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Categorizing numeric nutrients criteria and implications for water quality assessment in the Pearl River Estuary, China

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Coastal eutrophication, the over-enrichment of water with nutrients, has become a global ecological problem. As coastal waters are subjected to great pressure due to anthropogenic influences and climate change, establishing numeric nutrient criteria for coastal waters has been exceedingly complex at present. To control and improve the water quality of the Pearl River Estuary (PRE), based on the data from 2015 to 2020, the nutrient criteria of the PRE and adjacent waters were established using frequency statistical analysis. Based on the spatiotemporal salinity patterns, the coastal waters of the PRE were divided in three subareas namely freshwater (Zone I), mixed (Zone II), and seawater (Zone III) using cluster analysis. The recommended criteria values of dissolved inorganic nitrogen (DIN) were 0.573, 0.312, and 0.134 mg·L^{-1} in Zones I, II, and III, respectively. The total nitrogen (TN) criterion for Zone III (0.222 mg·L^{-1}) was much lower than those for Zone I (0.902 mg·L^{-1}) and Zone II (0.885 mg·L^{-1}).The dissolved inorganic phosphorus (DIP) criteria were different for the three Zones, ranging from 0.004 to 0.009 mg·L^{-1}, and the total phosphorus (TP) recommended criteria in Zones I, II, and III were 0.039, 0.028, and 0.020 mg·L^{-1}, respectively. In the water quality assessment, the categorizing numeric nutrients criteria can be referred and applied into fresh, mixed, and seawater zones of PRE. The results of this study provide a new nutrient reference condition in the PRE, which could be helpful in establishing integrated land-ocean unified nutrient criteria and water quality assessment, and implementing effective coastal eutrophication control in the future.

KEYWORDS

coastal eutrophication, nutrient criteria, water quality, assessment, Pearl River estuary

Introduction

Nitrogen (N) and Phosphorus (P) are the material foundations for the growth and reproduction of phytoplankton, and their composition, concentration, and distribution directly affect the primary productivity of the ocean and food chain (Ryther and Dunstan, 1971, Anderson et al., 2002; Conley et al., 2009; Davidson et al., 2014; Lin G. and Lin X., 2022). In marine environments, N and P nutrients exist in various forms of inorganic and organic matter (Zhang et al., 2019a, Ge et al., 2021). Dissolved inorganic nitrogen (DIN, including NO_3^--N, NO_2^--N and NH_4^+-N) is the main form of N, and its concentration change is the main factor controlling the change in total nitrogen (TN) content (Wei et al., 2014; Zhang et al., 2020b; Hong et al., 2019). In addition, dissolved organic nitrogen (DON) is an important potential direct or indirect nitrogen source for microorganisms and phytoplankton (Lu et al., 2016; Luo, 2017; Luo et al., 2017; Zhang et al., 2020c). As ocean is one of the largest nitrogen pools, its important role in the marine nitrogen cycle has been widely recognized (Luo et al., 2017). The main form of P is dissolved inorganic phosphorus (DIP), which accounts for more than 50% of total phosphorus (TP), and is not only the most preferred phosphorus source for organisms, but also a limiting factor for most of the marine ecosystems (Li, 2017; Zhang et al., 2017; Zhang et al., 2019a; Zhang et al., 2020). When DIP cannot meet the needs of phytoplankton, heterotrophic bacteria in the ocean supplement DIP by hydrolyzing dissolved organic phosphorus (DOP) using alkaline phosphatase (Zhang et al., 2021). With the rapid development of industry and agriculture, the use of pesticides, fertilizers, and fossil fuels has increased significantly, and a large amount of nitrogen and phosphorus nutrients have been transported to the estuaries and coasts through river flow, atmospheric deposition, and groundwater discharge, resulting in severe eutrophication (Herbeck et al., 2013; Li et al., 2014; Ménesguen, 2014; Wang et al., 2018; Zhang et al., 2019b; Zhang et al., 2020a; Peng et al., 2021). Nutrient criteria representing the enrichment status of surface waters that are minimally impacted by human development maybe defined as the threshold value that supports a particular beneficial designated use (US EPA, 1998; US EPA, 2001). Coastal eutrophication has caused serious damage to the ecosystem and disturbed the balance of original ecosystem, resulting in outbreak of harmful algal blooms and the formation of hypoxic or anoxic zones (DePinto and Verhoff, 1977; Boynton et al., 1982; Burkholder et al., 1992; Arai, 2001; Wu, 2007; Lin et al., 2018; Xu, 2020; Ke et al., 2022; Zhang et al., 2022; Cai et al., 2011). Therefore, developing and categorizing the numeric nutrients criteria played an important role in assessing the attainment of designated uses and measuring progress toward achieving water quality goals (US EPA, 1998; US EPA, 2001; Huo et al., 2015; Yang et al., 2019).

Estuaries are mixed areas of freshwater and seawater, and the interaction of hydrodynamic environment, biochemical processes, and human activities here is very complex (Zhang et al., 2017; Li et al., 2018; Lu et al., 2018; Li et al., 2013; Liu et al., 2011). Estuaries and coastal waters are particularly vulnerable to pollution because they are the final receiving waters of most rivers in the upper reaches. Estuarine zoning is the most effective tool for coastal marine resource management, which can identify different types of estuarine, so as to different standards can be used to manage estuarine resources (Hume and Herdendorf, 1988). It is also the premise for the formulation of nutrient criteria (Liu et al., 2011). To reduce and prevent the further aggravation of eutrophication, it is necessary and urgent to develop estuarine nutrient criteria. In the 1960s, the United States carried out research on water quality criteria, published a series of literatures on water quality criteria, and issued technical guidelines for nutrient water criteria for different water bodies (such as lakes, rivers, wetlands), which had a profound impact on the formulation of nutrient criteria or standards for every American state and other countries around the world (Xu, 2020; Department of Ecology and Environment of Jiangsu Province, 2021b). The reference condition approach, mechanistic modelling, and stressor-response analysis are commonly recommended for setting nutrient criteria globally (US EPA, 2000). The US EPA applied the reference state method and mechanical model method to formulate the TN nutrient criteria values for the Tampa Bay and Florida Bay, respectively, while the European Union pioneered the transition node identification method to determine the Baltic Sea nutrient criteria (Florida Department of Environment Protection, 2007; Florida Department of Environment Protection, 2011; Wang et al., 2020; Xu, 2020). The research on nutrient criteria in China started relatively late and began at the beginning of the 20th century (Xu, 1981; Xia and Zhang, 1990). Some progress has been made in establishing nutrient criteria for lakes based on foreign experience (Huo et al., 2009; Huo et al., 2017; Liang et al., 2021). However, systematic studies on nutrient criteria in estuaries are limited (Su et al., 2016; Liu et al., 2018a; Yang et al., 2019). In addition, the existing standards involving nutrient indicators include Environmental quality standards for surface water (GB3838-2002) applicable to freshwater and Seawater Quality Standard (GB3097-1997) applicable to seawater, but they are not applicable to the estuarine area where salt and fresh water are mixed, and the latter only includes two indicators of DIN and DIP, lacking the assessment of TN and TP (Douglas and Mclaughlin, 2014; Su et al., 2017). Some studies have used reference state method to establish nutrient criteria in the Liaohe Estuary and coastal waters (Hu et al., 2011; Su et al., 2016). On the basis of the segmentation of the Jiulong River Estuary, whose nutrient criteria was determined using reference conditions, statistical model analysis, and cumulative frequency distributions (Liu et al., 2018a), a nutrient criteria for the Yangtze Estuary and coastal waters was developed using population distribution approach (Yang et al., 2019). Therefore, categorizing numeric nutrients criteria for the estuarine transitional zones can be developed based on the segmentation of estuary.

