



Carbon removal, sequestration and release by mariculture in an important aquaculture area, China

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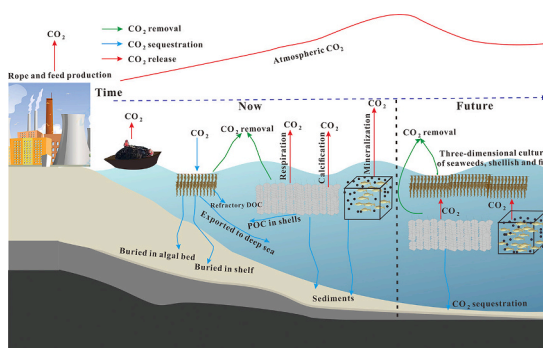
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HIGHLIGHTS

- Seaweeds removed 172 Gg C and sequestered 62 Gg C in 2022.
- CO₂ removal and release by shellfish demonstrated an increase trend during 2003–2022.
- The added CO₂ by fish culture achieved the peak of 60 Gg C in 2011 and then decreased until 2022.
- *Gracilariopsis* requires the smallest area to neutralize CO₂ release by mariculture.
- Area is enough for culturing seaweeds to neutralize total CO₂ emission in Shandong.

GRAPHICAL ABSTRACT



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ABSTRACT

To combat with climate change, most countries have set carbon neutrality target. However, our understanding on carbon removal, release and sequestration by mariculture remains unclear. Here, carbon removal, release and sequestration by maricultured seaweeds, shellfish and fish in Shandong Province during 2003–2022 were assessed using a comprehensive method that considers the processes of biological metabolism, seawater chemistry and carbon footprint. *Saccharina japonica* productivity has been largely enhanced since 2014, resulting in increased production and CO₂ removal and sequestration. Seaweeds removed 172 Gg C and sequestered 62 Gg C in 2022. CO₂ removal and release by shellfish demonstrated a slow increase trend, ranging from 231 to 374 Gg C yr⁻¹ and 897 to 1438 Gg C yr⁻¹ during 2003–2022, respectively. Contrary to seaweed and shellfish, maricultured fish added CO₂ to seawater due to the use of feeds. The added CO₂ by fish culture achieved the peak of 60 Gg C in 2011 and decreased to 25 Gg C in 2022. Most of this added CO₂ was released to atmosphere by microbial mineralization and it was in the range of 21–52 Gg C yr⁻¹ during 2003–2022. After summing up the contribution of seaweeds, shellfish and fish, both total CO₂ removal (from 110 to 259 Gg C yr⁻¹) and total CO₂ release (from 929 to 1429 Gg C yr⁻¹) increased remarkably during the past 20 years. To neutralize CO₂ release by shellfish and fish, *Pyropia yezoensis* needs the largest culture area ($1.65 \pm 0.15 \times 10^6$ ha) while *Gracilariopsis lemaneiformis* requires the smallest area ($0.11 \pm 0.03 \times 10^6$ ha). In addition, there are enough available areas for culturing *G. lemaneiformis*, *Ulva prolifera* and *Sargassum fusiforme* to neutralize total CO₂ emission in Shandong Province.

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This study elucidates carbon removal, release and sequestration capacities of mariculture and indicates that seaweed culture has a tremendous potential to achieve carbon neutrality target in Shandong.

1. Introduction

Due mainly to human activities, atmospheric CO₂ level has increased from 280 ppm in the pre-industrial period to the current 422 ppm according to NOAA (2024, www.CO2.earth). Continuous CO₂ rise is causing global warming that has been affecting every inhabited region across the globe, leading to a series of environmental, social, and economic problems. Risks to water supply, food security, human health, livelihoods, and economic growth are projected to increase with global warming (IPCC, 2021). For instance, with only 1.5 °C of global warming, twice as many megacities could become heat stressed, exposing >350 million more people to deadly heat by 2050 under a midrange population growth scenario (Matthews et al., 2017). Extreme weather events, like heatwave, are increasing along with global warming. Marine heatwaves can lead to coral bleaching, occurrence of harmful algal blooms and decline of biodiversity (Donovan et al., 2021; Gao et al., 2021b). In addition, continuous dissolution of CO₂ into seawater also results in ocean acidification, which can reduce the level of calcium carbonate saturation and calcification of coral and other calcifying organisms, threatening their survival (Orr et al., 2005). Given the multifaced consequences caused by rising CO₂ and associated climate change, many countries have pledged to be carbon neutral by 2050. As the country accounting for the biggest CO₂ emission, China also promised to achieve carbon neutrality by 2060.

To achieve carbon neutrality target, two aspects must be implemented simultaneously, reducing CO₂ emission and increasing CO₂ sequestration. To date, most studies focus on terrestrial emission and sequestration. Ocean based CO₂ emission assessment and removal approaches receive less attention although ocean covers 71 % of the earth surface. To meet rising population- nutrition- and income-driven demand, aquaculture has been intensively developed and become one of the fastest growing sectors of food production (FAO, 2023). World aquaculture production increased by 180 % between 2000 and 2019, from 43.0 to 120.1 million metric tonnes (Mt) (Verdegem et al., 2023). Furthermore, a near doubling of global fish demand by mid-century is projected assuming continued growth in aquaculture production (Naylor et al., 2021). Despite supplying blue food, environmental performance of aquaculture, particularly its contribution to carbon release and sequestration, is raising concerns (Gephart et al., 2021; Song et al., 2023).

China accounts for around 60 % of aquaculture production in the world and has a long history of culturing seaweeds and aquatic animals (FAO, 2023). In 2022, the production of maricultured seaweeds, fish and shellfish are 2.71, 1.93, 15.70 million tonnes, respectively. Some studies have been conducted to investigate the CO₂ sequestration by mariculture in China. However, different studies show discrepant results. For instance, Ren (2021) showed that cultivated seaweeds could sequester 1.21–2.14 Tg C yr⁻¹ during 2010–2017 while it was 0.96–1.41 Tg C yr⁻¹ during a similar period according to Liu et al. (2022). Gao et al. (2021a) reported a much lower range (0.15–0.30 Tg C yr⁻¹). The reasons for these differences come from different calculation methods or parameters. In terms of shellfish, there are larger divergences among previous studies. Some of them show that shellfish culture is a net carbon sink. For instance, Tang et al. (2011) showed that shellfish mariculture could increase atmospheric CO₂ absorption by seawater. In contrast, Song et al. (2023) reported a carbon release by shellfish culture in China. The main reason that leads to different results could be attributed to the misunderstanding of carbon removal, release and sequestration processes of mariculture. For instance, Ren (2021) deems that carbon removal from seas by seaweeds is the same as carbon sequestration and ignores that the latter refers to long-term carbon storage (>100 years)

(Chuan et al., 2020; Gao et al., 2021a). Ren (2021) considers that carbon removal from seas by shellfish aquaculture is equal to carbon uptake from atmosphere while shellfish calcification can enhance pCO₂ in seawater and lead to CO₂ release into atmosphere rather than absorption (Han et al., 2021; Yang et al., 2021).

Shandong Province is an important aquaculture area with long history, accounting for the second biggest production of maricultured seaweeds and shellfish in China (CFSY, 2023). In addition, Shandong has very strong science and technology support from aquaculture related universities and institutes in China. However, the contribution of cultured seaweeds, shellfish and fish in Shandong to carbon removal, release and sequestration remains unclear. To correctly assess the contribution of mariculture to carbon neutrality, clarifying the processes of carbon source and sink is very essential. In this study, by establishing references and timescale, we distinguished carbon removal, release and sequestration processes of seaweeds, shellfish and fish mariculture for the first time. Furthermore, a comprehensive method that integrates the processes of biological metabolism, chemical equilibrium and human input was used. We also hypothesized that seaweed culture can play a critical role in achieving carbon neutrality in Shandong. Accordingly, the potential of seaweed culture to neutralize CO₂ emission by mariculture and all sections in Shandong was assessed. This study supplies important insights into how mariculture contributes to carbon removal, release and sequestration and thus how mariculture should be adjusted to achieve carbon neutrality.

