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Increasing eutrophication driven by the increase of phosphate discharge in a subtropical bay in the past 30 years

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Although great efforts have been made to decrease nutrient (notably nitrogen and phosphorus) loads and lighten related environmental damages, coastal eutrophication remains a persistent environmental crisis. To estimate whether the rapid development of the local economy has accelerated the eutrophication in Zhanjiang Bay, a newly developing industry in South China, the combination of the seasonal nutrients and other physicochemical parameters from twenty cruises during 2017-2021 and historical data (past 30 years) was analyzed in this study. The results showed that the eutrophication in the upper bay is significantly higher than that in the lower bay (more than 8 times), which is mainly related to the terrestrial input and weak hydrological conditions in the upper bay. Also, eutrophication is more severe in the rainy seasons than that in the dry seasons (nearly 2 times) because abundant nitrogen and phosphorus were brought into the bay by terrestrial discharge and river water. From a long-term perspective, dissolved inorganic nitrogen concentrations have been effectively controlled in Zhanjiang Bay, while phosphate concentration increased sharply in the past 30 years. Correspondingly, the eutrophication in Zhanjiang Bay significantly increased over the past 30 years, and the eutrophication index in the recent 5 years (2017-2021) is nearly 10 times that of the 1990s, suggesting that the increase of phosphate discharge from the increasing industrial factories around Zhanjiang Bay, rather than nitrogen discharge is the culprit causing the aggravation of eutrophication. Our study is essential to effectively implement a land-ocean integrated nitrogen and phosphorus control strategy to improve water quality and mitigate eutrophication in the bay.

KEYWORDS

eutrophication, nutrients, chemical oxygen demand, Zhanjiang Bay, nitrogen, phosphorus

Introduction

Coastal regions, situated between the land and the ocean, are rich in biological resources and diverse ecosystems, and are also the center of global economic development and human activities (Winther et al., 2020; Dai et al., 2022; Dai et al., 2023). However, the rapid development of socio-economy and climate change have continuously intensified the environmental loads on this vital area since the Industrial Revolution, which in turn has had a negative impact on human society and has damaged the sustainability of the region and global oceans (Doney et al., 2012; Winther et al., 2020; Dai et al., 2023). Among many factors, pollution of nutrients, primarily due to massive discharges from agriculture and industry, has caused worldwide expansion of eutrophication in the coastal regions, which can directly endanger the biodiversity, habitat and even mass mortality of impacted marine organisms, and thus eventually impair marine ecosystem and human society (Carpenter, 2008; Deegan et al., 2012; Breitburg et al., 2018; Malone and Newton, 2020; Maúre et al., 2021). Although great efforts have been made to reduce nutrient (mainly nitrogen and phosphorus) loads and lighten related environmental damages, eutrophication and its eco-environmental effects remain a persistent environmental crisis in the coastal oceans (Kelly et al., 2021; Lao et al., 2021; Wang et al., 2021b; Dai et al., 2023).

Over the past decades, coastal nutrient structure and loads have substantially changed due to increased terrestrial anthropogenic discharges (Justić et al., 1995; Howarth and Marino, 2006; Malone and Newton, 2020; Wang et al., 2021b). Global phosphorus and nitrogen loads elevated by 40-50% from 1980 to 2015 (Beusen et al., 2022). Investigations suggested that artificial fertilizers are the dominant component of anthropogenic nitrogen input in many basins (Cui et al., 2020; Zhou et al., 2021), and 22-40% of the anthropogenic nitrogen is poured into the coastal waters through riverine export (Howarth et al., 2012; Chen et al., 2016). In addition, fossil fuel combustion, planting nitrogen-fixing crops and wastewater inputs increased the nitrogen loads in the coastal waters (Glibert et al., 2014; Peñuelas and Sardans, 2022). However, due to the implementation of phosphorus emission control policies, there is no increased trend in phosphorus level in most coastal regions (Howarth and Marino, 2006). But in some regions, due to the impact of human activities, such as industrial discharges, coastal phosphate concentrations have significantly increased in the past few decades (Lao et al., 2021). These large amounts of anthropogenic nitrogen are directly discharged into the coastal waters, which increases the N:P ratio, and thus potentially creates phosphorus-limited water and the change of dominant algal species (Glibert et al., 2014; Lin et al., 2016; Peñuelas and Sardans, 2022). Therefore, most researches indicated that elevating nitrogen loads is a dominant factor for eutrophication in most coastal oceans (Sinha et al., 2017; Paerl, 2018; Kelly et al., 2021).

Zhanjiang Bay, located in the northwestern South China Sea (SCS), is one of the most important mariculture grounds, and aquaculture production and export bases in South China. However, due to intense human activities, the ecological environment of the bay is gradually deteriorating (Li et al., 2014; Li et al., 2020; Zhang et al., 2021; Chen et al., 2022b; Lao et al., 2022). Among the environmental problems, eutrophication is the most prominent problem in Zhanjiang Bay (Shi et al., 2015; Zhang et al., 2020; Zhang et al., 2021). Because there are many sewage outlets of municipal sewage treatment plants along the coast of the bay, an enormous number of urban and industrial wastewater is directly discharged into the bay, increasing the nutrient loads in the bay (Zhang et al., 2021; Zhou et al., 2021). Additionally, river runoff around the bay can directly input domestic and agricultural nutrients from the basin into Zhanjiang Bay (Li et al., 2020; Zhang et al., 2021; Lao et al., 2022). In addition, intensive mariculture activities contribute to nutrient loads of the bay (Li et al., 2020; Lao et al., 2022). However, since there is only a narrow channel that connects the bay to outer seawater, the hydrodynamic conditions of the bay are weak, which is not conducive to the diffusion of pollutants (Wang et al., 2021c). More importantly, due to intensified human activities (e.g., dredging activity and artificial dams), the runoff in the bay has dramatically decreased, but increased intrusion of high salinity (increased by 23%) water from the outer bay, resulting in the pollutants discharged from human activities being seriously retained in the bay (Lao et al., 2022). Thus, the water quality of the Bay is gradually deteriorating (Shi et al., 2015; Lao et al., 2022). However, the long-term trend and driving factors of eutrophication in Zhanjiang Bay remain unclear. To address this issue, the combination of the seasonal nutrients and other physicochemical parameters from twenty cruises from 2017 to 2021 and historical data were analyzed in this study.

