https://doi.org/10.5194/egusphere-2023-3155
Preprint. Discussion started: 20 February 2024

Assessment of nutrient cycling in an intensive mariculture system

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Abstract. Rapid expansion of mariculture during past decades has raised substantial concerns about impacts on the coastal environment, notably eutrophication. This study focuses on one of the world's highest density mariculture sites, Sansha Bay, Fujian Province, China, featuring integrated multi-trophic aquaculture practices involving croaker, kelp and oyster, based on examination of nutrient distributions and releases. A two-endmembers-mixing model showed significant addition of dissolved inorganic nitrogen (DIN; 6.9 ± 4.1 μmol L^{-1}) and phosphorus (DIP; 0.45 ± 0.29 μmol L^{-1}) associated with mariculture activities in spring 2020. A mass balance model estimated an annual release of N and P from cage fish farming systems fed with mixed trash fish feed and formulated feed of $(2.42 \pm 0.15)\times10^4$ tons and $(5.33 \pm 0.37)\times10^3$ tons, respectively. Of the total feed input, 52.8 ± 4.7 % of DIN and 33.0 ± 3.7 % of DIP were released into seawater, values much higher than the riverine input and exchange with offshore coastal waters. A co-culture strategy involving kelp and oyster production in 2020 removed $(1.08 \pm 0.01)\times10^3$ tons of N and $(1.56 \pm 0.08)\times10^2$ tons of P, respectively. Therefore, adjusting feed strategies and improving feed conversion rates could alleviate eutrophication caused by mariculture expansion in this ecosystem.

1 Introduction

The global demand for aquatic products during past decades has fueled the rapid development of coastal aquaculture, making it the fastest growing sector of food production worldwide (Chen and Qiu, 2014; Subasinghe et al., 2009). Global aquaculture production has increased by approximately 600 %, from 17.3 million tons (MT) in 1990 to 120.1 MT in 2019 (Fao, 2021). Mariculture accounts for ~48 % of total aquaculture production with a growth rate of 2.36×10^6 tons yr^{-1}, which outcompetes that of freshwater culture (1.92×10^6 tons yr^{-1}) over the past decade (Fao, 2021). Notably, China emerged as the largest global aquaculture producer since 1989, contributing to ~57 % of total world production in 2019, i.e., a tenfold increase over the past two decades (Fao, 2021). This growth trend is expected to continue globally, most notably in developing countries (Diana et al., 2013).

Mariculture development has provided great economic and social benefits; the environmental impacts of using formulated and trash fish feeds are of critical concern (Cao et al., 2015; Cao et al., 2017). Cage farming for example, has been reported to contribute additional pressures to the already impacted coastal marine environment (Chopin et al., 2001; Dai et al., 2023). Many studies have reported eutrophication in waters used for mariculture (Bouwman et al., 2013; Schneider et al., 2005; Wang et al., 2012; Skriptsova and Miroshnikova, 2011). While the causes of eutrophication may be complex, they are often attributed to the low utilization efficiency of fish feeds (Nederlof et al., 2021). Although the Norwegian salmon farming industry has taken important steps to reduce nutrient release, such as the optimization of feed composition and improvement of the feed conversion rate (FCR), which has attained a range of 1.06-1.17. However, such FCR values still translate into the release into the environment of 58 %-62 % nitrogen (N), 79 %-81 % phosphorus (P) and 58 %-62 % carbon (C) of the total feed input (Wang and Olsen, 2023). These unutilized nutrients can accumulate in both the water and sediment, eventually altering the aquatic and sedimentary environment, especially in closed and semi-closed bays where water exchange is limited. Consequently, harmful algal blooms (HABs) and seasonal hypoxia frequently occur in these coastal waters (Anderson et al., 2002; Aure and Stigebrandt, 1990; Li et al., 2014b; Breitburg et al., 2018). Mariculture can also result in varying environmental impacts. Thus, He et al. (2022) reported that suspended aquaculture weakens the onshore current near the aquaculture boundary and upwelling within the offshore aquaculture area, resulting in a reduction of ~60 % in the nutrient supply. Therefore, the effects of mariculture systems on coastal nutrient dynamics still remain inconclusive and quantitatively uncertain.

A novel development in the mariculture sector to increase its sustainability is to adopt an integrated multi-trophic aquaculture (IMTA) approach (Jiang et al., 2012; Granada et al., 2016; Chopin et al., 2012; Campanati et al., 2021; Wu et al., 2015; Wei et al., 2017). Macroalgal cultivation is often included in IMTA systems to alleviate environmental impacts, as it allows efficient nutrient recycling and transformation, because of its pronounced nutrient absorption and storage capacity in tissues, while also offering an ecologically friendly option (Yang et al., 2006; Marinho-Soriano et al., 2009). Bivalve

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suspension-feeders provide another effective trophic pathway for the removal of suspended particulates and phytoplankton from the water in IMTA systems, leading to the conversion of nutrients into biomass (Martinez-Porchas and Martinez-Cordova, 2012; Carboni et al., 2016). However, the IMTA system is a complex ecosystem, in which the efficiency of nutrient removal by macroalgae and nutrient addition from fish cage farming depend on the feed type, the species under cultivation and the feeding strategy, variables which have generally not been fully studied. It is thus crucial to assess nutrient cycling in IMTA systems, and determine the nutrient budget by also considering other sources of nutrients, as these have important policy implications (Campanati et al., 2021; Carballeira Braña et al., 2021).

We here focus on Sansha Bay located in Fujian Province, China, one of the highest density IMTA system worldwide, featuring the world's largest croaker (*L. crocea*) cage culture as a case study to shed light on the interactions between a mariculture system and the environment (Song et al., 2023). Production of this fish species accounts for 72.3 % of total fish aquaculture production in Sansha Bay, requiring millions of tons of feed annually (Xie et al., 2020). Therefore, the development of this mariculture industry and the associated use of feeds might be a major contributor to nutrient pollution in this bay. However, to date, little is known about how the N and P budgets in the fish farming system are affected by different types of feeds. Finally, the role of macroalgae and bivalve mollusks in nutrient removal in Sansha Bay remains unknown.

This study aims to provide a scientific basis for assessing the role of mariculture as a driver of changes in the coastal environment and proposes to support science-based decision-making for transforming the mariculture activities in coastal waters into a more sustainable model. We examine semi-quantitatively the variation in nutrient characteristics in spring. Subsequently, we establish a mass balance of N and P to assess the release of nutrients from fish farming systems using different feeds. We also analyze the external nutrient input/removal in this ecosystem that is affected by intensive mariculture (fish, kelp and oyster farming), riverine input and exchange with offshore coastal waters.

