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The contribution of fish and seaweed mariculture to the coastal fluxes of biogenic elements in two important aquaculture areas, China

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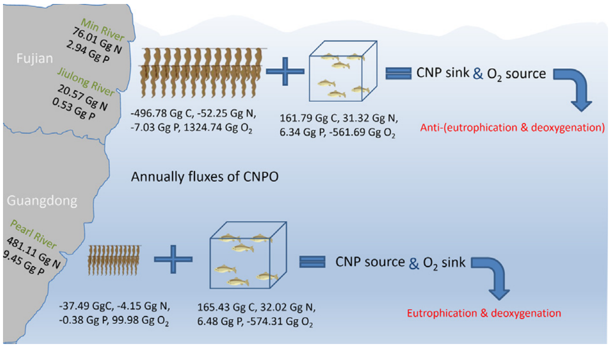
HIGHLIGHTS

- The P flux due to fish mariculture in Fujian is 2.2 folds of the Min River.
- The P flux due to fish mariculture in Guangdong is 69 % of the Pearl River.
- The N flux due to seaweed culture in Fujian is 69 % of the Min River.
- The N flux due to seaweed culture in Guangdong is 0.86 % of the Pearl River.
- The combination of fish and seaweed culture leads to a sink of CNP in Fujian but a source in Guangdong.

GRAPHICAL ABSTRACT



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ABSTRACT

Carbon, nitrogen, phosphorus and oxygen (CNPO) are essential biogenic elements, driving life activities in marine environments. However, the integrated research of fish and seaweed culture on the fluxes of CNPO is scarce. To bridge the research gap, the contribution of mariculture of fish and seaweeds to the fluxes of CNPO in two important mariculture provinces, Fujian and Guangdong, in China, was investigated for the first time. Data from published literature and this study were integrated to calculate the CNPO fluxes using relative formulas. CNP release and O_2 loss caused by fish mariculture increased with year (2003 – 2020) and reached 185.55 ± 3.18 Gg C, 35.92 ± 0.51 Gg N, 7.27 ± 0.24 Gg P and 644.18 ± 11.05 Gg O_2 for Fujian and 215.81 ± 2.51 Gg C, 41.77 ± 0.40 Gg N, 8.47 ± 0.19 Gg P and 749.23 ± 8.71 Gg O_2 for Guangdong in 2020. The averaged P fluxes due to fish mariculture in Fujian and Guangdong during 2016–2020 are 2.2 folds of the Min River and 69 % of the Pearl River, respectively. CNP removal and O_2 generation by seaweed culture in Fujian also increased with year (2003–2020) and reached 555.74 ± 16.45 Gg C, 58.44 ± 4.83 Gg N, 7.80 ± 1.41 Gg P and 1481.97 ± 43.86 Gg O_2 in 2020. In contrast, seaweed culture in Guangdong resulted in maximal C (39.81 ± 1.43 Gg), N (4.33 ± 0.26 Gg) removal and O_2 (106.15 ± 3.82 Gg) release in 2013 and maximal P (0.41 ± 0.03 Gg) removal in 2019. The averaged N and P fluxes due to seaweed culture in Fujian during 2016–2020 are 69 % and 2.4 folds of the Min River, respectively. The different mariculture structure leads to a net CNP sink in Fujian but a net CNP source in Guangdong. The net CNP source may lead to seawater acidification, eutrophication and deoxygenation in coastal areas. These findings supply solid data for adjusting mariculture structure to achieve CNPO neutrality within mariculture.

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1. Introduction

Food demands are rising due to population explosion. Blue foods, defined as aquatic foods captured or cultivated in marine and freshwater systems, are considered as a potential solution due to the limited land area combined with increase environmental pressures from terrestrial agriculture (Springmann et al., 2018; Naylor et al., 2021a, b). With the increased consumption of blue foods, wild stocks had been not enough to meet the demand. Therefore, aquaculture has been developed fast and global aquaculture production has grown at a compound annual rate of 8.2 % since 1950 (FAO, 2020). In 2013, annual aquaculture production exceeded capture fishery landings (FAO, 2020). Global fish production reached 179 million tonnes in 2018, with a total first sale value estimated at USD 401 billion, of which 82 million tonnes, valued at USD 250 billion, came from aquaculture production (FAO, 2020).

Aquaculture provides sustainable, nutrient-rich animal protein for human and relieves food security to a large extent (Crona et al., 2020). However, with the increase of aquaculture scale, the environmental impacts it brings about are causing concerns (Gephart et al., 2021). Aquaculture, particularly for those with feeds, can cause eutrophication and hypoxia when uneaten feeds are decomposed by microbes (Chislock et al., 2013; Herbeck et al., 2013; Gephart et al., 2021). In 2016, 0.94 Tg P was released into aquatic environments due to fish aquaculture (Huang et al., 2020). Shrimp and fish ponds covering an area of 39.6 km^2 in the Northeastern coast of Hainan, tropical China, discharges 612 t total dissolved nitrogen (TDN) and 680 t particulate nitrogen (PN) annually into coastal waters, leading to coastal eutrophication and thus threatening seagrasses and corals (Herbeck et al., 2013). In addition to NP release, aquaculture also generates greenhouse gas emissions. For instance, farmed diadromous fishes can generate about 20,400 kg CO_2 t^{-1} edible weight. The released CO_2 can come from both on-farm activities and off-farm feed production (Gephart et al., 2021). Contrary to fish aquaculture, seaweed cultivation is considered to be able to remove CNP from seawater and mitigate deoxygenation in the seas in addition to supplying food and biofuels for humans (Gao et al., 2018, 2020, 2021; Duarte et al., 2022). Xiao et al. (2017) found that seaweed aquaculture in China annually removes approximately 75,000 t nitrogen and 9500 t phosphorus. Gao et al. (2021) estimated that cultivated seaweeds in China could remove 605, 830 t of carbon, sequestrated 344, 128 t of carbon and generated 2533, 221 t of oxygen, apart from removing 70, 615 t of nitrogen and 8, 515 t of phosphorus annually. However, these studies only calculated the CNP in harvested seaweeds and excluded those in deep oceans and refractory form that can be sequestered in seawater for a long term (Gao et al., 2022a, b).

China is a key player in global blue-food production and trade. Both animal and seaweed aquaculture in China accounted for about 60 % of global production, representing one of the largest producers, consumers, importers, and exporters of blue-food in the world (FAO, 2020). Previous studies investigated the impacts of fish or seaweeds mariculture on aquatic environments separately, but little is known about the combined effects of these two, particularly for a comprehensive influence on the fluxes of CNPO that are essential biogenic elements, driving life activities and regulating ecosystems in marine environments (Jiang et al., 2015; Gu et al., 2021; Han et al., 2021a, b). In addition, the information regarding the CO_2 release by fish mariculture is very scarce. Gao et al. (2021) investigated the NP fluxes of fish and seaweed mariculture in a national level and found current cultivated seaweeds can only absorb about half of N and one-third of P released from fish mariculture in China. To adjust mariculture structure, action must be taken from a regional level. However, relative studies in a regional level in China are limited. Guangdong and Fujian provinces rank Nos. 1 and 2 for fish production, respectively. Additional, Fujian province accounts for the biggest seaweed production in China while the scale of seaweed cultivation in Guangdong is much smaller than Fujian. It is hypothesized that different mariculture structure may lead to contrasting CNPO fluxes and thus environment impacts in this study. A new method that includes CNP in seaweeds that exist in deep oceans and in the refractory form is used to assess the contribution of farmed seaweeds to CNP fluxes. Therefore, the present study, investigating the combined effects of fish and seaweed mariculture on coastal CNPO fluxes in these two provinces, could provide helpful data for adjusting mariculture structure to achieve CNPO neutrality within mariculture.