2. Materials and methods

2.1. Production of mariculture

The data on production and cultivation area of seaweed, shellfish and fish were obtained from the China Fishery Statistical Yearbook for the years of 2004–2023. The census method was used to compile the data of production and cultivation area for seaweeds, shellfish and fish in Shandong Province, in which data were collected from each local farmer and then compiled (Song et al., 2023). Although there may be some errors during the data collection and compilation, census is considered to a reliable statistical method (Baffour et al., 2013). Annual productivity of seaweed, shellfish and fish was calculated by dividing production with cultivation area. Due to the data unavailability of single fish species, only mean productivity of fish was calculated.

2.2. CO₂ removal and sequestration by maricultured seaweeds

Carbon removal represents CO₂ removed by maricultured seaweeds from seawater. It was calculated by the following formula: R_c (tonne carbon yr⁻¹) = $P \times C$, where R_c is carbon removal amount, P is annual production of seaweeds, and C is carbon content of seaweeds. Carbon content of different seaweed species are based on our previous measurements and literature (Table S1). Carbon sequestration is defined as the carbon that can be stored in the ocean for 100 years at least (Gao et al., 2021a). The amount of sequestered carbon (Sc) = $POC_{b1} + POC_{b2} + POC_e + RDOC - R_c$, where POC_{b1} is the POC buried in the algal bed, POC_{b2} is the POC buried in the continental shelf, POC_e is the POC exported to the deep sea, $RDOC$ is the refractory DOC, and R_c released CO₂ during culture. According to the published data (Song et al., 2023), the average ratios of POC_{b1} , POC_{b2} , POC_e , $RDOC$ to harvested POC (POC_h) are 0.031, 0.024, 0.063 and 0.293, respectively. Based on Lian et al. (2023), released CO₂ mainly comes from consumed diesel and rope throughout the life cycle of seaweed culture, which emitted 1.92 and 137.01 kg C for each tonne seaweed (DW), respectively. It is worth

noting that these ratios are generalized while they may be different for different seaweeds and vary with year. However, the specific ratios for each seaweed species and each year are unavailable and we have to use the generalized ones in this study.

2.3. CO₂ removal, release and sequestration by maricultured shellfish

Same as carbon removal by maricultured seaweeds, carbon removal by maricultured shellfish represents CO₂ removed from seawater when harvesting shellfish. It was calculated by the following formula: R_C (tonne carbon yr⁻¹) = $P \times K_1 \times C_{sh} + P \times K_2 \times C_{tissue}$, where P is annual production of shellfish, K_1 is the ratio of dry shell to total shellfish (FW), C_{sh} is carbon content in shell, K_2 is the ratio of dry soft tissue to total shellfish (FW), and C_{tissue} is carbon content in soft tissue. The values of K_1 , C_{sh} , K_2 and C_{tissue} are based on our previous measurements and literature (Tables S2–S5).

Carbon release by shellfish consist of two parts. One comes from culture-related aspects and the other is caused by shellfish *per se*. For the first part, on-farm energy use and aquatic N₂O also contribute to GHG (Greenhouse Gas) emission. According to Xu et al. (2022), GHG emission is 1.2 t CO₂e for each tonnes shellfish. For the second part, calcification and respiration of shellfish result in carbon release from seawater to atmosphere. The formula for carbon release is thus as follows: R_c (tonne carbon yr⁻¹) = $R_{cul} + R_{cal} + R_{res}$, where R_{cul} , R_{cal} and R_{res} represent CO₂ released by culture-related aspects, shellfish calcification and respiration, respectively. According to the reaction equation of calcification, $Ca^{2+} + 2HCO_3^- \leftrightarrow CaCO_3 \downarrow + CO_2 \uparrow + H_2O$, one mole CO₂ is generated when one mole CaCO₃ is synthesized by shellfish calcification. Therefore, the same amount CO₂ as CaCO₃ synthesized in the shells is released into seawater. However, not all generated CO₂ from calcification can be released to atmosphere because calcification alters total alkalinity and carbonate system of seawater. Therefore, the coefficient Φ is introduced, which represents moles of CO₂ released to atmosphere per mole of CaCO₃ formed. $R_{cal} = (P_i \times K_i \times C_i \times \Phi)$, where P_i is production of each shellfish species, K_i is the ratio of dry shell to total weight for each species, C_i is the carbon content in shell for each species, and Φ represents moles of CO₂ released to atmosphere per mole of CaCO₃ formed in shellfish culture areas of Shandong. The coefficient Φ is 0.773 after being calculated according to the carbonate parameters supplied by Jiang et al. (2014), Zhu et al. (2017) and Yang et al. (2021) and the equation supplied by Humphreys et al. (2018).

The CO₂ released by shellfish respiration was calculated with the formula according to Schwinghamer et al. (1986): $\log_{10} R = 0.367 + 0.993 \log_{10} P$, where P and R are productivity (kcal m⁻² yr⁻¹) and respiration rate (kcal m⁻² yr⁻¹) of shellfish, respectively. The coefficient of 18.85 J mg SFDW (shell free dry weight)⁻¹ was used to convert shellfish productivity from weight to energy (Rumohr et al., 1987). In addition, 1 J = 0.239 cal and 1 gC = 11.4 kcal were used for unit conversion (Chauvaud et al., 2003). CO₂ sequestration by shellfish, $S_c = C_{sediment} + C_{shell}$ (Fodrie et al., 2017; Song et al., 2023), where $C_{sediment}$ is carbon sequestered in sediment, C_{shell} is organic carbon in shellfish shells. $C_{sediment} = F \times (1-r)$, where F is carbon in feces of shellfish and r is the remineralization coefficient of feces and sediments. F is calculated based on carbon budget equations of shellfish (Schwinghamer et al., 1986; Song et al., 2023). The value of r is 0.87 based on Gao et al. (2008). Organic carbon in shells of shellfish can be sequestered in the long-term and was calculated according the following formula: $C_{shell} = W_{shell} \times f_1 \times f_2$, where C_{shell} is organic carbon in shells, W_{shell} is weight of shells, f_1 is the fraction of organic matter in shells, and f_2 is the fraction of carbon in organic matter. Based on Fodrie et al. (2017), the numbers of 0.0136 and 0.36 were used for f_1 and f_2 , respectively.

2.4. CO₂ removal and release by maricultured fish

Fish mariculture in China relies on wild (fresh trash fish) or artificial feeds. Some of feeds are not eaten by fish but are released into seawater.

Therefore, carbon in fish comes from feeds that are input from lands. Fish mariculture does not remove carbon from seawater but increase carbon in seawater by uneaten feeds. Therefore, carbon removal by fish mariculture is a negative value. In the meantime, most of carbon in uneaten feeds are released to atmosphere via microbial mineralization, with the remaining in sediments. In addition, like shellfish culture, culture-related aspects of fish also contribute to GHG emission. According to Xu et al. (2022), GHG emission caused by feed production, on-farm energy use and aquatic N₂O is 2.4 t CO₂e for each tonne fish. Removed and released carbon by fish mariculture were calculated by the following formulas: $C_{removal} = -P \times F_c \times C_{feed} \times (1-R)$, $C_{release} = P \times F_c \times C_{feed} \times (1-R) \times r + C_{cul}$, where $C_{removal}$ is removed C, P = fish production, F_c = feed coefficient, the feed consumption per unit weight increase of fish, R = retention rate of feed C in fish, $C_{release}$ = released C, C_{feed} = content of C in feeds, r = the remineralization coefficient of uneaten feeds, and C_{cul} = culture-related carbon release. Half of the fish increase in weight is from artificial feeds and the other half is from wild feeds (fresh trash fish) based on recent investigations (Wang, 2016; Li et al., 2021). The contents of C in artificial and wild feeds are 39.64 % and 13.67 %, respectively (Xiong et al., 2023). The retention rate of feed carbon in fish is set at 30 % based on previous studies (Xu et al., 2007; Lazzari and Baldissarotto, 2008; Herath and Satoh, 2015). The feed coefficient is 1.52 and 6.49 for artificial and wild feeds, respectively (Gao et al., 2021a). The value of 0.87 was used for r according to Hao et al. (2008).