2 Materials and methods

2.1 Study area and maricultural practices

Sansha Bay, located on the northeast coast of Fujian Province, China, is a semi-enclosed bay comprised of several secondary bays, such as Baima Harbor, Yantian Harbor, Dongwuyang, Guanjingyang, and

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Sandu'ao (Fig. 1) (Lin et al., 2017a). The surface area of the bay is ~675 km^2 (Lin et al., 2016), with a tortuous 553 km coastline (Yan and Cao, 1997). It has only one outlet, ca. 2.9 km wide, to the southern East China Sea (ECS) (Wang et al., 2018). Due to its geographical features, Sansha Bay has historically served as a natural sheltered bay (Lin et al., 2017a), that fostered the vigorous development of the mariculture industry (Xie et al., 2020; Wei et al., 2017; Wu et al., 2015). Sansha Bay is surrounded by mountains with outcroppings of medium acidic volcanic rocks(Yan and Cao, 1997). It receives several mountain rivers, among which the Jiaoxi and Huotongxi Streams provide the largest runoff. The annual freshwater discharge of the Jiaoxi Stream is 6.97×10^{10} m^3 (Huang and Ding, 2014), while that of the Huotongxi Stream is 2.73×10^9 m^3 (Li et al., 2014a). The bay is characterized by strong semidiurnal tides, with an average tidal range exceeding 5 m (Lin, 2014). The tidal prism is ~2.68×10^9 m^3 (Wang et al., 2011), and the water depth within the bay ranges from a few meters to 90 m. The region experiences prevailing seasonal monsoons, influenced by the winter northeast wind and summer southwest wind. As a result, the Bay water is a dynamic mixture of the China Coastal Current (CCC), the Taiwan Warm Current (TWC), and the South China Sea Warm Current (SCSWC), with seasonal variation in their proportions (Xu and Xu, 2013; Wang et al., 2018; Yang et al., 2008; Huang et al., 2019). The unique natural geomorphology and intensive mariculture activities have influenced the flow field and water exchange in Sansha Bay. Cage aquaculture weakens the overall flow within the bay, but appears to strengthen the water flow among individual cages (Lin et al., 2019), while the difference in residual water level between the outer and inner bay is more influenced by cross-shore winds (Lin et al., 2017b). Moreover, the seawater half-exchange time in the main channel is less than 10 days, while it exceeds 30 days at the head of the bay (Wu et al., 2015; Lin et al., 2017a; Lin et al., 2019).

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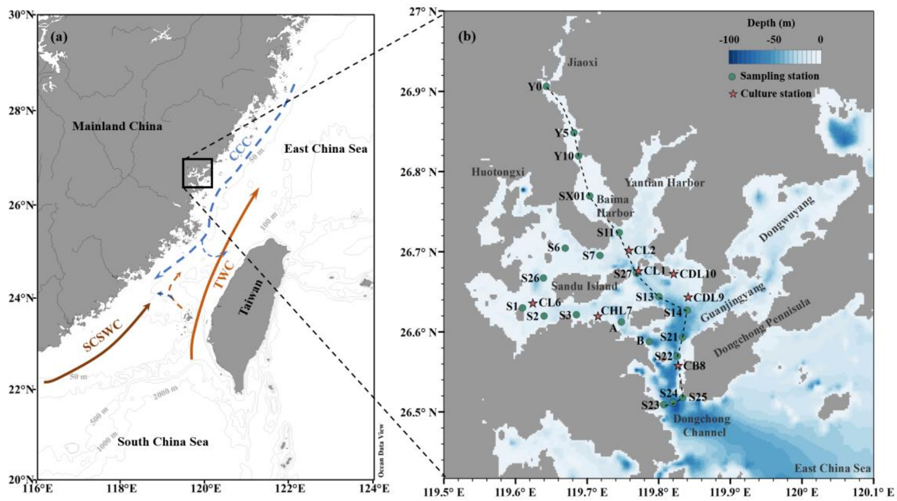

Figure 1. (a) Map of the study area and schematic of local currents. Bathymetric data reflect the latest General Bathymetric Chart of the Oceans (GEBCO) grid data. Blue dashed lines, red and brown solid lines represent the China Coastal Current (CCC), Taiwan Warm Current (TWC) and South China Sea Warm Current (SCSWC). (b) Sampling stations (dark green circles) in Sansha Bay during May 2020. Also shown are mariculture zones (red stars). Jiaoxi and Huotongxi are two main streams discharging into Sansha Bay.

Aquaculture activities utilize ~69 % of Sansha Bay's area. The main cultured species include the *L. crocea*, oyster (*C. angulate*), kelp (*L. japonica*), and others. Cages used in shallow and deep-water have an area of 4.5×10^6 m^2 and 1.39×10^6 m^3, respectively as of 2020 (data from Xingchun Wang, Min Dong Fisheries Research Institute, Fujian Province). Specifically, the farming densities for fish 100-200 g and 500 g wet weight approach ca. 600 and 160 fish m^{-2}, respectively (Liu et al., 2022). Trash fish feeds constitute the primary bait for breeding *L. crocea*, accounting for ca. 80 % of the feed source (Song et al., 2023). Based on remote sensing data from 1999 to 2020 (detailed in supplementary text S1), mariculture gradually expanded from nearshore to offshore waters. Cage culture began sporadically around several islands in deep-waters as early as 1999 and then expanded linearly and widely in the nearshore and surrounding areas, while macroalgal culture was scattered throughout the region in 2020 (Fig. 2(a)). The productions of *L. crocea* and kelp were gradually increasing in the marine area off Ningde City (production data from the Statistical Yearbooks of Ningde City, http://tjj.ningde.gov.cn/xxgk/tjxx/tjnj/). The total production of *L crocea* increased from 56 tons in 1990 to 1.785×10^5 tons in 2020. Due to differences in the statistical methodology for *L. crocea* and oyster (the latter includes the shell weight) since 1997, oyster production showed a linear increase (Fig. 2(b)). A synthesis of literature data(Cai, 2007; Zheng, 2017; Liu, 2013; Wang et al., 2009) reveals that nutrient

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concentrations increased steadily from 1984 to 2020, with an annual mean growth rate of 3.42 ± 1.50 μmol L^{-1} for dissolved inorganic nitrogen (DIN) and 0.15 ± 0.01 μmol L^{-1} for dissolved inorganic phosphorus (DIP). Thus, DIN and DIP concentrations have increased more than two and three folds, respectively, since the 1980s (Fig. 2(c)).

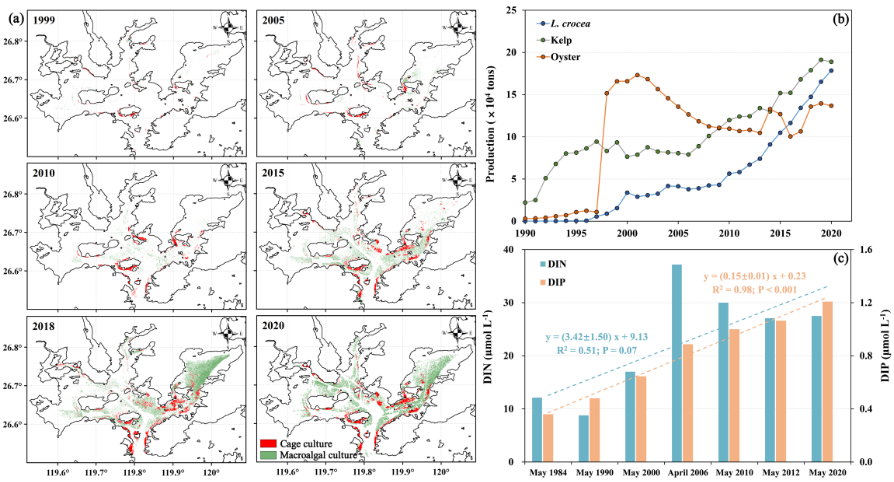

Figure 2. (a) Classification of cage culture and macroalgal culture in Sansha Bay from 1999 to 2020 based on Lansat and Sentinel-2 remote sensing data used to perform support vector machine (SVM) classification, in which red represents cage culture and green represents macroalgal culture. (b) Changes in annual production of *L. crocea*, kelp and oyster from 1990 to 2020. (c) Dissolved inorganic nitrogen (DIN) and phosphorus (DIP) concentrations in Sansha Bay from 1984 to 2020. Light blue and orange dashed lines represent the linear regression lines over time of DIN and DIP concentrations, respectively; linear regression equations are also shown (R^2 = coefficient of determination).