2. Materials and methods

2.1. Production of maricultured fish and seaweeds

The data on production and cultivation area of fish and seaweeds in Fujian and Guangdong are from the China Fishery Statistical Yearbook for the years of 2000–2020. The census method is used to calculate the production and cultivation area of seaweeds in Fujian and Guangdong, in which data are collected from each local farmer and then compiled (Gao, 2015). Mariculture of fish and seaweeds in Fujian and Guangdong is conducted in the coastal seawaters (Fig. 1). Annual productivity of fish and seaweeds was calculated by dividing production by cultivation area.

2.2. CNP release by fish mariculture

Released C, N and P by fish mariculture were calculated by the following formulas:

$$R_{C/N/P} = F \times C_{C/N/P} \times (1 - R) \tag{1}$$

$$F = P \times Fc \tag{2}$$

The Eqs. (1) and (2) are based on Lazzari and Baldisserotto (2008) and Gao et al. (2021), where $R_{C/N/P}$ is released C, N or P, F = feed amount, $C_{C/N/P}$ = content of C, N or P in feeds, R = retention rate of feed C, N or P in fish, P = fish production, and Fc = feed coefficient, the feed consumption per unit weight increase of fish. Here, the assumption is that 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, N and P in artificial feeds are 39.64 %, 7.40 % and 1.62 %, respectively (Table S1) and for wild feeds they are 13.67 %, 2.71 % and 0.53 %, respectively (Table S2), after integrating published data and our own data. The retention rate of feed C, N and P in fish is set at 30 % based on previous studies (Xu et al., 2007; Lazzari and Baldisserotto, 2008; Herath and Satoh, 2015). The feed coefficient is 1.52 and 6.49 for artificial and wild feeds, respectively (Gao et al., 2021). The data of fish production are from the China Fishery Statistical Yearbook for the years of 2004–2021. Please see Section 2.5 for the collection of published data on the contents of CNP in fish feeds.

In terms of the data on the contents of CNP in fish feeds from the present study, artificial fish feeds for different growth stages of *Larimichthys crocea* were obtained from State Key Laboratory of Large Yellow Croaker Breeding, China in April 2021. Content of carbon and nitrogen in feeds was measured by an Elementar Vario EL Cube (Elementar, Germany). Phosphorus content in feeds was determined according to the method in Solórzano and Sharp (1980).

2.3. CNP removal by farmed seaweeds

Removed C, N and P by farmed seaweeds were calculated by the following formula:

$$RA_{C/N/P} = (P_1 + P_2) \times C_{C/N/P} \tag{3}$$

where $RA_{C/N/P}$ is the removed amount of C/N/P by farmed seaweeds, P_1 is the production of harvested seaweeds, P_2 is the production of POM and DOM that are from cultivated seaweeds and sequestered in seawater for a long term, which also leads to the decreased of dissolved inorganic nitrogen (DIN) and dissolved inorganic phosphorus (DIP) in seawater, and $C_{C/N/P}$ represents the content of C/N/P in seaweed tissues. The estimation of P_2 is according to Gao et al. (2021). Therefore, removed CNP in this study

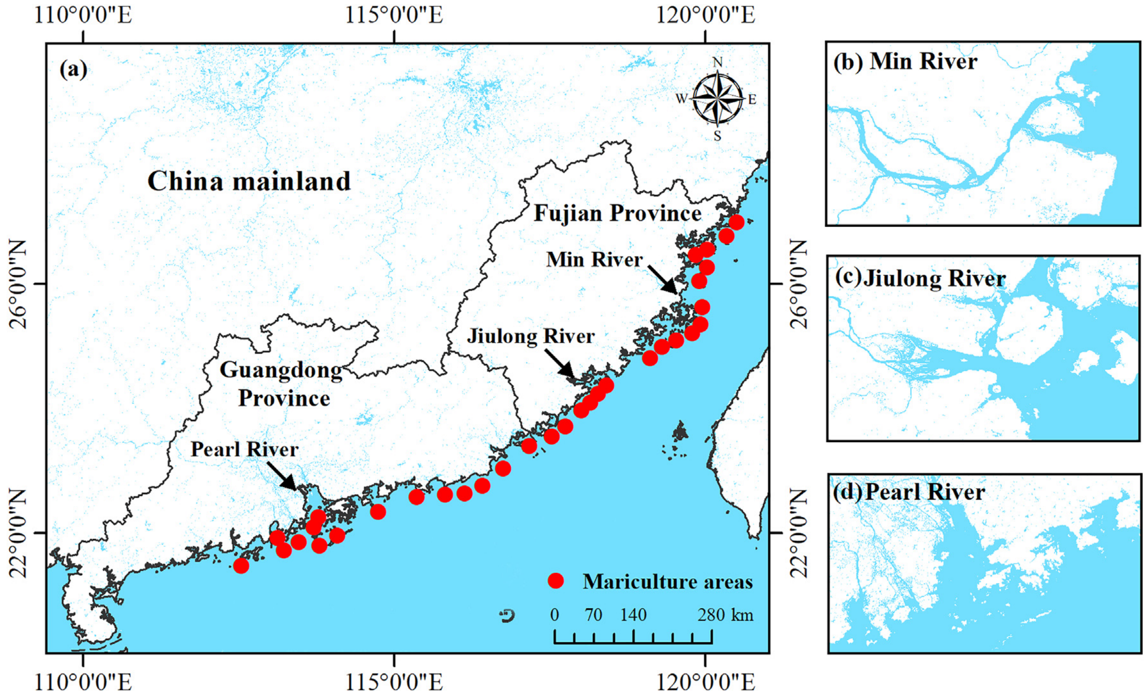

Fig. 1. Mariculture of fish and seaweeds in Fujian and Guangdong, China. The distribution of mariculture area is based on Fu (2020).

includes both CNP in harvested seaweeds and those in sequestered POM and DOM in seawater. The $C_{C/N/P}$ of seven farmed seaweeds was obtained based on the published data and our own data (Tables S3-S5). Please see Section 2.5 for the collection of published data.

In terms of the data from the present study, *Saccharina japonica* was collected in Sansha Bay in Fujian Province, China in May 2020 and *Betaphycus gelatinum* was collected in a seaweed farm in Changjiang, Hainan Province, China in May 2021. Seaweed samples were washed three times with autoclaved seawater and then dried for 24 h at 60 °C. The dry seaweeds were fumed with 12 M HCl for 24 h to remove PIC and DIC. Content of tissue carbon and nitrogen in seaweeds was measured by an Elementar Vario EL Cube (Elementar, Germany). Tissue phosphorus content was determined according to the method in Solórzano and Sharp (1980).

Net CNP fluxes were calculated by the removed CNP due to farmed seaweeds minus released CNP due to farmed fish.

