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Long-Term Variations of Biogenic Elements and Nutritional Status in Daya Bay, Northern South China Sea

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Abstract: This study explored the variations in the characteristics of the trophic structure of Daya Bay island waters over the last four decades based on the survey findings and research data on biogenic elements (dissolved inorganic nitrogen (DIN), NO_2^-, NO_3^-, NH_4^+, PO_4^{3-}, and SiO_3^{2-}) in Daya Bay during 1985–2021. At this time, the DIN concentration increased from 21.14 $\mu g \cdot L^{-1}$ to 558.42 $\mu g \cdot L^{-1}$ (26.41-fold increase), whereas the SiO_3^{2-} concentration increased by only 3.6-fold. The PO_4^{3-} concentrations attained a peak in 2004 and experienced a steady decline over the rest of the survey period. The fractions of NH_4^+, NO_3^-, and NO_2^- in DIN changed from 0.45, 0.40, and 0.15 in 1986 to 0.26, 0.74, and 0.003 in 2021, respectively. Overall, the mean values of NH_4^+, NO_3^-, and NO_2^- accounted for 45.2%, 42.5%, and 12.3%, respectively. The N/P(DIN/PO_4^{3-}) ratio in Daya Bay increased from 28.08 in the 1980s to 51.63 in the 2010s. Meanwhile, the nutrient limitation conditions showed a gradual shift from N-limited to P-limited conditions. According to the nutrient quality index (NQI) analysis, the trophic state level of Daya Bay waters fell into the oligotrophic category 30 years ago (1985–2002, NQI < 2), whereas it increased from the mesotrophic level in 2005 (NQI = 2.03) to the eutrophic level in 2019 (NQI = 3.33) over the last 20 years. The results based on the eutrophication index (EI) of Daya Bay waters were generally consistent with those based on the NQI, displaying that the trophic level of Daya Bay waters indicated an increasing trend from 2005 to 2019. Moreover, the assessment data in 2021 indicated a decrease in the NQI to 0.90, thereby attaining the oligotrophic level again. This may be related to the decrease in aquacultural area in the bay over the last two years. The correlation analysis among the DIN, PO_4^{3-}, and nutrient levels of Daya Bay waters indicated that the input of nitrogen and phosphorus was the primary reason for the higher nutrient levels in the water bodies; among them, municipal sewage discharge, aquaculture, and atmospheric deposition from industry are the main factors for the over importation. This indicates that the changes in the biogenic element concentrations led to variations in the trophic structure and level of Daya Bay and may be attributed to population growth and the development of the seaside industry and agriculture in the region.

Keywords: biogenic elements; trophic level; long-term changes; eutrophication

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1. Introduction

The biogenic elements in marine water and the essential nutrients for the growth of marine organisms [1] form the basis of the material cycle in marine ecosystems [2]. Biogenic elements are the first link in the marine ecosystem and play a fundamental role in the energy flow, material cycle, and information transfer of the marine ecosystem, which is important for maintaining the balance and stability of the marine ecosystem [3]. Human development activities in the coastal zone of Daya Bay and the rapid economic development in the Pearl River Delta region have significantly changed the nutritional composition of Daya Bay island reef waters [4]. This has led to a large impact on the biological structure of Daya Bay waters [5]. Studying the variation in the biogenic elements in Daya Bay waters is significant for conducting fundamental research on subtropical fishery resources in the South China Sea and promoting the exploration and analysis of habitat characteristics near the coastal waters in the South China Sea. The bay is a link between land and sea and has a superior development environment, so it occupies a very prominent position in all socioeconomic development and has high socioeconomic and ecological service value, and it is also a sensitive area for environmental change and a fragile zone of ecosystems [6]. Daya Bay has undergone significant changes because of its unique geographical location since the 1980s, when the reform and opening up policy was formulated in China [7], which profoundly affected the local fishery habitat [8].