The Pearl River Estuary (PRE) is characterized by superior natural conditions, dense population, and developed economy (Yin et al., 2000; Zhou et al., 2018; Li et al., 2020; Huang et al., 2021). With the development of social and economic activities and high-intensity human activities, the wastewater and sewage discharge to the PRE continues to increase, which intensifies the enrichment of nutrients in local waters (Lu et al., 2009). This results in the frequent occurrence of red tide disasters and the increasing degree and expansion of hypoxia phenomenon (Glibert et al., 2018; Geeraert et al., 2021; Ke et al., 2022). The amount of TN discharged from land sources in the PRE is still high, and the water quality has been inferior to Grade IV (DIN > 0.5mg mg·L^{-1} or DIP > 0.045mg/L) for a long time. DIN and DIP are the main factors exceeding the standard, among which DIN exceeds the standard by 1.7–5.9 times (Moffat, 1998; Tong et al., 2015; South China Sea Branch of the State Oceanic Administration, 2017; State of Oceanic Administration, 2018; Yan and Li, 2018; Department of Ecology and Environment of Guangdong Province, 2022a). In the summer of 2019, the concentrations of DIN and DIP in the PRE were 0.168-1.247 mg·L^{-1} and 0.011-0.044 mg·L^{-1}, respectively, and 25% of the monitoring stations were over-eutrophication (Ma and Zhao, 2021). In addition, from 1981 to 1998 and 2000 to 2009, nearly 116 red tides were recorded in the PRE affecting more than 2850 km^2 of coastal waters (Wei et al., 2012). Many red tides had a long duration and high toxicity, endangering the habitat environment of aquatic organisms (Lai et al., 2018). At present, the PRE has been listed as one of the three major areas in the "14th Five-Year Plan" national key sea area of comprehensive management battle, which will drive the overall improvement of the quality of marine ecological environment (Ministry of Ecology and Environment of the People's Republic of China, 2022b). The government and Guangdong Province have also successively put forward plans to prevent and control pollution in estuaries and coastal waters, comprehensively implementing the principle of "coordinating land and sea, taking into account river and sea", strengthening the linkage between land pollution control and comprehensive treatment of marine environment, adhering to both pollution prevention and control and ecosystem protection, so as to realize the whole chain of pollution control from source to end (Ministry of Ecology and Environment of the People's Republic of China, 2022a; Department of Ecology and Environment of Guangdong Province, 2022b). Therefore, to control and improve water quality, provide effective management measures, it is urgent and critical to establish effective nutrients criteria.

Based on the seasonal data of the PRE and adjacent waters from 2015 to 2020, the study objectives were (1) to establish nutrient criteria using the frequency distribution method, (2) to establish water quality standards based on nutrient criteria to evaluate and verify it, and (3) to estimate the background concentrations of N and P and implement zoning management in the PRE. This study can provide a scientific basis for effectively solving the problem of eutrophication, evaluating water quality in coastal waters, and joint prevention and control of water pollution land-ocean coordination.

Materials and methods

Study areas

The Pearl River originates from Mashen Mountain in the Wumeng Mountain System of the Yunnan-Guizhou Plateau, with a total drainage area of 453690 km^2, and flows through Yunnan, Guizhou, Guangxi, Guangdong, Hunan, Jiangxi, and northern Vietnam (Zhao, 1989; Cui et al., 2020). The Pearl River is rich in water, with an annual inflow of 326 billion steres and an annual runoff of over 330 billion steres, followed by the Yangtze River. It is the largest water system in southern China (Zeng et al., 2020). The runoff of the Pearl River varies greatly, and the annual distribution of runoff is corresponding to the rainy season. The runoff in the wet season accounts for 70-80% of the total amount of the whole year, which is characterized by the wet > normal > dry season, indicating that the runoff is of the type of precipitation replenishment (Zhao, 1989; Yuan, 2005). The PRE is more than 80 km long, from north to south, and covers an area of 2100 km^2. It borders Guangzhou in the north, Shenzhen and Hong Kong in the east, Zhuhai and Macao in the west, and is located in the core of the Guangdong-Hong Kong-Macao Greater Bay Area (Huang X. and Huang L., 2002) (Figure 1A). The tidal nature of the PRE belongs to irregular semi diurnal mixed tide. The surface flow velocity is higher than the bottom flow velocity, and the summer flow velocity is higher than the winter flow velocity (Xia and Zhou, 2021). With the rapid development of industry and agriculture along the PRE and the continuous influx of population, pollutants such as nitrogen, phosphorus, and other nutrients enter the PRE through artificial discharge, river carrying, precipitation, and other means, resulting in eutrophication and deterioration of water quality (Niu et al., 2020a, Niu et al., 2020b, Qiu et al., 2010, Yin et al., 2013). The environmental quality of the PRE ecosystem needs to be improved urgently (Strokal et al., 2017; Zeng et al., 2020).

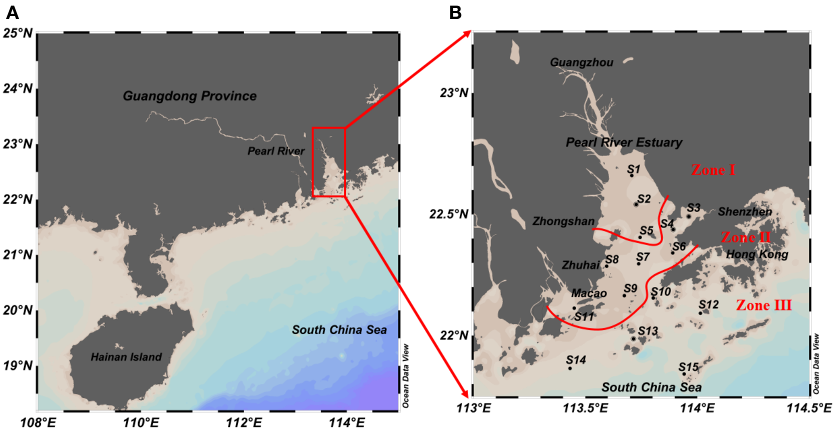

FIGURE 1
Study area **(A)** and monitoring stations **(B)** of the Pearl River Estuary coastal water.