Notes: Nutrient 1984 data are from the *National Comprehensive Survey of Coastal Zone and Tiled Resources*; 1990 data are from volume 7 of *Gulf Annals of China*; 2000 data are from Cai (2007); 2006 data are from Wang et al. (2009); 2010 data are from Liu (2013); 2012 data are from Zheng (2017); 2020 data are from this study. Due to the lack of data in May 2006, the substitution of April data could be a potential reason for the elevated DIN concentration.

2.2 Sampling and methods of analysis

The research cruise was conducted onboard the R/V *Funing 11* during May 18-28, 2020. Water temperature and chlorophyll *a* (Chl-*a*) concentrations were measured continuously using a multi-parameter instrument (YSI Model 5065, YSI Co., USA). Discrete water samples for salinity (S) determination were collected using Niskin bottles and analyzed in the laboratory using a portable salinometer (Model Multi 340i, WTW Co., Germany). Simultaneously, water samples for analysis of

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nitrate (NO_3^-), nitrite (NO_2^-), ammonium (NH_4^+), phosphate (PO_4^{3-} or DIP), and silicate ($Si(OH)_4$) concentrations were taken and filtered through a 0.45 μm pore size cellulose acetate membrane filter into 125 mL high density polyethylene plastic bottles.

Nutrient samples, except those for $Si(OH)_4$, were frozen onboard and then measured at a land-based laboratory in Xiamen University. Samples for $Si(OH)_4$ analysis were preserved with 100 μL of chloroform and were refrigerated at 4 °C until analysis. Nutrient concentrations were measured using an Auto Analyzer 3 (AA3) instrument (Bran+Luebbe, Germany) with a detection limit of 0.04 μmol L^{-1} for NO_2^-, 0.1 μmol L^{-1} for NO_2^-+NO_3^-, 0.08 μmol L^{-1} for PO_4^{3-} and 0.16 μmol L^{-1} for $Si(OH)_4$. NH_4^+ concentrations were determined using an integrated syringe-pump-based environmental-water analyzer (iSEA) based on the improved indigo phenol blue spectrophotometric method, with a detection limit of 0.15 μmol L^{-1} (Li et al., 2019). Samples for total alkalinity (TA) analysis were stored in 250 mL PYREX® borosilicate glass bottles and preserved with 250 μL of saturated $HgCl_2$ solution (Guo and Wong, 2015). Total alkalinity was measured using potentiometric titration with 0.1 mol L^{-1} hydrochloric acid, and certified reference material for carbon dioxide from Andrew G. Dickson (the Scripps Institution of Oceanography, University of California, San Diego, USA) was adopted to standardize measurements (Cai et al., 2004). Both precision and accuracy were ± 2 μmol kg^{-1}. Dissolved oxygen (DO) concentration from discrete water samples was measured onboard using the spectrophotometric Winkler method (Labasque et al., 2004). Meanwhile, biological samples were also collected, such as those of *L. crocea*, kelp, oyster and fish feed, which were freeze-dried and used for analysis of N and P components ($C_{N,P}$). C_N was determined using an elemental analyzer (Model Vario EL cube, Elementar Co., Germany), while C_P was analyzed using an AA3 analyzer after ashing the samples.

2.3 Endmember mixing model

A two-endmember mixing model was used to construct the conservative mixing schemes between different water masses based on the TA-S diagram and to quantify the addition or removal of nutrients on top of conservative mixing (Fig. 3a) because TA is assumed to be quasi-conservative in the absence of organic matter production/degradation and the exclusion of biogenic calcium carbonate production/dissolution processes (Zhai et al., 2014; Zhao et al., 2020). Given that freshwater discharge into the bay mainly occurs via the Jiaoxi Stream, we selected station Y0, located at the most upstream of the surveyed section of the Jiaoxi, with a salinity of 0.3 and high nutrient concentrations, as the optimal

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candidate for the freshwater endmember, represented by the symbol "FW". The seawater endmember is defined as East China Sea offshore water, represented by the subscript "SW". The average value of station S25 characterized by high salinity and low nutrient concentrations was selected as the seawater endmember. Therefore, the mixing model is based on mass balance equations for S and the water fractions (f), as follows:

$$f_{FW} + f_{SW} = 1, \tag{1}$$

$$f_{FW} \times S_{FW} + f_{SW} \times S_{SW} = S_{meas}, \tag{2}$$

Based on the percent contribution of various water masses, the predicted conservative concentrations of nutrients ($Nutrient_{pre}$) could be calculated by Eq. (3). The difference between the field measured value ($Nutrient_{meas}$) and the predicted conservative value was denoted as Δ (ΔNutrient, i.e., ΔDIN, ΔDIP, and $\Delta Si(OH)_4$), reflecting the amount of nutrient production (positive) or removal (negative) associated with non-mixing processes:

$$Nutrient_{pre} = Nutrient_{FW} \times f_{FW} + Nutrient_{SW} \times f_{SW}, \tag{3}$$

$$\Delta Nutrient = Nutrient_{meas} - Nutrient_{pre}, \tag{4}$$

where $Nutrient_{FW}$ and $Nutrient_{SW}$ represent the nutrient concentrations of the two endmembers, respectively.

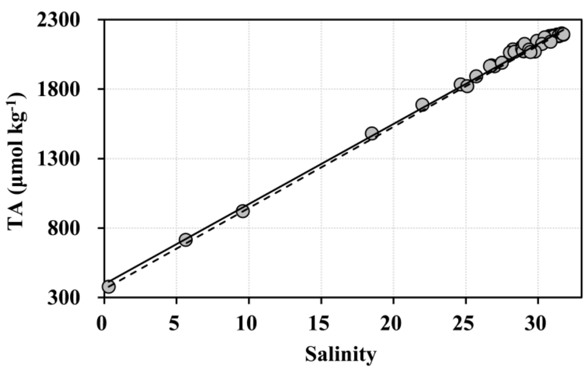

Figure 3. Total alkalinity (TA) versus salinity (S) diagram at all Sansha Bay stations. The solid black line represents the fitted linear regression line, and the dashed black line represents the conserved mixing line of freshwater and seawater endmembers.

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2.4 Mass balance of N and P in fish farming systems

Nitrogen and phosphorus budgets were established based on a mass balance principle (Wang et al., 2012; Olsen and Olsen, 2008); parameter values in the model are listed in Table S1. In the context of fish farming, the total amount of feed input (I) is equivalent to the sum of assimilated feed by fish (A) and the total waste discharge (L). It should be emphasized that waste emission from a fish farming operation is distinct from that of individual fish due to the loss of feed as particulates and the occurrence of fish mortality. In our calculations, we assumed that mortality is insignificant because dead fish are usually removed directly. Thus, the budget was calculated as:

$$\mathrm{I} = \mathrm{A} + \mathrm{L}, \tag{5}$$

The amount of N and P in feed input ($I_{N, P}$) was obtained by multiplying fish production (P_f), by the feed conversion ratio (FCR), and the N and P content of feed ($C_{N, P}$):

$$\mathrm{I}_{\mathrm{N,P}} = P_f \times \mathrm{FCR} \times \mathrm{C}_{\mathrm{N,P}}, \tag{6}$$

The feed loss (L) is associated with a different feed loss ratio (LR) in the different feeding systems. Feed losses previously represented a relatively important source of particulate waste from fish farming, but feeding is currently better controlled such that losses are lower than in the past. In our calculations, 20 % of fish production was fed with formulated feed, with an LR of 4 ± 1.4 % (Bureau et al., 2003; Corner et al., 2006; Cromey et al., 2002); while 80 % of production was fed with conventional trash fish with an LR of 13 % (Qi et al., 2019).