The nutrients in seawater play a key role in phytoplankton growth, and the concentrations of DIN, PO_4^{3-}, and SiO_3^{2-}, as well as the trophic structure, are closely related to phytoplankton growth [9]. The elemental ratio of nitrogen, silicon, and phosphorus (N/Si/P) is a major indicator for characterizing the trophic structure of water bodies and plays a crucial role in phytoplankton growth and reproduction [10]. High or low variations in the nutrient content in the seawater environment can lead to an abnormal N/P/Si ratio and disruptions in the trophic structure of seawater [11], thereby limiting the growth and reproduction of phytoplankton and, ultimately, leading to an adverse effect on the phytoplankton population structure [12]. According to Redfield [13], a Si/N/P ratio of 16:16:1 and Si/N > 1 indicate that the phytoplankton growth in seawater is N limited. Si/N < 1 indicates that it is Si limited. In addition, N/P > 16 indicates that the phytoplankton growth in seawater is P limited and N limited and vice versa (N/P $\leq$ 16) [14].

In this study, historical survey data on Daya Bay from 1985 to 2021 were collected and compiled to evaluate and analyze the changes in the nutrient characteristics of Daya Bay, South China Sea. Generally, the findings of this study lay the foundation for the study of biogenic elements in the Daya Bay waters and provide a research reference for the conservation of fisheries habitats and the exploitation of fisheries resources.

2. Materials and Methods

2.1. Study Areas

Daya Bay (114°33′–114°53′ E, 22°26′–22°32′ N), located on the east side of the Pearl River estuary and adjacent to Dapeng Bay in the west and Honghai Bay in the east, is an important part of the Pearl River Delta economic zone [15]. The Daya Bay sea area is in the south of the Tropic of Cancer, i.e., in the subtropical region (Figure 1b). The Guangdong provincial government approved the establishment of the Daya Bay Aquatic Resources Provincial Nature Reserve in 1983, and the core area of the Daya Bay Aquatic Resources Provincial Nature Reserve currently has a total area of 126 km^2 [16]. At present, Daya Bay is currently the only typical bay in China with three nuclear power plants operating simultaneously [17], along with a 150,000-ton crude oil terminal and the Daya Bay Petrochemical Industrial Zone, led by the China Shipping Shell petrochemical project (Figure 1a). The developed socioeconomic and petrochemical industries in the area [18], frequent human activities [19], large amounts of domestic and aquacultural wastewater [20], and the afflux of industrial wastewater [21] have led to problems such as a reduced self-purification capacity and the degradation of germplasm resources in Daya Bay. Furthermore, the biogenic elements of the island reef waters in the bay, as a region that enriches the biodiversity of biological resources, are subject to considerable changes.

2.2. Data Sources and Collection Criteria

Surface and bottom water samplings were collected and onsite formatting was performed at all survey stations using organic glass water harvesters. For the chlorophyll a (*Chl-a*) analysis, seawater samples were filtered with a glass–fiber GF/F filter (0.7 µm, Whatman), then transferred to acid-washed polyethylene bottles, subpackaged, and stored at −20 °C in a refrigerator. Further, the samples were then sent to the laboratory of the South China Sea Fisheries Research Institute (Guangzhou) of the Chinese Academy of Fisheries Sciences for further analysis. In addition, filtered seawater (250 mL) from each station was stored in acid-washed polyethylene bottles and placed in a refrigerator at −20 °C for the analysis of the major nutritive salts in the seawater.