2.5. Total CO₂ removal and release by mariculture

Total CO₂ removal by mariculture = $C_{removal1} + C_{removal2} + C_{removal3}$, where $C_{removal1}$, $C_{removal2}$, $C_{removal3}$ are CO₂ removed by seaweeds, shellfish and fish, respectively. Total CO₂ release by mariculture = $C_{release1} + C_{release2} - C_{sequestration}$, where $C_{release1}$, $C_{release2}$, $C_{sequestration}$ are CO₂ released by shellfish and fish and sequestered by seaweeds, respectively.

2.6. CO₂ removal, release and sequestration intensity of mariculture

CO₂ removal, release and sequestration intensity of seaweeds, shellfish and fish was calculated by annual CO₂ removal, release and sequestration amount divided by culture area. Due to the unavailability of culture areas for single fish species, only mean CO₂ removal, release intensities for fish were calculated.

2.7. Required area to achieve carbon neutrality

The required area (A_{Ri}) for each seaweed species to achieve carbon neutrality was calculated according to the formula of $A_{Ri} = C_T / C_{si}$. C_T is the total CO₂ amount that is needed to be sequestered annually by seaweed to achieve mariculture carbon neutrality in Shandong or Shandong's carbon neutrality. Mariculture carbon neutrality in Shandong is based on the total CO₂ release by mariculture in Section 2.6. China's 2060 carbon neutrality goal will require up to 2.5 Gt CO₂ to be sequestered annually (Fuhrman et al., 2020). CO₂ emission of Shandong accounted for 9.14 % of the national emission. Therefore, Shandong needs to sequester 0.23 Gt CO₂ annually considering its current CO₂ emission. C_{si} (kg C m⁻² yr⁻¹) is the carbon sequestration intensity for each seaweed species. To represent the latest carbon sequestration capacity and minimize the impact of climate change, the mean values of 2018–2022 were used.

2.8. Data analysis

Data for CO₂ removal and sequestration by maricultured seaweeds, CO₂ removal and release by maricultured shellfish and fish, and required seaweed area to achieve carbon neutrality were subjected to error propagation analysis and expressed as means ± SE. The initial SE came

from carbon content in seaweed thalli, shell and soft tissue of shellfish, and fish feeds, etc.

3. Results

Production of cultured seaweeds had a small fluctuation of $323\text{--}336 \times 10^3 \text{ t DW yr}^{-1}$ during 2003–2013 (Fig. 1a). It increased sharply to $663 \times 10^3 \text{ t DW yr}^{-1}$ in 2014, maintained stable in the following seven years but decreased to $549 \times 10^3 \text{ t DW yr}^{-1}$ in 2022. *Saccharina japonica* was the dominant cultured species during 2003–2021, accounting for 73–94 % of the total seaweed production. In 2022, it was replaced by *Gracilariopsis lemaneiformis* due to a rapid decrease. From 2005 to 2012, *Undaria pinnatifida* contributed the second biggest production but was replaced by *G. lemaneiformis* after 2012.

Mariculture area of seaweeds increased from $19 \times 10^3 \text{ ha}$ in 2003 to $23 \times 10^3 \text{ ha}$ in 2007, then decreased to $16 \times 10^3 \text{ ha}$ in 2009 (Fig. 1b). It changed in a narrow range of $17\text{--}20 \times 10^3 \text{ ha}$ during the years of 2010 to 2016, and then increased to $24 \times 10^3 \text{ ha}$ in 2017. Afterwards, it decreased to $20 \times 10^3 \text{ ha}$ in 2019 and maintaining stable until 2022. Most culture areas (72–94 %) were occupied by *S. japonica* during the past 20 years and *Pyropia yezoensis* ranked the second in most years. The mean productivity of cultured seaweeds showed a small fluctuation of $1.26\text{--}1.73 \text{ kg DW m}^{-2} \text{ yr}^{-1}$ during 2003 and 2012, and then increased to $3.43 \text{ kg DW m}^{-2} \text{ yr}^{-1}$ in 2014, followed by a slowly decrease until 2017 (Fig. 1c). Afterwards, it was relatively stable. In most years, *G. lemaneiformis* showed the biggest productivity among the seven seaweeds, followed by *U. pinnatifida*.

The production of shellfish steadily increased from $2.55 \times 10^6 \text{ t FW yr}^{-1}$ in 2003 to $3.98 \times 10^6 \text{ t FW yr}^{-1}$ in 2017 (Fig. 2a). It decreased to $3.63 \times 10^6 \text{ t FW yr}^{-1}$ in 2019 and then slowly increased to $4.10 \times 10^6 \text{ t FW yr}^{-1}$ in 2022. Clam contributed the biggest production, followed by oyster and scallop. The mariculture area of shellfish was $272 \times 10^3 \text{ ha}$ in

2003 and decreased to $178 \times 10^3 \text{ ha}$ in the following year (Fig. 2b). Afterwards, it showed a rising trend and achieved the largest area of $377 \times 10^3 \text{ ha}$ in 2017. The mariculture area fluctuated in a range of $317\text{--}360 \times 10^3 \text{ ha}$ during 2018–2022. The mean productivity of shellfish was $0.94 \text{ kg FW m}^{-2} \text{ yr}^{-1}$ in 2003 and increased to $1.47 \text{ kg FW m}^{-2} \text{ yr}^{-1}$ in 2004 (Fig. 2c). It fluctuated in a narrow range of $1.01\text{--}1.21 \text{ kg FW m}^{-2} \text{ yr}^{-1}$ during 2005–2022. Oyster had the highest productivity in most years, but was replaced by pen shell in 2021 and by clam in 2022.

From 2003 to 2011, fish production showed an increasing trend (Fig. 3a). After achieving the peak ($115 \times 10^3 \text{ t FW yr}^{-1}$) in 2011, it maintained a higher production range of $105\text{--}109 \times 10^3 \text{ t FW yr}^{-1}$ during 2012–2015. Then it decreased with time and had the lowest production ($47 \times 10^3 \text{ t FW yr}^{-1}$) in 2022. Lefteye flounder dominated the fish production in all years, accounting for 49–72 % of the total production. Sea bass was the second biggest species, accounting for 15–33 % of the total production. Mariculture area of fish was $5.60 \times 10^3 \text{ ha}$ in 2003 and increased to $9.11 \times 10^3 \text{ ha}$ in 2004 (Fig. 3b). After a stable period during 2004–2008, it reached the peak of $17.65 \times 10^3 \text{ ha}$ in 2009. Afterwards, it showed a decrease trend until 2019. Therefore, fish production and area shrank in recent years, which is different from seaweed and shellfish. Mean productivity of maricultured fish went up and down during the past 20 years (Fig. 3c). During 2003–2007, it showed a rising trend but then decreased to $0.72 \text{ kg FW m}^{-2} \text{ yr}^{-1}$ in 2009. Afterwards, it increased with year until 2012. After a stable period during 2012–2017, it increased to $2.36 \text{ kg FW m}^{-2} \text{ yr}^{-1}$ in 2019. During the last four years, it showed a decrease trend.