Materials and methods

Study area and sampling

Zhanjiang Bay, located in the western Guangdong Province, is a nearly-enclosed bay northwest of the SCS (Figure 1). Zhanjiang city surrounds the bay, and the area is 2091.5 km^2. The depth of the bay ranged widely (2-32 m), the deeper in the central parts and shallower in the upper bay. Zhanjiang Bay is geographically and hydrodynamically complex. There are two water masses affecting the seawater of the bay. At the top of the upper bay is the Suixi River, which is approximately 80 km long with an average annual flow of 10.4×10^8 m^3, flowing into the bay. At the end of the mid-bay, there is only a narrow channel (~2 km width) that connects the bay to the SCS. Zhanjiang Bay is a typical mariculture bay. The upper bay is mainly for oyster row culture, and the mid-bay is mainly for cage culture due to the deep water (Chen et al., 2022b; Lao et al., 2022). As the upper bay is surrounded by the main urban area of Zhanjiang city, and the intensive cultivation of oyster activities, it is subject to a high pollution burden (Li et al., 2020; Chen et al., 2022b). The mid bay is broad waters between two islands, mainly affected by domestic sewage, intensive mariculture and nonpoint sources (Li et al., 2020; Chen et al., 2022b). Zhanjiang Bay is a typically tropical meteorological climate due to the influence of the East Asian Monsoon. The mean air temperature in this region is 23.5°C. The annual rainfall is 1731 mm, and the precipitation almost occurs in the summer monsoon period (April to October defined as rainy seasons, >85% of the annual rainfall) (Chen et al., 2021).

Twenty cruises were conducted during the spring (March), summer (June), fall (September), and winter (December) from 2017 to 2021 in Zhanjiang Bay. The sampling stations of each cruise are extended from the top of the upper bay (areas above the black dotted line) to the lower bay (Areas below the black dotted line) (Figure 1). Seawater samples were collected from the upper layer (0-5 m) of the bay using a 12 L Niskin sampler. During the sampling, seawater temperature, salinity, dissolved oxygen (DO), and pH were determined on-site. Temperature and salinity were measured using an RBR maestro multiparameter water quality monitor (RBRmaestro3, RBR, Canada). pH was measured by a pH meter (Thermo Model 868). Seawater for chlorophyll-a (Chl-*a*) samples was filtered by the glass fiber filters (GFF, 0.7 µm, Whatman), and the GFF was stored at -20°C. Seawater for nutrient samples were filtered by pre-acid clean acetate cellulose Blters (0.45 µm), and the filtrate were transferred into acid-cleaned polyethylene bottles and stored at −20°C.

Chemical analysis

The DO and chemical oxygen demand (COD) measurements were in accordance with National Marine Monitoring Code Part 4-Seawater Analysis (GB 17378.4–2007). DO concentration was measured by the Winkler titration method with a precision of 0.07 mg L^{-1}. COD concentrations were measured by the potassium