2.4. Oxygen loss and generation

Oxygen loss caused by fish mariculture was calculated by the following equation (Redfield et al., 1963): $(CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 138O_2 \leftrightharpoons 106CO_2 + 16HNO_3 + H_3PO_4 + 122H_2O$ (4), which suggests that 138 mol O_2 are consumed when 106 mol carbon are mineralized. The mineralized/released carbon is based on the calculation in Section 2.2.

The amount of oxygen released by farmed seaweeds was calculated according to the photosynthesis equation: $6CO_2 + 6H_2O \rightarrow C_6H_{12}O_6 + 6O_2$ (5), when every tonne of carbon is fixed, 2.67 t of O_2 are generated. Here the involved carbon includes both harvested carbon and sequestrated carbon, which can be obtained from Section 2.3.

Net O_2 flux was calculated by the generated O_2 due to farmed seaweeds minus consumed O_2 due to farmed fish.

2.5. Data collection and calculation

The published data of CNP content in fish feeds and seaweed tissues were obtained through a search of ISI Web of Science, Scholar Google, and CNKI on July 15, 2022, using the terms "carbon, nitrogen, phosphorus, fish feeds and China" or "carbon, nitrogen, phosphorus, seaweeds and China" as keywords, respectively. Hence, all literature published July 15, 2022 was screened. Fifteen and 39 papers were finally selected respectively for fish feeds and seaweeds after double-checking the validity of the data. Data were expressed as means ± standard error. For those calculated parameters, e.g., CNPO release and removal, net CNPO fluxes, error propagation analysis was conducted.

3. Results and discussion

3.1. Production of maricultured fish and seaweeds in Fujian and Guangdong

Annual production of maricultured fish increased with year for both Fujian and Guangdong provinces (Fig. 2a). It increased from 0.09×10^6 to 0.36×10^6 t FW for Fujian and from 0.11×10^6 to 0.41×10^6 t FW for Guangdong during the years of 2003–2020. There was higher production in Guangdong than Fujian for each year. The pattern of farmed seaweeds is quite different from fish (Fig. 2b). Annual production of seaweeds in Fujian also increased with year, from 0.37×10^6 t DW in 2003 to 1.19×10^6 t DW in 2020. However, annual production of seaweeds in Guangdong reached the plateau in 2013 (0.074×10^6 t FW) and decreased to 0.064×10^6 t FW in 2020. More importantly, annual production of seaweeds in Fujian is 10–19 folds higher than that in Guangdong. Mariculture area of fish showed a noticeable fluctuation in Guangdong province (Fig. 2c). It maintained in a range of 19.54–20.44×10^3 ha during the years of 2003–2006, but sharply decreased to 14.48×10^3 ha in 2007 and increased to 18.51×10^3 ha in 2020 after fluctuating in a small range (12.21–15.08×10^3 ha). Compared to Guangdong, the mariculture area of fish in Fujian was much more stable, which slowly increased from 6.96×10^3 ha in 2003 to 11.22×10^3 ha in 2020 (Fig. 2c). Farming area of seaweeds in Fujian showed an increase firstly followed by a decrease during 2003–2007 (Fig. 2d). Afterwards, it steadily increased until 2020. The farming area of seaweeds in Guangdong showed a slowly decreasing trend, from 3.29×10^3 ha in 2003 to 1.89×10^3 ha in 2020 (Fig. 2d). Farming area of fish in Guangdong was higher than that in Fujian but farming area of seaweeds in Guangdong was much lower than that in Fujian.

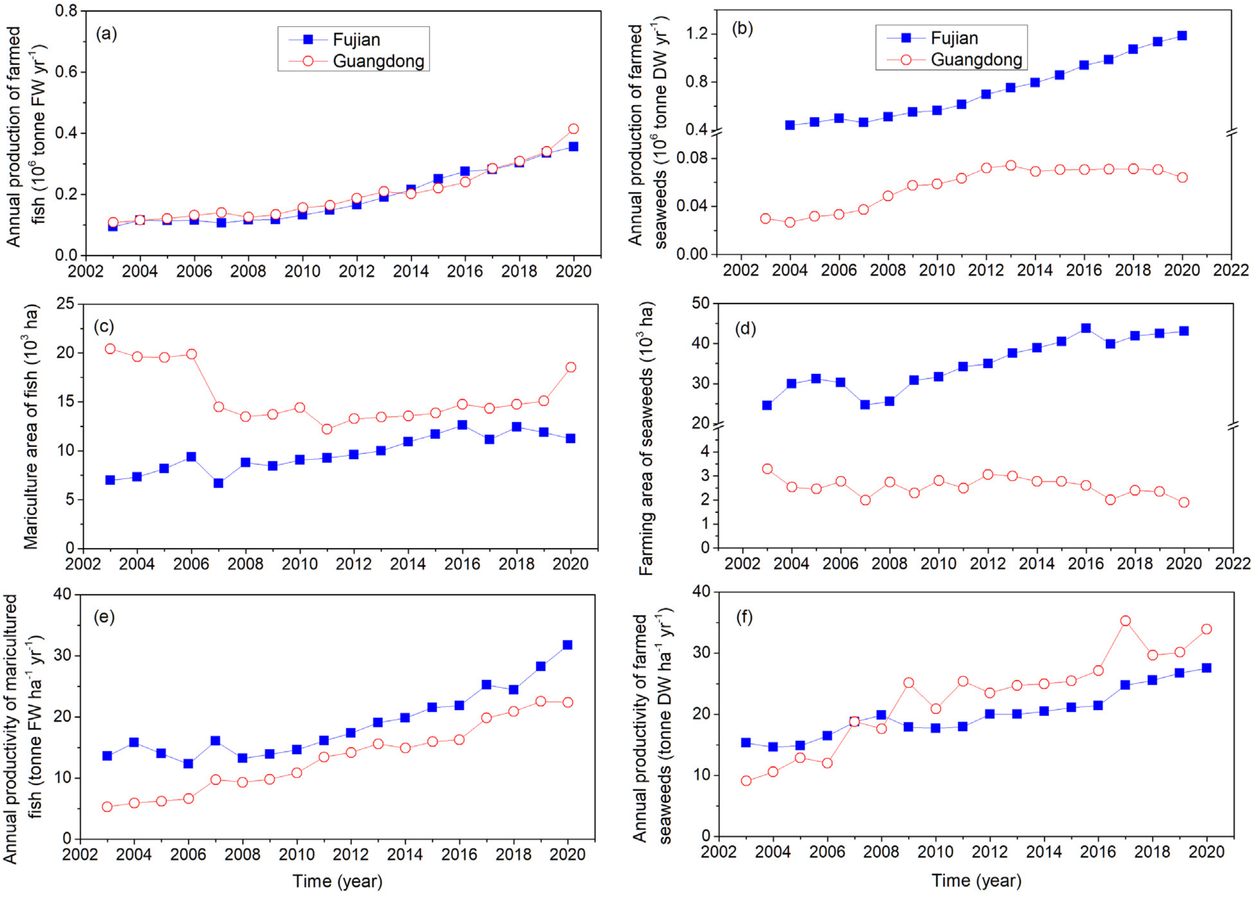

Fig. 2. Production, area and productivity of maricultured fish and seaweeds in Fujian and Guangdong, China during the period of 2003–2020. The data were based on fish and seaweeds with specific names in China Fishery Statistical Yearbook are included.