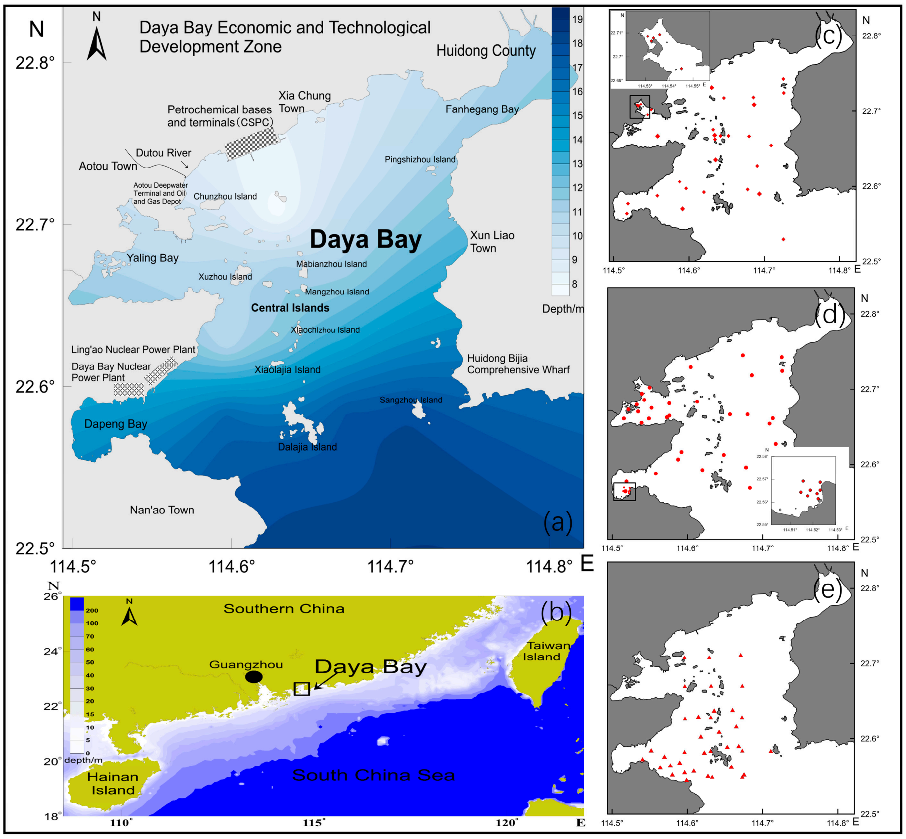

Figure 1. Geographical location and sample collection sites in Daya Bay: (**a**) cities and industries adjacent to Daya Bay; (**b**) schematic representation of Daya Bay; (**c**) sample collection stations during 1985–2000; (**d**) sample collection stations during 2000–2010; (**e**) sample collection stations during 2010–2021.

The data used in this study were obtained from three sources: (1) sea-going field surveys from 37 survey stations (Figure 1) within the waters of Daya Bay conducted in the spring (April), summer (August), autumn (October), and winter (December) of 2015; spring (April) and autumn (November) of 2019; summer (August) of 2020; and spring (April) of 2021 by the South China Sea Fisheries Research Institute of the Chinese Academy of Fisheries Sciences; (2) data collected from published papers and monographs; and (3) data from historical archival materials from the South Chi na Sea Fisheries Research Institute of the Chinese Academy of Fisheries Science and unpublished information on Daya Bay.

2.3. Sample Analysis and Research Methods

The analysis of nutrients, including dissolved inorganic nitrogen (DIN), nitrite (NO_2^-), nitrate (NO_3^-), ammonia nitrogen (NH_4^+), phosphate (PO_4^{3-}), and silicate (SiO_3^{2-}), was conducted according to the relevant requirements in the *National Marine Monitoring Code* (GB 17378.3-2007). NO_3^-, NH_4^+, NO_2^-, and PO_4^{3-} were analyzed by the cadmium column reduction method, indophenol blue method, diazo–azo method, and phosphomolybdenum blue method, respectively. The standard edition and recovery experiments were performed for each batch of the water samples to ensure the accuracy of the analytical results. The parallel measurements were conducted on 10–15% of each batch of seawater samples to test the accuracy of the analytical results. In addition, the relative standard deviation was set to less than 10% for parallel samples and less than 5% for quality-control samples. The chemical oxygen demand (COD) in the samples was measured following the *National Marine Monitoring Code Part 4-Seawater Analysis* (GB 17378.4-2007). Dissolved oxygen in the samples was measured by the Winkler titration method, with an accuracy of 0.07 mg·L^{-1}. Moreover, COD in the samples was measured by the potassium permanganate oxidation method, with an accuracy of 0.15 mg·L^{-1}. *Chl-a* was determined by the fluorometric method according to the relevant requirements in GB/T12763.6-2007 using a Turner Designs 10-AU fluorometer.

According to the Three-Year Action Plan for Marine Environmental Protection in Huizhou Daya Bay Economic and Technological Development Zone (www.dayawan.gov.cn, accessed on 1 January 2017 to 31 December 2019), as the central archipelago is in the core area of Daya Bay Aquatic Resources Provincial Nature Reserve, the water quality protection management standard in this region should be following the first-class standard for seawater quality (seawater quality standard GB 3097-1997).

