



Mariculture structure adjustment to achieve China's carbon neutrality and mitigate climate change



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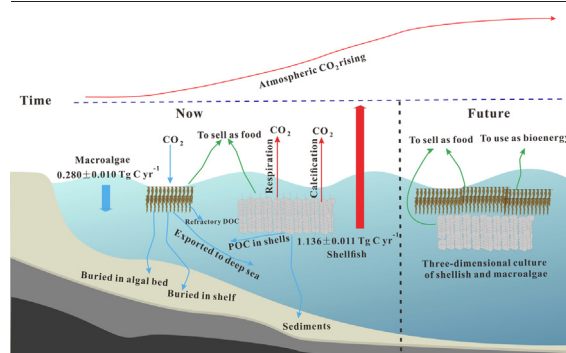
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HIGHLIGHTS

- Cultured shellfish in China release $0.741 \pm 0.008 \text{ Tg C yr}^{-1}$ through calcification.
- Cultured shellfish in China release $0.580 \pm 0.004 \text{ Tg C yr}^{-1}$ by respiration.
- Cultured shellfish in China sequester $0.184 \pm 0.001 \text{ Tg C yr}^{-1}$ in sediments and shells.
- Cultured macroalgae in China sequester $0.280 \pm 0.010 \text{ Tg C yr}^{-1}$ in seawater.
- It is feasible to achieve China's carbon neutrality by expanding macroalgae culture.

GRAPHICAL ABSTRACT



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ABSTRACT

China is responsible for the biggest shellfish and macroalgae production in the world. In this study, comprehensive methods were used to assess the CO_2 release and sequestration by maricultured shellfish and macroalgae in China. Through considering CaCO_3 production and CO_2 release coefficient (Φ , moles of CO_2 released per mole of CaCO_3 formed) in different waters, we find that cultured shellfish released $0.741 \pm 0.008 \text{ Tg C yr}^{-1}$ through calcification based on the data of 2016–2020. In addition to calcification, maricultured shellfish released $0.580 \pm 0.004 \text{ Tg C yr}^{-1}$ by respiration. Meanwhile, shellfish sequestered 0.145 ± 0.001 and $0.0387 \pm 0.0004 \text{ Tg C yr}^{-1}$ organic carbon in sediments and shells, respectively. Therefore, the net released CO_2 by maricultured shellfish was $1.136 \pm 0.011 \text{ Tg C yr}^{-1}$, which is about four times higher than that maricultured macroalgae could sequester ($0.280 \pm 0.010 \text{ Tg C yr}^{-1}$). To achieve carbon neutrality within the mariculture system, shellfish culture may need to be restricted and meanwhile the expansion of macroalgae cultivation should be carried out. The mean carbon sequestration rate of seven kinds of macroalgae was $174 \pm 6 \text{ g C m}^{-2} \text{ yr}^{-1}$ while some cultivated macroalgae had higher CO_2 sequestration rates, e.g. $356 \pm 24 \text{ g C m}^{-2} \text{ yr}^{-1}$ for *Gracilaria lemaneiformis* and $331 \pm 17 \text{ g C m}^{-2} \text{ yr}^{-1}$ for *Undaria pinnatifida*. In scenario 0.5 (CCUS (Carbon Capture, Utilization and Storage) sequesters 0.5 Gt CO_2 per year), using macroalgae culture cannot achieve China's carbon neutrality by 2060 but in scenarios 1.0 and 1.5 (CCUS sequesters 1.0 and 1.5 Gt CO_2 per year, respectively) it is feasible to achieve carbon neutrality using some macroalgae species with high carbon sequestration rates. This study provides important insights into how to develop mariculture in the context of carbon-neutrality and climate change mitigation.

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1. Introduction

Due to high nutritive content and delicious taste, there is a rising demand for seafood. To meet this demand, aquaculture has been intensively developed and become one of the fastest growing sectors of food production (FAO, 2020). Global seafood consumption, including finfish and shellfish, was growing at a mean annual rate of 3.1 % between 1961 and 2017, which is faster than other livestock and animal production sectors at 2.1 % per year (FAO, 2020). Shellfish is the main cultured aquaculture group, accounts for 42.6 % of global aquatic production (FAO, 2020). With the increase of global population, the development of shellfish aquaculture can play an important role in sustainable food supply. In addition, bivalve shellfish culture can reduce the occurrence of algal blooms via top-down control of phytoplankton (Galimany et al., 2020). On the other hand, shellfish release CO₂ when calcifying; they generate the same amount of CO₂ from HCO₃⁻ when transform HCO₃⁻ in seawater to form their shells (Ca²⁺ + 2HCO₃⁻ ⇌ CaCO₃↓ + CO₂↑ + H₂O), which drives the increase of pCO₂ in seawater and decrease of total alkalinity (Humphreys et al., 2018). In addition, respiration of shellfish also generates CO₂. On the other hand, shellfish can sequester organic carbon in sediments through feces sinking and in shells (Fodrie et al., 2017). Therefore, shellfish can be a carbon source or sink, which depends on the balance of CO₂ release and sequestration. The contribution of shellfish aquaculture to CO₂ release or sequestration has been assessed in some regions; however, most studies have focused on one side (CO₂ release or sequestration), and little has considered both sides (Tang et al., 2011; Morris and Humphreys, 2019; Liu et al., 2022).

Macroalgae inhabit along the coasts of all continents and dominate rocky shores, forming the most extensive vegetated coastal areas in the world (Gattuso et al., 2006; Krause-Jensen and Duarte, 2016). Macroalgae serve as refugia, habitat and food for marine animals, sustaining coastal ecosystems. They are also used as raw materials for the industries of food, chemical, pharmaceutical and biofuel (Gao et al., 2018, 2020). In spite of the most productive marine macrophytes on a global scale (Duarte and Cebrián, 1996), macroalgae were excluded from the conception of blue carbon because most macroalgae grow on rocks in the intertidal zones where algal debris is easily washed away by tides and carbon burial is considered impossible. However, particulate organic matter (POM) of macroalgae were detected worldwide at up to 5000 km from coastal areas and 24 % of macroalgae available at the surface were expected to reach the seafloor at a depth of 4000 m, suggesting that carbon in macroalgae can be sequestered in oceans for a long term (Ortega et al., 2019). Krause-Jensen and Duarte (2016) calculated carbon sequestration of wild macroalgae based on a model integrating those in the deep ocean and sediments, finding that macroalgae could sequester 173–199 Tg C yr⁻¹, which exceeds the sum of traditional blue carbon (mangroves, salt marshes and seagrasses) (Krause-Jensen and Duarte, 2016; Gao et al., 2022a). Previous studies investigated the carbon fixation of cultivated macroalgae in China (Tang et al., 2011; Ren, 2021); however, more work is needed to assess the carbon sequestration of cultivated macroalgae.

China is responsible for the biggest aquaculture for both shellfish and macroalgae in the world (FAO, 2020) and meanwhile China accounts for approximately one-third of global CO₂ emission. To combat climate change and achieve the Paris temperature target, China has pledged to reach peak emissions of CO₂ by 2030 and carbon neutrality by 2060. Therefore, it would be very essential to investigate the contribution of farmed shellfish and macroalgae on carbon release and sequestration in China. However, how much CO₂ is released or sequestered by maricultured shellfish and macroalgae remains controversial and unclear (Ren, 2021; Yang et al., 2021; Liu et al., 2022), which prevents optimizing culture structure to enhance carbon sink by mariculture. We hypothesize that shellfish may release more CO₂ than macroalgae can sequester, given the larger culture scale of shellfish compared to macroalgae. In this study, we estimated the amount of CO₂ released and sequestered by maricultured shellfish and macroalgae during the past 18 years in China using comprehensive methods. The potential of farming macroalgae to reach carbon neutrality of China in different scenarios was also assessed. This study can make a

solid contribution to assessing the carbon source and sink of mariculture and optimizing culture structure to achieve carbon neutrality.