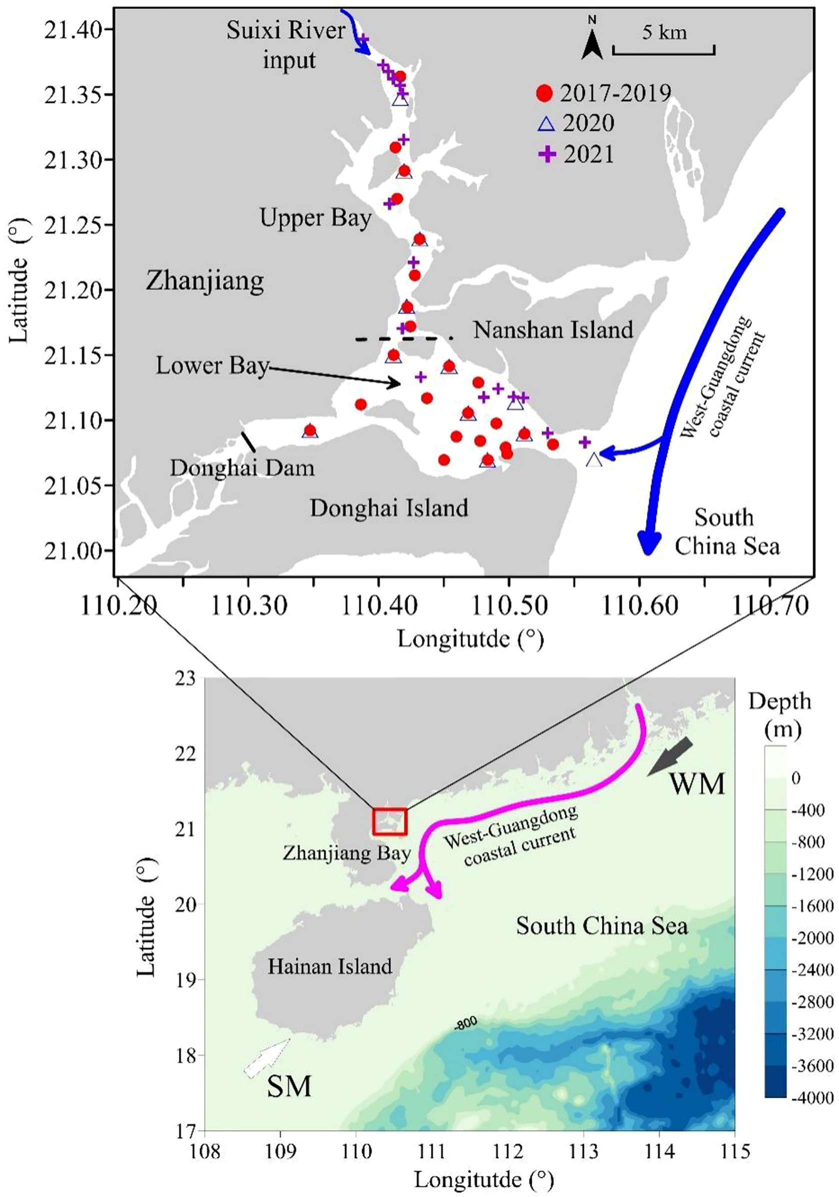

FIGURE 1
Study area and the sampling sites in the Zhanjiang Bay during 2017-2021. The hydrodynamic characteristics of Zhanjiang Bay is modified from Lao et al., 2022. SM, summer monsoon; WM, winter monsoon.

permanganate oxidation method, and the precision was 0.15 mg L^{-1}. The GF/F sample for the measurement of Chl-*a* was extracted using 90% acetone solution (v/v) at 4 °C in darkness for one day (24 h), and then determined using the fluorometric method. The concentrations of NO_2^-, NO_3^- and PO_4^{3-} were determined by colorimetric method using a San++ continuous flow analyser (Skalar, Netherlands). The concentrations of NH_4^+ were determined by spectrophotometry after treatment with Nessler's reagent. NO_3^- was determined using the cadmium–copper reduction method. PO_4^{3-} was determined using the phosphomolybdate blue method. The detection limits of NO_2^-, NO_3^-, NH_4^+ and PO_4^{3-} were 0.0006 mg L^{-1}, 0.0006 mg L^{-1}, 0.00042 mg L^{-1} and 0.00062 mg L^{-1}, respectively.

Eutrophication assessment

The eutrophication in Zhanjiang Bay was evaluated using the eutrophication index (EI), the formula as follow (Huang et al., 2022; Ke et al., 2022):

$$EI = \frac{COD \times DIP \times DIN}{4500} \times 10^6 \quad (1),$$

the units of COD, DIP (PO_4^{3-}) and DIN are mg L^{-1}. Generally, if the value of EI > 1, the seawater in the bay is considered eutrophic. A total of five classifications can be divided to assess the eutrophication, which is widely used to evaluate eutrophication of coastal waters (Andersen et al., 2017; Lao et al., 2021; Huang et al., 2022; Ke et al., 2022), that is as following: EI < 1 represents no eutrophication, 1.0 ≤ EI < 2.0 represents light eutrophication, 2.0 ≤ EI < 5.0 represents moderate eutrophication, 5.0 ≤ EI <15.0 represents high eutrophication, EI ≥ 15.0 represents severe eutrophication.

Results

Seasonal variations of physiochemical parameters and eutrophication in Zhanjiang Bay

The seasonal variations of physiochemical parameters from 2017 to 2021 are presented in Figures 2, 3. Seawater temperature was higher in rainy seasons (summer and fall), whereas a lower value was observed in dry seasons (winter and spring) (Figure 2A). The lower salinity generally occurred in the rainy seasons, whereas higher salinity occurred in the dry seasons (Figure 2B). The lowest salinity in a year generally occurred in the fall. The pH values ranged from 7.12 to 8.33, and the higher values occurred in winter whereas lower values occurred in rainy seasons (Figure 2C). The DO level in the rainy seasons (most < 6 mg L^{-1}) was significantly lower than that in the dry seasons (*t*-test, p<0.001). The seasonal variations of Chl-*a* were not obvious, but there was a remarkably high level in the fall of 2021 (Figure 2E). Except for the remarkably high COD level in the winter of 2019, the level was generally higher in the rainy seasons (Figure 2F). Additionally, concentrations of COD in the upper bay were significantly higher than that in the lower bay (Figure 4, *t*-test, p<0.001).

Nutrient concentrations show significantly seasonal variation, but the trend is prominent among different years (Figure 3). The concentrations of PO_4^{3-} in the rainy seasons were significantly higher than that in the dry seasons in 2017, 2018 and 2021 (*t*-test, p<0.01). Additionally, PO_4^{3-} concentrations in 2017 were significantly higher than those in the same period of other years (except for summer in 2021, *t*-test, p<0.001). In 2019 and 2020, higher PO_4^{3-} concentrations were observed in spring, summer and fall, whereas the lower concentrations occurred in winter. The highest DIN concentration was observed in the fall of 2021 (0.85 ± 0.55 mg L^{-1}), and the concentration in 2021 was significantly higher than those in the same period of other years (*t*-test, p<0.001). The DIN concentrations were significantly higher in rainy seasons (*t*-test, p<0.001), whereas lower concentrations occurred in dry seasons (except for 2019). In 2019, the higher DIN concentration occurred in the dry seasons (spring), whereas lower concentration occurred in summer. Similarly, the concentration of DIN in the spring of 2019 was significantly higher than the other three seasons (*t*-test, p<0.001). These considerable differences in seasonal changes between years reflect the impact of intense human activities and multi-point source discharges in Zhanjiang Bay. Additionally, the degree of eutrophication in rainy seasons was nearly 2 times higher than that in the dry seasons. The highest EI value was observed in the fall, followed by the summer, whereas the lower EI occurred in the spring and winter. However, in 2019, there was no significant seasonal change in EI value in the bay (*t*-test, p>0.05). Additionally, EI values in the upper bay were significantly higher than that in the lower bay (more than 8 times).