There was a small fluctuation for CO_2 removal ($68\text{--}99 \text{ Gg C yr}^{-1}$) by maricultured seaweeds during 2003–2013 (Fig. 4a). It increased to 190 Gg C yr^{-1} in 2014 and then slowly decreased until 2019 (178 Gg C yr^{-1}). It then increased to 189 Gg C yr^{-1} in 2021 and decreased to 173 Gg C yr^{-1} in 2022. *S. japonica* contributed the most CO_2 removal during 2003–2021, accounting for 69–93 % of the total CO_2 removal. In 2022, it

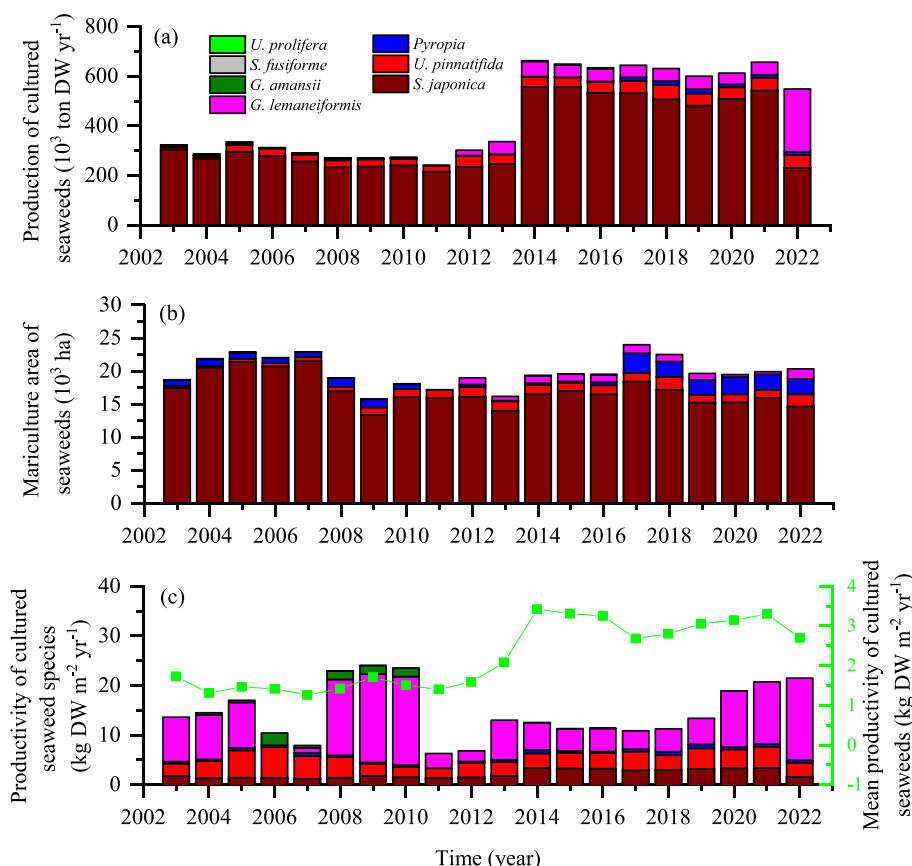


Fig. 1. Seaweed production (a), area (b) and productivity (c) in Shandong Province of China during 2003–2022.

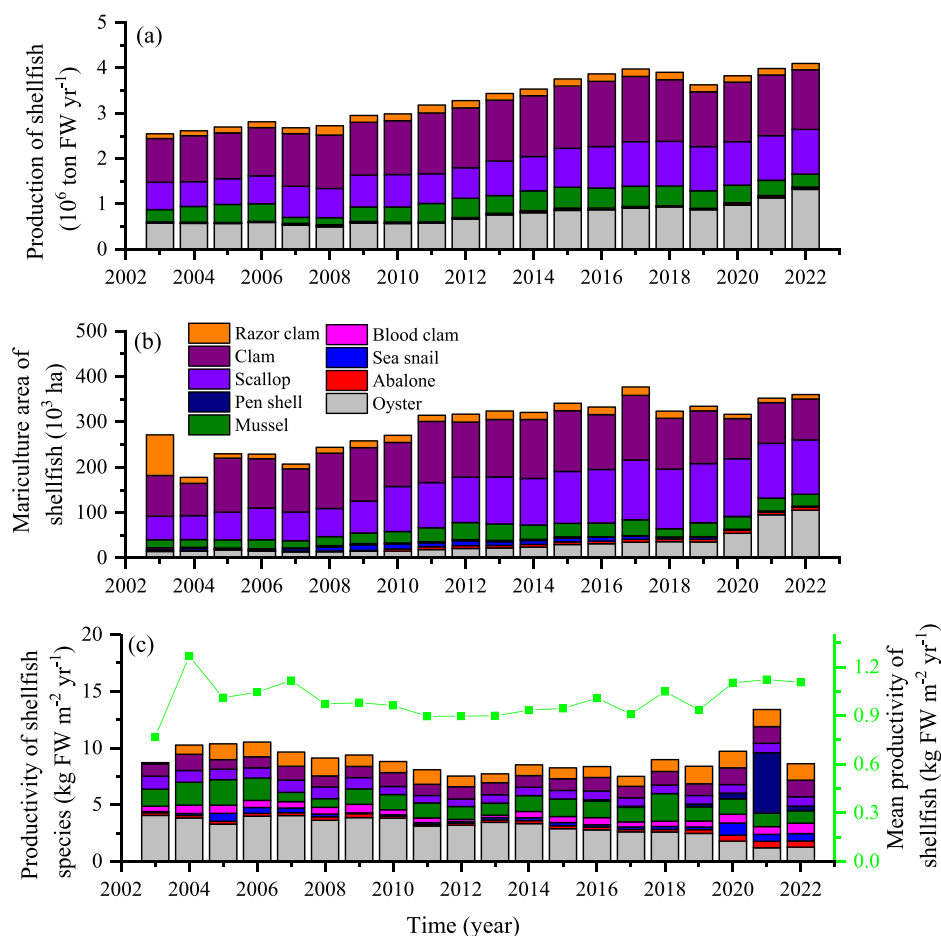


Fig. 2. Shellfish production (a), area (b) and productivity (c) in Shandong Province of China during 2003–2022.

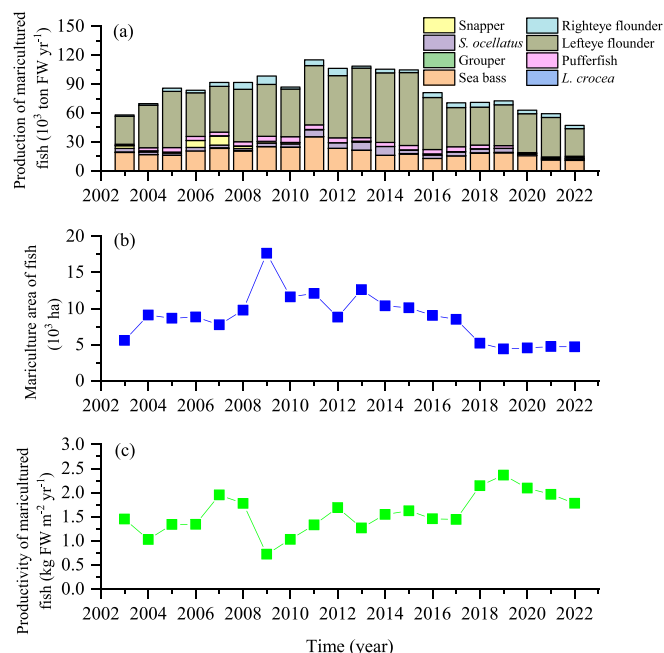


Fig. 3. Fish production (a), area (b) and productivity (c) in Shandong Province of China during 2003–2022. Due to data unavailability of culture area for specific fish species, only total culture area (b) and mean productivity (c) are represented.

was replaced by *G. lemaneiformis* that contributed 50 % of the total CO_2 removal. The trend of CO_2 sequestration by maricultured seaweeds was the same as CO_2 removal but the amount was lower than CO_2 removal (Fig. 4b). It ranged from 24 to 67 Gg C yr^{-1} during the past 20 years.