Data sources and chemical analysis

Monitoring data in the PRE were acquired from the Department of Ecology and Environment of Guangdong Province. During the six years from 2015 to 2020, the study area was sampled in the dry (April), wet (July), and normal (October) water flow seasons every year, totaling to 18 sampling seasons. A total of 15 stations were set in the study area (Figure 1B) and 921 datasets were collected, including 262 groups of DIN, 90 groups of TN, DIP groups of 262, TP groups of 90, and 217 groups of salinity. Water samples were collected using cleaned polyethylene bottles or hard glass bottles and promptly brought back to the laboratory for filtration within 24 h. The relative standard deviations (RSD) of repeated determinations of selected samples were less than 5%. The details of analytical methods, processes and instruments in this study had been described in the Specification of Oceanographic Survey (China National Standardization Management Committee, 2007; China National Standardization Management Committee, 2007b; Ministry of Environmental Protection of the PRC, 2009).

Analytical approach of nutrients criteria

Combined with domestic and foreign experience, the frequency distribution method was used to establish nutrient water criteria in the PRE. In the case of sufficient data or good habitat condition, the corresponding value of the 25th percentile (or other suitable percentile) of the nutrient index frequency distribution curve was used as the criteria value. The selection of specific percentage point is only a suggestion and can be determined according to the habitat of the study area. If the area is heavily polluted, the corresponding value of the 5th percentile (or other appropriate percentile) on the frequency distribution curve of nutrient indicators can be selected as the criteria value (Hu et al., 2011; McLaughlin, 2014; Yang, 2015). After processing and integrating the experimental data, the K-S test (Kolmogorov-Smirnov test) was conducted to determine whether the data conformed to a normal distribution. If it conformed ($P \geq 0.05$), a frequency distribution diagram was drawn. If not ($P < 0.05$), the partial maximum or minimum values were removed or logarithmic transformation was carried out before testing, and then the corresponding frequency distribution diagram was drawn. Based on the figure, reference to the historical data and the data of this study, water quality criteria were determined, and then compared with Seawater Quality Standard to evaluate, interpret, and correct the recommended water quality criteria. The detailed process is illustrated in Figure 2 (US EPA, 1998; US EPA, 2001; Meng et al., 2008; Li et al., 2019).

Data processing methods

Microsoft Excel 2016 was used to record and organize the data, and Statistical Product and Service Solution (SPSS) software was used for salinity clustering analysis to complete the estuary division. The establishment of the nutrient criteria adopted the frequency distribution method, and the normal distribution test of each nutritional index was performed using SPSS to determine the criteria. Ocean Data View (4.0) (Schlitzer, 2002) was used to draw a schematic diagram of the monitoring stations and a spatiotemporal distribution map. Other images,

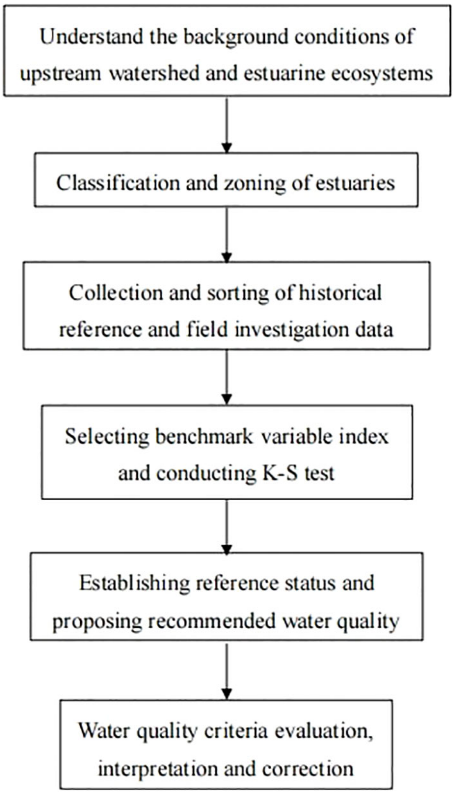

FIGURE 2
Framework of establishing water quality criteria in the PRE coastal water (Li et al., 2019).

such as the frequency distribution curve and salinity cluster analysis dendrogram, were drawn using Origin Lap 2021 software. The average data used in this study are expressed as mean ± standard deviation (mean ± SD).

Results

Spatiotemporal salinity variations and segmentation solutions in the PRE coastal water

Among the three periods in the PRE, the average salinity was 18.48 ± 8.86 PSU, and the maximum and minimum salinity were 32.48 PSU in the dry season and 2.23 PSU in the wet season, respectively. The salinity in the PRE decreased from dry to wet water flow period, and then remarkable increased during the normal water flow period. The salinity range in surface seawater in the dry season was 5.47–32.48 PSU, with an average value of 19.92 ± 8.74 PSU. In the wet season, salinity in the surface water had a range from 2.23–27.99 PSU, and the average was 14.31 ± 8.06 PSU, which differed from the other two seasons and was the lowest. The average salinity in the normal season was 21.21 ± 8.73 PSU, which ranged from 5.71–32.15 PSU. Spatially, the distribution of salinity was similar during the three water flow periods, and it was low in the upper part of the estuary and gradually increased to the open sea (Figure 3).

Applying the SPSS cluster analysis model with salinity as the reference condition, the PRE can be divided in three zones (Figure 4). The first subsection contained S1, S2, and S5, which included the upper estuary, whose water characteristics are mainly freshwater, and the average salinity value was 6.93 ± 2.56 PSU. The second subsection contains S3, S4, S6, S7, S8, S9, and S11. This area belongs to the middle of the estuary and is the largest area where salt water and fresh water are mixed in different proportions, with an average salinity of 16.71 ± 1.49 PSU. The third subsection contained S10, S12, S13, S14, and S15, which are the ocean-dominated areas in the lower part of the estuary with an average salinity of 27.96 ± 2.56 PSU. As a result, based on the morphological characteristics of the estuary and the

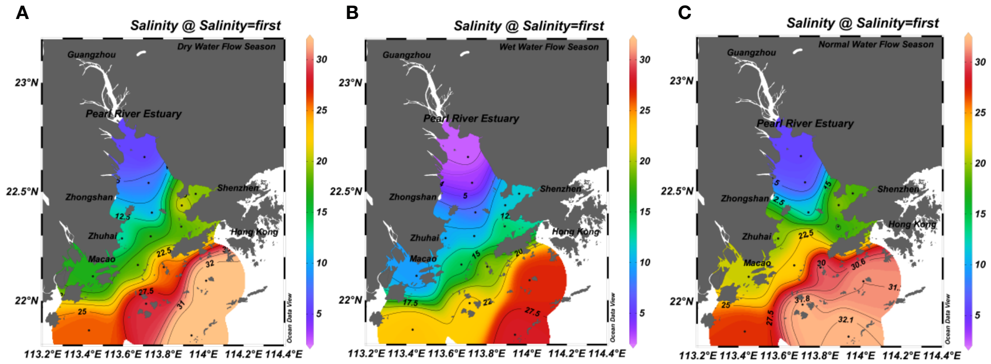

FIGURE 3
Spatial and temporal distribution of salinity along the PRE coastal water in the dry **(A)**, wet **(B)**, normal **(C)** season.