The mass balance for individual fish (f) can be represented by:

$$I_f = A_f + F_f = G_f + E_f + F_f, \tag{7}$$

where I_f is the food intake, equal to total assimilation (i. e., A = I - L), F_f is feces production. A_f is food assimilation, which can be calculated from the food intake and assimilation efficiency (AE) as $A_f = I_f \times AE$. Food assimilation consists of two parts, G_f and E_f, where G_f represents somatic growth or retention in biomass that can be calculated as fish production multiplied by the N and P content in fish ($G_f = P_f \times C_{N, P}$), and E_f is excretion through urine discharge. Nitrogen and P are excreted mainly in the form of ammonium released at the gills and PO_4^{3-} in urine, respectively. The amount of E_f is equal to the amount of nutrients assimilated minus that retained in fish biomass. Excreted N and P are released into the environment in dissolved inorganic form, while N and P from uneaten feed and feces are released as

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particulate organic nitrogen (PON-Feed and PON-Fecal) and particulate organic phosphate (POP-Feed and POP-Fecal), respectively. Approximately 15 % of particles will be dissolved in organic form (dissolved organic nitrogen, DON, and dissolved organic phosphate, DOP, respectively) according to Chen et al. (2003) and Sugiura et al. (2006).

2.5 Kelp and oyster removal

Kelp and oyster culture are effective in removing N and P from the environment. For kelp, the amount removed can be calculated as kelp production multiplied by the N and P content in kelp. For oysters, the amount removed includes the harvest of shell and soft tissue, which can be expressed as: removal = production × soft tissue ratio × soft tissue $C_{N,P}$ + production × shell ratio × shell $C_{N,P}$.

3. Results and discussion

3.1 Nutrient distribution in Sansha Bay

During late spring in Sansha Bay, the temperature showed a spatially variable distribution. It attained a maximum of 23.6 °C in Baima Harbor, upstream of the bay, while a lower temperature (~21.9 °C) was observed near the Dongchong Channel, at the Sansha Bay mouth (Fig. 4(a)). The salinity increased gradually from the inner bay to the mouth of the bay with values of 0.3 and 31.7 in the Jiaoxi Stream and the Dongchong Channel, respectively (Fig. 4(b)). Therefore, Sansha Bay is characterized by lower temperature and higher salinity at the mouth of the bay than upstream in the bay, mainly due to the influence of river runoff from the northwest of the bay and coastal ECS waters.

Nutrient concentrations generally decreased from the inner to outer portion of the bay. The highest NO_3^-+NO_2^- and $Si(OH)_4$ concentrations, 76.1 μmol L^{-1} and 153.5 μmol L^{-1}, respectively, were found in the Jiaoxi Stream, and they decreased linearly with increasing salinity (Fig. 4(e) and (f)). The highest DIP and NH_4^+ concentrations were not observed in the Jiaoxi Stream, but rather in waters near Sandu Island (stations S1, S2, S6 and S26) and Guanjingyang (station S14) (Fig. 4(g) and (h)), which may result from the release of fish feeds (Fig. 4(i)). Chl-*a* concentration were low (<0.1 μg L^{-1}) and uniform throughout the study area (Fig. 4(c)), indicating relatively low phytoplankton productivity.

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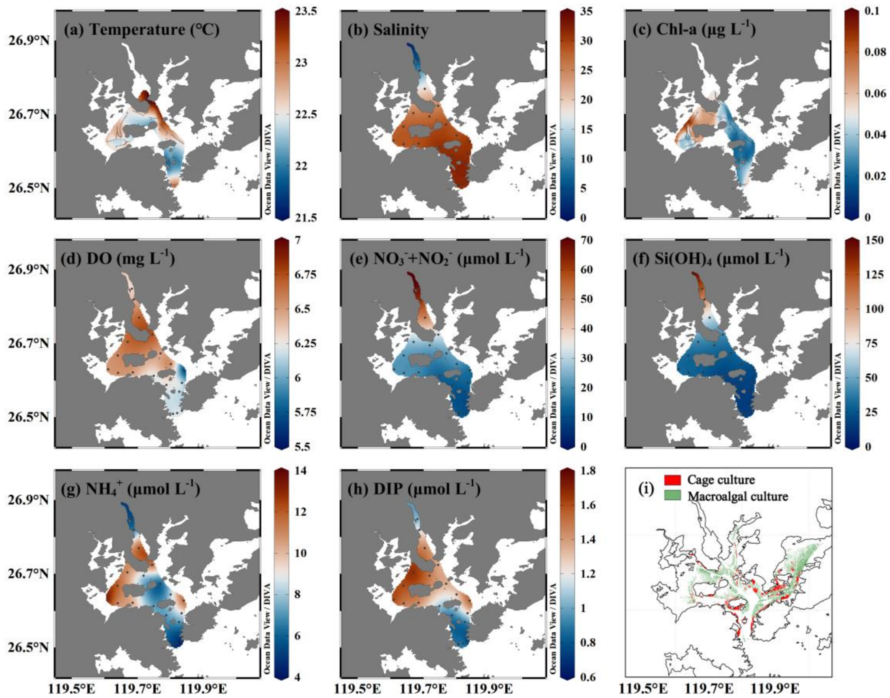

Figure 4. Distribution of temperature, salinity, and concentrations of chlorophyll-*a* (Chl-*a*), dissolved oxygen (DO), nitrate + nitrite (NO_3^-+NO_2^-), silicate ($Si(OH)_4$) ammonium (NH_4^+), and dissolved inorganic phosphorus (DIP) in surface waters in Sansha Bay (a-h). The distribution of macroalgae and cage culture in surface waters (i) is shown in green and red, respectively.

The water was well-mixed throughout the water column, with only weak stratification in the estuarine plume, as shown in Fig. 5. Overall, nutrient concentrations followed a similar pattern to their distribution in the surface layer, with zonal variation from the inner to the outer portion of the bay. This may explain the absence of high Chl-*a* concentrations during the cruise due to weak stratification. The NO_3^-+NO_2^- concentration decreased from 62.4-76.1 μmol L^{-1} at the river estuary to 9.3-11.9 μmol L^{-1} at the Dongchong Channel, at the mouth of the bay; $Si(OH)_4$ concentrations exhibited a similar spatial distribution, gradually decreasing seaward. Lower concentrations were observed, however, near the Dongchong Channel, ranging from 0.71-0.92 μmol L^{-1} for DIP, and 3.99-5.46 μmol L^{-1} for NH_4^+, except for the high values in Baima Harbor (Station SX01). Additionally, higher DIP and NH_4^+ concentrations as well as reduced DO levels were noted in station S14, which may result from organic matter remineralization.