CO_2 removal by shellfish increased from 231 Gg C yr^{-1} in 2003 to 257 Gg C yr^{-1} in 2006, and then decreased to 238 Gg C yr^{-1} in 2007 (Fig. 5a). It then showed a rising trend until it reached the peak of 362 Gg C yr^{-1} in 2017. Afterwards, it decreased to 331 Gg C yr^{-1} in 2019 and then increased with year until 2022. Clam contributed the most CO_2 removal until 2020, accounting for 29–39 % of the total CO_2 removal. In 2021 and 2022, oyster replaced clam, being the biggest contributor. The pattern of CO_2 release by shellfish was similar to CO_2 removal (Fig. 5b) but it had higher values due mainly to calculation and respiration. It ranged from 897 to 1438 Gg C yr^{-1} . Clam also contributed the most CO_2 release in all years except for 2022.

In contrary to seaweed and shellfish, fish culture did not remove CO_2 from seawater but added CO_2 due to the use of feeds. CO_2 addition increased from 30 Gg C yr^{-1} in 2003 to 45 Gg C yr^{-1} in 2005 and then fluctuated between 44 and 48 Gg C yr^{-1} during 2005–2010 (Fig. 6a). It increased to the peak of 60 Gg C yr^{-1} in 2021 and then showed a decrease trend, reaching the minimum of 25 Gg C yr^{-1} in 2022. The pattern of CO_2 release by maricultured fish was the same as CO_2 addition, in a range of 52–128 Gg C yr^{-1} (Fig. 6b).

After summing up the CO_2 removal by seaweed and shellfish and CO_2 addition by fish, total CO_2 removal by mariculture in Shandong was presented (Fig. 7a). The total CO_2 removal was 110 Gg C yr^{-1} in 2003 and it then showed a decrease trend, reaching the minimum of 57 Gg C yr^{-1} in 2011. It rapidly increased to 203 Gg C yr^{-1} in 2014 and then slowly increased with year, achieving the peak of 254 Gg C yr^{-1} in 2022.

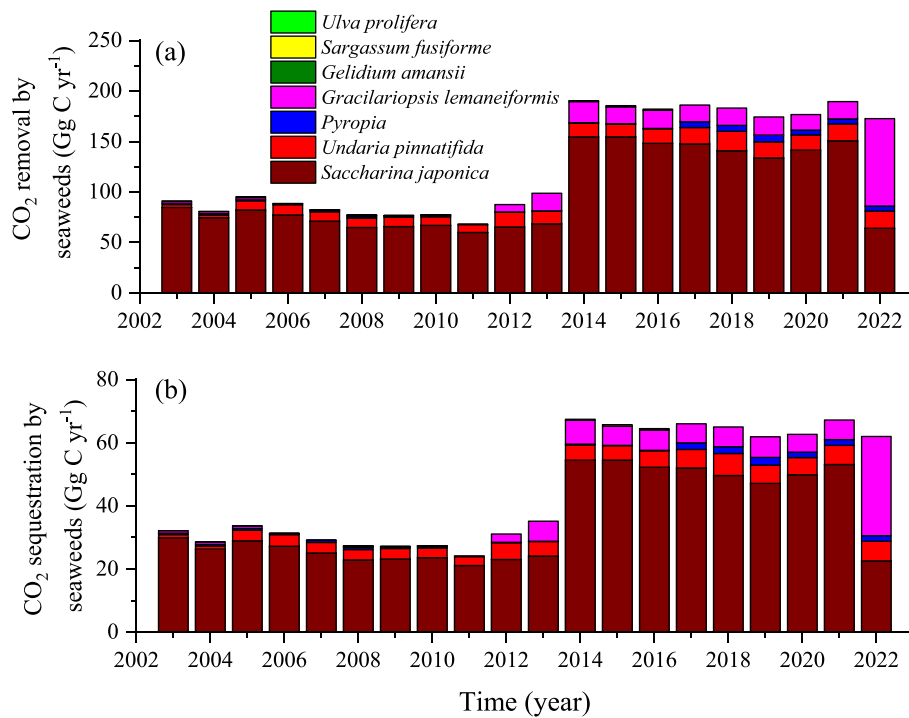


Fig. 4. CO₂ removal (a) and sequestration (b) by maricultured seaweed in Shandong Province of China during 2003–2022. The standard error of data is shown in Table S6.

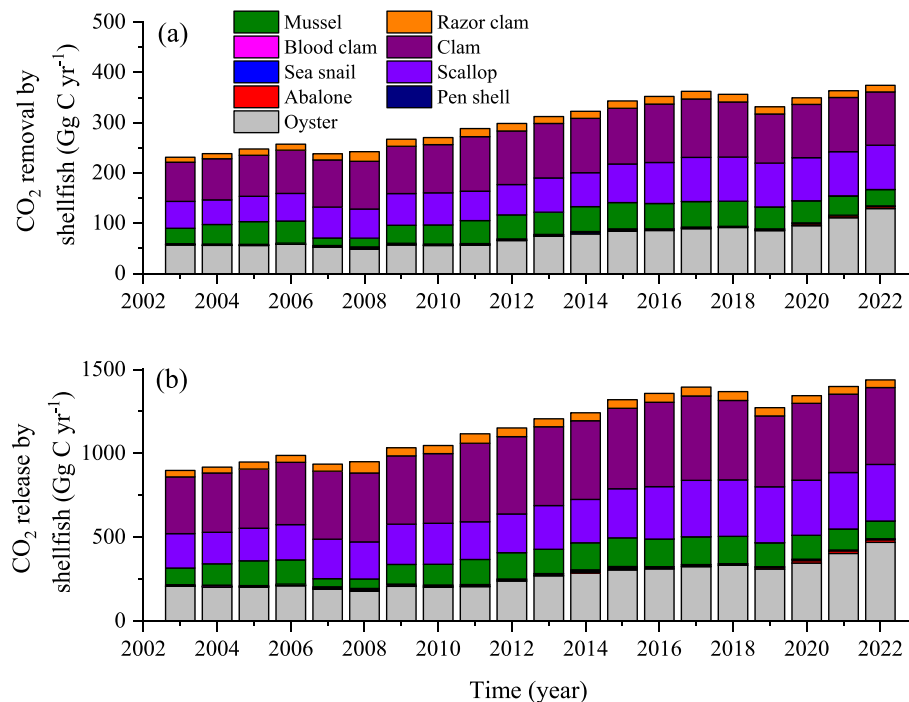


Fig. 5. CO₂ removal (a) and release (b) by shellfish in Shandong Province of China during 2003–2022. The standard error of data is shown in Table S7.

Total CO₂ release by mariculture increased from 930 ± 17 Gg C yr⁻¹ in 2003 to 1408 ± 24 Gg C yr⁻¹ in 2017 (Fig. 7b) and then decreased to 1291 ± 22 Gg C yr⁻¹ in 2019. Afterwards, it showed a rising trend and reached 1249 ± 26 Gg C yr⁻¹ in 2022. To sum up, total CO₂ removal by mariculture experienced a sharp increase in 2014 while total CO₂ release showed a gradual increase during the past 20 years.

