu mol\ kg^{-1}$)	Bivalve	101	166	148
	Kelp	152	207	178
	Non-farming	147	200	176

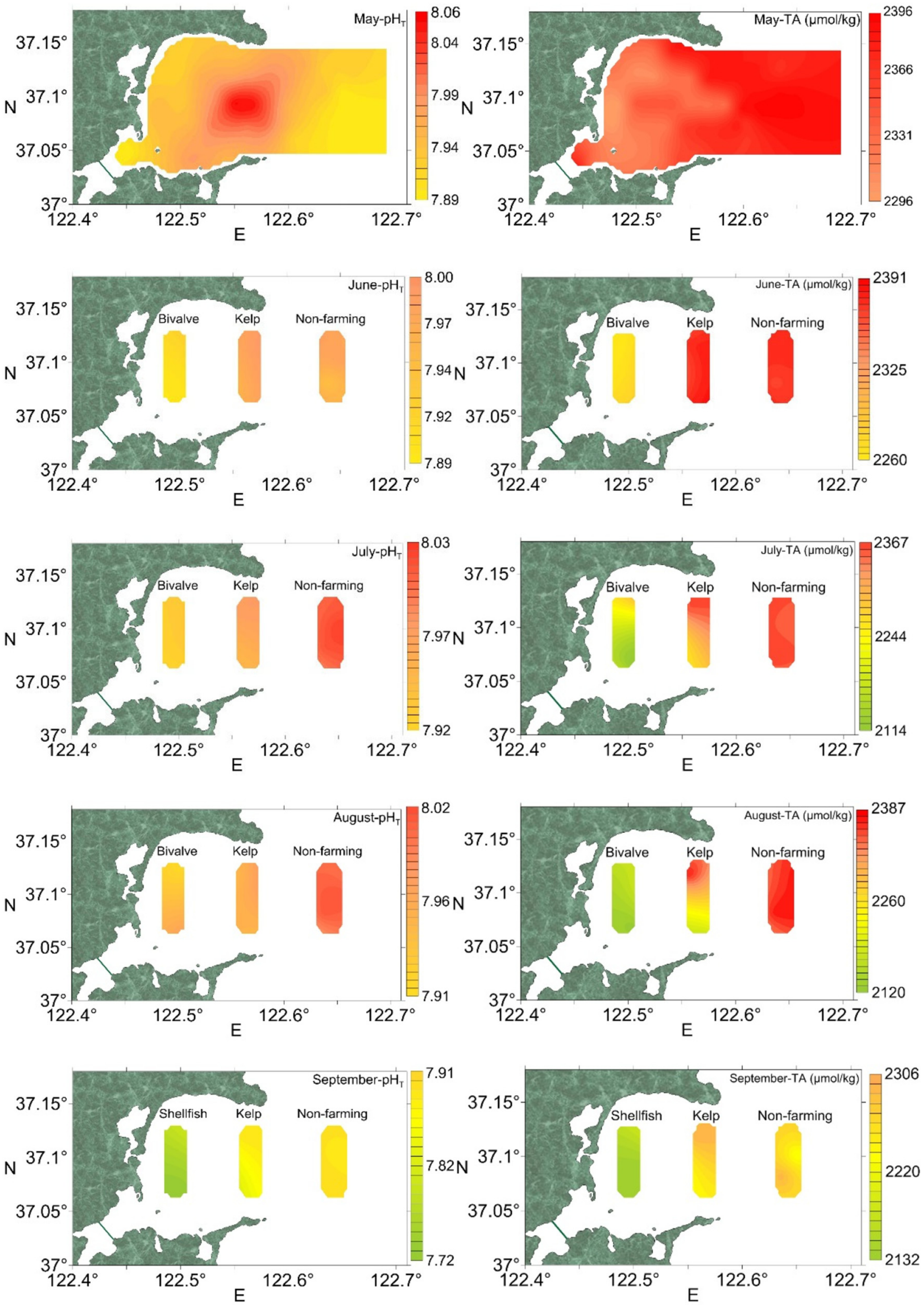

Fig. 2. Contour plots of pH$_T$ and total alkalinity (TA) (μmol kg^{-1} seawater) of the surface seawater from different farming areas (bivalve farming and kelp farming areas) in the Sanggou Bay and the outer bay (non-farming) area from May to September. All of the measured seawater pH$_T$ were normalized to 25 °C.