The nutrient quality index (NQI) [22] is a composite index and one of the most commonly used eutrophication evaluation methods in China. The NQI is calculated from COD, DIN, dissolved inorganic phosphorus (DIP), and *Chl-a* using the following equation:

$$\mathrm{NQI} = \frac{COD^c}{COD^s} + \frac{DIN^c}{DIN^s} + \frac{DIP^c}{DIP^s} + \frac{Chl\_a^c}{Chl\_a^s}$$

where COD^c, DIN^c, DIP^c, and Chl_a^c are the monitoring concentrations of COD_{Mn}, DIN, DIP, and *Chl-a*, respectively, and COD^s, DIN^s, DIN^s, and Chl_a^s are the evaluation standard values of 3.0 mg·L^{-1}, 0.3 mg·L^{-1}, 0.03 mg·L^{-1}, and 0.05 mg·L^{-1}, respectively. According to the relationship between the NQI and nutrient levels in the sea, an NQI $\leq$ 2 indicates oligotrophic seawater, 2 < NQI < 3 indicates mesotrophic seawater, and NQI $\geq$ 3 indicates eutrophic seawater.

The eutrophication index (EI) [23], proposed by Japanese scholar Morihiro Aizaki (1981) and later introduced by Jingzhong Zou et al. (1983) in China, reassigned the constant a. Presently, EI has been widely used in eutrophication research and is calculated as follows:

$$\mathrm{EI} = \frac{COD_{Mn} \times DIN \times DIP \times 10^6}{4500}$$

where COD_{Mn}, DIN, and DIP are the measured values of COD, DIN, and DIP (mg·L^{-1}), respectively. The eutrophication degree of seawater was evaluated as follows: E < 1.0, poor eutrophication; 1.0 $\leq$ E < 2.0, mild eutrophication; 2.0 $\leq$ E < 5.0, moderate eutrophication; 5.0 $\leq$ E < 15.0, heavy eutrophication; and E $\geq$ 15.0, severe eutrophication.

If there were data from multiple surveys in the same year, the mean value of each index in this study was blurred; if there were data from only one survey per year, these data were used. When homogenizing the data, considering that historical data acquisition will inevitably be affected by tides, seasons, currents, seasonal influences, and other factors, to make the research results more clear and powerful, during the data processing, we tried to take a weighted average to reflect the phased Daya Bay environmental nutrient data values.

3. Results

3.1. Long-Term Variation in Nutrient Concentrations in Daya Bay

The DIN concentrations in Daya Bay started to increase during 1989–1990 and stabilized at 550 μg·L^{-1} during 1991–2003. Since 2004, sharp fluctuations were observed in the DIN content, which continued to increase until 2013. During 2013–2017, the DIN concentration decreased linearly. Over the past 15 years, the lowest DIN concentration was observed in 2017. These concentrations returned to their normal levels in the early 20th century. From 2017 onwards, there was a linear increase in the DIN content in Daya Bay, reaching a record high in 2020, followed by a sharp decline to the 2005 level. The variations in the DIN concentrations in Daya Bay from the 1980s to the present can be divided into four stages: sub-volatile period (1985–1990), stable low-value period (1990–2005), volatile period (2005–2015), and highly volatile period (2015–2021), with an overall increasing trend (Figure 2a).