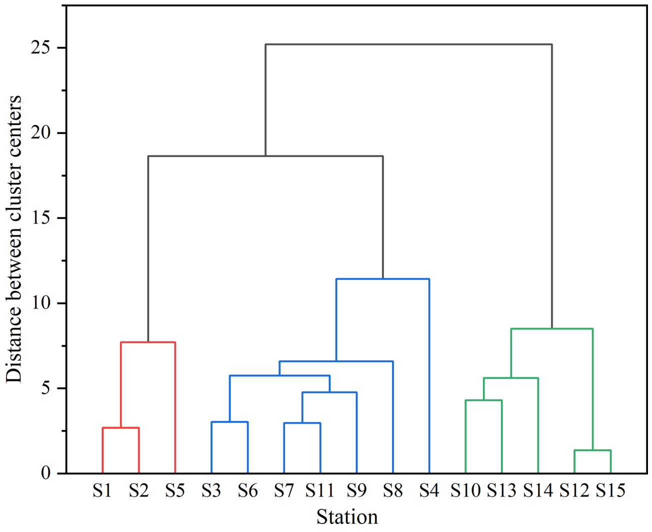

FIGURE 4
Cluster analysis tree of salinity.

spatial distribution of salinity (Figure 3), the PRE can be divided in Zone I (S1, S2, S5), Zone II (S3, S4, S6, S7, S8, S9, S11), and Zone III (S10, S12, S13, S14, S15).

DIN and TN criteria in the PRE coastal water

The frequency distributions of nitrogen and phosphorus nutrients in the PRE was obtained through a normal distribution test and frequency distribution analysis of the partitioned data (Figure 5, Table 1, and Table A1).

In Zone I, the average value of DIN in the three water flow seasons was 1.459 mg·L^{-1}, with a median of 1.445 mg·L^{-1}, varying from 0.098 to 3.237 mg·L^{-1}. The average value and median of TN were 1.832 mg·L^{-1} and 1.783 mg·L^{-1}, respectively, and the variation range was 0.177–3.220 mg·L^{-1} from the dry to the normal season. According to the Bulletin on Marine Environment of Guangdong Province from 2015 to 2017 and the Bulletin on Ecological Environment of Guangdong Province from 2018 to 2020, the water quality of coastal waters along the PRE is poor. Zone I is located in the estuary, and the water quality of most sea areas was Grade IV or inferior. In recent years, DIN has exceeded standard limits. Therefore, for DIN and TN the corresponding value of the 5th percentage point of the frequency distribution curve was selected as the reference state to propose the criteria value, which were 0.573 mg·L^{-1} for DIN and 0.902 mg·L^{-1} for TN (Figure 6).

In Zone II, the average value of DIN in the three water flow seasons was 0.953 mg·L^{-1} and the median was 0.912 mg·L^{-1}, varying from 0.116 to 2.379 mg·L^{-1}. The average and median values of TN were 1.287 mg·L^{-1} and 1.270 mg·L^{-1}, respectively, and the variation range was 0.286–2.480 mg·L^{-1}. Based on the Bulletin on Marine Environment of Guangdong Province from 2015 to 2017 and the Bulletin on Ecological Environment of Guangdong Province from 2018 to 2020, the water quality of coastal waters along the PRE was poor, but it was Grade I or Grade II in most of the sea area of Zone II, which was better than Zone I. The 15th percentage point of the water quality frequency distribution curve was selected as the reference state to propose the recommended water quality criteria, which were 0.312 mg·L^{-1} for DIN and 0.885 mg·L^{-1} for TN (Figure 6).

In Zone III, the average value of DIN in the three water flow seasons was 0.217 mg·L^{-1} and the median was 0.250 mg·L^{-1}, varying from 0.020 to 1.049 mg·L^{-1}. The average and median values of TN were 0.432 mg·L^{-1} and 0.347 mg·L^{-1}, respectively, and the variation range was 0.038–1.340 mg·L^{-1}. It can be seen from the data that nutrient pollution in Zone III was not as serious as that in Zones I and II. It may be due to the distance from the estuary, the reduction of the influence of terrestrial input and the physical mixing process of ocean currents (Liu et al., 2020). However, it is possible that there is a negative correlation between salinity and nitrogen and phosphorus nutrients (Hu et al., 2016; Zhou et al., 2022; Ke et al., 2022). Therefore, the 25th percentile under the water quality frequency distribution curve was selected as the reference state, and the recommended water quality criteria were proposed as 0.134 mg·L^{-1} for DIN and 0.222 mg·L^{-1} for TN (Figure 6).