Moreover, in the bivalve farming area, the average seawater TA decreased from May (2323 ± 17 μmol kg^{-1} seawater) to September (2152 ± 28 μmol kg^{-1} seawater), as the seawater temperature increased.

3.2. Spatial and temporal variability of pCO$_2$ and DIC

Kelp and bivalve farming also strongly influenced the spatial and temporal variation of pCO$_2$ and DIC in the Sanggou Bay (Fig. 3). The

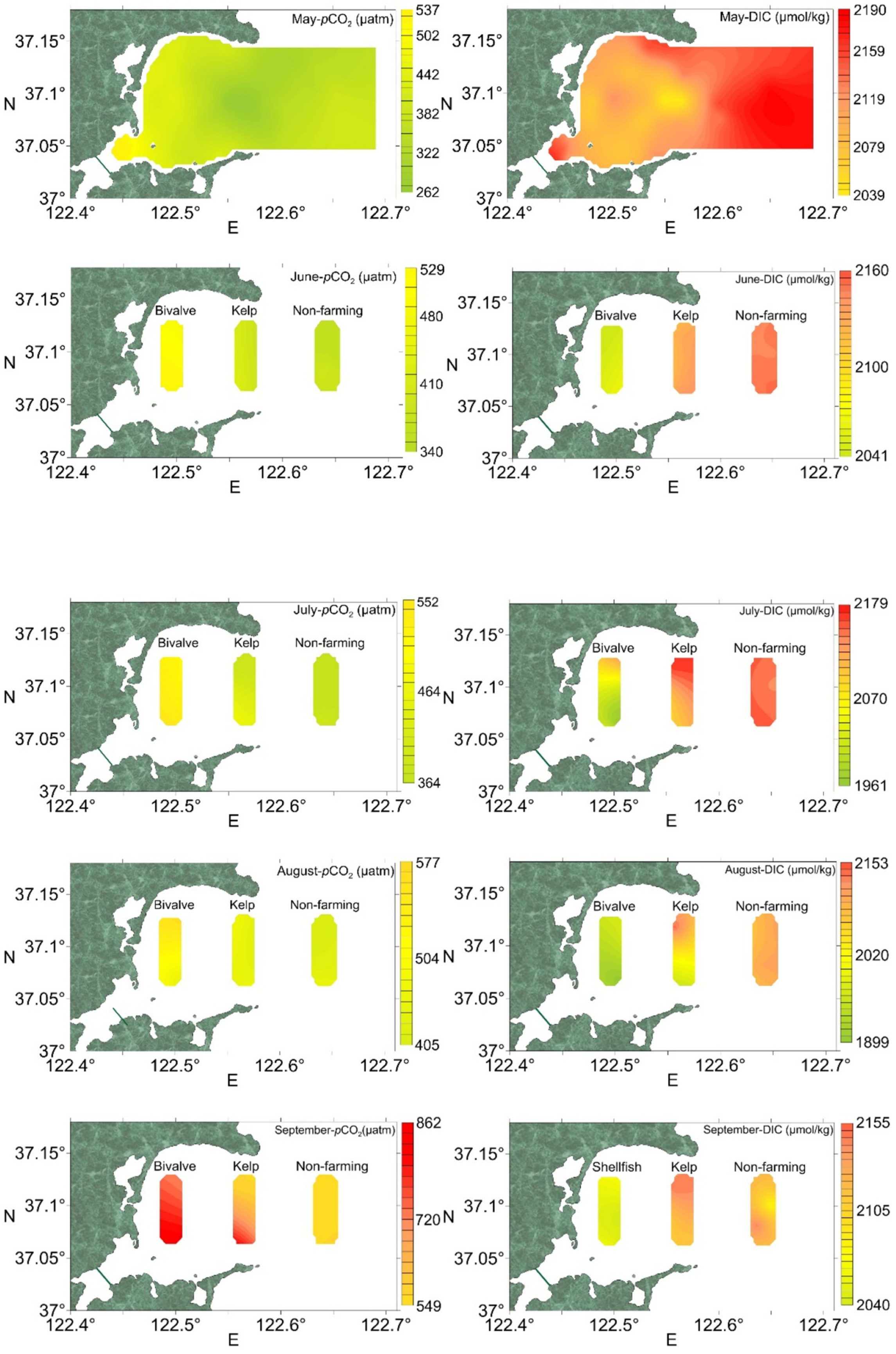

Fig. 3. Contour plots of pCO$_2$ (μatm) and dissolved inorganic carbon (DIC) (μmol kg^{-1} seawater) of the surface seawater from different farming areas (bivalve farming and kelp farming areas) in the Sanggou Bay and outer bay (non-farming) area from May to September. pCO$_2$ and DIC were calculated based on pH$_T$, TA, water temperature and salinity.

spatial variation of pCO_2 showed opposite trend compared to that of the seawater pH_T. In the May and June cruises, when kelp was being cultivated, the lowest average pCO_2 level appeared in the kelp farming area (335 ± 34 and 370 ± 43μatm, respectively), however, in the July, August and Septembercruises, the lowest average pCO_2 level was found in the outer bay area (367 ± 5, 432 ± 3 and 570 ± 3μatm, respectively). In general, the temporal variation of the pCO_2 level in all of the three areas increased with the rising seawater temperature form May to September.

In the May cruise, the lowest DIC level was found in the kelp farming area (2102.67 μmol kg^{-1} seawater), while the lowest DIC level appeared in the bivalve farming area in the other four cruises. In terms of temporal variation, the DIC level increased from May to July and decreased from August and September in the kelp farming area, while the DIC level decreased from May to August and increased again in September in the bivalve farming area. In the outer bay area, the DIC level generally decreased from May to September.