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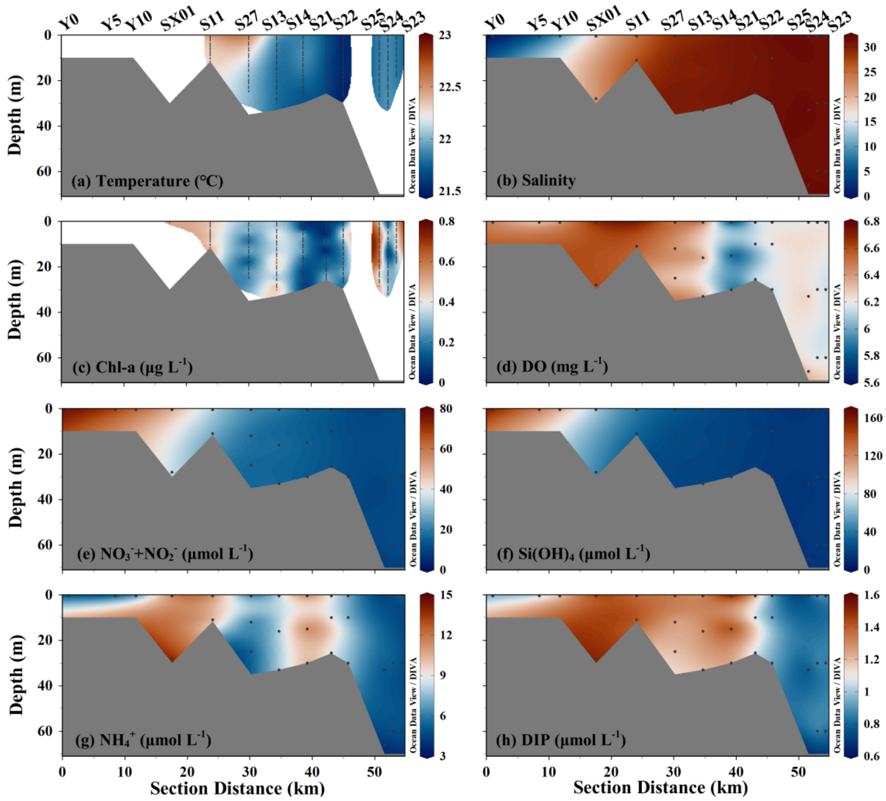

Figure 5. Distribution of temperature, salinity, and concentrations of chlorophyll-*a* (Chl-*a*), dissolved oxygen (DO), nitrate + nitrite (NO_3^-+NO_2^-), silicate ($Si(OH)_4$), ammonium (NH_4^+), and dissolved inorganic phosphorus (DIP) along the main section of Sansha Bay (a-h) from the Jiaoxi Stream (0 km) to the Dongchong Channel. Black circles within the transect indicate stations.

3.2 Nutrient changes due to biogeochemical processes and nutrient stoichiometric ratios

The DIN concentration gradually decreased with increasing salinity and exhibited obvious addition within the bay, attaining 6.9 ± 4.1 μmol L^{-1} (34.5 % ± 16.3 %) throughout the bay area. However, there was no anomalous enrichment near Sandu Island and the mariculture zone (Fig. 6(a)). However, the behavior of DIP differed in that the variation in DIP concentrations was more complex with a S value of 25 as the boundary. We observed a 58.8 % ± 37.0 % (0.45 ± 0.29 μmol L^{-1}) DIP addition with a maximum addition of 1.19 μmol L^{-1} in the mariculture zone. DIP concentrations gradually increased at $S < 25$, while they decreased when S exceeded 25. This pattern was likely induced by a buffering mechanism of DIP (Froelich, 1988), and/or may be attributable to variable feed flows in different zones (Fig. 6(b)).

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Additionally, we observed that $Si(OH)_4$ generally behaved conservatively with only minor removal. Although diatoms were dominant in this bay during spring, their abundance was relatively low, as indicated by the low Chl-*a* concentrations. As a result, individual stations deviated from the conservative mixed line at salinity below 25 (Fig. 6(c)).

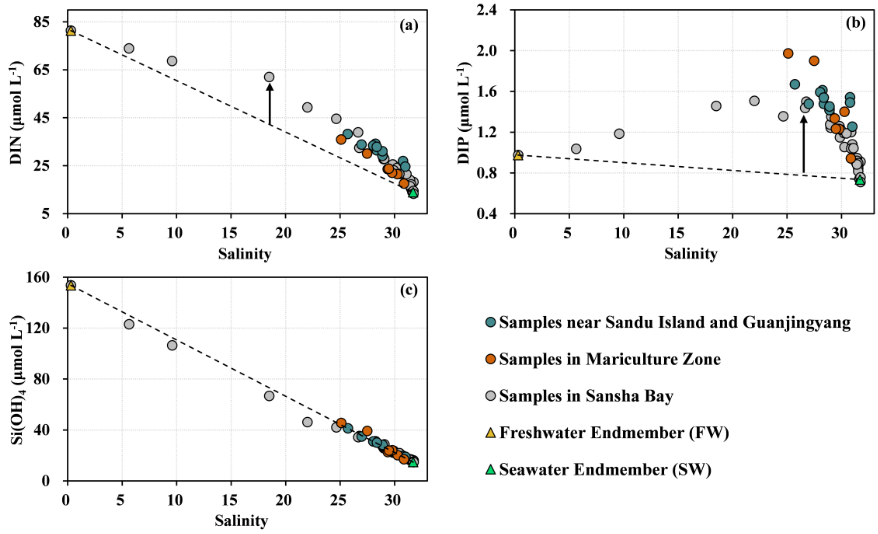

Figure 6. Relationship between nutrient concentrations vs. salinity in Sansha Bay in May 2020. Dark green, red and gray circles indicate stations near Sandu Island (station S1, S2, S6 and S26) and Guanjingyang (station S14), stations in the mariculture zone and freshwater and seawater endmember stations (yellow and green triangles). The black dashed line represents the conservative mixing line, and the black arrow indicates the obvious addition of dissolved inorganic nitrogen (DIN) and phosphorus (DIP) based on the conservative mixing lines.

Overall, distinct DIN:DIP ratios associated with different water masses resulted in a variable effect on nutrient stoichiometry. Owing to the significant inflow of DIN from the Jiaoxi Stream, the plume water ($S < 25$) exhibited a high DIN:DIP ratio. When the salinity was > 25, DIN and DIP showed a strong linear association with a ratio of 23.38 ± 1.19 (Fig. 7(a)), which exceeded the Redfield ratio (Redfield et al., 1963), while, the DIN:DIP ratio in the mariculture zone was 15.49 ± 2.37, i.e., close to the Redfield ratio (Redfield et al., 1963), and consistent with previous observations in other mariculture waters in China (Bouwman et al., 2013). The $Si(OH)_4$:DIN ratio, however, exhibited different characteristics: in the plume region it was 3.08 ± 0.36, and thus significantly higher than the diatom absorption ratio of 1:1 (Martin‐Jézéquel et al., 2003; Brzezinski, 1985). This might be mostly due to weathering of volcanic rock, which is dominant in Ningde, and resulted in relatively high silicate concentrations. The

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$Si(OH)_4$:DIN ratio was approximately 1:1 in the region with salinity > 25, while it was 1.69 ± 0.16 in the mariculture zone (Fig. 7(b)), which may be related to the lack of diatom dominance and preference for N in this region.

Results of the endmember mixing model showed significant DIN and DIP additions (positive values) at most stations. The ΔDIN:ΔDIP ratios in the study area were lower than the DIN:DIP ratios. Specifically, the ΔDIN:ΔDIP addition ratio was 17.49 ± 4.66 at S > 25 and 7.91 ± 1.48 in the mariculture zone, indicating nutrient addition with a lower N:P ratio. There was no obvious pattern for the $\Delta Si(OH)_4$:ΔDIN ratio because $Si(OH)_4$ is basically conservative in Sansha Bay. Given the intensity of mariculture activity in this bay, the lower ΔDIN:ΔDIP ratio may be related to bait feeding. Previous studies have shown that 51 % of both N and P in bait were dissolved in water and the mass ratio of N and P was only 6.7 (Jia et al., 2003). Alternatively, this may be due to the addition of non-Redfield ratio nutrients from sediment. A study conducted in tidal flats of the Dutch Eastern Scheldt revealed that the DIN:DIP ratio in sediment porewater was notably lower than the Redfield ratio (Rios-Yunes et al., 2023).