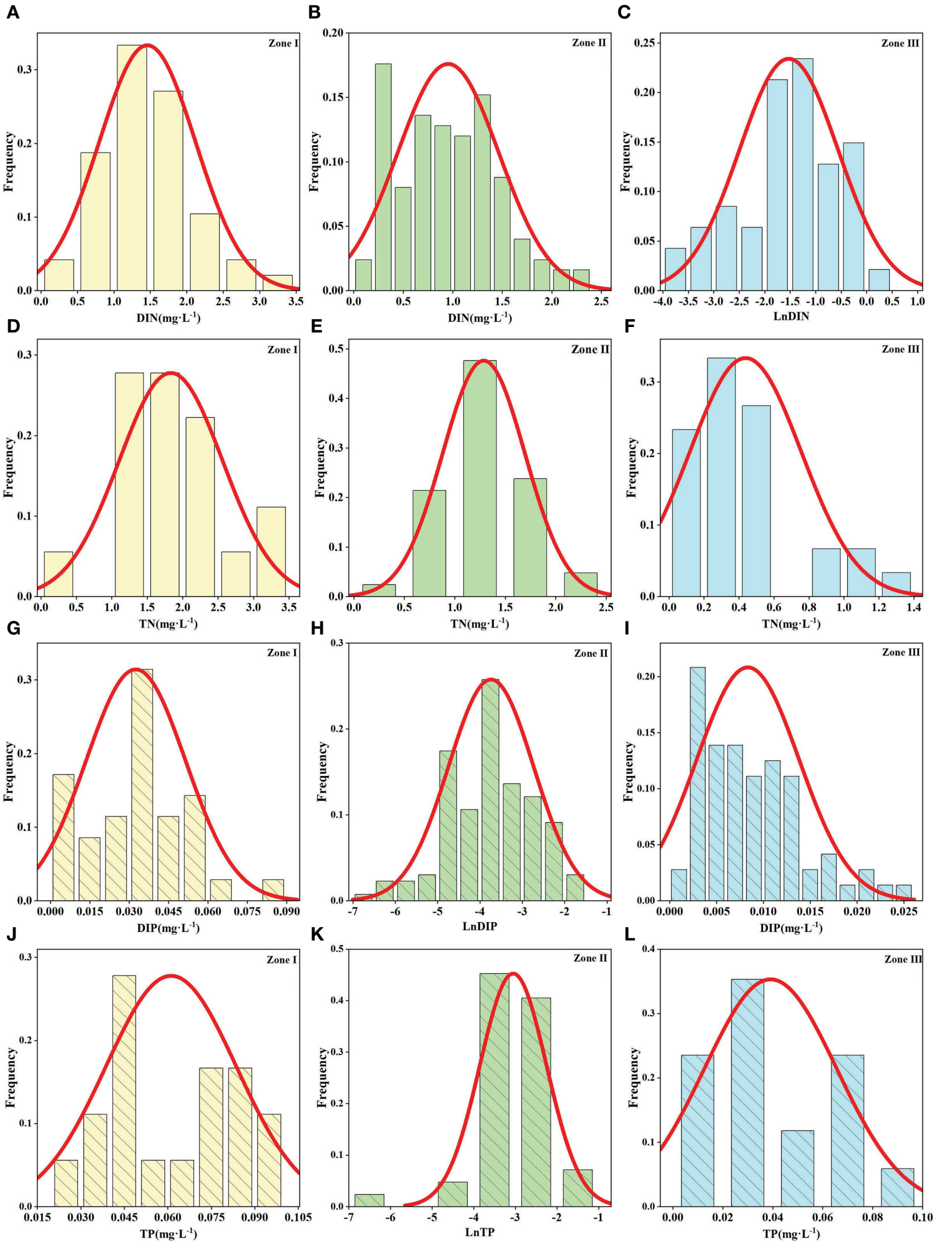

FIGURE 5
Frequency distributions of DIN **(A–C)**, TN **(D–F)**, DIP **(G–I)**, and TP **(J–L)** in the PRE coastal water.

DIP and TP criteria in the PRE coastal water

In Zone I, the average value of DIP in all three seasons was 0.033 mg·L^{-1}, with a median of 0.033 mg·L^{-1}, ranging from 0.002 to 0.086 mg·L^{-1}. The mean value of the TP concentration during the three seasons was 0.039 mg·L^{-1}, the median was 0.059 mg·L^{-1}, and the variation range was 0.028–0.098 mg·L^{-1}. For DIP and TP the corresponding value of the 15th percentage point of the frequency distribution curve was selected as the reference state to propose the criteria value, which were 0.009 mg·L^{-1} for DIP and 0.039 mg·L^{-1} for TP (Figure 7).

In Zone II, during the three water flow seasons, the average value of DIP was 0.024 mg·L^{-1}, with a median of 0.024 mg·L^{-1}, ranging from 0.001 to 0.201 mg·L^{-1}. The mean value of the TP concentration was 0.047 mg·L^{-1}, the median was 0.049 mg·L^{-1}, and the variation range was 0.002–0.259 mg·L^{-1}. The 15th percentage point of the water quality frequency distribution

TABLE 1 Frequency distribution results of DIN, TN, DIP and TP in the PRE coastal water (mg·L^{-1}).

Area	Nutrient	Number	5%	15%	20%	25%	50%	75%	85%
Zone I	DIN	48	0.573	0.790	0.924	1.019	1.445	1.798	2.108
	TN	18	0.902	1.247	1.289	1.377	1.783	2.166	2.402
	DIP	35	0.004	0.009	0.015	0.019	0.033	0.046	0.051
	TP	18	0.035	0.039	0.040	0.041	0.059	0.082	0.085
Zone II	DIN	125	0.230	0.312	0.402	0.565	0.912	1.320	1.492
	TN	42	0.745	0.885	0.935	1.035	1.270	1.513	1.626
	DIP	132	0.005	0.009	0.010	0.011	0.024	0.049	0.067
	TP	42	0.018	0.028	0.032	0.033	0.048	0.076	0.102
Zone III	DIN	47	0.038	0.073	0.127	0.134	0.249	0.414	0.628
	TN	30	0.117	0.160	0.190	0.222	0.347	0.521	0.743
	DIP	72	0.002	0.002	0.003	0.004	0.008	0.012	0.013
	TP	17	0.001	0.016	0.017	0.020	0.035	0.063	0.070

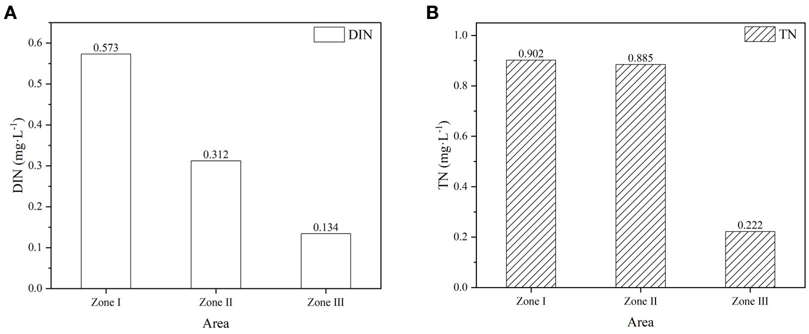

FIGURE 6
Recommended criteria for DIN **(A)** and TN **(B)** in the PRE coastal water.

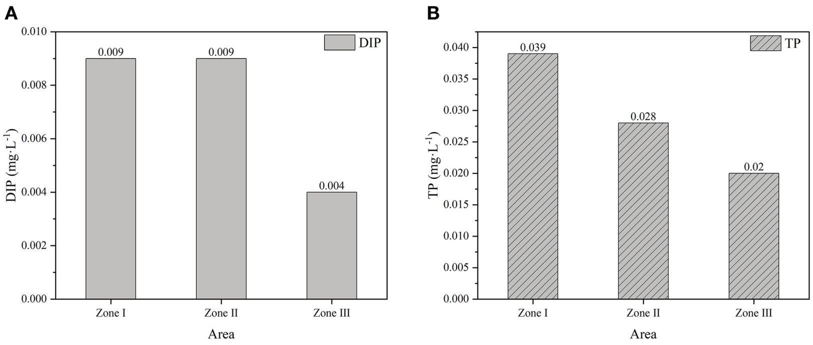

FIGURE 7
Recommended criteria for DIP **(A)** and TP **(B)** in the PRE coastal water.

curve was selected as the reference state to propose the recommended water quality criteria, which were 0.009 mg·L^{-1} for DIP and 0.028 mg·L^{-1} for TP (Figure 7).