Annual productivity of maricultured fish steadily increased with year for both Fujian and Guangdong (Fig. 2e). Interestingly, Fujian had higher annual productivity of maricultured fish than Guangdong. In terms of annual productivity of farmed seaweeds, it also increased with year for the two provinces. Guangdong had a lower productivity than Fujian before 2009 but became higher afterwards until 2020 (Fig. 2f).

With more population living in the coastal areas, China's consumption of blue foods is shifting toward marine species (Crona et al., 2020). Delicious flavor and high nutrition also contributes to increased demand for seafood. Therefore, the fish production rapidly increased both in Guangdong and Fujian. The mariculture area for fish did not show a significant increase, which results in the increased productivity. The increased productivity can be attributed to improved culture technique and new strains with higher growth rate (Guan et al., 2020).

The patterns of seaweed production are different between Fujian and Guangdong. The increase in Fujian is higher. The earliest cultivated seaweeds, such as *Saccharina japonica* and *Undaria pinnatifida*, are cold temperate species, which were not suitable to be cultivated in subtropical zones, including Fujian and Guangdong (Xing et al., 2019). With the breakthrough in new strains that can be grown in hot waters, the production in Fujian has rapidly increased. The shrinkage of seaweed cultivation in Guangdong may be due to higher market prices for fish compared to seaweeds, which drives the shift toward fish mariculture.

3.2. CNP fluxes caused by fish and seaweed mariculture in Fujian and Guangdong

Carbon release by maricultured fish in Fujian slowly increased from 49.12 ± 0.74 Gg in 2003 to 61.04 ± 0.94 Gg in 2009 (Fig. 3a). Afterwards, it entered a track of rapid increase, from 68.91 ± 1.13 Gg in 2010 to 185.55 ± 3.18 Gg in 2020. *L. crocea* is the dominant species that was responsible for 49–57 % carbon release in Fujian. Snapper and sea bass were the second and third contributors, which contributed to 11–15 % and 10–13 % carbon

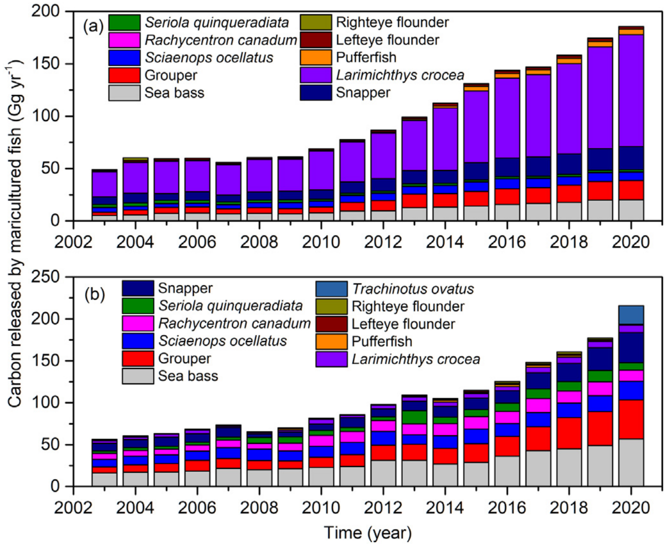

Fig. 3. Annual carbon release by maricultured fish in Fujian (a) and Guangdong (b), China during the period of 2003–2020.

release, respectively. Carbon release by maricultured fish in Guangdong increased from 56.28 ± 0.66 Gg in 2003 to 73.47 ± 0.89 Gg in 2007 and then from 65.20 ± 0.82 Gg in 2008 to 215.81 ± 2.51 Gg in 2020 (Fig. 3b). Different from Fujian, sea bass (25–32 %) contributed most in Guangdong, followed by grouper (13–23 %) and snapper (7–18 %).

The pattern of nitrogen release by maricultured fish is similar to carbon release (Fig. 4). Nitrogen release in Fujian slowly increased from 9.51 ± 0.12 Gg in 2003 to 11.82 ± 0.15 Gg in 2009 and rapidly increased from 13.34 ± 0.18 Gg in 2010 to 35.92 ± 0.51 Gg in 2020 (Fig. 4a). *L. crocea* was the biggest contributor, releasing 4.62 ± 0.11–20.65 ± 0.48 Gg N yr^{-1}, followed by sea bass (1.00 ± 0.02–3.90 ± 0.09 Gg N yr^{-1}) and grouper (0.58 ± 0.01–3.55 ± 0.08 Gg N yr^{-1}). For Guangdong, nitrogen release increased from 10.89 ± 0.11 Gg in 2003 to 13.53 ± 0.13 Gg in 2009 and rapidly increased from 15.76 ± 0.15 Gg in 2010 to 41.77 ± 0.40 Gg in 2020 (Fig. 4b). Sea bass released the most N (3.15 ± 0.07–10.98 ± 0.26 Gg yr^{-1}), followed by grouper (1.38 ± 0.03–9.02 ± 0.21 Gg yr^{-1}) and snapper (1.77 ± 0.04–6.97 ± 0.16 Gg yr^{-1}). Phosphorus release by maricultured fish in Fujian increased from in 1.92 ± 0.06 Gg 2003 to 7.27 ± 0.24 Gg in 2020 (Fig. 5a). *L. crocea* contributed 0.94 ± 0.05–4.18 ± 0.23 Gg P yr^{-1}, followed by snapper (0.29 ± 0.02–0.87 ± 0.05 Gg P yr^{-1}) and sea bass (0.20 ± 0.01–0.79 ± 0.04 Gg P yr^{-1}). Phosphorus release by maricultured fish in Guangdong increased from 2.21 ± 0.05 Gg in 2003 to 8.47 ± 0.19 Gg in 2020 (Fig. 5b). Sea bass contributed the most (0.64 ± 0.04–2.22 ± 0.12 Gg P yr^{-1}), followed by grouper (0.28 ± 0.02–1.83 ± 0.10 Gg P yr^{-1}) and snapper (0.17 ± 0.01–1.41 ± 0.08 Gg P yr^{-1}).

Fish mariculture in China is fed systems. For fed aquaculture, feed production can account for more than 70 % of CO_2 emissions for most species (Gephart et al., 2021). Additionally, uneaten feeds, faeces and excretion products can be mineralized by bacteria and CNP in feeds can be released into seawater. Due to low retention rate of feeds, most CNP in feeds are not assimilated by fish but released into the aquatic environments. Chatvijitkul et al. (2017) reported that around 60–80 % of nitrogen and phosphorus in feeds released into the aquatic environments. The released CNP could affect water quality. Han et al. (2021a, b) found that nutrient levels and pCO_2 in the fish-mariculture area were significantly higher than non-culture areas in Shen'ao Bay, a typical subtropical aquaculture bay in Guangdong, China. The released CNP amount also depends on fish production and feed coefficient. It has been documented that wild feeds usually has a much higher feed coefficient of 3.18–10.21 compared to artificial feeds (0.68–2.64) (Gao et al., 2021). High production and feed coefficients lead to a large number of feeds that are used in fish mariculture. This, combined with low retention rates of feeds, result in a large amount of CNP released into seawater. High production of *L. crocea*, snapper, sea bass, grouper lead to large release of CNP from feeds with these fishes. The N and P fluxes caused by fish mariculture in Fujian represent 41 % and 2.2 folds of Min River that is the biggest river in Fujian (Table 1). Meanwhile, the N and P fluxes caused by fish mariculture in Guangdong represent 7 % and 69 % of Pearl River that is the biggest river in Guangdong (Table 1). Furthermore, the fluxes of NP due to fish mariculture in the two provinces are both more than the fluxes of Jiulong River that is the second biggest river in Fujian (Table 1). Harmful algal blooms (HABs) commonly occur in these estuaries due to the input of nutrients from rivers (Li et al., 2019). Accordingly, the comparable nutrient fluxes of fish mariculture could also lead to HABs. The NP fluxes caused by fish mariculture in Fujian in this study is 8–28 % lower than those reported in Wang et al.'s (2019) study. The key reason for this difference is that different fish production was used. In this study, production of ten main fish species was used while production of all fish including unknown species was used in Wang et al.'s (2019) study. From this point of view, the present study may underestimate the CNP release by maricultured fish. The production of unknown species was not used in this study because this production is doubtfully high.