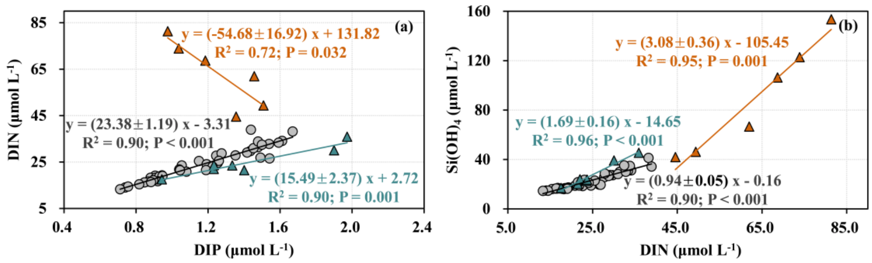

Figure 7. Dissolved inorganic nitrogen to phosphorus (DIN:DIP), and silicate to DIN ($Si(OH)_4$:DIN) ratios in Sansha Bay. Red triangles, gray circles and dark green triangles represent samples with salinity < 25, salinity > 25 and the mariculture zone, respectively. Solid lines of the corresponding colors are the fitted linear regression lines. Linear regression equations are also shown.

3.3 Nutrient release from the fish farming system

The production of *L. crocea* in 2020 reached 1.785×10^5 tons (Statistical Yearbooks of Ningde; http://tjj.ningde.gov.cn/xxgk/tjxx/tjnj/). The N and P budget model for the cage system using both trash fish feed and formulated feed is shown in Fig. 8. Specifically, in the trash fish feed system, only 21 ± 1 % of N and 10 ± 1 % of P was incorporated into fish tissues, corresponding to $(5.54 \pm 0.26)\times10^3$ tons of N and $(5.33 \pm 0.009)\times10^2$ tons of P, and the remaining nutrients were released and lost to seawater or sediment (Fig. 8(a) and (b)). The N released to seawater was mainly excreted as DIN (53 ± 6 %), and 26 ± 7 % of N sank to the bottom as particulate organic nitrogen (PON) in the N budget model fed by trash

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fish (Fig. 8(a)). In the P budget model, however, ca. $(2.90 \pm 0.41)\times10^3$ tons (57 ± 9 %) and $(1.70 \pm 0.18)\times10^3$ tons (33 ± 4 %) of P were lost as particulate organic phosphorus (POP) and DIP, respectively, whereas about $(4.34 \pm 0.62)\times10^2$ tons of POP were released in the water column as DOP (Fig. 8(b)).

For the formulated feed system, only 33 ± 2 % of N and 15 ± 0.5 % of P in feed were absorbed and utilized for incorporation in fish biomass. The remaining 67 ± 2 % of N ($(2.87 \pm 0.08)\times10^3$ tons) and 85 ± 4 % of P ($(7.33 \pm 0.28)\times10^2$ tons) were released into the water in both inorganic and organic form or settled to bottom sediments (Fig. 8(c) and (d)). In the N budget model, about half of N (49 ± 2 %) was lost as DIN (Fig. 8(c)). Similarly, 52 ± 4 % of P ($(4.50 \pm 0.36)\times10^2$ tons) was deposited as particulates in the P budget model (Fig. 8(d)). Results indicated that the majority of the N waste generated by the fish farming system was excreted as DIN, whereas the majority of P waste settled as POP. For both feeding systems, the retention rate of N in fish biomass was higher than that of P, suggesting that fish excrete a lower proportion of N than P. Additionally, the released DIN:DIP ratios showed inconsistencies. Molar DIN:DIP ratios were ~18.72 and ~16.39 in the trash fish feed and formulated feed systems, respectively, and thus quite consistent with the Redfield ratio (Redfield et al., 1963), which might partially explain the variable ΔDIN:ΔDIP ratios in the waters.

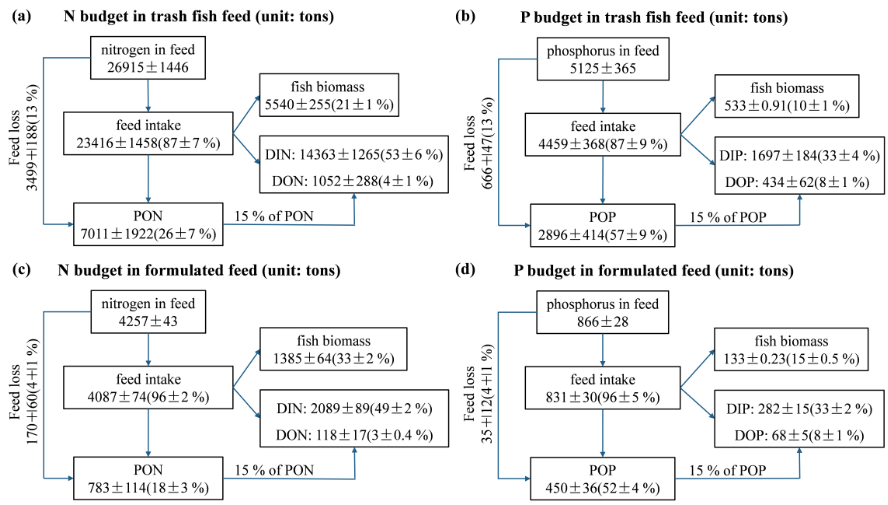

Figure 8. Nitrogen (N; (a) and (c)) and phosphorus (P; (b) and (d)) budget for the cage systems using formulated and trash fish feeds in Sansha Bay. Percentage values represent the proportion of each data to N and P in feed. DIN and DIP: dissolved inorganic nitrogen and phosphorus, respectively; DON and DOP: dissolved organic nitrogen and phosphorus, respectively; PON and POP: particulate organic nitrogen and phosphorus, respectively.

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Our budget model showed that N and P released from the system fed by trash fish feed was much greater than that of the system fed by formulated feed due to the higher feed loss ratio in trash fish feed. Additionally, Fig. 9(a) demonstrates a progressive rise in the release of DIN and loss of PON-Feed with an increasing percent of trash fish feed, while DON and PON-Fecal were less variable. Therefore, formulated feed with varying particle size may be used for different fish sizes in comparison to the trash fish feed, which can reduce feed loss during farming. In addition, N and P retention rates of trash fish feed were lower than those of formulated feed. According to Verdegem et al. (1999), the higher feed protein content (generally higher in trash fish feed), the higher the proportion of N waste excreted. Moreover, the lower nutritional value and digestibility of trash fish feed may also explain its lower N retention rate (Hasan et al., 2016). The impacts of various feed losses and assimilation efficiencies are shown in Fig. 9(b) and (c). In the above calculations for the trash fish feed system, a 13 % feed loss value and 85 % N assimilation efficiency were used. The percentage of DIN excretion decreases as feed loss increases, and the percentage of combined organic waste from feed (PON-Feed) increases in a comparable manner (Fig. 9(b)). In contrast, a rise in the percent of DIN and a decline in the similarity between PON-Fecal are found with an increase in assimilation efficiency (Fig. 9(c)). Therefore, minor changes in feed loss and assimilation efficiency are unlikely to result in significant deviations in model predictions.