3.3. Kelp and bivalve farming manipulated the seawater carbonate system in the Sanggou Bay

Large scale aquaculture altered the carbonate system of seawater that flew through the kelp and bivalve farming areas. Seawater pH_T and $[CO_3^{2-}]$ in the kelp farming area increased, while seawater pCO_2 and DIC decreased when seawater flowed through the cultivated kelp (Fig. 4). We also found that pH_T of seawater in the bivalve farming area was higher than that of the seawater in the outer bay area in May when the cultivated kelp reached its maximum biomass. The opposite results were observed in July, August and September after the cultivated kelp had been completed harvested. When seawater entered the bivalve farming area, seawater pH_T, $[CO_3^{2-}]$ and DIC declined, while seawater pCO_2 increased. Generally, the effects of bivalve farming on these carbonate variables became stronger from May to September (Fig. 4).

These activities shapde the distinctive inorganic carbon characteristics of the bivalve farming, kelp farming and outer bay areas. The carbonate system of surface seawater was highly correlated between all three sites with in the same area (Fig. 5). The bivalve farming area was far less correlated with the kelp farming and outer bay areas. It was also easy to distinguish the sampling sites of the kelp farming area from that of the outer bay area. These results also demonstrated the differential effects of different farming species on seawater inorganic system variation in the Sanggou Bay.

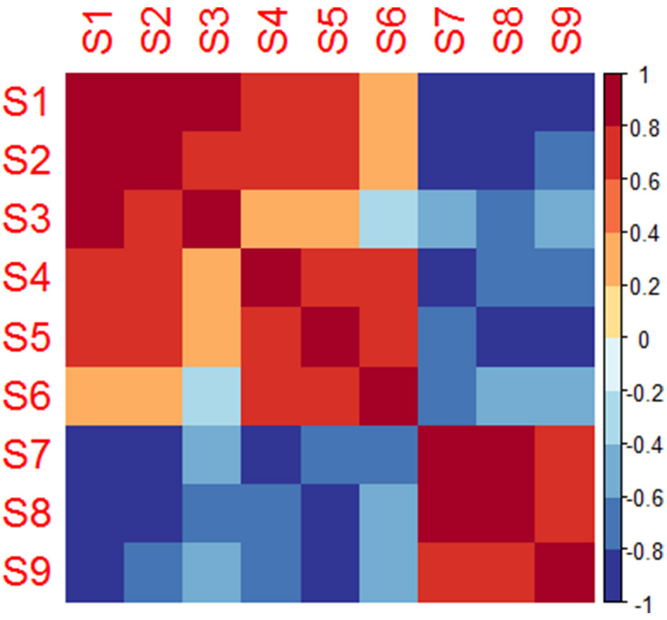

Fig. 5. Correlation heat maps of seawater carbonate system variables (pH, TA, DIC, and $pCO2$) between the nine sampling sites. S1, S2 and S3 were from the bivalve farming area, S4, S5, and S6 were from the kelp farming area, while S7, S8 and S9 were from the outer bay non-farming area.