Based on the data in the past 20 years, *G. lemaneiformis* had the highest CO₂ removal intensity (2.89 ± 0.42 kg C m⁻² yr⁻¹), followed by

S. fusiforme (1.65 ± 0.15 kg C m⁻² yr⁻¹) and *U. prolifera* (1.64 ± 0.00 kg C m⁻² yr⁻¹) (Fig. 8a). *P. yezoensis* had the lowest CO₂ removal intensity (0.17 ± 0.01 kg C m⁻² yr⁻¹). The order of CO₂ sequestration intensity was similar to the CO₂ removal: *G. lemaneiformis* (1.00 ± 0.16 kg C m⁻² yr⁻¹) > *U. prolifera* (0.61 ± 0.00 kg C m⁻² yr⁻¹) > *S. fusiforme* (0.59 ± 0.05 kg C m⁻² yr⁻¹) > *U. pinnatifida* (0.42 ± 0.03 kg C m⁻² yr⁻¹) > *S. japonica* (0.21 ± 0.02 kg C m⁻² yr⁻¹) > *Gelidium amansii* (0.20 ± 0.03 kg C m⁻² yr⁻¹) > *P. yezoensis* (0.061 ± 0.006 kg C m⁻² yr⁻¹) (Fig. 8b). In

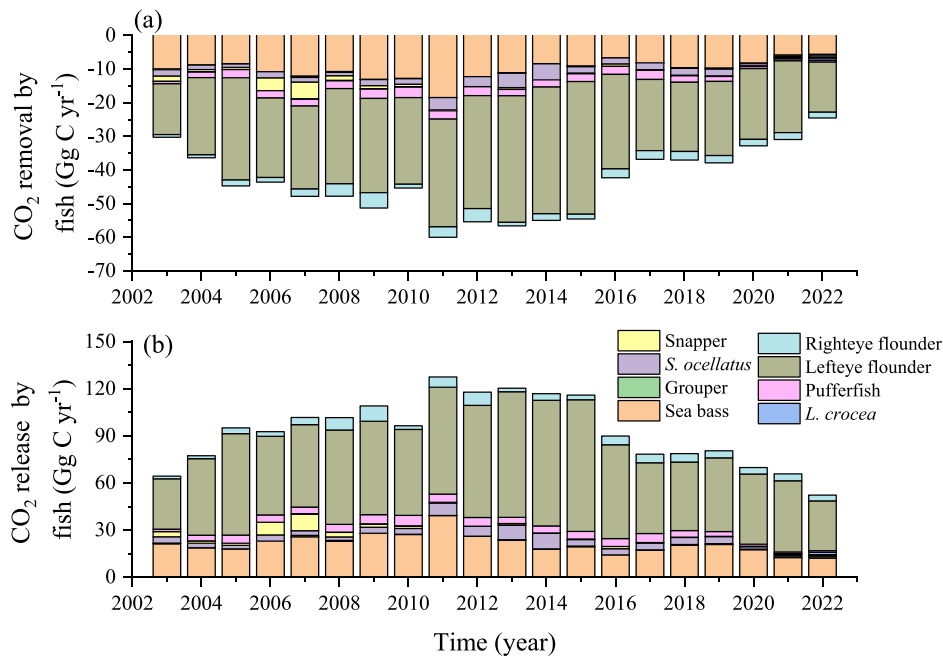


Fig. 6. CO₂ removal (a) and release (b) by fish in Shandong Province of China during 2003–2022. The standard error of data is shown in Table S8.

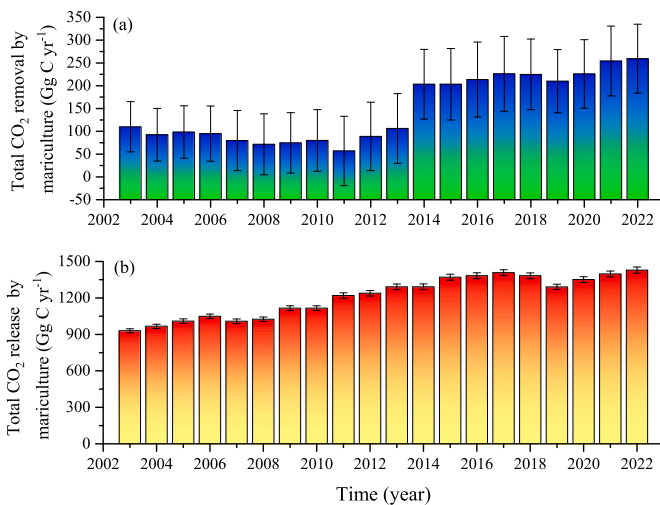


Fig. 7. Total CO₂ removal (a) and release (b) by mariculture in Shandong Province of China during 2003–2022. Error bar means standard error.

terms of shellfish, oyster had the highest CO₂ removal intensity ($0.30 \pm 0.02 \text{ kg C m}^{-2} \text{ yr}^{-1}$), followed by mussel ($0.16 \pm 0.01 \text{ kg C m}^{-2} \text{ yr}^{-1}$) and razor clam ($0.10 \pm 0.01 \text{ kg C m}^{-2} \text{ yr}^{-1}$) (Fig. 8c). Oyster also had the highest CO₂ release intensity ($1.09 \pm 0.07 \text{ kg C m}^{-2} \text{ yr}^{-1}$), followed by mussel ($0.53 \pm 0.04 \text{ kg C m}^{-2} \text{ yr}^{-1}$) and clam ($0.40 \pm 0.02 \text{ kg C m}^{-2} \text{ yr}^{-1}$) (Fig. 8d). Compared to shellfish, fish had much higher CO₂ release intensity ($1.74 \pm 0.10 \text{ kg C m}^{-2} \text{ yr}^{-1}$).

Required areas of seaweed culture to neutralize CO₂ release by fish and shellfish were calculated (Fig. 9a). The largest area ($1.65 \pm 0.15 \times 10^6 \text{ ha}$) comes from *P. yezoensis*, followed by *Gelidium amansii* ($0.55 \pm 0.11 \times 10^6 \text{ ha}$) and *S. japonica* ($0.49 \pm 0.06 \times 10^6 \text{ ha}$). Culturing *G. lemaneiformis* needs the smallest area ($0.11 \pm 0.03 \times 10^6 \text{ ha}$). All areas are less than the available marine area ($14.06 \times 10^6 \text{ ha}$) in Shandong. To meet the target of carbon neutrality by 2060, the total CO₂ emission amount of Shandong Province will be $62 \times 10^6 \text{ t C}$. To neutralize this amount of CO₂ emission, required areas of seaweeds culture were calculated (Fig. 9b). The required areas for *G. lemaneiformis*

($4.96 \pm 1.11 \times 10^6 \text{ ha}$), *S. fusiforme* ($10.51 \pm 0.93 \times 10^6 \text{ ha}$) and *U. prolifera* ($10.19 \pm 0.00 \times 10^6 \text{ ha}$) are below the available area, indicating that there are enough areas for culturing these seaweed species to achieve carbon neutrality of Shandong.

4. Discussion

4.1. Changes of mariculture production

There was a rapid increase in seaweed production in 2014, which was mainly caused by *S. japonica* culture. The use of new strain with higher productivity contributed to the large increase since culture area did not significantly vary (Zhang et al., 2018). The decrease of seaweed production in 2022 was also related to *S. japonica* culture. From November 2021 to April 2022, *S. japonica* cultured in a main farm of Shandong Province, Rongcheng, suffered from an unprecedented loss, which may be caused by high water transparency and exhausted nutrients resulting from a red tide (Li et al., 2023a, 2023b). This natural disaster covered an area of 9,300 ha and led to a direct economic loss of $20 \times 10^8 \text{ yuan}$ (Li et al., 2023a, 2023b). Compared to other seaweeds, *G. lemaneiformis* had higher productivity. This can be attributed to its longer culture period. *G. lemaneiformis* can be year around cultured in China while other seaweeds cannot make it (Gao et al., 2021a). For instance, *Pyropia* can only be cultured for 3–5 months in a year, which resulted in lower productivity. Therefore, new strain with longer culture period will be a direct for improving seaweed yield.