2. Materials and methods

2.1. Production of shellfish and macroalgae

The data on production and cultivation area of shellfish and macroalgae are from the China Fishery Statistical Yearbook for the years of 2004–2021. Annual productivity of shellfish and macroalgae was calculated by dividing production by cultivation area. Shellfish and macroalgae are farmed in coastal seawaters along the coastline of China from Liaoning Province in the North of China to Hainan Province in the South of China. The census method is used to estimate the production and cultivation area of shellfish and macroalgae in China, in which data are collected from each local farmer and then compiled (Gao, 2015). Although there may be some errors during the data collection and compilation, census is considered to a reliable statistical method because all the items are taken into account (Baffour et al., 2013).

2.2. Weight of shellfish and carbon content in shell

The data of shell and soft tissue weight of shellfish and carbon content in shell were from the combination of literature and the measurements done during this study (Tables S1–S3). The published data were obtained through a search of ISI Web of Science, CNKI and Scholar Google on December 15, 2022, using the terms “shellfish, shell, soft tissue, carbon content and China” as keywords, and hence all literature published before December 15, 2022 was screened. Forty two papers were finally selected after double-checking whether the reported shellfish were from Chinese seas.

Ten species (*Magallana angulata*, *Sinonovacula constricta*, *Solen grandis*, *Haliotis discus hannai*♀ × *H. fulgens*♂, *Ruditapes philippinarum*, *Mytilus coruscus*, *Azumapecten farreri*, *Atrina pectinata*, *Babylonia lutosus*, and *Scapharca subcrenata*) that are some of the main shellfish cultured in China were collected from coastal farms of China during November–December 2021. Please see Tables S1–S3 for details. Ten to twenty samples of each species were randomly selected for the following measurements. Shells and soft tissues of shellfish were dried at 60 °C in an oven (DHG-9146A, Jing Hong, China) till constant weight (~48 h). Shells and soft tissues of each species were then weighed. Dried shells were ground by a mortar and the power was passed through a sieve with a mesh aperture of 0.15 mm. The carbon content in the filtered shell power was measured by an Elementar Vario EL Cube (Elementar, Germany).

2.3. Estimate CO₂ generated from calcification of shellfish

According to the reaction equation, Ca²⁺ + 2HCO₃⁻ ⇌ CaCO₃↓ + CO₂↑ + H₂O there is one mole CO₂ generated when one mole CaCO₃ is synthesized. Therefore, the amount of generated carbon is equal to that of calcified carbon. However, not all generated CO₂ is released to atmosphere and thus the coefficient Φ is introduced, which represents moles of CO₂ released to atmosphere per mole of CaCO₃ formed. The amount of carbon released to atmosphere due to calcification, C_{cal} = Σ(P_i × R_i × C_i × Φ_i), where P_i is production of each shellfish species in each province of China, R_i is the ratio of shell to total weight for each species, C_i is the carbon content in shell for each species, and Φ_i represents moles of CO₂ released to atmosphere per mole of CaCO₃ formed for each shellfish species. The coefficient Φ was calculated according to Humphreys et al. (2018). Since shellfish are produced in different provinces in China and carbonate systems vary in different provinces. Each province in China was allocated an Φ based on the carbonate parameters in coastal seawater of each province (Table S4).

2.4. Estimate CO₂ release from respiration of shellfish

The CO₂ released from shellfish respiration was estimated using the relation established by Schwinghamer et al. (1986): log₁₀R = 0.367 + 0.993

$\log_{10} P$, where P and R are production and respiration ($\text{kcal m}^{-2} \text{yr}^{-1}$) of shellfish, respectively. The conversion factor of $18.85 \text{ J mg SFDW}^{-1}$ was used for shellfish energetic content (Rumohr et al., 1987). SFDW means shell free dry weight. In addition, $1 \text{ J} = 0.239 \text{ cal}$ and $1 \text{ gC} = 11.4 \text{ kcal}$ (Chauvaud et al., 2003).

2.5. CO₂ sequestration by shellfish

CO₂ sequestration by shellfish can be composed by two parts, those in sediments and shells (Fodrie et al., 2017). $C_{\text{sediment}} = F \times (1-r)$, where C_{sediment} is carbon sequestered in sediment, F is carbon in feces of shellfish, r is the remineralization coefficient of feces and sediments, which is 0.87 (Hao et al., 2008). F is calculated based on carbon budget equations of shellfish (Table S5). Organic carbon in shells of shellfish can be sequestered in the long-term and they were calculated based on Fodrie et al. (2017). $C_{\text{shell}} = W_{\text{shell}} \times f_1 \times f_2$, 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. According to Fodrie et al. (2017), the numbers of 0.0136 and 0.36 were used for f_1 and f_2 , respectively. Inorganic carbon in shells is not considered as carbon sink because they come from seawater rather than atmosphere. Therefore, the net CO₂ influx caused by maricultured shellfish = $C_{\text{cal}} + C_{\text{res}} - C_{\text{sediment}} - C_{\text{shell}}$, where C_{res} represents carbon release by respiration (Fig. 1).

2.6. CO₂ sequestered by cultivated macroalgae

Carbon sequestration represents the carbon that is able to be stored in the ocean for a long term (>100 years). The calculation of CO₂ sequestered (C_s) by cultivated macroalgae is according to the formulas $C_s = \text{POC}_{\text{b1}} + \text{POC}_{\text{b2}} + \text{POC}_e + \text{RDOC}$ (Fig. 1), 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. The respired CO₂ is not involved in this calculation as it is part of fixed CO₂ by macroalgae through photosynthesis. The harvested POC is not involved in this calculation either because CO₂ in it can be returned to atmosphere in a short term. Although there are studies investigating the feasibility of sinking macroalgae to deep oceans for carbon sequestration, this approach is ahead of science and beyond the ethics for now (Ricart et al., 2022). According to the literature (Table S6), 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; therefore, the ratio of total sequestered carbon to harvested POC is 0.410. It is worth noting that the ratio is generalized and different macroalgae may have different values. The specific ratios for each macroalgae species are unavailable for now. The average values of carbon

contents in tissue of the seven farmed macroalgae are used (Table S7). The published data of carbon content in macroalgae were obtained through a search of ISI Web of Science, Scholar Google and CNKI on 30 December 2022, using the terms 'carbon content, macroalgae, seaweed and China' as keywords. CO₂ gap between cultured shellfish and macroalgae means CO₂ released by shellfish minus CO₂ sequestered by shellfish and macroalgae.

2.7. Required area to achieve carbon neutrality

The required area (A_{Ri}) for each macroalgae species to achieve carbon neutrality was calculated according to the formula of $A_{\text{Ri}} = C_{\text{T}}/C_{\text{Si}}$. C_{T} is the total CO₂ amount that is needed to be sequestered annually by macroalgae to achieve carbon neutrality in China. China's 2060 carbon neutrality goal will require up to 2.5 GtCO₂ to be sequestered each year and three scenarios are set up here based on Fuhrman et al. (2020) and Yu et al. (2022), in which CCUS (Carbon Capture, Utilization and Storage) contributes 0.5, 1.0 and 1.5 Gt CO₂ per year, encoded as scenario 0.5, 1.0 and 1.5, respectively. The remaining CO₂ will be sequestered by afforestation, in which terrestrial and marine afforestation account for 60 % and 40 % respectively (Fuhrman et al., 2020; Yu et al., 2022; Zhao et al., 2023); C_{Si} ($\text{g C m}^{-2} \text{yr}^{-1}$) is the carbon sequestration rate for each macroalgae species, which was calculated as the annually sequestered carbon divided by culture area.

2.8. Data analysis

To better represent the current status, the data of CO₂ sequestration rates during 2016–2020 were averaged to calculate the required areas to achieve China's carbon neutrality. Data were expressed as means \pm SE. The required area was subjected to error propagation analysis.