In Zone III, the average DIP value in all three seasons was 0.008 mg·L^{-1}, with a median of 0.008 mg·L^{-1}, ranging from 0.001 to 0.024 mg·L^{-1}. The mean value of the TP concentration was 0.039 mg·L^{-1}, the median was 0.035 mg·L^{-1}, and the variation range was 0.001–0.085 mg·L^{-1}. The 25th percentile under the water quality frequency distribution curve was selected as the reference state, and the recommended water quality criteria were proposed as 0.004 mg·L^{-1} for DIP and 0.020 mg·L^{-1} for TP (Figure 7).

Water quality assessment standard for DIN, TN, DIP and TP in the PRE coastal water

According to the sea water quality standard and different applicable functions and protection objectives, the coastal waters of the PRE were divided in four functional areas (Table A2), and the marine water quality standards of DIN, TN, DIP, and TP were formulated.

For the water quality standard limits of nitrogen and phosphorus applicable to class I marine functional areas, the seawater quality standard needs to be stricter because it involves areas with special needs to be protected, such as marine nature reserves and rare and endangered marine biological reserves. The sea water and environmental quality standards for surface water stipulate relatively strict water quality requirements for areas requiring special protection, such as nature reserves. Therefore, it was formulated with reference to the water quality criteria for coastal waters in the PRE (Table 2).

For the water quality standard limits of nitrogen and phosphorus applicable to class II marine functional areas, considering the accessibility and economic factors of the standards and the large amount of nutrients required for aquaculture biological bait in aquaculture areas, the standard values of nitrogen and phosphorus should be relaxed appropriately. The nutrient water quality standards for fishery waters in the seawater quality standard and the environmental quality standards for surface water are considerably higher than those for nature reserves. The nutrient indicators in Zones I and II can be formulated with reference to a cumulative frequency of 25% in the results of the frequency distribution curve, and Zone III can be formulated with reference to a cumulative frequency of 50% in the results of the frequency distribution curve (Table 2).

Classes III and IV include general industrial water use areas, coastal scenic tourism areas, marine port waters, and marine development operation areas. As the nutrient index has relatively little impact on the use function and protection objectives of the two types of marine functional areas and considering the accessibility and economic factors of the standard, the nutrient standard limit should be more relaxed. Similar situations are reflected in the seawater quality standards and environmental quality standards for surface water. The standard was developed with reference to the values of 50% and 70% of the cumulative frequency in the statistical results of the frequency distribution curve in Zones I and II, whereas Zone III is formulated with reference to the cumulative frequencies of 75% and 85% in the statistical results of the frequency distribution curve method (Table 2).

Discussion

Comparison of nutrients by reference condition

During the 40 years from 1980 to 2020, nutrients in the PRE showed different trends (Long et al., 2020). The concentration of DIN showed a very significant increasing trend, while the DIP

TABLE 2 Sea water quality standard in coastal waters of the PRE (mg·L^{-1}).

Area	Nutrient	I Grade	Ⅱ Grade	III Grade	IV Grade
Zone I	DIN	0.573	1.019	1.445	1.789
	TN	0.902	1.377	1.783	2.166
	DIP	0.009	0.019	0.033	0.046
	TP	0.039	0.041	0.059	0.082
Zone II	DIN	0.312	0.565	0.912	1.320
	TN	0.885	1.035	1.270	1.513
	DIP	0.009	0.011	0.024	0.049
	TP	0.028	0.033	0.048	0.076
Zone III	DIN	0.134	0.249	0.414	0.628
	TN	0.222	0.347	0.521	0.743
	DIP	0.004	0.008	0.012	0.013
	TP	0.020	0.035	0.063	0.070

concentration began to decrease after a significant increase, and then remained unchanged (Howarth, 2008; Huang, 2008; Long et al., 2020; Ma et al., 2009). In the early 1980s, the concentration of DIN was only 0.120 mg·L^{-1} (Yuan, 2005), and 0.270 mg·L^{-1} in 1986 (He et al., 2004), which is close to the DIN criteria of the PRE seawater area (0.134 mg·L^{-1}) and the mixing area (0.312 mg·L^{-1}); therefore, the DIN concentration in the PRE without pollution was roughly between 0.120 mg·L^{-1} and 0.312 mg·L^{-1}. Before 1990, the DIN concentration in the PRE increased slowly and reached as high as 0.510 mg·L^{-1} in 1995 (Dang et al., 2019). Then, it began to increase explosively and reached a peak of 1.256 mg·L^{-1} in 2017, increasing more than 10 times. The concentration of DIP was maintained between 0.008 and 0.015 mg·L^{-1} before 1990 (Yuan, 2005), which was comparable to the DIP criteria proposed in this study for the PRE seawater area (0.004 mg·L^{-1}), mixing area (0.009 mg·L^{-1}), and freshwater area (0.009 mg·L^{-1}). The DIP concentration in the PRE without pollution was approximately 0.004–0.015 mg·L^{-1}. After 1990, the DIP began to increase remarkably, reaching 0.070 mg·L^{-1} in 2010 (Dang et al., 2019), which had increased more than eight times in the past 30 years. It then began to fluctuate at a high level, showing an oscillatory downward trend.

Some studies have shown that the water quality of the PRE has gradually changed from exceeding the standard of heavy metals to nutrients around 1990, and excessive pollutants were mainly DIN and DIP (Yuan, 2005; Dang et al., 2019). However, owing to the lack of long-term continuous monitoring data and seawater quality assessment for TN and TP, the phenomenon of exceeding the standard may have occurred earlier. In recent years, the Guangdong Provincial Department of Ecology and Environment and many studies have monitored TN and TP in the PRE. The results suggested that the average concentrations of TN and TP were 1.113 and 0.050 mg·L^{-1} in 2019-2020, respectively (Department of Ecology and Environment of Guangdong Province, 2020; Department of Ecology and Environment of Guangdong Province, 2021). Compared with 2014, TN decreased, while TP was relatively constant (Zhang et al., 2015). In addition, TN/TP can also reflect the demand of phytoplankton for nutrients, reflecting the total amount of nutrients that may be absorbed and utilized by phytoplankton in the sea area (Redfield et al., 1963; Huang and Hong, 1999; Chen et al., 2013; Jin and Liu, 2013). Therefore, it is necessary to establish the relationship between DIN and TN, DIP and TP, and to calculate the background concentration of TN and TP to evaluate the criteria of TN and TP, to better understand their impact on eutrophication.

There is a negative correlation between DIN and DIP and salinity in the PRE, indicating that the terrigenous nutrients are strongly influenced by the mixing of sea and fresh water (Lan et al., 2014; Ke et al., 2022; Hong, 2022; Smith et al., 1999). The runoff input regulates the distribution of N and P to the sea, resulting in a gradual decrease in DIN and DIP from south to north. Therefore, the N and P nutrient criteria values (except DIP) are shown as Zone I > Zone II > Zone III.