4. Discussion

In the Sanggou Bay, the photosynthetic activity of kelp removes inorganic carbon from seawater and reduces DIC and pCO_2 level, while increasing pH_T level. On the contrary, respiration of bivalves releases CO_2 into seawater and increases DIC and pCO_2 level, while reducing pH_T levels. In addition, the calcification of bivalves uses bicarbonate (or carbonate) and reduces the TA of the surrounding

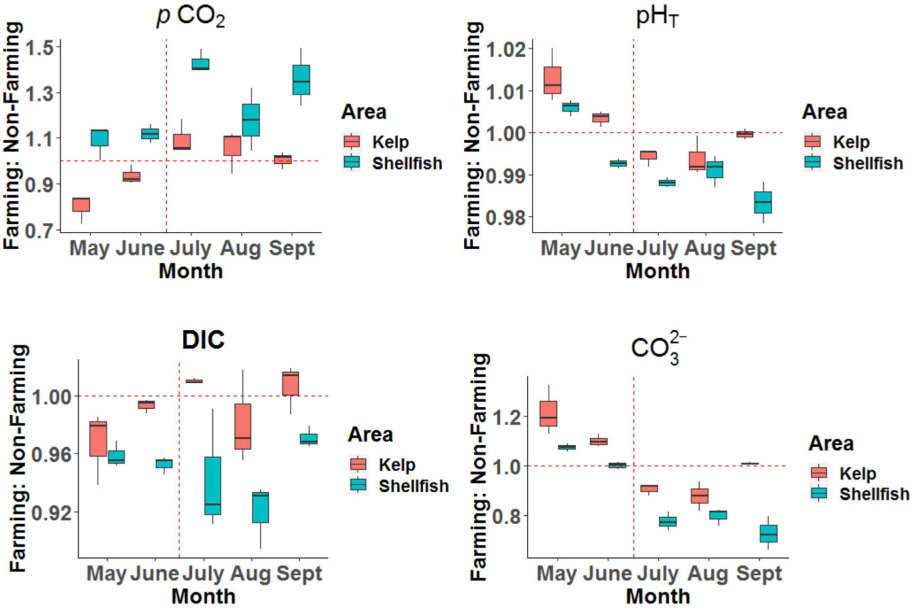

Fig. 4. Variation in the ratios of inorganic carbon variables from the farming areas to the average value of the corresponding variables from the non-farming areas among the five surveys.

seawater. Based on the findings of this work, it is clear that these metabolic activities are the main drivers that shaped the spatiotemporal variation of the seawater carbonate system in this semi-enclosed bay.

4.1. The effect of kelp farming on the seawater carbonate system

Large-scale and high-density of kelp farming is one of the main drivers of seawater carbonate system variation in the Sanggou Bay, especially in May when kelp reaches its maximum biomass. In mariculture ecosystem, the biomass density is usually much higher than that of natural habitats. According to the data provided by the local Fishery Technology Extension Station, the average annual production of kelp in the Sanggou Bay is approximately 15 tons (wet weight) within 1380 m^2. This means that, in May when the cultivated kelp reached its maximum biomass, the average kelp biomass within 1 m^2 meter was close to 11 kg, which is far greater than that in natural kelp forests (around 1.5 kg m^{-2}, Koweek et al. (2017) and in seagrass meadows (1 to 2 kg m^{-2}, (Lopez et al., 2019; Sousa et al., 2019)).