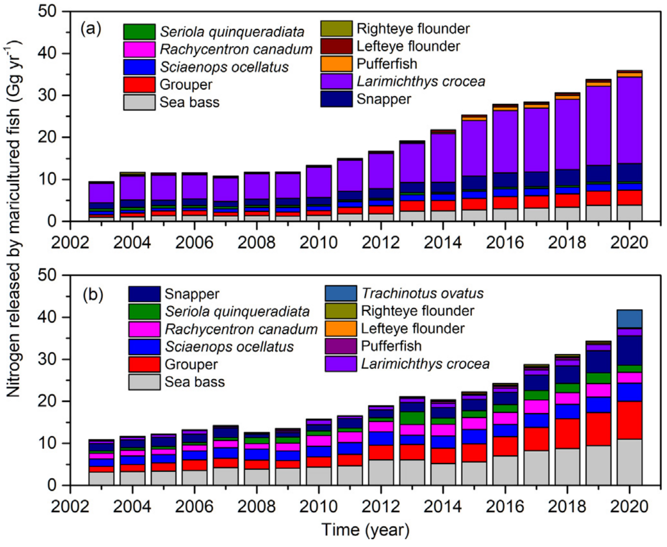

Fig. 4. Annual nitrogen release by maricultured fish in Fujian (a) and Guangdong (b), China during the period of 2003–2020.

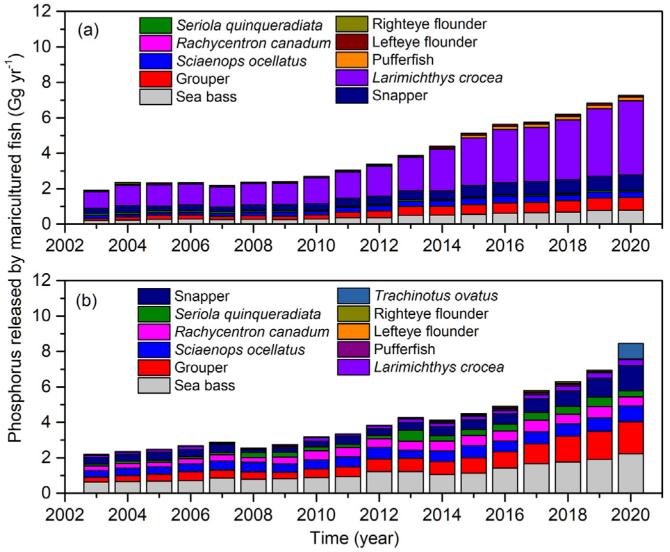

Fig. 5. Annual phosphorus release by maricultured fish in Fujian (a) and Guangdong (b), China during the period of 2003–2020.

Carbon removal by cultivated seaweeds in Fujian gradually increased during the past 18 years, from 167.95 ± 6.11 Gg in 2003 to 555.74 ± 16.45 Gg in 2020, which is an about 3-folds increase (Fig. 6a). The proportions of *S. japonica* and *Pyropia* decreased from 88 % and 11 % to 65 % and 8 %, respectively while the proportion of *G. lemaneiformis* increased from

Table 1
Comparison of nutrient fluxes of main rivers and mariculture in Fujian and Guangdong, China. The fluxes caused by mariculture are the averaged values ± SD of 2016–2020. DIN and DIP represents dissolved inorganic nitrogen and dissolved inorganic phosphorus, respectively.

Source	DIN flux (Gg yr^{-1})	DIP flux (Gg yr^{-1})	References
Min River	76.01	2.94	Liu et al., 2009
Jiulong River	20.57	0.53	Liu et al., 2009
Pearl River	481.11	9.45	Liu et al., 2009
Fish mariculture in Fujian	31.32 ± 1.55	6.34 ± 0.31	This study
Fish mariculture in Guangdong	32.02 ± 2.94	6.48 ± 0.59	This study
Seaweed culture in Fujian	52.25 ± 2.33	7.03 ± 0.28	This study
Seaweed culture in Guangdong	4.15 ± 0.08	0.38 ± 0.01	This study

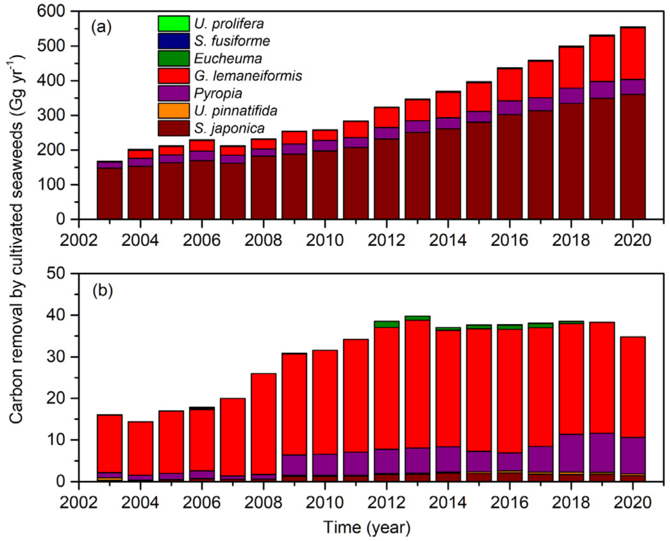

Fig. 6. Annual carbon removal by maricultured seaweeds in Fujian (a) and Guangdong (b), China during the period of 2003–2020.

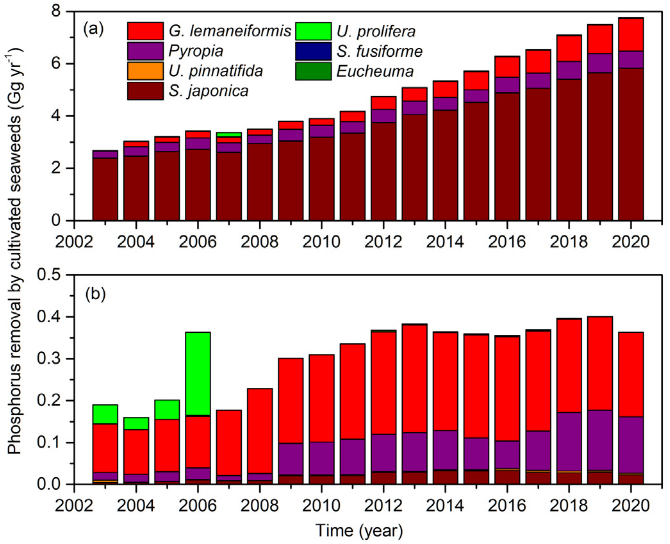

Fig. 8. Annual phosphorus removal by cultivated seaweeds in Fujian (a) and Guangdong (b), China during the period of 2003–2020.