Comparison of numeric nutrient criteria

The numerical nutritional standards for the different estuaries and coastal waters were different (Table 3). This may be due to the natural characteristics of different estuaries, such as dilute water, water residence time, and vertical stratification (US EPA, 2001), as well as differences in methods of deriving nutrient criteria in different countries (Wang et al., 2020). The recommended nutrient criteria in the PRE were comparable to those found in most of the estuaries as listed in the Table 3. The research on water criteria in China mainly focuses on DIN and DIP, while the international studies focus on TN and TP (Raymond et al., 2011; Evans-white et al., 2013), and the criteria values of TN and TP compared with those in the coastal waters of estuaries of China, are relatively strict. Among these the TN criteria of the coastal waters of the Yangtze River Estuary are more than 10 times that of the coast of Hawaii, and that of the freshwater area of the PRE is about six times (Table 3). Compared to other estuaries in China, the Yangtze River Estuary is the largest estuary in China and is located in the core area of the Yangtze River Delta economy. It is also the most developed area for industry and agriculture in China. In recent years, the impact of rapid economic development on near-shore water quality has become increasingly evident. Moreover, coastal waters of the Yangtze River estuary are affected by water masses such as the Yangtze River flushing fresh water, Jiangsu-Zhejiang coastal current, northern Jiangsu coastal current, and Taiwan warm current, and the hydrological environment is relatively complex (Li et al., 2022). They all resulted in lower DIN, TN, DIP, and TP nutritional criteria than those for other estuarine areas in China. There are few differences in the DIP and TP criteria between the PRE and Liaohe River Estuary, which may indicate that the two estuaries have similar phosphorus distribution characteristics, both of which are phosphorus limited (Wu et al., 2017; Zeng et al., 2020; Chen et al., 2020; Ke et al., 2022). Some studies have shown that nitrogen in the PRE ranks first among many estuaries in China (Huang et al., 2003; Lai et al., 2018). To alleviate nitrogen pollution, the recommended standards for DIN and TN are relatively strict.

Recommended standard certification and analysis of results

In this study, the monitoring data of DIN and DIP in the near-shore waters of the PRE from 2015 to 2020 were used to evaluate the new standards proposed in this study, while the

TABLE 3 Reference condition or criteria in different coastal water and estuaries worldwide (mg·L^{-1}).

Zone		DIN	TN	DIP	TP	References
Coastal water of Jiangsu Province	mixed area		0.95		0.074	Department of Ecology and Environment of Jiangsu Province, 2021a
	seawater area		0.41		0.042	
Jiulong River Estuary	fresh water area	0.896		0.028		Liu et al., 2018
	mixed area	0.294		0.024		
	seawater area	0.196		0.028		
Yangtze River Estuary		1.05-1.24	1.56-1.85	0.030-0.036	0.060-0.072	Yang et al., 2019
Liaohe River Estuary		0.77	1.11	0.005	0.037	Su et al., 2016
Hawaii	coastal waters		0.15		0.020	US EPA, 2009
	estuarine area		0.20		0.025	
Biscayne Bay			0.24-0.38		0.005-0.009	Briceo et al., 2010
Europe	Baltic coast		0.013-0.033			Helcom, 2015; Helcom, 2017; Herrero et al., 2019
	Mediterranean coast		0.0115-0.0186			
	estuary		0.089-0.105			
Pearl River Estuary	fresh water area	0.573	0.902	0.009	0.039	This study
	mixed area	0.312	0.885	0.009	0.028	
	seawater area	0.134	0.222	0.004	0.020	

exceedances of DIN and DIP in the same period were analyzed using the seawater quality standard (GB3097-1997). If the seawater quality standard was adopted for assessment, the DIN concentration in the three seasons did not meet the Grade IV water quality standard by more than 50%, especially in the dry season, which was 56.67%. At the same time, only approximately 40% of the DIN concentration met the Grade I, II, or III water quality standards. Using the recommended water quality standard proposed in this study, the concentration of DIN did not meet the Grade IV water quality standard decreased by nearly 75% in the normal period, while DIN concentration met Grade I, Grade II, and Grade III water quality standards and increased to more than 60%, and up to 76% in the normal water flow season (Figure 8A). As for DIP, if the seawater quality standard was employed, the DIP concentration in the dry, wet, and normal seasons met the water quality standards of Grade I and Grade II, reaching approximately 70%, and the water quality in the inferior Grade IV was less than 20%. If the water quality standard recommended in this study was used, the proportion of DIP concentration that met Grade I and Grade II water quality standards during the three seasons considerably decreased and dropped by half in the dry season, while the proportion of DIP concentration in Grade IV or inferior increased by 59% in the dry season (Figure 8B).

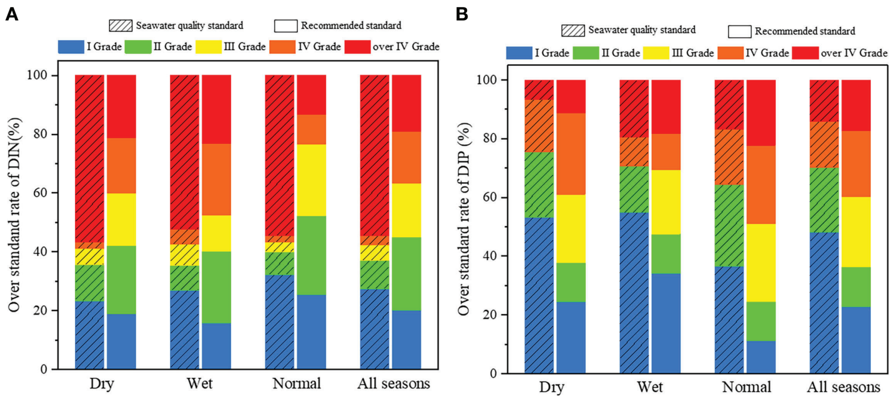

FIGURE 8
Seawater quality standard and the recommended standard assessment DIN **(A)** and DIP **(B)**.

Currently, there are no seawater assessment standards for TN and TP in China. The study also used monitoring data from the PRE in 2019 and 2020 to verify the TN and TP specified in this standard. The proportions of TN and TP failed to meet the Grade IV water quality standard, which was the highest in the wet season. Overall, TN and TP met the Grade III water quality standard (Figure 9). The compliance rate of TP meeting the Grade III water quality standard was higher than that of DIN, TN, and DIP. As shown in the picture, using the recommended standard to evaluate DIN, TN, DIP and TP, the annual compliance of each index was almost the same, the proportion of each nutrient standard to reach Grade III water quality standard was more than 60%. Therefore, it can be tentatively judged that there may be “over-protection” for DIN and “under-protection” for DIP in the current seawater quality standard of the PRE. Therefore, from the management perspective, the new standard had no obvious impact on the assessment of seawater quality in the coastal waters of the PRE. From a scientific point of view, the standard is formulated based on the monitoring data of PRE over the years and conforms to the status of the environmental quality of PRE. Since the standard is a recommended local standard and was first explored in China, it is scientifically reasonable and operable, although it cannot consider all aspects.