The long-term trend of eutrophication in Zhanjiang Bay

Eutrophication in Zhanjiang Bay exhibited a significantly increased trend over the recent five years (2017-2021) (Figure 5). A total of 71-87% of the seawater in Zhanjiang Bay was in eutrophication (EI>1) during 2017-2021, most of which were in moderate to severe eutrophication (Table 1). Significantly, the contribution of severe eutrophication in the seawater of Zhanjiang Bay increased remarkably over the recent five years, nearly half of the seawater in the bay was in severe eutrophication in 2021 (46%). The fluctuating but no obvious trend of COD concentration, and the decrease of PO_4^{3-} concentration but remarkably increase of DIN concentration indicate that the increase of nitrogen discharge may be the main driving factor for the aggravation of eutrophication in Zhanjiang Bay over the recent five years. Compared with the past 30 years, the EI values increased significantly in Zhanjiang Bay (R^2=0.71, p<0.01) (Figure 5). The EI values exhibited a significantly positive correlation with PO_4^{3-}concentration (R^2=0.68, p<0.01), but no correlation with DIN and COD concentration (p>0.05). In addition, the seawater in Zhanjiang Bay was at phosphorus-limited (nitrogen excess) in the 1990s (N:P ratio of 61). However, with the increase of PO_4^{3-} concentration, the seawater in Zhanjiang Bay changed to nitrogen-limitation after entering the 21st century (an average N:P ratio of 7.5), especially in the 2010s (an average N:P ratio

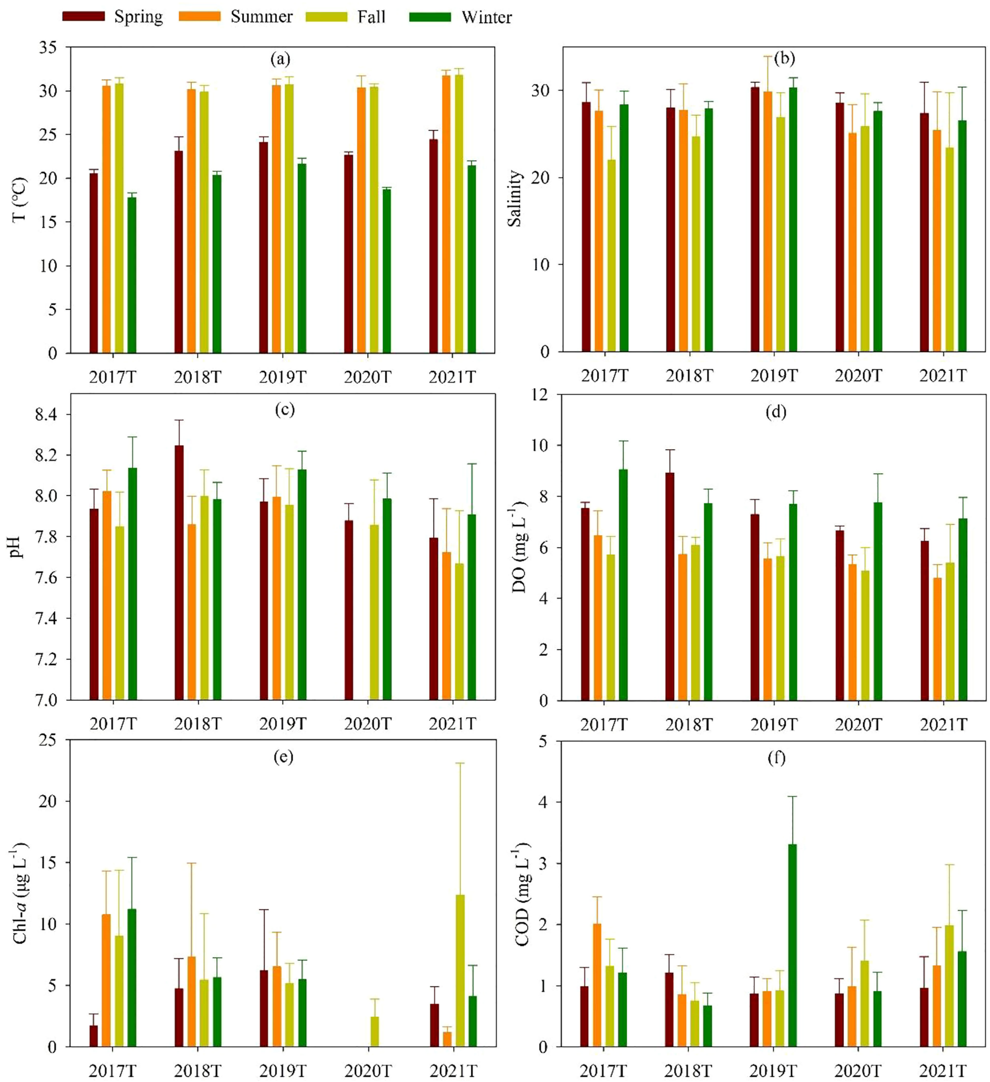

FIGURE 2
Seasonal distributions of temperature, salinity, pH, DO, Chl-*a* and COD in Zhanjiang Bay from 2017 to 2021.

of 5.0). This suggests that increased phosphate discharges may be responsible for the aggravation of eutrophication in Zhanjiang Bay over the past 30 years. Overall, the increase in phosphate discharges is the main reason for the aggravation of eutrophication in Zhanjiang Bay over the past 30 years. However, with the decrease in phosphate discharges and the increase in nitrogen discharges in recent 5 years, the driving factor has changed to the increase in nitrogen discharges.