0.03 % to 27 %. Different from Fujian, carbon removal by cultivated seaweeds in Guangdong reached the peak (39.81 ± 1.43 Gg) in 2013, then fluctuated in a narrow range (37.07 ± 1.31–38.51 ± 1.25 Gg), and decreased to 34.79 ± 1.13 Gg in 2020 (Fig. 6b). *G. lemaneiformis* is the dominant species for carbon removal in Guangdong during the past 18 years, followed by *Pyropia*. *S. japonica* ranked third in the years 2009–2020. Nitrogen removal by cultivated seaweeds in Fujian increased by ~3 fold (from 17.44 ± 1.89 Gg in 2003 and 58.44 ± 4.83 Gg in 2020) during the past 18 years (Fig. 7a). *S. japonica*, *G. lemaneiformis* and *Pyropia* almost covered all nitrogen removal in Fujian. Nitrogen removal by cultivated seaweeds in Guangdong increased from 2.11 ± 0.13 Gg in 2003 to 4.33 ± 0.26 Gg in 2013, and then slowly decreased to 3.89 ± 0.23 Gg in 2020 (Fig. 7b). *G. lemaneiformis* accounted for 46–93 %, followed by *Pyropia* (5–29 %) and *S. japonica* (1–5 %). Phosphorus removal by seaweed cultivation in Fujian also increased with year, from 2.68 ± 0.58 Gg in 2003 to 7.75 ± 1.41 Gg in 2020 (Fig. 8a). Phosphorus removal in Guangdong increased from 0.19 ± 0.02 Gg in 2003 to 0.36 ± 0.03 Gg in 2006, and then slowly to 0.38 ± 0.03 Gg in 2013 after a sharp decrease in 2007 (Fig. 8b). The peak (0.40 ± 0.03 Gg) occurred in 2019 and then decreased to 0.36 ± 0.03 Gg in 2020 (Fig. 8b).

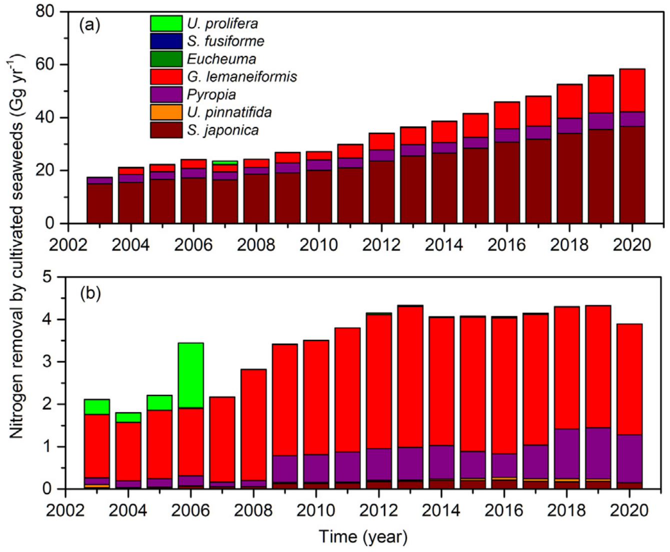

Fig. 7. Annual nitrogen removal by cultivated seaweeds in Fujian (a) and Guangdong (b), China during the period of 2003–2020.

Photosynthesis of seaweeds can uptake CO_2 in seawater and transform it into organic carbon. When seaweeds are harvest, these CO_2 are removed from seawater. Therefore, seaweed cultivation can alleviate coastal acidification, reduce pCO_2 in seawater and enhance carbon sink of seas (Jiang et al., 2020; Gao et al., 2021, 2022a, b; Xiao et al., 2021). The CO_2 removal amount rests mainly with production of seaweeds although different carbon contents among seaweeds also contribute (Gao et al., 2021). The highest production of *S. japonica* and *G. lemaneiformis* in Fujian and Guangdong resulted in highest CO_2 removal in these two provinces, respectively. Different from continuous increase of CO_2 removal by seaweed cultivation in Fujian, CO_2 removal stopped increasing after 2013 in Guangdong, which can be attributed to stagnant production of seaweed. Apart from CO_2 removal, seaweeds can assimilate DIN and DIP in seawater during growth and removal them from seawater when they are harvested, which can counteract eutrophication caused by effluent discharge from lands or animal mariculture (Gao et al., 2022a, b). The lower NP removal amount in Guangdong is mainly caused by lower seaweed production. Although the CNP fluxes in Guangdong are lower than those in Fujian, these values are higher than those reported in Hu et al.'s (2022) study. For instance, the CNP fluxes by farmed seaweeds in Guangdong in 2019 are 34.79, 3.89 and 0.36 Gg in this study, while they are 21.51, 2.89 and 0.32 Gg in Hu et al.'s (2022) study. The main reason for these gaps is that different methods are used. Hu et al.'s (2022) study excludes the CNP in seaweeds that are transported to deep oceans and in refractory form, which may underestimate the CNP removal by farmed seaweeds. In addition to seaweed cultivation, the combination of microalgae and nitrifier-enriched-activated-sludge is deemed as an effective approach to remove CNP in wastewater (Sepehri and Sarrafzadeh, 2018; Sepehri et al., 2020).

Net CNP fluxes combining mariculture of fish and seaweeds were calculated (Fig. 9). The carbon flux in Fujian showed a net sink and it increased from 118.84 ± 6.15 Gg in 2003 to 370.19 ± 16.75 Gg in 2020 (Fig. 9a). In contrary, the carbon flux in Guangdong showed a net source. It also increased with year, from −40.25 ± 0.92 Gg in 2003 to −181.02 ± 2.75 Gg in 2020. The pattern of nitrogen flux is similar to carbon (Fig. 9b). The nitrogen flux increased from 7.93 ± 1.90 Gg in 2003 to 22.52 ± 4.86 Gg in 2020 in Fujian and from −8.78 ± 0.17 Gg in 2003 to −37.88 ± 0.46 Gg in 2020 in Guangdong. The pattern of phosphorus flux is different from carbon or nitrogen (Fig. 9c). The P flux in Fujian in around zero, suggesting that P release by fish mariculture is neutralized by P removal by seaweeds. Seaweeds can neutralize P released by fish