3. Results

Annual production of maricultured shellfish increased from 8.99×10^6 ton FW yr^{-1} in 2003 to 14.00×10^6 ton FW yr^{-1} in 2020 (Fig. 2a). Oyster contributed 35–41 % of the total production, followed by clam with 28–34 % of the total production. Among nine kinds of maricultured shellfish, pen shell has the lowest production (0.06–0.45 % of the total production), followed by abalone (0.12–1.45 % of the total production). The culture area of shellfish increased from 845 to 965 $\times 10^3$ ha during the years 2003–2006 and then decreased to 728 $\times 10^3$ ha in 2007 (Fig. 2b). Afterwards, it rapidly rose until it hit the peak of 1404 $\times 10^3$ ha in 2013. Then it slowly decreased and reached 1085 $\times 10^3$ ha in 2020. Based on the

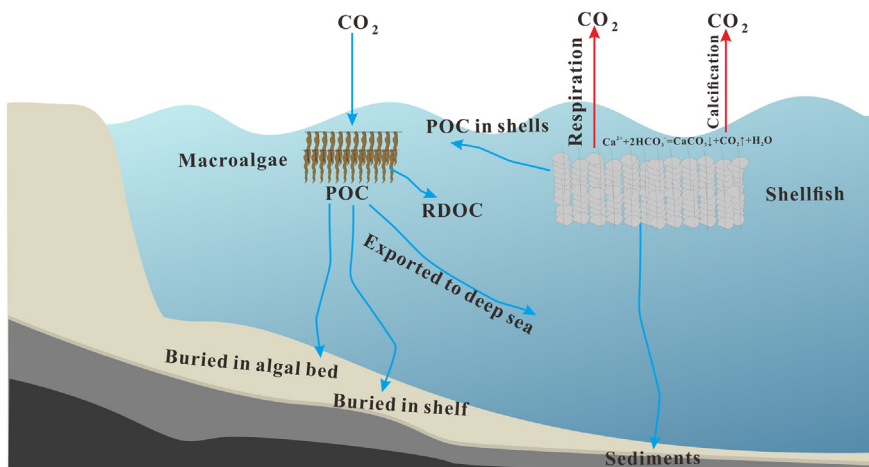


Fig. 1. Carbon pathways of cultivated shellfish and macroalgae. Sequestered carbon is marked in blue and released carbon is marked in red. POC means particle organic carbon and RDOC means refractory dissolved organic carbon.

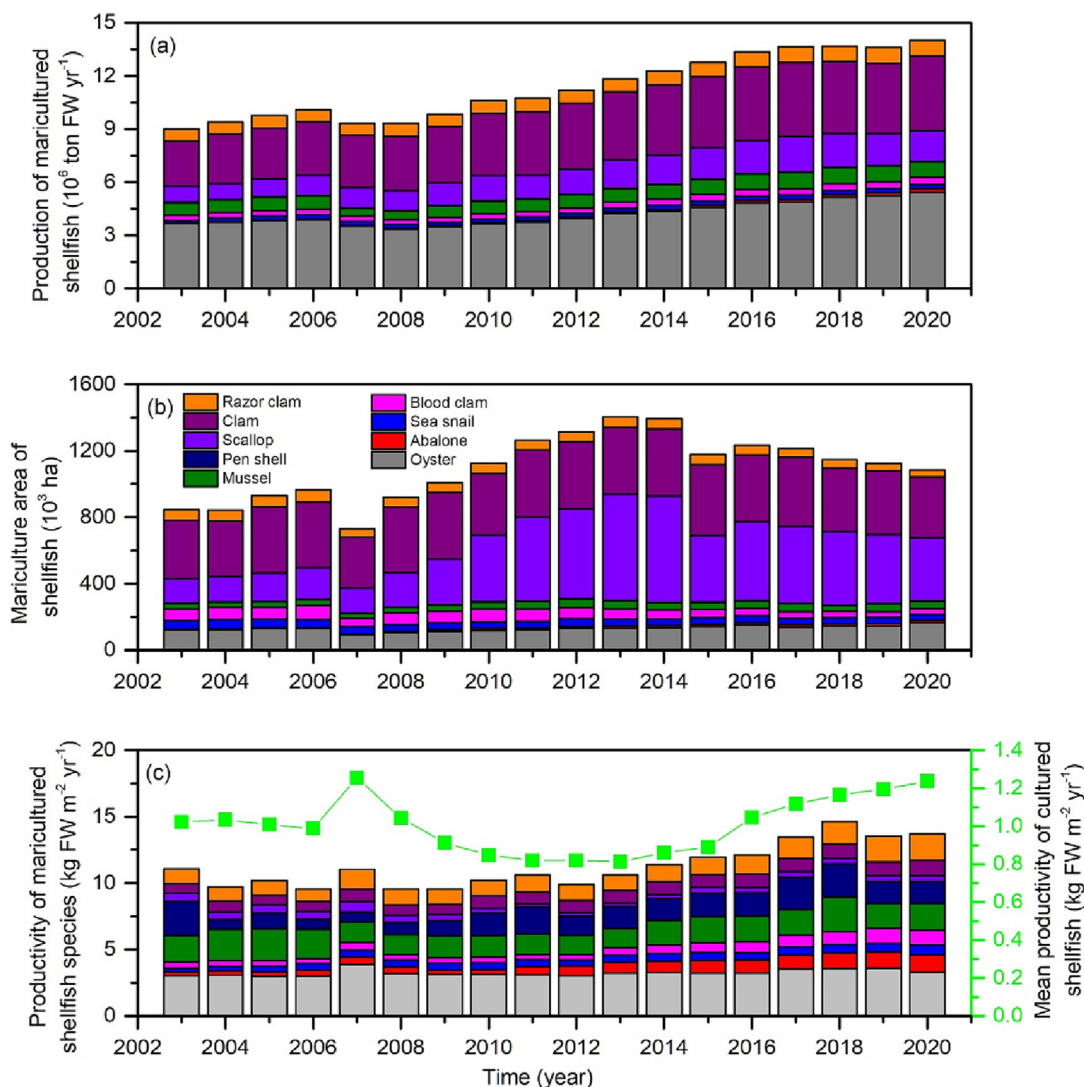


Fig. 2. Production (a), area (b) and productivity (c) of maricultured shellfish during 2003–2020 in China.

average values during 2003–2020, clam has the largest culture area ($386 \pm 7 \times 10^3$ ha), followed by scallop ($367 \pm 39 \times 10^3$ ha). Pen shell has the smallest culture area ($1.17 \pm 0.11 \times 10^3$ ha), followed by abalone ($10.95 \pm 1.02 \times 10^3$ ha). In terms of productivity (Fig. 2c), oyster leads the list (2.97 – 3.88 kg FW m^{-2} yr^{-1}), followed by mussel (1.45 – 2.61 kg FW m^{-2} yr^{-1}). Abalone has the largest variation in productivity that increased by about 5 folds (from 0.27 to 1.30 kg FW m^{-2} yr^{-1}) during the past 18 years. The mean productivity of cultured shellfish ranged from 0.81 to 1.25 kg FW m^{-2} yr^{-1} during the past 18 years.

Annual production of farmed macroalgae increased from 1.13×10^6 ton DW yr^{-1} in 2003 to 2.50×10^6 ton DW yr^{-1} in 2020 (Fig. 3a). *Saccharina japonica* dominates the production, accounting for 60–68 % of the total production. The proportion of *U. pinnatifida* decreased from 17 to 20 % during 2003–2005 to 9–10 % during 2018–2020. In contrast, the proportion of *G. lemaneiformis* increased 5 % in 2003 to 17 % in 2020. Among seven kinds of farmed macroalgae, *Ulva prolifera* has the lowest production (0.007 – 0.15 % of the total production), followed by *Euclima* (0.08 – 0.96 % of the total production). The culture area of macroalgae increased from 79×10^3 ha in 2003 to 138×10^3 ha in 2020 (Fig. 3b). Based on the average values during 2003–2020, *Pyropia* has the largest culture area ($56 \pm 4 \times 10^3$ ha), followed by *S. japonica* ($41 \pm 0.8 \times 10^3$ ha). *U. prolifera* has the smallest culture area ($0.09 \pm 0.02 \times 10^3$ ha), followed by *Euclima* ($0.58 \pm 0.09 \times 10^3$ ha). In terms of productivity (Fig. 3c),

G. lemaneiformis (1.17 – 3.71 kg DW m^{-2} yr^{-1}) and *S. japonica* (1.95 – 3.65 kg DW m^{-2} yr^{-1}) lead the list, followed by *U. pinnatifida* (1.29 – 3.19 kg DW m^{-2} yr^{-1}). Productivity of *Sargassum fusiforme* and *Euclima* has increased by about 4 folds (from 0.44 to 1.93 kg DW m^{-2} yr^{-1} for *Sargassum fusiforme* and from 0.75 to 3.31 kg DW m^{-2} yr^{-1} for *Euclima*) during the past 18 years. The mean productivity of cultured macroalgae increased from 1.44 kg DW m^{-2} yr^{-1} to 1.82 kg DW m^{-2} yr^{-1} during the past 18 years.