Discussion

Terrestrial and river diluted-water input dominate the eutrophication in Zhanjiang Bay

The eutrophication in the upper bay is remarkably higher than that in the lower bay (Figure 4), which is not only related to the terrestrial input, but also to the natural topography and tidal current in the bay. Additionally, increasing intrusion of higher salinity water from the outer bay can retain the contaminants in the inner bay (Lao et al., 2022). Suixi River at the top of the upper bay is the main river entering into Zhanjiang Bay, which can directly carry pollutants discharged from agricultural production, domestic sewage and industrial production into the bay (Shi et al., 2015; Li et al., 2020; Lao et al., 2022; Tang et al., 2022). Additionally, due to the different topography between the upper and lower bays in Zhanjiang Bay, the tidal range and tidal current are quite different (Wang et al., 2021c). The upper bay is narrow and curved, and the tidal velocity is slow, which leads to slow water exchange and is not conducive to the diffusion of pollutants. In addition, due to intensive human activities in recent years, such as the dredging for the transportation of ships and artificial dams for farmland irrigation, the intrusion of seawater from the outer bay has elevated remarkably over the past two decades (increased by 23%), resulting

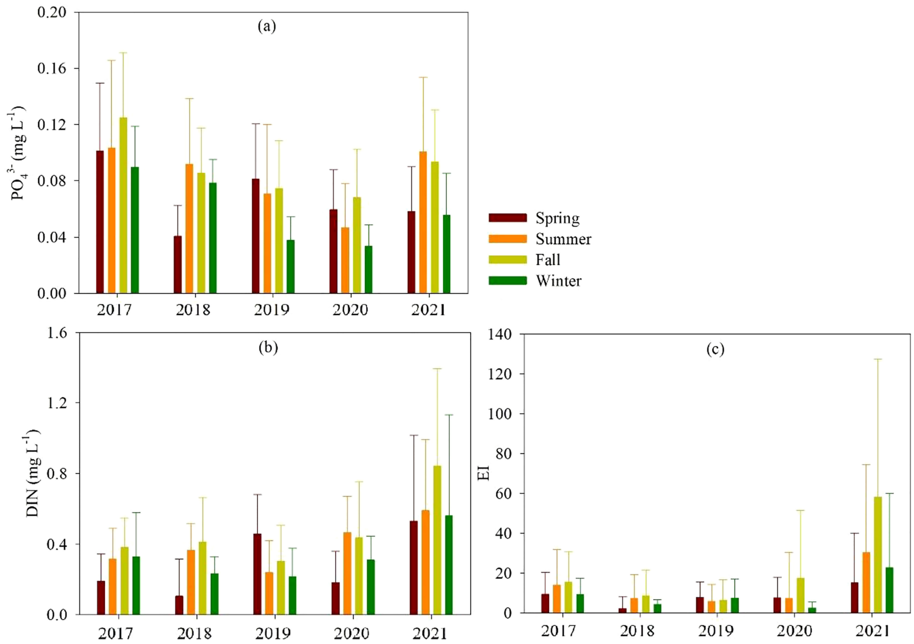

FIGURE 3
Seasonal distributions of nutrients and eutrophication index in Zhanjiang Bay from 2017 to 2021.

that the pollutants of freshwater are retained in the inner bay (Lao et al., 2022). Thereby, the concentrations of DIN, COD and PO_4^{3-}, and EI values were significantly higher in the upper bay than that in the lower bay (Figure 4). However, in the lower bay, a broader area strengthens seawater's self-purification ability of seawater stronger (Shi et al., 2015; Wang et al., 2021c). In addition, the intrusion of high-salinity but relatively oligotrophic water can dilute the nutrients in the lower bay (Lao et al., 2022). Thus, compared to the upper bay, the eutrophication in the lower bay is much lighter.

Seasonally, the EI values were generally higher in the rainy seasons (except for 2019), which are depended on the occurrence of DIN, COD and PO_4^{3-} (Table 2). Indeed, except for 2019, the concentrations of DIN, and PO_4^{3-} were significantly higher in the rainy seasons (Figure 3, *t*-test, $p<0.01$), during which is

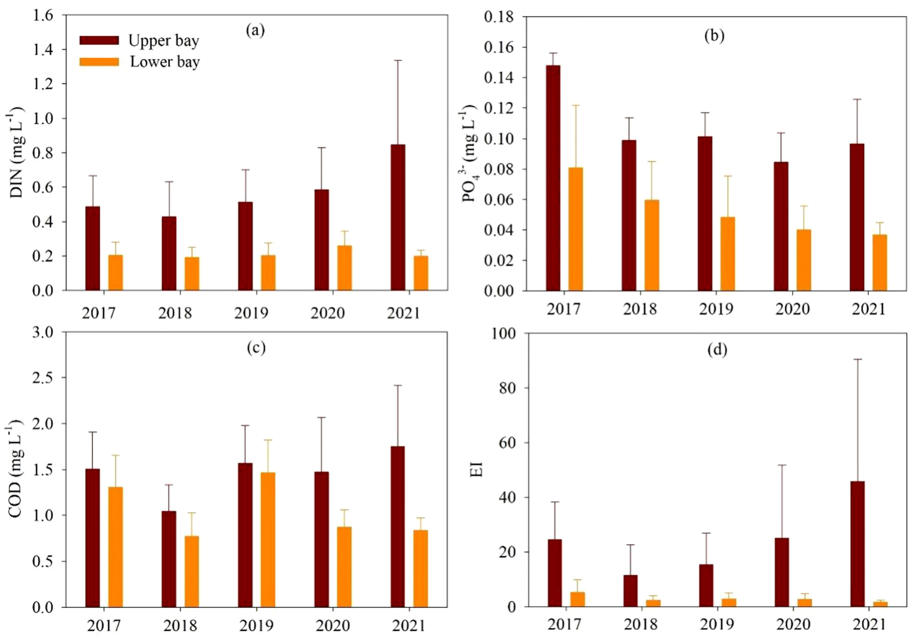

FIGURE 4
The distribution of nutrients, COD and EI in upper and mid bay from 2017 to 2021.