It is known that the natural habitats of macrophyte in coastal waters play an important role in global carbon uptake and stock (Fourqurean et al., 2012; Rohr et al., 2016; Tokoro et al., 2014). In these habitats, significant variations in the carbonate system in kelp forest ecosystems (Koweek et al., 2017; Pfister et al., 2019) and seagrass meadows (Bergstrom et al., 2019; Chou et al., 2018; Cyronak et al., 2018) have been frequently reported. Although the global annual production of farming algae is very large (FAO, 2020), the effect of these algae in carbonate system variations in local waters did not attract much attention.

In the May cruise of our investigation, the lowest DIC and pCO_2 levels and highest pH_T value were observed in the center of the kelp farming area, and the largest differences in DIC, pCO_2 and pH_T between the kelp farming and outer bay areas were 156 μmol kg^{-1}, 102 μmol kg^{-1} and 0.15 pH units, respectively.Referring to studies that research the monthly growth and carbon content of kelp cultivated in the Sanggou Bay (Mao et al., 2018; Wang et al., 2020), we calculated the inorganic carbon fixation rate of these kelp in May. The calculated date showed that approximately 0.19-mol of inorganic carbon was fixed in 1 m^2 from the kelp farming area within one day. If we assume that the kelp grew in seawater within 0-3 m below the surface, then around 63 μmol inorganic carbon would have been removed from one liter of seawater within 24 h.

Our results demonstrated that seawater must be 'trapped' in the kelp farming area for more than one day before it entered the bivalve farming or returned to the outer bay area. It had been reported that the high density of kelp significantly reduced the tidal flux in the upper water layers 2–5 m below the surface, where kelp grew (Zeng et al., 2015). As a result, water exchange between the inner bay and the outer bay area was hindered by the 'kelp forest'. Due to the lowered water exchange rate, it was also much easier for the kelp to reduce seawater DIC level within the Sanggou Bay.

Large-scale kelp farming also occurs to the north of the Sanggou Bay, in a nearshore open environment, in which water exchange is supposed to be easier than that in the Sanggou Bay. Jiang et al. (2013) reported that the DIC level in those kelp farming areas were also lower compared to those in non-farming areas, however, no significant difference in DIC level was detected. Although the average densities of kelp biomass in these two study sites were approximately the same, it would be harder to observe significantly altered carbonate system if seawater did not reside in kelp farms long enough. There was a special situation reported by Martz et al. (2009), who found significantly altered carbonate system in surface seawater during a phytoplankton bloom in open waters. Although the seawater did not reside in open waters, phytoplankton drifted along with the seawater, so it was easy for the blooming phytoplankton to manipulate the carbonate system in the surrounding seawater.

4.2. The effect of bivalve farming on seawater inorganic carbon variation

Large-scale and high-density of bivalve farming is another main driver of seawater carbonate system variation in the Sanggou Bay. In all of the five cruise, the lowest pH_T, TA, DIC and the highest pCO_2 level were usually found in the bivalve farming area. In some natural habitats, the calcification of corals and other reef-building organisms also contribute to the significant spatiotemporal variability in seawater carbon chemistry in coral reef flats (Page et al., 2019; Watanabe et al., 2006). Even, calcifying algae (coccolithophore) bloom can reduce seawater TA and shift seawater carbonate system in a large bay (Harlay et al., 2010). However, the carbonate system variation within bivalve farming area may be different from that in these natural habitats, as the biomass density of calcifying organisms in mariculture systems are usually much higher. The annual production of bivalves within the Sanggou Bay is around 75×10^3 t and the growth rates of these organisms reach peak in summer. It was estimated that the carbon sequestration rate in shell forming of cultivated oyster in the Sanggou Bay was 2.14 t C hm^{-2} yr^{-1} (Ren, 2014), and approximately 80% of $CaCO_3$ in the shell was deposited from May to October. Based on the data from the work of (Ren, 2014), we recalculated the carbon deposition rate from May to October and the result was 0.079 mol C m^{-2} d^{-1}. TA reduction is synchronized with the use of HCO_3^- in the calcification process. When one mole of $CaCO_3$ is produced by calcifying organisms, the TA decreases by two moles. Accordingly, the TA reduction rate should be 0.158 mol $m^{-2}d^{-1}$ in this case. Oysters also grew in seawater 0–3 m under the surface, which would make the seawater reduction rate 52 μmol kg^{-1} within 24 h. The greatest TA reduction in bivalve farming area was 244 μmol kg^{-1}. Therefore, bivalves should be the main calcifying organisms that were responsible for the TA reduction in the Sanggou Bay. We can also expect that the water residence time in the bivalve farming area must have been even longer than that in the kelp farming area. The prolonged residence time of seawater also made it easy to observe significant variation in the seawater carbonate system in coral reefs. Page et al. (2019) reported that seawater carbonate system variation in coral reef flat was strongly affected by water depth through its effects on the benthic biomass to seawater volume ratio, seawater flow rates, and residence time.