CO₂ released by calcification of shellfish increased from 0.501 Tg yr^{-1} in 2003 to 0.765 Tg yr^{-1} in 2020 (Fig. 4a). Oyster contributed the most to the CO₂ release, accounting for 43–50 % of the total release, followed by clam (22–27 %). Pen shell and abalone contributed the least (0.03–0.28 %), followed by abalone (0.07–0.90 %). Compared to calcification, respiration released less CO₂, which also increased from 0.368 Tg yr^{-1} in 2003 to 0.592 Tg yr^{-1} in 2020 (Fig. 4b). Clam and oyster contributed the most to CO₂ release by respiration, accounting for 31–35 % and 22–27 %, respectively. Pen shell contributed the least (0.05–0.35 %), followed by blood clam (1.6–2.3 %) and abalone (0.4–4.5 %). CO₂ sequestration in sediments by maricultured shellfish increased from 0.09 Tg yr^{-1} in 2003 to 0.15 Tg yr^{-1} in 2020 (Fig. 4c). Scallop (0.024 – 0.053 Tg yr^{-1}) and oyster (0.023 – 0.036 Tg yr^{-1}) were the two biggest contributors, followed by clam (0.022 – 0.036 Tg yr^{-1}). In terms of organic carbon sequestered in shells of shellfish, it increased from 0.026 Tg yr^{-1} in 2003 to 0.040 Tg

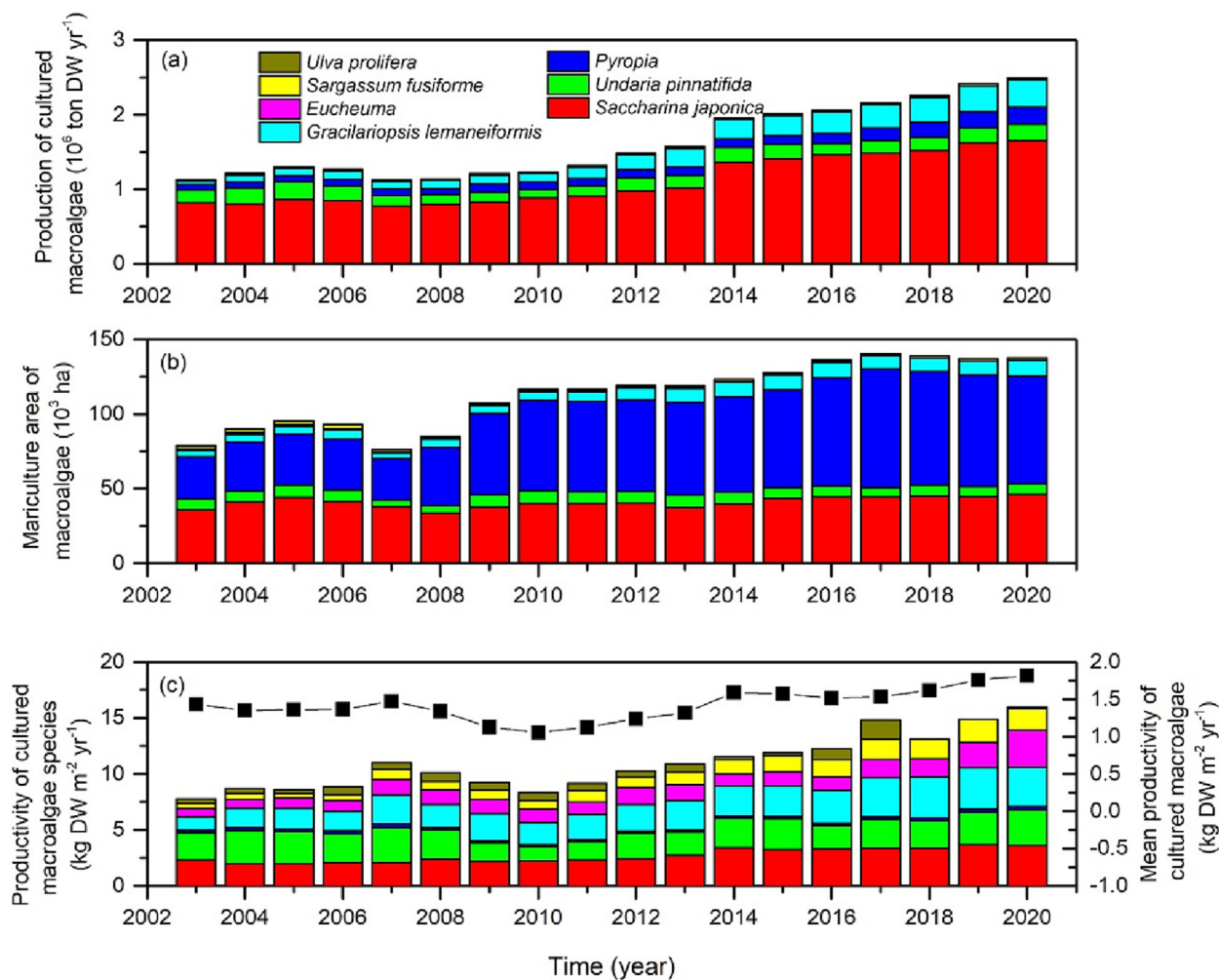


Fig. 3. Production (a), area (b) and productivity (c) of maricultured macroalgae during 2003–2020 in China.

yr^{-1} in 2020 (Fig. 4d). Oyster and clam contributed the most, accounting for 44–51 % and 23–28 % of the total CO_2 sequestration, respectively. Pen shell and abalone contributed the least, accounting for 0.03–0.25 % and 0.06–0.56 % of the total CO_2 sequestration, respectively.

When integrating CO_2 released by calcification and respiration with CO_2 sequestered in sediments and shells, it turns out to be a significant net CO_2 source because sequestered CO_2 is much less than released CO_2 (Fig. 5a). The net CO_2 release ranged from 0.75 to 1.17 Tg yr^{-1} , with the lowest in 2003 and highest in 2020. Oyster (35–41 %) and clam (26–31 %) were still the top two contributors while pen shell contributed the least (0.03–0.29 %), followed by abalone (0.22–2.19 %) and sea snail (1.96–3.18 %). CO_2 sequestered by cultivated macroalgae annually more than doubled (from 0.137 to $0.308 \text{ Tg C yr}^{-1}$) during the past 18 years (Fig. 5b). *Saccharina japonica* is the biggest contributor although its contribution decreased from 68 % in 2003 to 61 % in 2020. The contribution of *Undaria pinnatifida* also decreased from 17 % in 2003 to 10 % in 2020. On the other hand, the contribution of *Gracilariopsis lemaneiformis* and *Pyropia* increased from 8 % and 5 % in 2003 and to 11 % and 17 % in 2020, respectively. After comparing CO_2 release by shellfish and sequestration by macroalgae, the gap between them was calculated (Fig. 5c). Due to higher production of shellfish, the combination of maricultured shellfish and macroalgae shows a net CO_2 source. The net CO_2 release increased from $0.617 \text{ Tg C yr}^{-1}$ in 2003 to $0.864 \text{ Tg C yr}^{-1}$ in 2020, the average of which in the past five years is $0.857 \pm 0.01 \text{ Tg C yr}^{-1}$.