Different from the rapid increase of seaweed production in 2014, the production of shellfish slowly increased with year. This increase was caused by culture area rather than productivity since mean shellfish productivity maintained a stable trend. A notable phenomenon is that oyster productivity decreased with time although culture area increased. This is related to transformation of oyster industry in Shandong, which aims to produce oyster with high quality by reducing culture density. Although reduced culture density led to lower productivity, the high quality brings in high market value (Tan et al., 2019).

The fish production has decreased since 2015, which was mainly caused by the reduced culture of lefteye flounder. The rumor that eating lefty flounder could cause decreases led to the shrinking market demand (Guan et al., 2020). Although culture area decreased rapidly, the mean

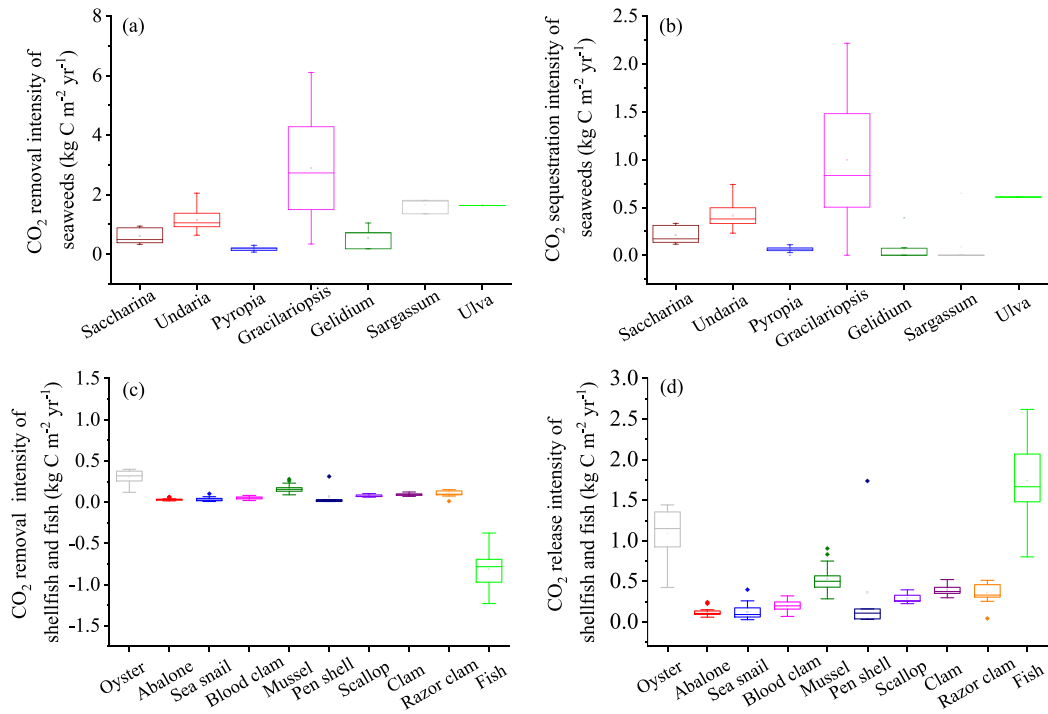


Fig. 8. CO₂ removal, sequestration and release intensity of seaweeds (a and b), shellfish (c and d) and fish (c and d) in Shandong Province of China during 2003–2022.

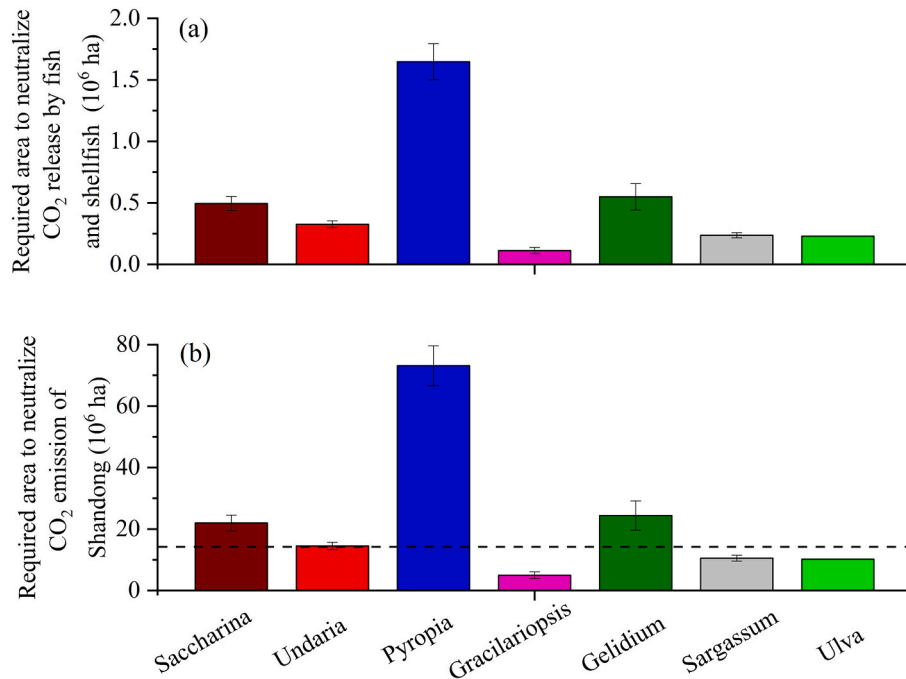


Fig. 9. Required area of culturing seaweeds to neutralize CO₂ release by fish, shellfish (a) and total CO₂ emission in Shandong Province in 2021 (b). The dashed lines represent available area (14.06×10^6 ha) for seaweed cultivation in China according to Marine Functional Zoning in Shandong Province (2011–2020) issued by People's Government of Shandong Province (MFZSD, 2012).

productivity of fish has showed in rising trend since 2009, which can be attributed to the improved culture techniques (Guan et al., 2020).

4.2. CO₂ removal, release and sequestration by mariculture

The changes of CO₂ removal and sequestration by seaweeds with year were associated with the production of seaweeds because carbon

content among culture seaweed species did not have large differences (Gao et al., 2021a). Shen et al. (2024) reported that CO₂ removal and sequestration rates of cultured seaweeds were 130–174 Gg C yr⁻¹ and 131–175 Gg C yr⁻¹ in Shandong during 2010–2020, respectively. The CO₂ removal rate is comparable to our result of 77–177 Gg C yr⁻¹. However, the CO₂ sequestration rate is much higher than our result (28–78 Gg C yr⁻¹). This is because carbon in harvested seaweeds is

deemed as carbon sequestration in their study. Carbon in harvested seaweeds will be transformed into CO₂ and released to atmosphere when seaweeds are consumed by human as food or animals as feed. Therefore, they cannot be defined as carbon sequestration that represents long-term carbon storage (> 100 years). In future, negative emission technology of BECCS (bioenergy with carbon capture and storage) can be deployed with harvested seaweeds and carbon in harvested seaweeds can be sequestered in this case (Hanssen et al., 2020; Gao et al., 2022).