Implications of establishing estuarine nutrients criteria for alleviating eutrophication

There are differences in the nutrient monitoring indicators stipulated in China’s current seawater quality standard and environmental quality standards for surface water, and they are applicable to seawater and freshwater, respectively (Ministry of Ecology and Environment of the People’s Republic of China, 1997; Ministry of Ecology and Environment of the People’s Republic of China, 2002). However, they may not be fully applicable to estuaries in which seawater and freshwater are mixed (Su et al., 2016; Li et al., 2020). Therefore, the significance of establishing estuarine nutrient criteria for eutrophication mitigation is as follows: first, the water quality index system is unified, supplementing the seawater quality assessment of TN and TP, and the nutrient pollution status and source of nutrient pollution are objectively assessed (Yang, 2015); second, the management of estuaries by zoning and classification has been realized, and the establishment of a joint mechanism for pollution prevention and control of watershed, estuary, and coastal sea area waters has been promoted (Song et al., 2021); finally, the establishment of estuarine nutrient criteria realizes the overall planning of land and sea, and takes into account rivers and seas, which promotes water pollution prevention and control, water ecological protection, and water resources management, which is of great significance to further realize the construction of beautiful estuaries and bays and high-quality development of the marine economy (Liu et al., 2017; Ministry of Ecology and Environment of the People’s Republic of China, 2022). In addition, land-based pollution will lead to nutrient enrichment in estuaries, which will cause great damage to the marine environment (Wu et al., 2019). Based on the fact that the PRE is a typical estuarine ecosystem with high N and low P, and has been in a P-limited state for a long term, it is appropriate to adopt the pollution control mode of land and sea integration (Redfield et al., 1963). Establishing estuarine nutrient criteria can estimate the background concentration of nutrient, and formulate scientific water quality standard for assessment, which can strictly control the discharge of land-based pollutant, reduce the total amount of nutrients entering to the sea, and reduce the contribution of land-based pollution to the PRE from the source (Hu et al., 2020). At the same time, it is necessary to strengthen the comprehensive management of

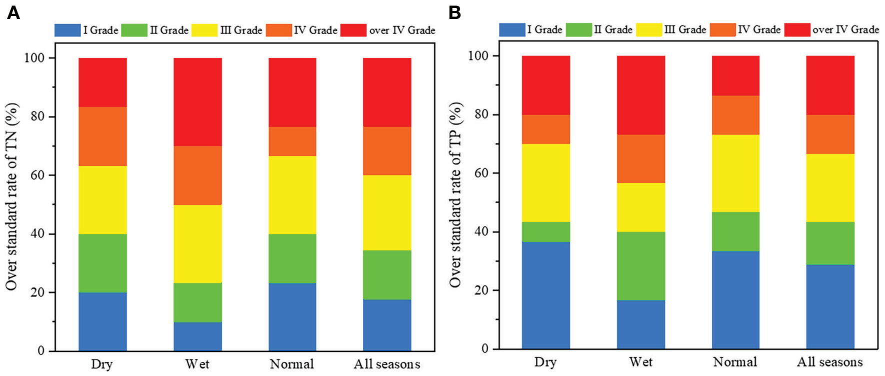

FIGURE 9
Recommended standard assessment TN **(A)** and TP **(B)**.

rivers flowing into the PRE, and reinforce the real-time monitoring of water quality in the PRE, accurately identify the sudden changes in water quality in the PRE, and provide a scientific basis for the improvement of water quality in the PRE (Hong, 2022).

Therefore, this study considers that long-term monitoring should be carried out in the coastal waters of various estuaries, such as the Pearl River Estuary, Yangtze River Estuary, and Yellow River Estuary, to accumulate rich reference data to provide data support for the formulation of nutrient criteria in China (Su et al., 2016). Simultaneously, corresponding nutrient control standards and management strategies should be put forward based on the actual situation of different estuaries (Huo et al., 2018; Zhou et al., 2020). Under the background of "Strengthening the environmental protection and restoration of estuaries and bays, implementing the comprehensive management of one bay and one policy in estuaries and adjacent bays, and building a number of beautiful bays by implementing classified policies", establish zoning management and control of nutrients in the basin-estuary-coastal water, achieve the goal of fine water quality management and control, make full use of the self-purification capacity of water bodies, and improve the eutrophication of estuarine coastal water (Department of Ecology and Environment of Guangdong Province, 2022b).

Conclusions

In this study, the PRE was divided in three segments based on salinity, corresponding to freshwater, mixed, and seawater areas, with increasing salinity from north to south. Frequency statistical analysis was applied to establish the recommended nutrient criteria. The recommended DIN, TN, DIP, and TN criteria in the freshwater area were 0.573, 0.902, 0.009, and 0.039 $mg{\cdot}L^{-1}$, respectively; the recommended DIN, TN, DIP, and TN criteria in the mixed area were 0.312, 0.885, 0.009, and 0.028 $mg{\cdot}L^{-1}$, respectively; and the recommended DIN, TN, DIP, and TP criteria in the seawater area were 0.134, 0.222, 0.004, and 0.020 $mg{\cdot}L^{-1}$, respectively. The DIN concentration in the PRE without pollution was approximately between 0.120 $mg{\cdot}L^{-1}$ and 0.312 $mg{\cdot}L^{-1}$, and the DIP concentration without pollution was approximately 0.004–0.015 $mg{\cdot}L^{-1}$. It is necessary to establish the relationship between DIN and TN, DIP and TP to better understand their impact on eutrophication. Under the integrated management mode of land and ocean, the emissions of land-based pollutants should be strictly controlled to reduce the impact on estuaries and coastal waters. At present, the Sea Water Quality Standard may have "over-protection" problems in the assessment of DIN of the PRE, while there may be "under-protection" problems in the DIP assessment. As a result, the establishment and application of nutrient water quality criteria unified the water quality monitoring indicator for land and ocean, is the basis for setting water quality standards, an effective measure to prevent eutrophication of water bodies, and a scientific basis for comprehensive monitoring, assessment, and management of nutrients in estuaries, which is of great significance for building beautiful estuaries and bays in China and worldwide. In the future, long-term monitoring should be carried out in the coastal waters of various estuaries, such as the Pearl River Estuary, Yangtze River Estuary, and Yellow River Estuary, to accumulate rich reference data to provide data support for the formulation of nutrient criteria in China.

Data availability statement

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

Author contributions

Conceptualization: PZ; Methodology: PZ and JBZ; Software: SO; Validation: PZ, JXZ, and LZ; Formal analysis: PZ, JXZ, and LZ; Writing-original draft preparation: PZ and SO; Writing-review and editing: PZ and SO; Visualization: PZ and JBZ; Supervision: PZ and JBZ; Project management: PZ and JXZ; Funding acquisition: PZ and JBZ. All listed authors made substantial, direct, and intellectual contributions to the work and are approved for publication.

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Conflict of interest

Publisher's note

Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fmars.2022.1004235/full#supplementary-material

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