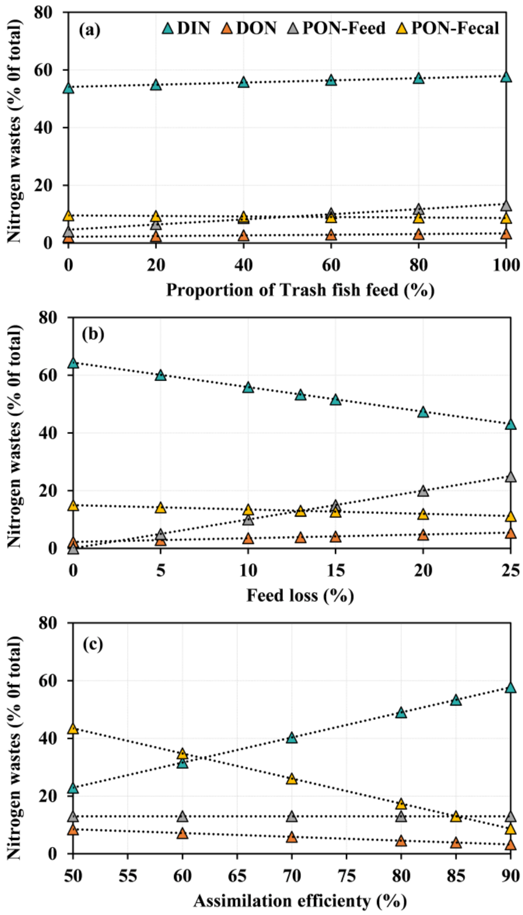

Figure 9. Relationship between the percentage of nitrogen released and the proportion of trash fish feed (a, in both feed systems), and feed loss rate (b, in the trash fish feed system) and assimilation efficiency (c, in the trash fish feed system). The PON-Feed and PON-Fecal represent PON in feed and feces, respectively (PON: particulate organic nitrogen). The dashed lines are fitted linear regression lines.

3.4 Nutrient budget in Sansha Bay

In summary, only 22.2 ± 1.3 % of N and 11.1 ± 0.7 % of P was assimilated by cultured fish, resulting in an annual release of $(2.42 \pm 0.15)\times10^4$ tons of N and $(5.33 \pm 0.37)\times10^3$ tons of P from the fish farming system co-fed with trash fish feed and formulated feed. Furthermore, 52.8 ± 4.7 % of DIN and 33.0 ± 3.7 % of DIP was released into the water, and ca. half (47.5 ± 6.6 %) of the P settled to the bottom in particulate form (Fig. 10).

To further clarify the nutrient contribution of the fish farming system to the water in Sansha Bay, we estimated the nutrient (DIN and DIP) fluxes contributed by river discharge and exchange with offshore coastal waters based on the mass balance model (Gordon et al., 1996), as detailed in Supplementary Text S2. For example, the river discharge in May 2020 was 71.3 m^3 s^{-1} (data from Nengwang Chen, Xiamen University). Results showed that the residual flow and the water exchange flux between the bay and offshore coastal water were -71.3 m^3 s^{-1} (negative values indicating outflow) and 720.1 ± 676.8 m^3 s^{-1}, respectively (Table 1), which was also consistent with previous data (~479.5 m^3 s^{-1}, unpublished data from Zhaozhang Chen, Xiamen University). Based on the water exchange rates and nutrient concentrations in different systems of this area, the DIN and DIP fluxes from the river were 2561 and 68 tons yr^{-1}, respectively, while nutrient fluxes from the bay to the offshore coastal water were 6229 ± 6090 tons yr^{-1} for DIN and 712 ± 648 tons yr^{-1} for DIP (Fig. 10). Due to the lack of field sediment data, biogeochemical processes at the sediment-water interface were not considered in the present study. As a result, without considering the contribution of sediment, the amount of dissolved inorganic nutrients (DIN and DIP) released by the fish farming system was much higher than that from river input. About 32.8 % of the DIN and 34.8 % of the DIP were transported from Sansha Bay to offshore coastal waters through the Dongchong Channel.

Furthermore, by analyzing the N and P components in kelp and oysters, it can be determined that each ton of dried kelp can absorb 22.5 kg N and 3.5 kg P from seawater, while oyster soft tissue and shell can absorb 80.7 and 1.6 kg of N, and 5.6 and 0.44 kg of P, respectively. We thus estimated the removal of N and P by combining the production of macroalgae (kelp, 1.888×10^5 tons) and bivalve mollusks (oysters, 1.368×10^5 tons) harvested in 2020 to assess nutrient removal by macroalgae and bivalve mollusks in the IMAT system. Our results showed that harvesting of kelp in Sansha Bay resulted in the removal of 708 ± 3 tons of N and 110 ± 8 tons of P, respectively. In the case of oysters harvesting, it removed 371 ± 11 tons of N and 46 ± 0.1 tons of P (Fig. 10). Therefore, the co-culture strategy removed $(1.08 \pm 0.01)\times10^3$ tons of N and $(1.56 \pm 0.08)\times10^2$ tons of P in total. As a result, total emissions of N and P from mariculture in 2020 were $(2.32 \pm 0.15)\times10^4$ tons and $(5.17 \pm 0.37)\times10^3$ tons, respectively. Results showed that the amount of N and P released from feed was much higher than that of fish, macroalgae and oysters harvest combined. Thus, the release of feeds in the aquaculture system is the main cause of eutrophication when other nutrient sources remain constant in Sansha Bay.

Table 1. Summary of salinity (S), dissolved inorganic nitrogen and phosphorus concentrations (DIN and DIP, respectively, in µmol L^{-1}), water discharge (V, m^3 s^{-1}) and calculated nutrient flux (F_{DIN} and F_{DIP}, mmol s^{-1}) in

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the mass balance model.

Salinity	S_{riv}	$S_{sys} \pm \sigma_{sys}$	$S_{oce} \pm \sigma_{oce}$	$S_{res} \pm u_{res}$
	0.3	29.1 ± 2.7	32.1 ± 0.8	30.6 ± 1.4
DIN (μmol L^{-1})	DIN_{riv}	$DIN_{sys} \pm \sigma_{DINsys}$	$DIN_{oce} \pm \sigma_{DINoce}$	$DIN_{res} \pm u_{DINres}$
	81.3	27.5 ± 9.0	9.7 ± 2.8	18.6 ± 4.7
DIP (μmol L^{-1})	DIP_{riv}	$DIP_{sys} \pm \sigma_{DIPsys}$	$DIP_{oce} \pm \sigma_{DIPoce}$	$DIP_{res} \pm u_{DIPres}$
	0.98	1.29 ± 0.27	0.36 ± 0.10	0.83 ± 0.14
V ($m^3 s^{-1}$)	V_{riv}	$V_{ex} \pm u_{ex}$		V_{res}
	71.3	720.1 ± 676.8		-71.3
F_{DIN} (mmol s^{-1})	F_{DINriv}	$F_{DINex} \pm u_{FDINex}$		$F_{DINres} \pm u_{FDINres}$
	5800	-12782 ± 13789		-1327 ± 335
F_{DIP} (mmol s^{-1})	F_{DIPriv}	$F_{DIPex} \pm u_{FDIPex}$		$F_{DIPres} \pm u_{FDIPres}$
	70	-670 ± 662		-59 ± 10

Note: S_{riv} represents the salinity of station Y0 in the Jiaoxi Stream; S_{sys} represents the average salinity in the bay, excluding stations Y0, Y5, Y10 in the stream and S23, S24, S25 at the bay mouth; S_{oce} represents the salinity of four stations (LJ21, LJ22, ND41 and ND42, not shown on the map) in offshore coastal waters; S_{res} represents the salinity of the residual flow; V_{riv}, V_{res} and V_{ex} represent the river discharge, residual flow discharge from the bay to offshore coastal waters and exchange flow between the bay and the offshore coastal water, respectively; σ represents the standard deviation, and u represents the uncertainty associated with each variable. Negative values indicate nutrient output.