Based on averaged value during past 18 years, shellfish species has a large range of CO_2 release rate due to calcification (Fig. 6a). Oyster has the highest rate of $218 \pm 4 \text{ g C m}^{-2} \text{ yr}^{-1}$, followed by mussel ($111 \pm$

$5 \text{ g C m}^{-2} \text{ yr}^{-1}$). Scallop has the lowest rate of $21 \pm 2 \text{ g C m}^{-2} \text{ yr}^{-1}$, followed by abalone $24 \pm 3 \text{ g C m}^{-2} \text{ yr}^{-1}$. CO_2 release rate due to respiration also varies with species (Fig. 6b). Mussel has the highest rate of $120 \pm 5 \text{ g C m}^{-2} \text{ yr}^{-1}$, followed by abalone ($93 \pm 11 \text{ g C m}^{-2} \text{ yr}^{-1}$). Blood clam has the lowest rate of $17 \pm 2 \text{ g C m}^{-2} \text{ yr}^{-1}$, followed by scallop ($24 \pm 2 \text{ g C m}^{-2} \text{ yr}^{-1}$). When integrating CO_2 release by calcification and respiration and sequestration in sediments and shells of cultured shellfish (Fig. 6c), oyster has the highest CO_2 release rate ($273 \pm 5 \text{ g C m}^{-2} \text{ yr}^{-1}$) and mussel ranks second ($185 \pm 8 \text{ g C m}^{-2} \text{ yr}^{-1}$). Scallop ($32 \pm 2 \text{ g C m}^{-2} \text{ yr}^{-1}$) and blood clam ($48 \pm 5 \text{ g C m}^{-2} \text{ yr}^{-1}$) are at the bottom of this list. In terms of macroalgae (Fig. 6d), *Gracilariopsis lemaneiformis* has the highest carbon sequestration rate of $356 \pm 24 \text{ g C m}^{-2} \text{ yr}^{-1}$, followed by *Undaria pinnatifida* ($331 \pm 17 \text{ g C m}^{-2} \text{ yr}^{-1}$), with *Pyropia* the lowest rate of $35 \pm 2 \text{ g C m}^{-2} \text{ yr}^{-1}$.

The required areas for macroalgae cultivation to achieve carbon neutrality of China based on different scenarios were calculated (Fig. 7). In scenario 0.5 (Fig. 7a), *Pyropia* has the largest required area ($561 \pm 49 \text{ ha}$), followed by *U. prolifera* ($139 \pm 67 \text{ ha}$). *G. lemaneiformis* having the smallest required area ($45 \pm 2 \times 10^6 \text{ ha}$), followed by *S. japonica* ($56 \pm 1 \times 10^6 \text{ ha}$). Neither area can be met by the available area of $39 \times 10^6 \text{ ha}$. In scenario 1.0 (Fig. 7b), required areas for all macroalgae species surpass the available area except for *G. lemaneiformis* that has a required area of $34 \pm 1 \times 10^6 \text{ ha}$. In scenario 1.5 (Fig. 7c), the required areas for all macroalgae decrease further and more species can meet the demand. For instance, the required areas for *G. lemaneiformis* ($22 \pm 1 \times 10^6 \text{ ha}$), *S. japonica* ($28 \pm 1 \times 10^6 \text{ ha}$) and *U. pinnatifida* ($31 \pm 2 \times 10^6 \text{ ha}$) are smaller than the available area.

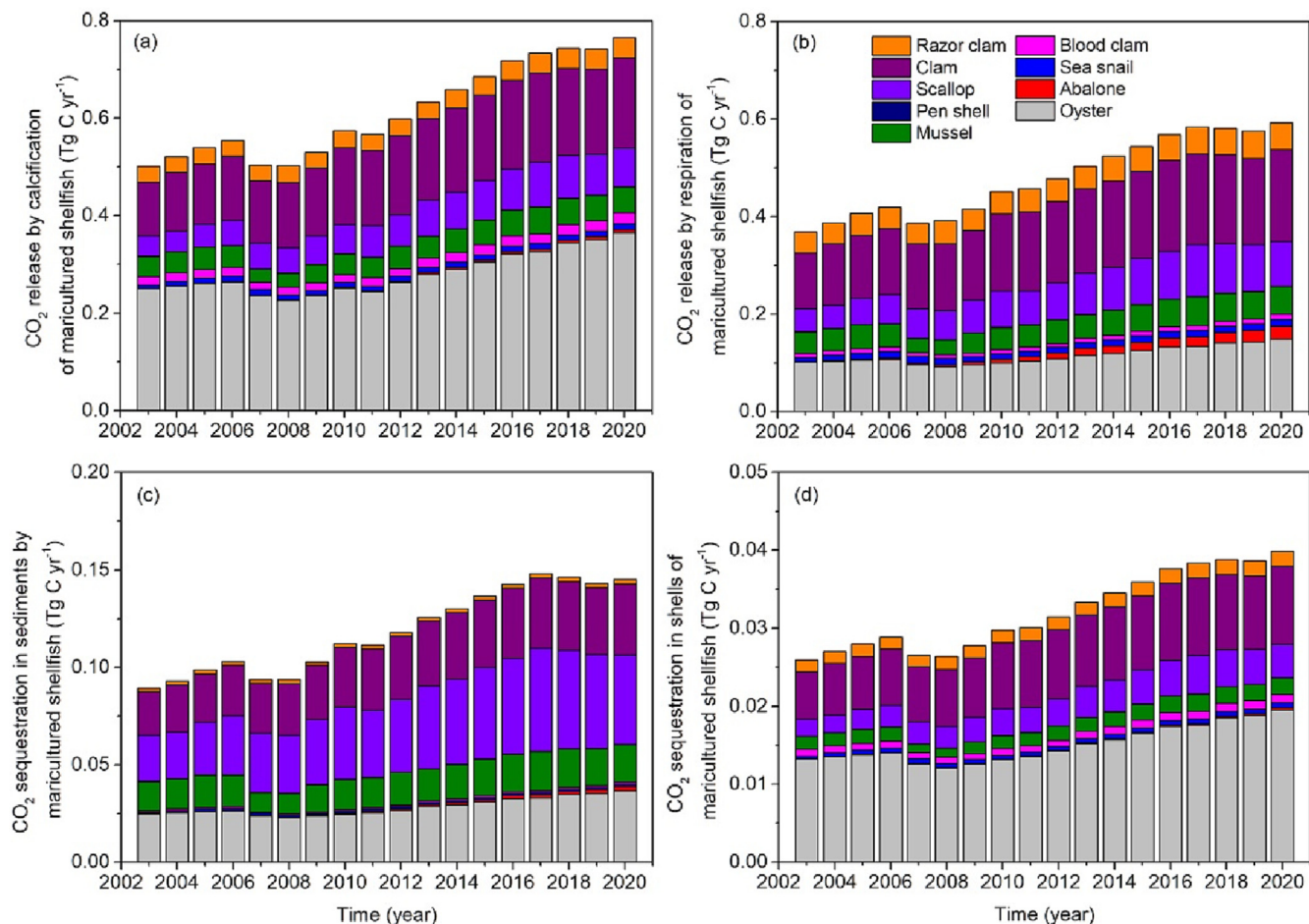


Fig. 4. Carbon release and sequestration by maricultured shellfish in China. (a) Carbon release by calcification; (b) carbon release by respiration; (c) carbon sequestration in shells as organic form; (d) carbon sequestration in sediments by maricultured shellfish.