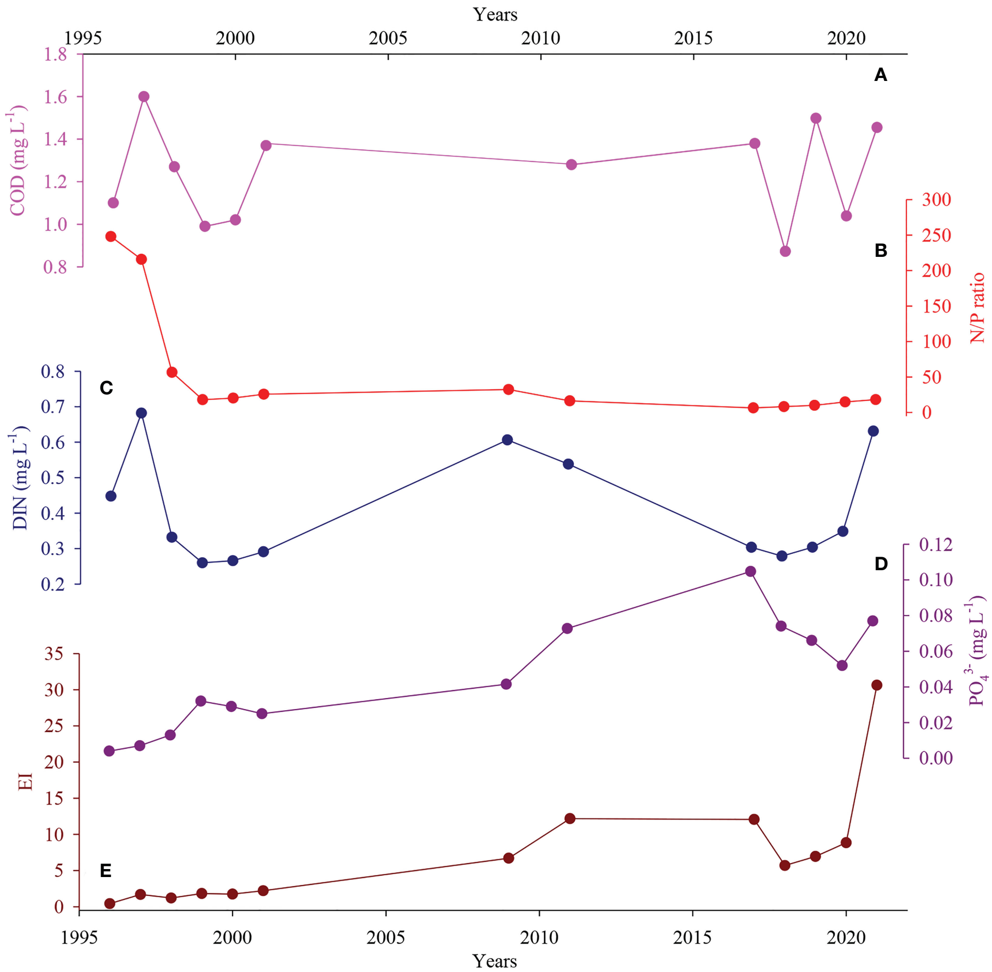

FIGURE 5
Annual variations of eutrophication index, N/P ratio, nutrients (DIN and PO_4^{3-}) and COD in Zhanjiang Bay over the past 30 years. The data for 1996 and 1997 were obtained from Cheng et al., 2012; the data of 1998-2001 obtained from Lu et al., 2002; the data of 2009 obtained from Zhang et al., 2012; the data of 2011 obtained from Shi et al., 2015.

characterized by lower salinity (Figure 2B). Additionally, significantly negative correlations were observed between EI values, DIN, and PO_4^{3-} with salinity (Table 2). This suggests that the DIN and inorganic phosphorus brought by terrestrial discharge and river freshwater maybe the main reasons for the high EI value in rainy seasons. The flood season in Zhanjiang is from April to October every year. The abundant rainfall can wash the point and non-point source pollutants in industrial and agricultural production into Zhanjiang Bay (Shi et al., 2015; Zhang et al., 2021; Zhong et al., 2022), particularly in extremely climates (e.g., heavy rainfall induced by typhoons) (Lao et al., 2023). In the Suixi River basin, massive pollutants can be brought from the wide basin area through watershed into the bay (Zhang et al., 2021; Zhong et al., 2022). The water mixing and related nutrient supply in the bay have been quantified using dual water isotopes, and the results also suggest that the nutrients in Zhanjiang Bay mainly originated from river freshwater input during the rainy seasons (Lao et al., 2022). Although the input of freshwater in Zhanjiang Bay has

TABLE 1 Contribution of different eutrophication status in Zhanjiang Bay during 2017-2021.

Eutrophication status	2017	2018	2019	2020	2021
Eutrophication (EI≥1)	87%	71%	85%	74%	77%
Light eutrophication	7%	9%	12%	19%	4%
Moderate eutrophication	19%	29%	32%	24%	15%
High eutrophication	33%	26%	30%	19%	12%
Severe eutrophication	28%	7%	10%	13%	46%

TABLE 2 Correlation between eutrophication index and environment factors in Zhanjiang Bay.