Since the lowest pH_T and highest pCO_2 levels were found in the bivalve farming area due to the metabolic activities of bivalves, one may expect that the respiration of large number of bivalves in the bay would elevate DIC level within the bivalve farming areas, especially when bivalves experience suitable seawater temperature in the summer. On the contrary, we found decreased DIC level in the bivalve farming area (Fig. 6). Although the pCO_2 level was elevated by the respiration of bivalves, the concentration of carbonate and bicarbonate was

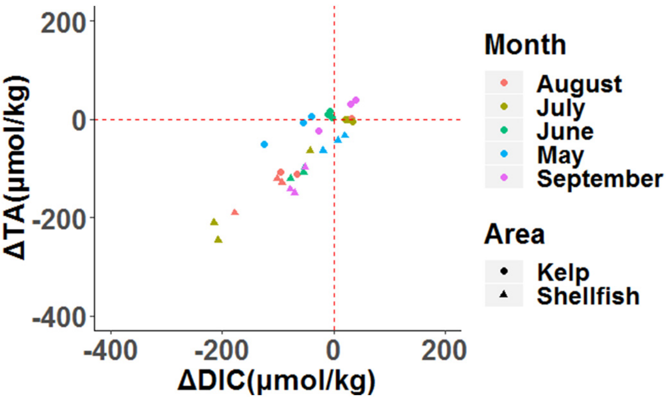

Fig. 6. Seawater ΔTA:ΔDIC regressions in kelp and bivalve farming areas. NCP represents net community primary production, while NCC represents net community calcification.

lowered by the calcification of these bivalves. As there was no algae that was cultivated within the bivalve farming area, the main reason for DIC reduction is that calcification surpassed respiration with respect to inorganic carbon regulation. It has been reported that oyster are capable of removing more inorganic carbon from seawater though calcification than by releasing inorganic carbon into seawater through respiration (Ren, 2014). However, photosynthesis is the main reason for DIC reduction in coral reefs; as seagrass and algae growth in the reef flat can greatly elevate seawater pH and decrease $p\mathrm{CO_2}$ during the day (DeCarlo et al., 2017). Moreover, the DIC level increase in the coral reef communities during the night when calcification and respiration continued and photosynthesis stops (Page et al., 2017). DIC reduction (ΔDIC) is used to evaluate the net community primary production (NCP) of coral reefs, but in our case it was not suitable. These differences may demonstrate that the calcification rate of large scale bivalve (mainly oyster) farming in the Sanggou Bay is far more intense than that of coral reefs.

4.3. The influence of altered carbonate system on marine ecosystem of Sanggou Bay

Large-scale kelp and bivalve farming influences the sea-air $\mathrm{CO_2}$ exchange in the whole Sanggou Bay. Macrophytes dominated ecosystems in shallow coastal waters showed a significantly high inorganic carbon capturing efficiency (Ikawa and Oechel, 2015; Kim et al., 2015), and could functionally be a sink or reservoir for atmospheric $\mathrm{CO_2}$ (Mazarrasa et al., 2015; Tokoro et al., 2014). Large-scale of kelp farming in the Sanggou Bay increases the $\Delta p\mathrm{CO_2}$ between air and surface seawater and promote the $\mathrm{CO_2}$ sink from the atmosphere to the sea. Using400 μatm as the atmospheric $p\mathrm{CO_2}$ level during our cruises (https://scripps.ucsd.edu/programs/keelingcurve/), the kelp farming area could have acted as a $\mathrm{CO_2}$ sink when the kelp had been cultivated, otherwise, this area would be a $\mathrm{CO_2}$ source after the kelp had been completely harvested (Table 2). The bivalve farming area was a $\mathrm{CO_2}$ source for air as the average seawater $p\mathrm{CO_2}$ level was higher than 400 μatm throughout the investigation (Table 2). Like the situation in some coral reef flats (Lonborg et al., 2019; Mcgowan et al., 2016), large-scale of bivalve farming in the Sanggou Bay was a net source of $\mathrm{CO_2}$ for the atmosphere. The calcification and respiration of cultured bivalves enlarges the $\Delta p\mathrm{CO_2}$ between sea and air and promoted the release of $\mathrm{CO_2}$ into the atmosphere. Overall, taking together the work of Ren (2014), Wang et al. (2020) and our calculations (Table 2), in May when kelp biomass was high, the $\mathrm{CO_2}$ sink rate in the kelp farming area was higher than $\mathrm{CO_2}$ releasing rate in the bivalve farming area.