4. Discussion

4.1. CO₂ release and sequestration by maricultured shellfish

Shellfish mariculture was considered as carbon sink since carbon in seawater is fixed in their shells as they grow (Tang et al., 2011; Ren, 2021). However, this fixation does not lead to decreased pCO₂ in seawater. Carbon sequestration in shells of shellfish cannot be considered as carbon sink since carbon sink represents CO₂ sequestration from atmosphere rather than from seawater. Instead, calcification results in increased pCO₂ in seawater and makes shellfish mariculture a carbon source. The increased pCO₂ has been found in many areas of extensive shellfish monoculture, indicating a carbon source for this activity (Han et al., 2021; Yang et al., 2021). The amount of CO₂ release by calcification depends on biomass and the ratio of shell to total weight. In this study, CO₂ released by shellfish calcification increased with year, which is mainly caused by increased production of shellfish. Although culture area decreased in recent years, the increased productivity due to improved strains results in continuous increase in shellfish production (Xiao et al., 2022). The higher production and shell: total weight of oyster, clam and scallop contribute to very large CO₂ release for these three categories. Meanwhile, lower production and shell proportion of pen shell and abalone lead to very small CO₂ release. Jiang et al. (2014) found that the CO₂ release rate of the scallop *Chlamys farreri* due to calcification was $53.95 \pm 3.98 \text{ g C m}^{-2} \text{ yr}^{-1}$, which is 2–4 times higher than those in this study. This can be attributed to higher CaCO₃ productivity ($650.53 \text{ g m}^{-2} \text{ yr}^{-1}$) used in Jiang et al. (2014) while it was only $126\text{--}308 \text{ g CaCO}_3 \text{ m}^{-2} \text{ yr}^{-1}$ in this study. The data in Jiang et al. (2014) was based on a mesocosm experiment in Sanggou Bay, China while the

data presented on this study is values based on the averaged values recorded at a national scale, which may lead to the difference. On the other hand, CO₂ release rate of oyster calcification in this study ($200\text{--}261 \text{ g C m}^{-2} \text{ yr}^{-1}$) is much higher than that ($11.11 \text{ g C m}^{-2} \text{ yr}^{-1}$) of *Crassostrea gigas* cultured in Sangou Bay, China (Jiang et al., 2015). Lower CaCO₃ productivity ($134.0 \text{ g CaCO}_3 \text{ m}^{-2} \text{ yr}^{-1}$) was recorded in Jiang et al. (2015) compared to this study ($2,186\text{--}2,856 \text{ g CaCO}_3 \text{ m}^{-2} \text{ yr}^{-1}$). Meanwhile, the CO₂ release rate of Portuguese oyster *Magallana angulata* ($153 \text{ g C m}^{-2} \text{ yr}^{-1}$) cultured in Daya Bay China is close to this study since its CaCO₃ productivity ($2150 \text{ g CaCO}_3 \text{ m}^{-2} \text{ yr}^{-1}$) is also very high (Han et al., 2017). Therefore, CO₂ release rates of oyster calcification depend on their CaCO₃ productivity that is related to biomass productivity and the ratio of shell to total weight of oyster, while these two parameters vary with stocking density and oyster species (Lejart et al., 2012; Han et al., 2017). The CO₂ release rate due to clam calcification ($31\text{--}50 \text{ g C m}^{-2} \text{ yr}^{-1}$) in this study is lower than the farmed short-neck clam *Ruditapes philippinarum* in the Marinetta lagoon (Italy) ($67 \text{ g C m}^{-2} \text{ yr}^{-1}$) (Mistri and Munari, 2012), but falls in the range of ($1\text{--}109 \text{ g C m}^{-2} \text{ yr}^{-1}$) the natural Asian clam, *Potamocorbula amurensis* in San Francisco Bay. It is worth noting that the calculation of CO₂ release to atmosphere in this study is based on Φ that represents the potential amount of CO₂ released to atmosphere by shellfish calcification. Although the increased pCO₂ in seawater drives the flow from seawater to atmosphere, the actual air-sea CO₂ exchange process may require several months to a year to re-equilibration (Jones et al., 2014).

In addition to calcification, respiration of shellfish also generates CO₂ and leads to increased pCO₂ in seawater. Compared to calcification, shellfish respiration generated less CO₂, which could be attributed to high

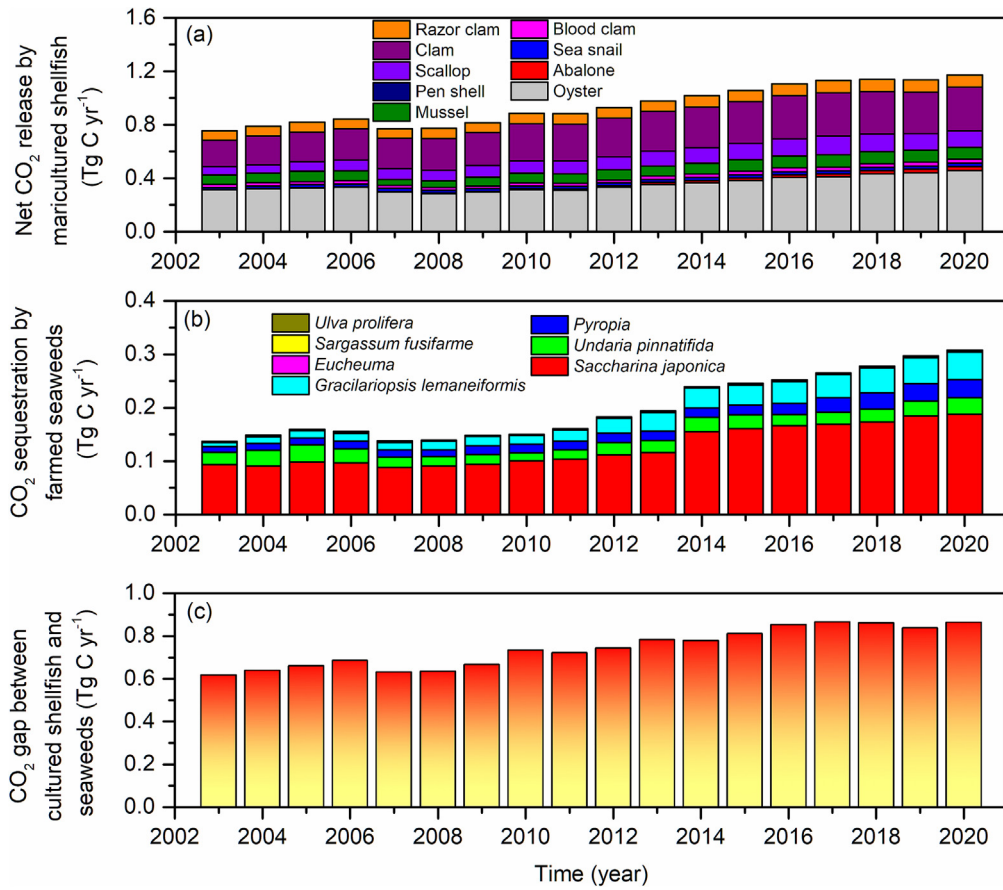


Fig. 5. Net CO₂ release by maricultured shellfish (a), carbon sequestration by cultivated macroalgae (b), and the CO₂ gap between cultured shellfish and sequestered CO₂ by macroalgae (c) during 2003–2020 in China.