	T	S	pH	DO	Chl-*a*	DIN	P	COD	EI
T	1.000	-0.086	-0.240	-0.884**	0.205	0.072	0.427	0.017	0.397*
S		1.000	-0.246	0.114	-0.067	-0.560*	-0.554*	0.013	-0.445*
pH			1.000	0.294	0.330	0.202	0.235	0.110	0.050
DO				1.000	0.086	-0.283	-0.328	0.064	-0.439*
Chl-*a*					1.000	0.129	0.534*	0.314	0.375*
DIN						1.000	0.208	0.018	0.794**
P							1.000	-0.109	0.354
COD								1.000	0.353
EI									1.000

*Correlation is significant at the 0.05 level; **Correlation is significant at the 0.01 level.

increased due to a large amount of precipitation in the rainy seasons, the contribution of freshwater only accounts for 7% of the total seawater in Zhanjiang Bay, while the nutrients from freshwater input account for 54-90% of the nutrient load in Zhanjiang Bay (Lao et al., 2022). The rainy seasons are periods of frequent agricultural activities. As Zhanjiang is a city dominated by industry and agriculture, the extensive use of chemical fertilizers in agricultural production and the discharge of industrial and domestic wastewater has brought a large amount of nutrients to Zhanjiang Bay (Li et al., 2020; Zhang et al., 2020; Zhou et al., 2021; Zhang et al., 2023). During rainy seasons, nitrogen fertilizer is the dominant nutrient input in the bay (accounting for 36%), following by soil nitrogen (32%) and sewage and manure (31%) (Li et al., 2020). These nutrients are directly inputted into the upper bay through the runoff from the upper river basin (Li et al., 2020). However, affected by climate events, the rainfall in Zhanjiang in 2019 dropped remarkably (the total rainfall is only about 1460 mm, decreasing by about 25%) (Chen et al., 2021), which would result in a decrease of runoff into the bay. This also is supported by the increase of salinity in Zhanjiang Bay in 2019, which is higher than salinity in the same period of other years (Figure 2B). The decrease in runoff leads to the reduction of the input of terrestrial pollutants, and thus the eutrophication is lighter in the bay during the rainy seasons (Figure 3C).

In dry seasons, the pollutants brought by terrestrial discharge and river freshwater have decreased due to the sharp decrease of runoff input (the contribution of freshwater only accounts for 1% of the bay) (Lao et al., 2022). However, this also increases the intrusion of high-salinity seawater with relatively oligo nutrients (Lao et al., 2022), which can dilute the pollutant loads in the bay. In addition, Zhanjiang Bay is located in the tropical region, with sufficient light in winter and spring and relatively high temperature (Figure 2A), which is conducive to the growth of algae, particularly for the macroalgae (Zhang et al., 2022; Zhang et al., 2023), and thus the Chl-*a* is still at a relatively high level during this period (Figure 2). In the case of a reduced external nutrient supply, algae growth will consume a certain amount of nutrients (Zhang et al., 2022; Zhang et al., 2023).

Aggravated eutrophication in Zhanjiang Bay caused by increase of phosphate discharges

Compared with historical data, the eutrophication in Zhanjiang Bay is significantly increased over the past 30 years (Figure 4D), and the EI values in the recent 5 years are nearly 10 times that of the 1990s. This could be caused by the increase of phosphate discharges despite the PO_4^{3-} concentration has declined in the recent 5 years (Figure 4C). Firstly, the Suixi watershed is primarily agricultural land use, the phosphate fertilizer used in cropland may be the important PO_4^{3-} source in Zhanjiang Bay (Zhang et al., 2021). Because Zhanjiang is a tropical coastal region with abundant rainfall, a large amount of rainfall can import these non-point sources of phosphorus into Zhanjiang Bay through runoff. Secondly, Zhanjiang Bay is surrounded by Zhanjiang city, which locates many industrial factories in the coastal regions. Massive wastewater from industrial factories with high phosphate concentrations is discharged into the bay (Zhang et al., 2021). Thirdly, the urban domestic wastewater discharge with high phosphorus concentration may be another important source of phosphorus in Zhanjiang Bay (Li et al., 2020; Zhang et al., 2021). Among these three sources, the domestic sewage and phosphate fertilizer should not be responsible for the increase of PO_4^{3-} concentration in Zhanjiang Bay over the past 30 years because the usage amount of phosphate fertilizer and the permanent population in Zhanjiang has not increased despite fluctuations in the past years (Figure 6). Additionally, to limit phosphorus discharge, China began to implement the policy of limiting and prohibiting phosphorus as early as the 1990s, and promoted the use of non-phosphorus products (Li et al., 2019). Thus, the increase in phosphorus discharged from industrial factories should be responsible for the increasing PO_4^{3-} concentration in Zhanjiang Bay. Indeed, after entering the 21st century, Zhanjiang's development has shifted from agriculture to industrialization, and its industrial scale has expanded rapidly (Figure 6). It is reported that the annual mean concentration of total dissolved phosphorus in the rivers adjacent to industrial factories, which directly input