Kelp not only influences the variation of the carbonate system in its own culturing area, but also affects the variation of that in the bivalve farming area. Outer bay seawater first flows through the kelp farming area and then enters bivalve farming area. Kelp reduce the DIC concentration of the seawater that flows through them; if we remove kelp, then the DIC concentration of the seawater from the bivalve farming area would be higher than it actually was. Assuming the seawater entered the bivalve farming area directly from the outer bay, the carbonate system would be rather different, especially in May when kelp biomass reaches its maximum (Fig. 7). We recalculated the seawater carbonate system in the bivalve farming area on the assumption that no kelp had been farmed. We added the ΔTA and ΔDIC between the seawater from the outer bay and kelp farming area to the observed value of TA and DIC of the seawater from the bivalve farming area and recalculated the carbonate system. The results showed that the average pH reduced by 0.12 pH units and the average pCO2 increased by 179 μatm in the bivalve farming area in May. In this case, the $\mathrm{CO_2}$ releasing rate in the bivalve farming area would be much stronger and bivalves have to grow in seawater with a lower pH level and a higher level of $p\mathrm{CO_2}$, which is not a favorable environment for these species (Gazeau et al., 2007). In June, when kelp harvesting was close to the end, the average pH reduced by 0.03 pH units and the average $p\mathrm{CO_2}$ increased by 44 μatm after recalculation. However, increased pH and decreased $p\mathrm{CO_2}$ levels were found in the last three cruises. The assumed effect of kelp farming may have been overestimated in the last three cruises, as we used ΔTA and ΔDIC from the May cruise, when kelp biomass reached its maximum. Nonetheless, if there were some kind of macroalgae that also had a similar biomass to that of kelp and it was cultivated from July to September, the carbonate system of seawater in the Sanggou Bay would surely be quite different.

Table 2
The average $p\mathrm{CO_2}$ of surface seawater and $\mathrm{CO_2}$ fluxes ($\mathrm{FCO_2}$) for five surveys categorized into three classes: Non-farming, Kelp farming, and Bivalve farming water. *The different upper-case letters indicate significant differences ($P < 0.05$) in variables among sampling areas within the same cruise. Positive $\mathrm{CO_2}$ flux values signify a $\mathrm{CO_2}$ exchange from seawater-to-air ($\mathrm{CO_2}$ source), while negative $\mathrm{CO_2}$ flux values signify an exchange from air-to-seawater ($\mathrm{CO_2}$ sink).

Variables	Cruise	Non-farming area	Kelp farming area	Bivalve farming area
$p\mathrm{CO_2}$ (μatm)	May	363 ± 3	289 ± 8	446 ± 2
	June	375 ± 6	370 ± 4	505 ± 3
	July	367 ± 2	405 ± 4	527 ± 6
	August	432 ± 2	452 ± 9	516 ± 6
	September	571 ± 3	573 ± 1	783 ± 8
$\mathrm{FCO_2}$ (mmol $\mathrm{m^{-2}\,d^{-1}}$)	May	-10.81 ± 5.01^{b}	-28.56 ± 5.77^{a}	8.90 ± 3.47^{c}
	June	-4.19 ± 3.46^{a}	-5.10 ± 6.70^{a}	13.47 ± 2.71^{b}
	July	-3.59 ± 0.53^{a}	0.45 ± 3.25^{ab}	10.92 ± 1.56
	August	2.68 ± 0.23^{a}	4.73 ± 3.66^{ab}	8.77 ± 4.49^{b}
	September	16.44 ± 0.2^{a}	16.71 ± 2.08^{a}	36.06 ± 6.78^{b}