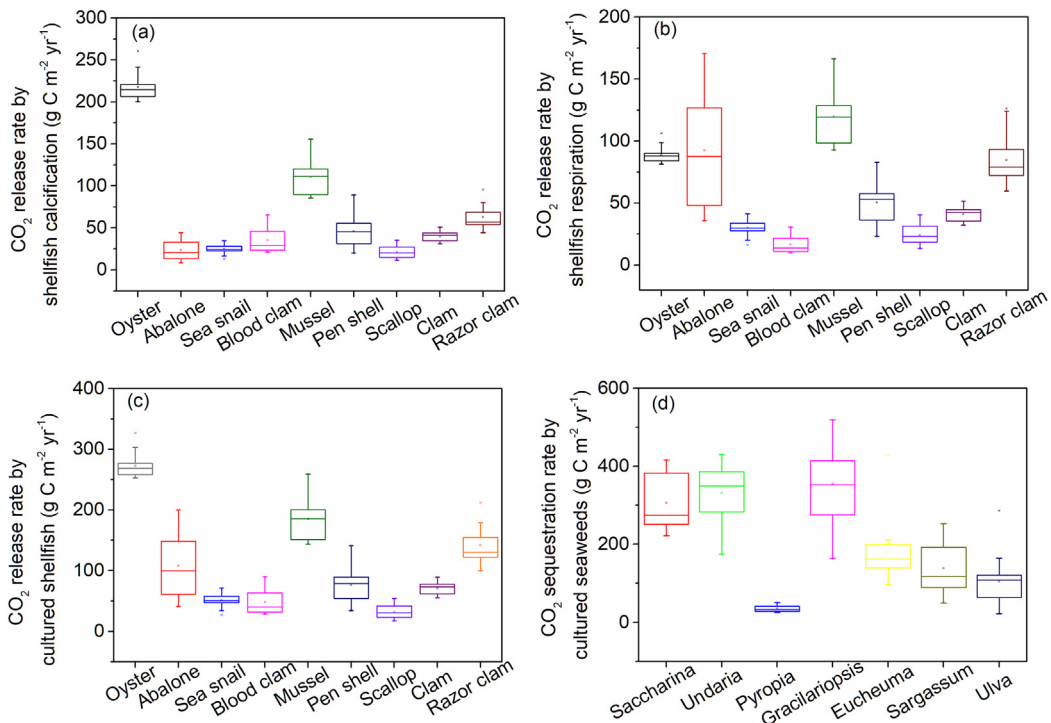


Fig. 6. Carbon release or sequestration rate of shellfish and macroalgae based on the data from 2003 to 2020 in China. (a) CO₂ release rate by shellfish calcification; (b) CO₂ release rate by shellfish respiration; (c) net CO₂ release rate after integrating calcification and respiration with organic carbon in sediments and shells of cultured shellfish; (d) CO₂ sequestration rate by cultured macroalgae.

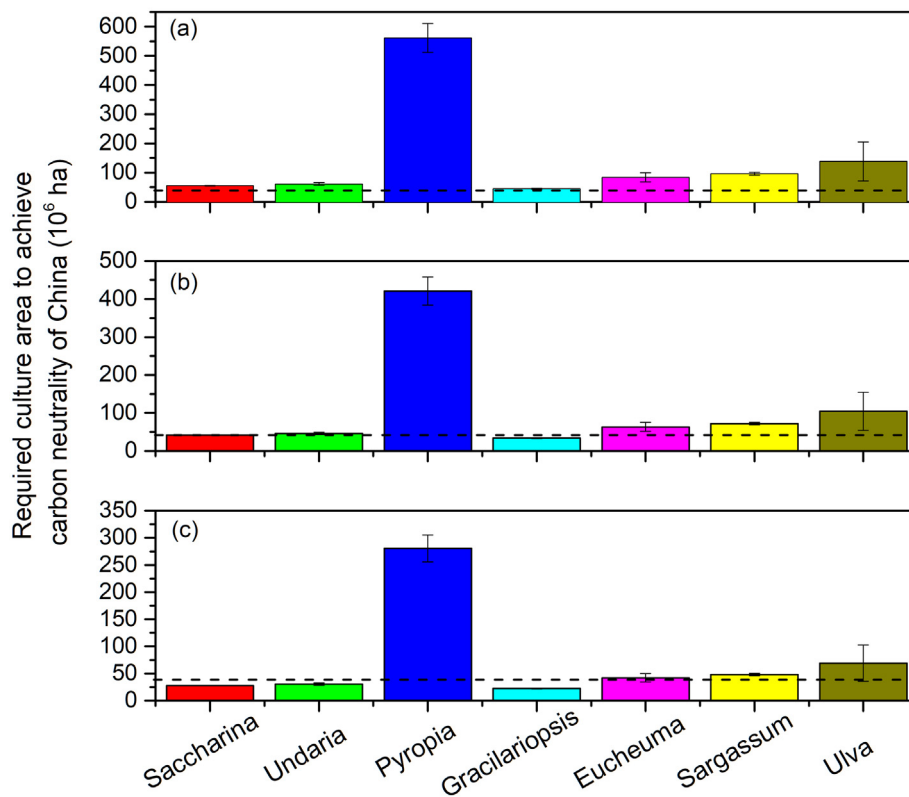


Fig. 7. Required culture area of macroalgae to achieve carbon neutrality of China based on different scenarios. (a) CCUS contributes 0.5 Gt CO₂ negative emissions per year, scenario 0.5; (b) CCUS contributes 1.0 Gt CO₂ negative emissions per year, scenario 1.0; (c) CCUS contributes 1 Gt CO₂ negative emissions per year, scenario 1.5. The dashed lines represent available area (39.4×10^6 ha) for macroalgae cultivation in China (Gao et al., 2021).

ratio of shell to soft tissue (Table S1). Contrary to calcification, clam rather than oyster contribute most CO₂ release by respiration as clam had higher ratio of soft tissue to shell compared to oyster (Table S1). In terms of CO₂ release rate due to respiration, CO₂ release rate of oyster calcification in this study ($81\text{--}106 \text{ g C m}^{-2} \text{ yr}^{-1}$) is higher than that ($30.8 \text{ g C m}^{-2} \text{ yr}^{-1}$) of *C. gigas* cultured in Sanggou Bay, China (Jiang et al., 2015) but lower than that ($251 \text{ g C m}^{-2} \text{ yr}^{-1}$) of *C. gigas* in the Bay of Brest, France and that ($258 \text{ g C m}^{-2} \text{ yr}^{-1}$) of the Portuguese oyster *M. angulata* cultured in Daya Bay China (Han et al., 2017). CO₂ release rate of clam due to respiration ($32\text{--}52 \text{ g C m}^{-2} \text{ yr}^{-1}$) in this study is lower than that ($227 \text{ g C m}^{-2} \text{ yr}^{-1}$) of the short-neck clam *Ruditapes philippinarum* in the Marinetta lagoon (Italy) (Mistri and Munari, 2012) but falls in the range of CO₂ release rate for the natural Asian clam, *Potamocorbula amurensis* in San Francisco Bay ($14\text{--}77 \text{ g C m}^{-2} \text{ yr}^{-1}$). The variation in CO₂ release rate among studies could be attributed to differences in the productivity of shellfish and the ratio of soft tissue to total weight.

Organic carbon in shells of shellfish and sediments can be sequestered for a long time and thus be considered as a carbon sink (Fodrie et al., 2017). All maricultured shellfish species show to be carbon sources in the present study. In contrast, shallow subtidal and saltmarsh-fringing oyster reefs in North Carolina were reported to be net carbon sinks (Fodrie et al., 2017). Two reasons may cause the different results. Firstly, Fodrie et al. (2017) excluded the CO₂ released by respiration of shellfish, which leads to the decreased CO₂ release. Secondly, Fodrie et al. (2017) assessed natural oyster reefs that have a tight contact with the sediment layer. This tight contact can reduce respiration and remineralization of shellfish feces and increase CO₂ sequestration, while most maricultured shellfish are hanged in surface seawater and harvested within 1–2 years, and thus most shellfish feces are remineralized during sinking and resuspension (Hao et al., 2008; Zhao and Zhang, 2022).