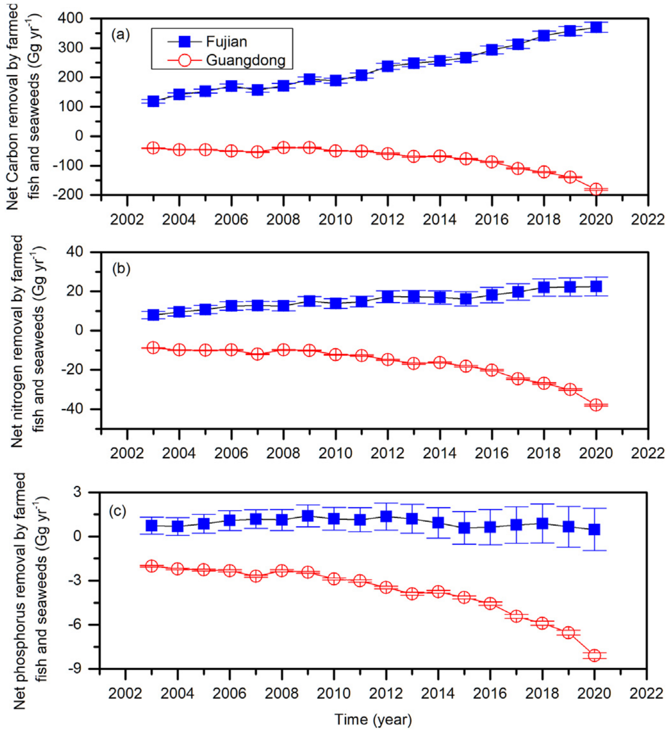

Fig. 9. Annual net flux for carbon (a), nitrogen (b), and phosphorus (c) caused by maricultured fish and seaweeds in Fujian and Guangdong, China during the period of 2003–2020.

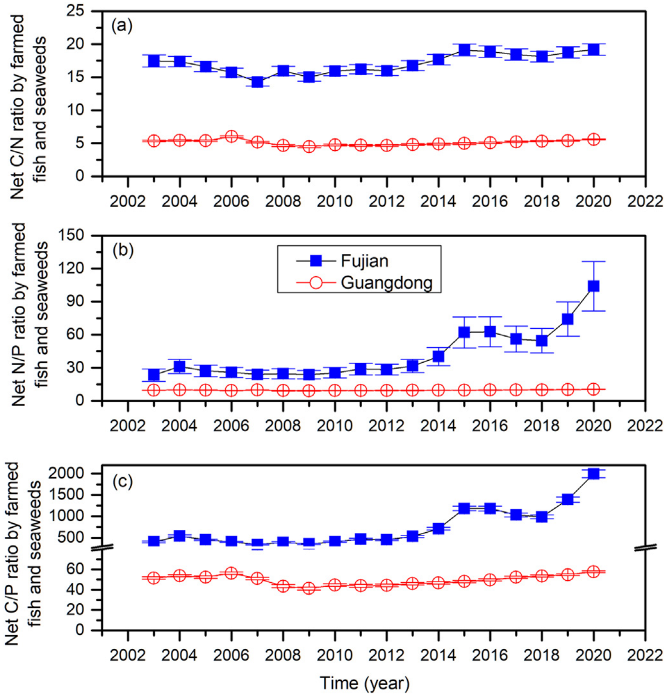

Fig. 10. Net C/N/P mole ratios by farmed fish and seaweeds in Fujian and Guangdong, China during the period of 2003–2020.

mariculture via direct or indirect ways. It has been shown that seaweeds can utilize organic P directly (Li et al., 2016), although the more common way that seaweeds utilize P is to uptake inorganic P (Roleda and Hurd, 2019). Organic P in fish feeds can be mineralized into inorganic P by bacteria and then be removed by seaweeds (Zhou et al., 2006). However, net P flux in Guangdong showed a remarkable source and increased from −2.02 ± 0.05 Gg in 2003 to −8.09 ± 0.19 Gg in 2020.

The combination of fish and seaweed mariculture leads to positive CNP removal in Fujian but net CNP release in Guangdong. This difference can be attributed to contrasting mariculture structure. In Fujian, production of seaweeds is 2.7–3.6 folds of fish production while it is 9–21 % of fish production in Guangdong. Therefore, to achieve CNP neutrality within mariculture, Guangdong needs to restrict its fish mariculture and/or expand its seaweed cultivation. Given lower market prices of seaweeds compared to fish, the transfer from fish to seaweeds mariculture may be difficult. Therefore, appropriate economic compensation is critical for this transfer (Duarte et al., 2017). Hu et al. (2022) has estimated that the economic compensations for *S. japonica*, *Gracilaria* spp. and *Eucheuma denticulatum* are $1.39/kg, $1.50/kg, and $1.27/kg respectively based on the market price in China.

The net C/N/P mole ratios by farmed fish and seaweeds based on the data of Fig. 9 were calculated (Fig. 10). The net C/N mole ratios in Fujian ranged from 14 to 19 while they were 5–6 in Guangdong (Fig. 10a). The net N/P mole ratios in Fujian had a large range of 24–104 while they were stable and lower (9–10) in Guangdong (Fig. 10b). The case for C/P ratios was similar to N/P; they changed from 343 to 1992 in Fujian but 41–58 in Guangdong (Fig. 10c). The results above show contrasting differences in C/N/P ratios between two areas; Fujian had much higher C/N/P ratios than Guangdong. The C/N/P ratios in Fujian are also higher than the Redfield ratio (106:16:1), which can be attributed to the dominant seaweed cultivations. Seaweeds usually have higher C/N/P ratios compared to the Redfield ratio (Bach et al., 2021; Gao et al., 2021). The higher C/N/P ratios mean seaweeds can remove more C and N when using 1 mol P, which is beneficial for the mitigation of ocean acidification and eutrophication because coastal seawaters in China are P limited (Wang et al., 2021). On the other hand, the lower C/N/P ratios in Guangdong compared to the Redfield ratio can be due to the dominant fish mariculture. Fish feeds have higher N and P contents compared to seaweeds (Table S1 & Gao et al., 2021), leading to the lower C/N/P ratios in Guangdong. This lower C/N/P ratios caused by farmed fish and seaweeds were consistent with the case of seawater samples in the study area (Yang et al., 2020).

3.3. O_2 fluxes due to fish and seaweed mariculture in Fujian and Guangdong

O_2 loss by maricultured fish in Fujian increased from 170.51 ± 2.57 Gg in 2003 to 644.18 ± 11.05 Gg in 2020 (Fig. 11a). *L. crocea* contributed most (49–57 %), followed by sea bass (10–13 %) and snapper (6–8 %).

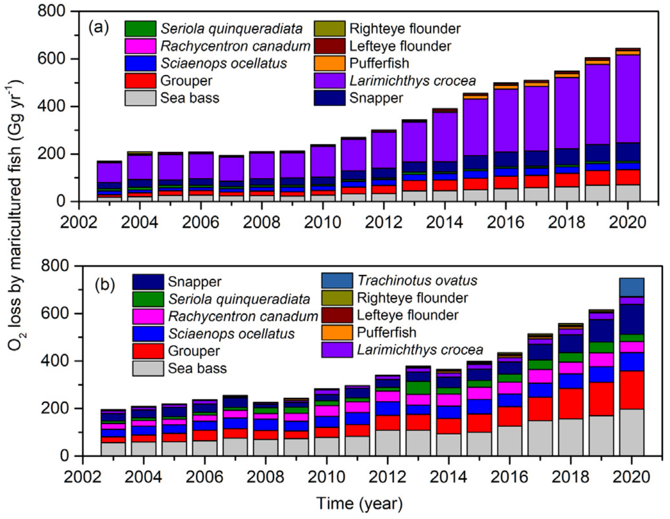

Fig. 11. Annual oxygen consumption by maricultured fish in Fujian (a) and Guangdong (b), China during the period of 2003–2020.