Alterating carbonate systems may influence other local organisms that inhabit the bay. The photosynthetic activity of seagrass increases seawater pH level and may buffer ocean acidification within the meadow (Hendriks et al., 2014). In these habitats, calcifying organisms may be benefited from the modification of the carbonate system. Hurd (2015) and Krause-Jensen et al. (2015) thought that wave-sheltered bays or the within canopies of macrophyte beds might provide inexpensive refugia from OA for vulnerable coastal calcifiers. It has been proven that the formation of larval shells is extremely susceptive to ocean acidification (Li et al., 2013; Li et al., 2018; Waldbusser et al., 2015). Besides oysters and mussels, there are many other wild populations of molluscs like abalone, scallops, clams and cockle dwells in the Sanggou Bay (Fang et al., 2016), and most of these species spawn in May and June. In addition, the larvae of these calcifying organisms can confine themselves to the upper water column. During this time, large-scale and high biomasses of kelp alter the local surface seawater carbonate system and favour the embryonic development and growth of these molluscs, which may provide essential refugee for these species during future ocean acidification (McNeil and Sasse, 2016; Stocker et al., 2013). It can be expected that macroalgae can act as a chemical refuge for shell-forming molluscs in co-culture marine farms under an ocean acidification scenario (Fernández et al., 2019).

Overall, large-scale kelp and bivalve farming are important biological drivers of seasonal carbonate system variation within the Sanggou Bay. The fluctuation of carbonate systems in farming areas were much larger than those in non-farming area. It was much easier for the kelp and bivalves to manipulate seawater carbonate systems within the Sanggou Bay, as water exchange between the inner bay and the outer bay area was hindered by these farmed species. Calcifying dwellers can benefit from carbonate systems manipulated by the kelp farming in the Sanggou Bay.

CRediT authorship contribution statement

Jiaqi Li: Conceptualization, Methodology, Writing - original draft. **Wenwen Zhang:** Investigation, Formal analysis. **Jingkun Ding:** Investigation, Formal analysis. **Suyan Xue:** Formal analysis. **Enze Huo:**

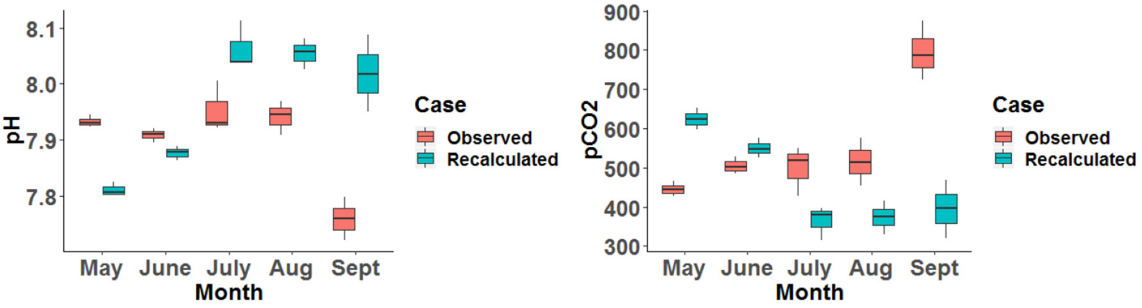

Fig. 7. Observed value of pH and $p\text{CO}_2$ in the bivalve farming area and the recalculated value on the assumption of removing (in May and June) or adding (in July, August and September) kelp farming. We added (May and June) or removed (July, August and September) the ΔTA and ΔDIC between seawater from the outer bay and the kelp farming area to the observed value of seawater TA and DIC from the bivalve farming area and recalculated its carbonate system.

Investigation. **Zhanfei Ma:** Investigation. **Wenhan Yu:** Investigation. **Zengjie Jiang:** Funding acquisition. **Janguang Fang:** Funding acquisition. **Yuze Mao:** Funding acquisition, Writing - review & editing.

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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