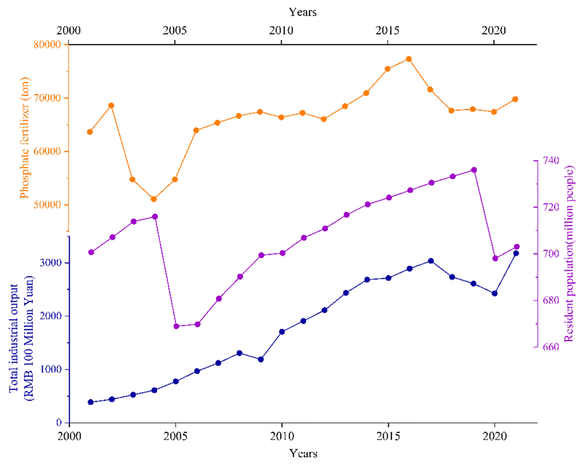

FIGURE 6
Temporal changes in phosphate fertilizer, resident population and industrial scale (total industrial output) in Zhanjiang from 2000 to 2021.

into Zhanjiang Bay, was nearly 6 times that of the coast of Zhanjiang Bay (Zhang et al., 2021). This suggests that the wastewater with high phosphorus discharged from the industrial factory in the city is the key source for increasing PO_4^{3-} concentration in Zhanjiang Bay. It is estimated that the annual total dissolved phosphorus flux in Zhanjiang Bay was 3848.07 tons, which dominated in the rainy seasons (accounting for 52.6%) (Zhang et al., 2021). The high phosphorus flux in the rainy seasons is mainly attributed to the large freshwater discharges induced by heavy precipitation, which can accelerate industrial wastewater flowing from rivers into the bay (Zhang et al., 2021). Thus, the continuous input of high-concentration phosphorus from industrial factories around Zhanjiang Bay could be responsible for the elevated PO_4^{3-} concentration in the bay over the past 30 years. However, due to implementation of a phosphorus restriction policy for industry, phosphorus discharges have decreased in recent 5 years (Wu et al., 2022). This may be responsible for the slight decrease in PO_4^{3-} concentration in the recent 5 years (Figure 4C). Nevertheless, the PO_4^{3-} concentration in the recent 5 years is still more than 5 times that in the 1990s.

In addition, the increase in anthropogenic nitrogen discharges is the main reason for severe nitrogen pollution in Zhanjiang Bay in 1990s. The application of nitrogen fertilizer increased dramatically after 1984, which resulted in remarkably increasing input of the net anthropogenic nitrogen into Zhanjiang Bay (Zhou et al., 2021), and the high DIN concentration in the 1990s (Cheng et al., 2012). However, since Zhanjiang began to implement soil testing and formula fertilization in 2006, the discharge of anthropogenic nitrogen began to exhibit a downward trend in Zhanjiang (Meng et al., 2015). This is consistent with the historical changes in net anthropogenic nitrogen input into Zhanjiang Bay, which exhibited a decreased trend after 2007 (Zhou et al., 2021). However, due to the intensive human activities (e.g., increasing industrialization) and rapid economic development, net anthropogenic nitrogen input into Zhanjiang Bay substantially increased again after 2015 (increased by ~16%). This could be responsible for the increase in DIN concentration in the recent (Figure 4B). Since the COD concentration has fluctuated but has no apparent trend over the past 5 years and the past 30 years (Figure 4A), the increase of eutrophication in Zhanjiang Bay can be related to the increasing phosphate discharge despite of the fact that it has slightly decreased in the recent 5 years. However, the rapid increase of anthropogenic nitrogen discharges can be responsible for the sharply elevated eutrophication in Zhanjiang Bay in the recent 5 years. Notably, DIN concentration has declined slightly in the recent 5 years (an average of 0.37 mg L^{-1}) compared with the 1990s (an average of 0.43 mg L^{-1}). In addition to the implementation of nitrogen emission control measures, the denitrification process may also have an impact on long-term changes in nitrogen. Because denitrification has become an important process in most estuaries, coastal waters, and oceans (Chen et al., 2009; Chen et al., 2020; Chen et al., 2022a; Lao et al., 2019; Zhuang et al., 2022), particularly in the Arctic Ocean, where a larger nitrate deficit has been found due to stronger shelf denitrification (Zhuang et al., 2022). However, such a process still needs further research to confirm in this region.

Overall, the increase of anthropogenic nitrogen and phosphorus discharges is the main factor causing the aggravation of eutrophication in Zhanjiang Bay, it is essential to effectively implement a land-ocean integrated nitrogen and phosphorus control strategy to improve water quality and mitigate eutrophication in the bay.

Conclusion

Seasonal change of nutrients (including NO_2^-, NO_3^-, NH_4^+ and PO_4^{3-}) and other environmental parameters were investigated from 2017 to 2021 to explore the long-term trend and driving factors of eutrophication in Zhanjiang Bay. The concentrations of DIN and PO_4^{3-}, and EI values in the upper bay were significantly higher than that in the lower bay. The eutrophication index in the rainy seasons was significantly higher than that in the dry seasons due to abundant nitrogen and phosphorus brought by terrestrial discharge and river freshwater into the bay during the rainy seasons. Compared with the historical data, the eutrophication in

Zhanjiang Bay significantly increased over the past 30 years, and the eutrophication index in the recent 5 years (2017-2021) is nearly 10 times that of the 1990s. This is mainly related to the increase in phosphate discharges from the increasing number of industrial factories in Zhanjiang. Increasing anthropogenic nutrient input is the key to casue the aggravation of eutrophication in Zhanjiang Bay. Thus, effectively implementing a control strategy is essential to improve water quality and mitigate eutrophication in the bay.

Data availability statement

The original contributions presented in the study are included in the article/supplementary materials. Further inquiries can be directed to the corresponding author.

Author contributions

FC was responsible for the conceptualization. GH and QL prepared and wrote the original draft. QL, FC, GJ and GH wrote, reviewed, and edited the manuscript. QZ, GH and QL were responsible for the data curation. QZ and GH were responsible for the experimental operation. GH, QZ and QL were responsible for field sampling. FC funded the acquisition. All authors contributed to the article and approved the submitted version.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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