4.2. Carbon sequestration by macroalgae

The cultivation production and area of macroalgae have been increasing at high rates during the past 18 years due to rising demands in the field of food, animal feed, chemical and pharmaceutical (Gao et al., 2021). The increased production contributes to increased carbon sequestration by macroalgae. The carbon sequestered by farmed macroalgae during 2010–2019 in this study ($0.15\text{--}0.30 \text{ Tg C yr}^{-1}$) is less than that ($0.96\text{--}1.41 \text{ Tg C yr}^{-1}$) reported by Liu et al. (2022). Ren (2021) showed even higher values ($1.21\text{--}2.14 \text{ Tg C yr}^{-1}$ during 2010–2017). The reasons for these differences come from different calculation methods or parameters. Liu et al. (2022) used a very high ratio of RDOC/DOC (0.855) while the ratio we used is 0.521. Meanwhile, Liu et al. (2022) assumed a 365 day of macroalgae cultivation and thus POC and DOC production was overestimated because most macroalgae species cannot be cultivated year around in China (Gao et al., 2021). Ren (2021) used harvested POC as removable carbon sink, which could be released to atmosphere in a short time when consumed and cannot be deemed as carbon sequestration. In fact, the carbon sequestration rates of some macroalgae species in this study, e.g. *G. lemaneiformis* ($356 \pm 24 \text{ g C m}^{-2} \text{ yr}^{-1}$) and *U. pinnatifida* ($331 \pm 17 \text{ g C m}^{-2} \text{ yr}^{-1}$) are even higher than rooted blue carbon plants, such as seagrasses ($117 \pm 19 \text{ g C m}^{-2} \text{ yr}^{-1}$), mangroves ($168 \pm 23 \text{ g C m}^{-2} \text{ yr}^{-1}$) and salt marshes ($224 \pm 34 \text{ g C m}^{-2} \text{ yr}^{-1}$) (Gao et al., 2022a). The higher carbon sequestration rates of farmed macroalgae compared to rooted blue carbon plants should due to higher growth rates and cultivation densities.

4.3. Adjustment of mariculture structure

While we cannot deny the contribution of shellfish mariculture to seafood supply and providing jobs, it does release a large quantity of CO₂ into seawater and thus into atmosphere during shell formation and

respiration. To deal with this problem, one direction is to constraint the development of shellfish aquaculture and replaces the species commonly cultivated with other species that release less CO₂, such as fish aquaculture that does not involve calcification. Another direction is to expand macroalgae cultivation. The mean CO₂ release and sequestration rate for shellfish and macroalgae are 87 ± 3 and $174 \pm 6 \text{ g m}^{-2} \text{ yr}^{-1}$, respectively. This is to say, to achieve carbon neutrality, one hectare of macroalgae cultivation can allow about two hectares of shellfish culture. More shellfish can be cultured if macroalgae with higher CO₂ sequestration rate are cultivated. For instance, *G. lemaneiformis* has a rate of $356 \text{ g m}^{-2} \text{ yr}^{-1}$, indicating that nearly four folds of area can be used for shellfish culture with zero CO₂ emission when *G. lemaneiformis* is cultivated. The current area for macroalgae cultivation is only 12 % of shellfish culture and therefore needs to be increased to completely neutralize CO₂ released by shellfish culture. While different mariculture structure can result in contrasting environmental consequences (Xiong et al., 2023), adjustment of mariculture structure is not an easy thing since shellfish culture commonly makes more profit than macroalgae. Therefore, subsidy needs to be provided to macroalgae farmers to propel the adjustment.

In addition to neutralizing CO₂ released by shellfish culture, macroalgae cultivation can also contribute to China's carbon neutrality by 2060. Based on the findings in this study, if CCUS (Carbon Capture, Utilization and Storage) contributes 1.5 Gt CO₂ per year, it is possible to achieve China's carbon neutrality by culturing *G. lemaneiformis*, *U. pinnatifida* or *S. japonica*. Even if CCUS sequesters 1.0 Gt CO₂ per year, cultivating *G. lemaneiformis* is also feasible. In terms of global scale, Froehlich et al. (2019) has concluded that the area suitable for macroalgae farming is approximately 48 million km², which is far more than required area to sequester 4 Gt CO₂ yr⁻¹ that is needed to limit warming to 2 °C above preindustrial conditions at a global scale in Representative Concentration Pathway 2.6 (Sanderson et al., 2016; Gao et al., 2022a). Therefore, macroalgae cultivation shows a huge potential of contributing to carbon neutrality both for China and the whole planet.

Meanwhile, it is worthy of noting that the calculations above are based on the current productivity. The productivity of farmed macroalgae may decrease as it expands to areas with lower nutrients. To enhance nutrients supply, integrated multi-trophic aquaculture (IMTA), in which species from two or more trophic levels are cultured together and the waste of one feeds another has been deemed as a balanced system for environment remediation (biomitigation) and production stability (Chopin et al., 2001; Gao et al., 2022b). This three-dimensional aquaculture system, usually autotrophic species up and heterotrophic species down, can also save culture area. Furthermore, IMTA can enhance productivity and functional and thus increase farm revenues, indicating an environmentally friendly and cost effective aquaculture mode (Gao et al., 2022b). Most of previous studies focus on the nutrient bioremediation of IMTA. This study indicates that IMTA may maintain the carbonate system of seawater, particularly pCO₂. The CO₂ released by calcification and respiration of shellfish can be utilized by macroalgae. Increased CO₂ can usually stimulate growth of macroalgae as CO₂ in seawater is limited for photosynthesis of macroalgae (Gao et al., 2016; Ji and Gao, 2021). Meanwhile, increased pH caused by photosynthesis of macroalgae can facilitate calcification of shellfish. Some trials have been conducted to determine the optimal ratio of shellfish to macroalgae to create a carbon sink (Jiang and Fang, 2021).

The increased biomass due to expansion of macroalgae cultivation may go beyond the demands as macroalgae are mainly consumed in Asian countries as marine vegetables (Araújo et al., 2021). In addition to using as food and chemicals, macroalgae can be used as biofuels. Bio-methane from macroalgae is very close to profitability (Gao et al., 2020). Therefore, high-volume demanding biofuel could be an ideal destiny for increased macroalgae biomass. In addition, the released carbon from biofuel can be sequestered if the negative emission technology of bioenergy with carbon capture and storage (BECCS) is deployed (Xing et al., 2021). These activities align strongly with a number of the United Nations Sustainable Development Goals (SDGs), such as UN SDG7 (“Ensure access to affordable, reliable, sustainable and modern energy for all”), UN SDG12 (“Ensure

sustainable production and consumption patterns”), UN SDG13 (“Take urgent action to combat climate and its impacts”), and UN SDG 14 (“Conserve and sustainably use the oceans, seas and marine resources for sustainable development”).

5. Conclusions

To reduce CO₂ emission and increase carbon sequestration are the necessary steps for achieving the Paris 1.5 or 2 °C target. While intensive studies focus on CO₂ emission on land, little attention is paid to CO₂ emission from ocean (Burandt et al., 2019). This study, for the first time, assessed the CO₂ emission from shellfish mariculture in China based on detailed data. Calcification and respiration of maricultured shellfish in China generates a huge quantity of CO₂. Therefore, to achieve carbon neutrality and generate a net carbon sink for mariculture, shellfish mariculture should be restricted and meanwhile macroalgae cultivation should be expanded. Although this adjustment and optimization of mariculture may decrease the income given the higher prices of shellfish, it can benefit the environments in terms of carbon neutrality and mitigating eutrophication. The co-culture of shellfish and macroalgae may be an effective solution to achieve carbon neutrality along with enhancing production of both shellfish and macroalgae, while the ratio of them and the selection of species need to be further studied in future to maximize carbon sequestration along with high biomass yield for both shellfish and macroalgae. In terms of China's carbon neutrality, it is feasible to achieve it through cultivating some macroalgae with higher productivity, e.g. *G. lemaneiformis*, *S. japonica* and *U. pinnatifida*, if CCUS can sequester 1.5 Gt CO₂ per year.

Author contributions

C.S. gathered preliminary data, conducted preliminary analysis, designed the formal analysis and investigation, developed data visualizations, developed the methodology, and contributed to writing the original draft and to reviewing and editing. Y.X. completed the formal analysis, developed the methodology, developed data visualizations, and contributed to writing and to reviewing and editing. P.J. developed data visualizations, and contributed to reviewing and editing. Y.S. gathered preliminary data, developed the methodology, contributed to reviewing and editing. Q.Z. gathered preliminary data, contributed to reviewing and editing. Z.M. gathered preliminary data, contributed to reviewing and editing. G.G. conceived and supervised the study, gathered preliminary data, conducted preliminary analysis, designed the formal analysis and investigation, developed the methodology, and contributed to writing the original draft and to reviewing and editing.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare no competing interests.

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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.2023.164986>.

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