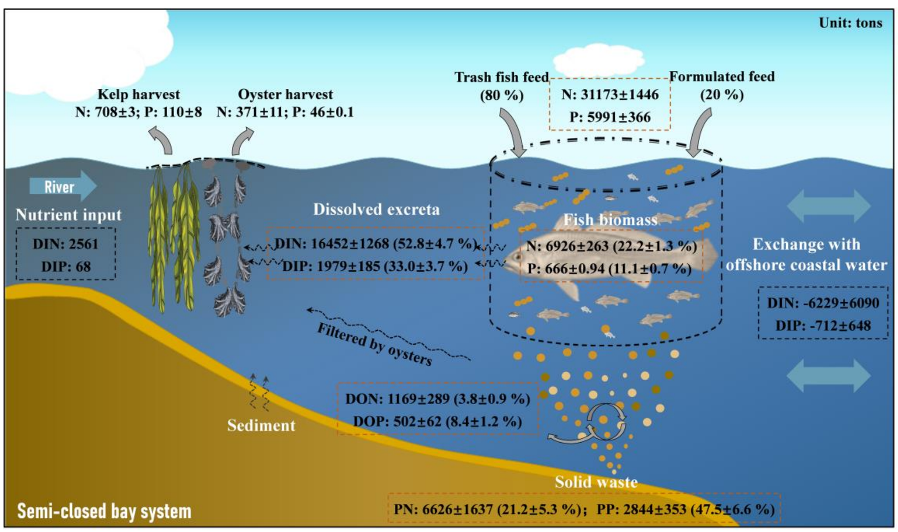

Figure 10. Conceptual diagram illustrating the nutrient transformation of different species resulting from

mariculture in the semi-enclosed bay system, which was influenced by riverine and offshore coastal water exchange. Briefly, only a small proportion of the feed was consumed in support of fish growth, and the rest was released into the water in dissolved forms or settled to the seafloor in particulate forms. Mixed farmed kelp and suspension-feeding oysters can effectively remove nutrients from the water. The units in the figure are given in tons. Negative values indicate nutrient output. Nutrient abbreviations as in Fig. 8.

3.5 Strategies for the sustainable development of mariculture

This study has revealed important differences in the amount of nutrients released between fish farming systems depending on the use of different types of feeds. These differences may involve substantial variation in the FCR and feed composition among other factors. Specifically, the FCRs for formulated feed and trash fish feed were 1.525 ± 0.007 and 6.745 ± 0.36, respectively (Gao et al., 2021; Qi et al., 2019). This indicates a notable 77.4 % increase in consumption when using trash fish feed compared to formulated feed, which requires an additional 5.22 tons of trash fish feed to produce one ton of *L. crocea*. As illustrated by the production in 2020, ~ 2.722×10^5 tons of formulated feed were required. In contrast, producing the same biomass of fish would demand 1.204×10^6 tons of trash fish feed. Despite the much higher consumption of trash fish feed, consideration of feed cost (with the price of formulated feed at 1.0×10^4 RMB per ton and trash fish feed at 1.2×10^3 RMB per ton (Shan et al., 2018)), the cost of formulated feed was higher by 1.277×10^3 million RMB. Therefore, in practical terms, many farmers in Sansha Bay prefer to use the lower-cost trash fish feed. However, it is critical to note that as the proportion of trash fish feed usage increases, so does the release of DIN and PON-Feed into the environment can thus potentially have negative impacts on water quality. Improving the FCR to reduce feed usage and altering feed type and composition to lower costs are thus crucial for the sustainable development of fish farming systems, which has important policy implications.

However, the release of inorganic and organic nutrient waste is considered one of the major environmental challenges in fish farming in Sansha Bay. According to China's current seawater quality standards (GB3097-1997), water quality in Sansha Bay was classified as Grade III. While the occurrence of surface algal blooms and bottom oxygen depletion were infrequent in the inner bay, harmful algal blooms (HABs) were a recurring issue in the outer regions of the bay. Currently, there is no clear evidence linking the occurrence of HABs to the release of inorganic nutrients from fish cages. Nevertheless, water quality may become a critical factor leading to oxygen reduction and the occurrence of HABs in sensitive locations as mariculture production continues to increase. Although the government of Ningde took

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measures to remove mariculture activities from the inner bay to the outer bay, it remains uncertain whether these measures will exacerbate HABs outbreaks outside the bay and reach a tipping point thus requiring long-term monitoring and research of water quality.

4 Conclusions

The spatial variability of nutrients in the Sansha Bay was mainly controlled by the water mass mixing, between the riverine water and seawater, as well as the effects of mariculture system. Based on the end-member mixing model, we estimated that the additions of DIN and DIP during spring were ~ 6.9 ± 4.1 μmol L^{-1} and ~ 0.45 ± 0.29 μmol L^{-1}, respectively. When using a mixture of formulated feed and trash fish feed at a 2:8 ratio in 2020, it was observed that approximately 52.8 ± 4.7 % of DIN and 33.0 ± 3.7 % of DIP from the feeds were released into the surrounding waters, significantly surpassing the river input and exchange with offshore coastal water.

Up to now, methods for nutrient removal from eutrophic seawater are very limited. The cultivation and harvesting of macroalgae and bivalve mollusks provide an effective technology for nutrient removal. For each ton of kelp (dry weight) harvested, 22.5 kg N and 3.5 kg P can be removed from the water. Therefore, we recommend judicious planning of the proportions of macroalgae, oysters and fish farming. Additionally, when compared to formulated feed, the consumption of trash fish feed for producing one ton of fish increased by 77.4 %. Consequently, enhancing feed conversion rate, improving feed composition, and reducing feed costs are advantageous for the sustainable development of fish farming systems and lowering the risk of oxygen depletion and harmful algal blooms in sensitive areas.

Data availability. Data for temperature, salinity, DO, Chl-*a* and nutrients are available on the National Science Data Bank (https://www.scidb.cn/en). The productions of *L. crocea*, kelp and oyster from 1990 to 2020 were obtained from the Statistical Yearbooks of Ningde City (http://tjj.ningde.gov.cn/xxgk/tjxx/tjnj/). The remote sensing data used to identify cage culture and macroalgal culture were obtained from Landsat (https://glovis.usgs.gov/app) and Sentinel-2 (https://scihub.copernicus.eu/dhus/#/home).

Supplement.

Author contributions. YW and MD co-designed the study. YW, LW, TH, YL and ZW contributed to sampling, data acquisition and analysis. YW and MD analyzed and interpreted the data and drafted the manuscript. XG and GW revised the manuscript and made constructive comments.

Competing interests. The authors declare that they have no conflict of interests.

Disclaimer.

Acknowledgements. We thank the captain, crew and scientific staff of the R/V *Funing 11* for their cooperation during the cruise, Kunning Lin for sample collection and ammonium analysis; Aiqin Han for assistance in sample collection, Yi Xu for the collection of biological samples, Nengwang Chen and Xuwen Fang for providing the YSI and river discharge data, Junhui Chen for guidance in sample analysis, Haixia Guo for processing the remote sensing data, Yanping Xu and Feifei Meng for her logistical support.

Financial support. This research was funded by the National Natural Science Foundation of China (NSFC #42188102).

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