CO₂ removal rate by shellfish ranged from 282 to 369 Gg C yr⁻¹ in Shen et al. (2024), which is comparable to our result of 270–362 Gg C yr⁻¹. However, shellfish can sequester 175–233 Gg C yr⁻¹ in Shandong Province in their study. Calcification of shellfish was deemed as carbon sink in Shen et al. (2024). However, one mole CO₂ is generated when one mole CaCO₃ is synthesized during shellfish calcification according to the reaction equation in Section 2.3. The carbon in CaCO₃ comes from HCO₃⁻ in seawater rather than CO₂ in atmosphere. The carbon transfer from HCO₃⁻ to CaCO₃ does not reduce pCO₂ in seawater but increases it, leading to the CO₂ release from seawater to atmosphere. The increased pCO₂ was reported in many shellfish culture areas (Han et al., 2021; Yang et al., 2021). Therefore, shellfish calcification is a carbon source rather than carbon sink process because the criteria for carbon source and sink is whether CO₂ is released to atmosphere or sequestered from atmosphere.

Previous study shows that fish culture can also remove CO₂ from seawater (Li et al., 2023a, 2023b). However, it did not consider carbon input from feeds. In fact, only a small fraction of feeds is used for fish growth. A large fraction is released into seawater and transformed into CO₂ by bacteria (Herath and Satoh, 2015). Fish culture areas usually have higher pCO₂ compared to atmospheric levels (Han et al., 2021). Therefore, fish culture is a carbon source rather than carbon sink. It is worth noting that not all uneaten feeds are mineralized to CO₂ and a small fraction of them can sink down to the bottom of seas, which can be sequestered for a long time (Adhikari et al., 2012). Therefore, the amount of CO₂ released by fish culture is lower than that of CO₂ added through feeds. However, the sequestered carbon come from land-based feeds and thus fish mariculture does not include real carbon sink process.

After integrating the CO₂ removal and addition by seaweeds, shellfish and fish, total CO₂ removal increased with time after 2011, which could be attributed to increased shellfish production and decreased fish production. Although this carbon was removed from seawater, they can be released to atmosphere after being consumed by human except shell from shellfish. Furthermore, removing this amount of carbon from seawater does not mean that ocean can absorb the same amount of CO₂ from atmosphere. As show in Fig. 7b, mariculture in Shandong led to net CO₂ release from seawater to atmosphere. Although photosynthesis of seaweed can reduce pCO₂ in seawater, respiration of shellfish and fish can increase pCO₂ in seawater, leading to the release of CO₂ from seawater to atmosphere. Furthermore, calcification of shellfish can reduce total alkalinity of seawater and thus the buffer capacity of seawater (Humphreys et al., 2018). This has been proved by in situ monitoring (Yang et al., 2021). The reduced buffer capacity can lead to more severe ocean acidification when atmospheric CO₂ dissolved in seawater.

4.3. Structure adjustment and mode improvement to achieve carbon neutrality

To achieve carbon neutrality, a lot of approaches have been proposed, including land-based and ocean-based techniques. Among marine based CO₂ removal approaches, mariculture is considered as a feasible approach because it can conduct carbon sequestration and produce seafood at the same time (Jiao, 2021). In addition, it does not need additional legal permission to conduct because it is a traditional industry with a long history in China and other Asian countries. Meanwhile, it is necessary to identify carbon removal, release and

sequestration processes of mariculture before being conducted in a large scale. The findings in the present study demonstrate that only seaweed culture can sequester carbon from atmosphere while shellfish and fish culture incur CO₂ release from seawater to atmosphere. In the case of Shandong Province, the sequestered CO₂ by seaweed is much lower than released CO₂ by shellfish and fish, leading to net carbon release. Therefore, expansion of seaweed culture is needed to neutralize the current CO₂ release amount by shellfish and fish. After the assessment based on carbon sequestration intensity, there are enough areas available for all seaweed species. Therefore, there is flexibility for culturing different seaweed species. On the hand other, to neutralize total CO₂ emission from all sectors in Shandong in 2021 (Guan et al., 2021), not all seaweed species can meet the target. For instance, culturing *P. yezoensis*, *S. japonica*, or *G. amansii* cannot sequester total CO₂ release in Shandong due to their lower CO₂ sequestration intensities. Previous studies also show the potential of seaweeds in achieving carbon neutrality but do not consider carbon footprint during seaweed culture (Gao et al., 2021a; Song et al., 2023). The present study integrates carbon release from consumed diesel and rope throughout the life cycle of seaweed culture with carbon sequestration by seaweeds. Therefore, the assessment of seaweed potential in achieving carbon neutrality is more accurate. It is worth noting that this assessment does not consider the effects of extreme weather and the strain improvement of cultivated seaweeds. Extreme weather may affect carbon sequestration capacity while strain improvement through crossbreeding and molecular editing can make them more robust to environmental stress (Jiang et al., 2024).

To accurately assess CO₂ absorption from and release into atmosphere, the efficiency of air-sea equilibration (i.e., the relative tendency of surface waters to reach equilibrium with the atmosphere) of carbon dioxide should be considered (Jones et al., 2014; Khatiwala et al., 2019). Equilibration efficiency can be defined as the ratio of surface residence time to air-sea equilibration time. If surface residence time is longer than air-sea equilibration time, seawater in the mixed layer will likely have sufficient time to come to equilibrium with the overlying atmosphere before returning to the ocean interior. If surface residence time is shorter than air-sea equilibration time, relaxation processes are relatively ineffective at getting rid of carbon anomalies, tending to favor air-sea disequilibrium. At present, the efficiency of air-sea equilibration in mariculture areas of Shandong Province is unknown and need to be explored in future studies.

It seems that seaweed culture should be expanded while shellfish and fish culture should be constrained to achieve the carbon neutrality within mariculture. To neutralize total CO₂ release in Shandong, very large areas are needed for seaweed culture. A three-dimensional mode is proposed here, with seaweeds up and shellfish and fish down. This three-dimensional mode can save culture space largely. In addition, released CO₂ by shellfish and fish could be efficiently absorbed by seaweeds in this mode and *in situ* assessment is needed to verify the effectiveness of this mode in carbon neutrality and biomass yield. It is worth noting that the assessment methods used in this study can be used in other mariculture areas in China or abroad. The mariculture structure is different in coastal provinces in China (Xiong et al., 2023), and thus different adjusting strategies must be conducted to achieve carbon neutrality of mariculture.

5. Conclusions

Due to the emergency of carbon emission reduction, the contribution of mariculture to carbon neutrality is gaining increased attention. However, the specific processes of carbon source and sink of mariculture remains unknown. This is the first study to investigate CO₂ removal, release and sequestration by maricultured seaweeds, shellfish and fish in Shandong Province. Furthermore, the potential of seaweed culture to achieve carbon neutrality in Shandong Province was assessed. The results show that CO₂ release from seawater to atmosphere caused by mariculture increased from 929 Gg C yr⁻¹ in 2003 to 1429 Gg C yr⁻¹ in

2022 although CO₂ removal also increased from 110 to 259 Gg C yr⁻¹. There are enough available areas for culturing any seaweed species to neutralize CO₂ release by shellfish and fish. However, only *G. lemaneiformis*, *U. prolifera* and *S. fusiforme* have the potential to neutralize total CO₂ emission in Shandong, with *G. lemaneiformis* having the minimum required area. Strain screening and improvement are needed to enhance carbon sequestration intensity. In addition, three-dimensional culture with seaweed up and shellfish or fish down is recommended for efficiently absorbing CO₂ released by shellfish and fish.

CRediT authorship contribution statement

Wei Li: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation. **Xu Li:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Chi Song:** Writing – review & editing, Visualization, Methodology, Investigation. **Guang Gao:** Writing – review & editing, Writing – original draft, Validation, Supervision, Methodology, Funding acquisition, Formal analysis, Conceptualization.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2024.172272>.

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