O_2 loss by maricultured fish in Guangdong increased from 195.39 ± 2.28 Gg in 2003 to 749.23 ± 8.71 Gg in 2020 (Fig. 11b). Sea bass contributed most (56.58 ± 1.60–196.98 ± 5.56 Gg), followed by snapper (31.71 ± 0.90–125.07 ± 3.53 Gg) and grouper (24.68 ± 0.70–161.76 ± 4.56 Gg). O_2 generation by farmed seaweeds in Fujian increased from 447.88 ± 16.29 Gg in 2003 to 1481.97 ± 43.86 Gg in 2020 (Fig. 12a). *S. japonica* contributed most O_2 generation (393.51 ± 16.27–961.59 ± 39.77 Gg), followed by *G. lemaneiformis* (0.14 ± 0.01–397.21 ± 18.44 Gg) and *Pyropia* (47.80 ± 0.64–127.24 ± 1.70 Gg). O_2 generation by farmed seaweeds in Guangdong increased from 42.74 ± 1.72 Gg in 2003 to 106.15 ± 3.82 Gg in 2013 and slowly decreased to 92.78 ± 3.02 Gg in 2020 (Fig. 12b). *G. lemaneiformis* accounted for 69–93 % of O_2 generation in Guangdong. The net O_2 flux combining maricultured fish and seaweeds showed a source in Fujian and a sink in Guangdong (Fig. 12c). The net O_2 flux increased from 277.37 ± 16.49 Gg in 2003 to 837.79 ± 45.23 Gg in 2020 in Fujian and from −152.65 ± 2.86 Gg in 2003 and to −656.44 ± 9.21 Gg in 2020 in Guangdong.

Due mainly to ocean warming, the global oceanic oxygen inventory has declined by 2.1 % during the past five decades (Schmidtko et al., 2017). Furthermore, coastal deoxygenation is proceeding at more rapid rates due to eutrophication and ocean circulation shift (Rabalais et al., 2014; Claret et al., 2018). Since the mid-20th century, over 700 coastal systems have reported low-oxygen areas, which are also referred to as “dead zones” (Limburg et al., 2020). Ocean deoxygenation can impose severe harmful impacts on survival and development of many organisms, including corals, fish, shellfish, etc. (Hughes et al., 2020; Limburg et al., 2020). The decomposition of uneaten feeds in fed aquaculture by bacteria can generate O_2 loss, leading to hypoxia environments that are adverse to growth of farmed aquatic animals (Gao et al., 2022a, b). In this study, fish mariculture reduced O_2 concentrations by 3.01 and 1.55 mg L^{-1} d^{-1} in Fujian and Guangdong in 2020, respectively, which is equivalent to reducing the dissolved oxygen in aquaculture waters (4 m in depth) by 19 % and 37 % daily if the dissolved oxygen (DO) level is 8.16 mg L^{-1} and the air-sea exchange is excluded (Gao et al., 2021). The higher production along with intensive photosynthesis of cultivated seaweeds completely neutralized the O_2 loss due to fish mariculture in Fujian and resulted in net O_2 generation. On the other hand, the small culture scale of seaweeds could not neutralize O_2 loss caused by fish mariculture in Guangdong, net O_2 loss increasing with year. Therefore, the adjustment of mariculture structure can alleviate not only eutrophication but also deoxygenation of coastal waters.

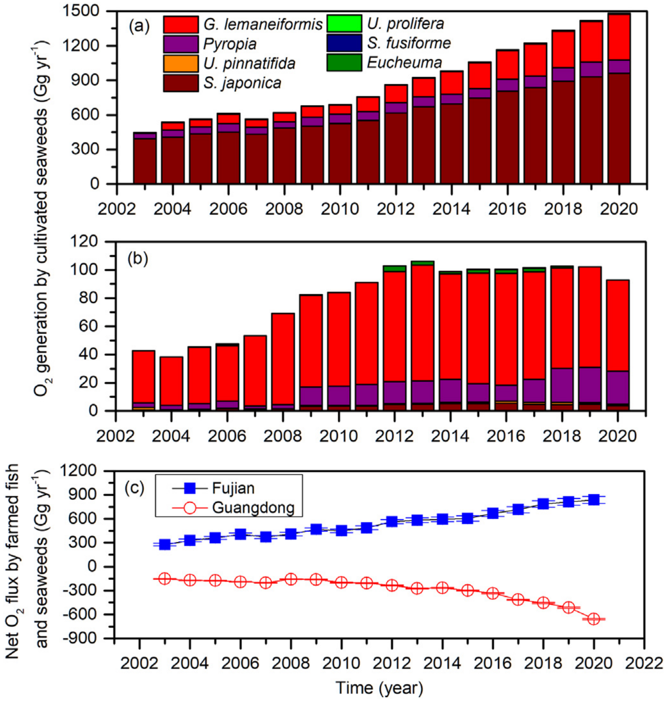

Fig. 12. Annual oxygen generation by farmed seaweeds in Fujian (a) and Guangdong (b), and net oxygen flux by farmed fish and seaweeds in these two provinces (c) in China during the period of 2003–2020.

4. Conclusions

The contribution of fish and seaweed mariculture to coastal CNPO fluxes in two important aquaculture areas, Fujian and Guangdong, China, has been investigated for the first time. Fish mariculture in the two areas both releases a large amount of CNP, which is comparable to the fluxes of the main rivers in the two provinces. In terms of seaweed cultivation, in Fujian it can completely remove/replenish all CNP/O released/consumed by fish mariculture, leading to a net CNP sink and an O_2 source, and contributing to mitigating eutrophication and de-oxygenation in Fujian. On the other hand, less seaweed cultivation in Guangdong cannot completely neutralize CNPO released/consumed by fish mariculture, resulting in a net CNP source and an O_2 sink. The increased CNP release and O_2 consumption after combining fish and seaweed mariculture in Guangdong may lead to or exacerbate eutrophication and de-oxygenation in Guangdong, disturbing healthy ecosystems. Although this is a case study in two provinces, China, the methodologies in the present study can be applied to mariculture in other areas in China or in the world. The findings and methodologies supply helpful information and tools for assessing environmental impacts of mariculture and adjusting mariculture culture to achieve the neutrality of essential biogenic elements. This study focuses on maricultured fish and seaweeds, excluding wild fish and seaweeds. Future study can include wild fish and seaweeds, assessing the contribution of them to the cycle of biogenic elements, and producing a more comprehensive picture.

CRediT author contributions statement

Yonglong Xiong: Investigation, Methodology, Formal analysis, Writing- original draft preparation, Writing- reviewing and editing. Lin Gao: Investigation, Methodology, Writing- reviewing and editing. Liyin Qu: Methodology, Visualization, Writing- reviewing and editing. Juntian Xu: Methodology, Writing- reviewing and editing. Zengling Ma: Methodology, Writing- reviewing and editing. Guang Gao: Conceptualization, Methodology, Formal analysis, Visualization, Writing- original draft preparation, Writing- reviewing and editing, Supervision, Funding acquisition.

Data availability

Data will be made available on request.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.159056.

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