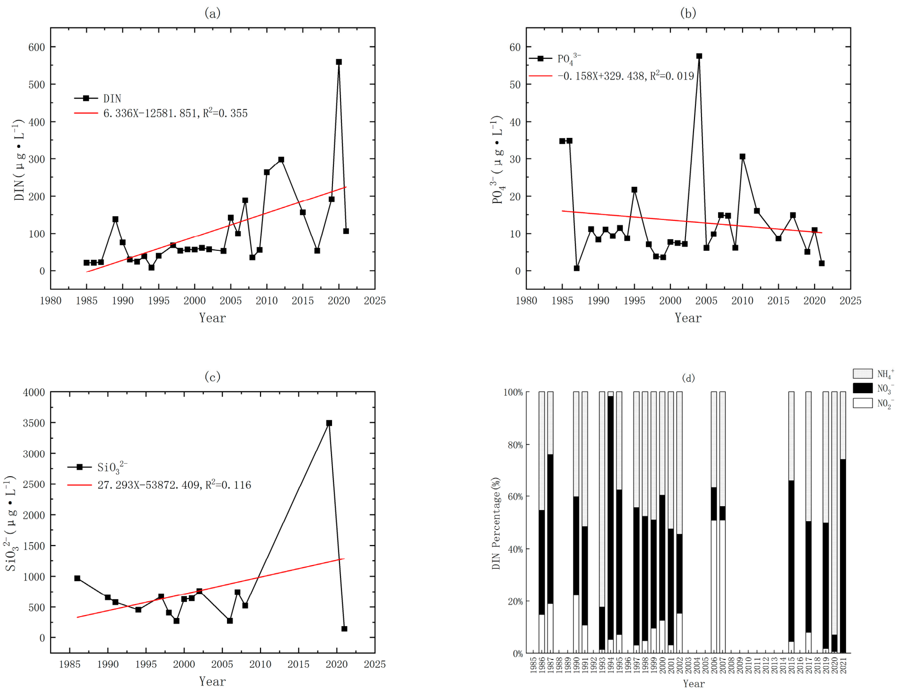

Figure 2. Long-term variations in the nutrient concentrations in Daya Bay (μg·L^{-1}): (**a**) variation in the DIN concentrations in Daya Bay during 1985−2021; (**b**) variation in PO_4^{3-} concentrations in Daya Bay during 1985−2021; (**c**) variation in SiO_3^{2-} concentrations in Daya Bay during 1985−2021; (**d**) variation in DIN fractions in Daya Bay during 1985−2021.

The PO_4^{3-} concentrations in Daya Bay were at relatively high levels in 1985–1986 and then declined to the lowest levels in history in 1987. The PO_4^{3-} concentrations remained relatively stable during 1989–2003, except for a slight increase in 1995. The PO_4^{3-} concentrations reached a record high in 2004 but returned to lower levels in the subsequent year. In addition, they returned to the initial levels during 2005–2010 after a slight increase. Moreover, the PO_4^{3-} concentrations in Daya Bay showed a continuous fluctuating downward trend from 2010 to 2021 and reached the second-lowest value in 2021. The variational trend in the PO_4^{3-} concentrations in Daya Bay can also be divided into four stages: stable fluctuation period (1985–2003), brief peak period (2003–2005), sub-stable fluctuation period (2005–2010), and stable decline period (2010–2021). Overall, the PO_4^{3-} concentrations showed cyclical fluctuations, with a decreasing trend over the last decade. However, PO_4^{3-} was relatively stable compared with other nutrients (Figure 2b).

The SiO_3^{2-} concentrations in Daya Bay declined steadily from 1985 to 1995. In addition, the concentrations underwent range fluctuations from 1995 to 2008 but remained relatively stable. The SiO_3^{2-} concentrations increased sharply from 2008 to 2019 and reached a record high level in 2019, followed by a sharp decrease and a record low level in 2021. The variational trend in the SiO_3^{2-} concentrations in Daya Bay can be divided into three stages: stable declining period (1985–1995), stable fluctuating period (1995–2008), and high fluctuating period (2008–2021). Despite the decrease in the concentrations and fluctuations in several instances, SiO_3^{2-} generally showed an increasing trend (Figure 2c).

The DIN composition in Daya Bay changed significantly over the last 40 years. In general, NH_4^+ dominated DIN (45.2%), followed by NO_3^- (42.5%), and NO_2^- (12.3%). NO_2^-, NO_3^-, and NH_4^+ remained relatively stable from 1985 to 1990. However, there was a continuous decrease in the proportion of NO_2^-, while the proportion of NH_4^+ continuously increased, and the proportion of NO_3^- remained generally stable during 1990–1993. In addition, the proportion of NO_3^- increased rapidly and reached a record high in 1994 (93.1%). Since then, the proportion of NO_3^- has been decreasing, whereas the proportions of NO_2^- and NH_4^+ steadily increased (1994–2002). The highest proportion of NO_2^- (51%) was observed in 2006 and 2007, whereas the proportion of NH_4^+ remained largely stable. The proportion of NH_4^+ increased steadily during 2015–2019, whereas the proportions of NO_3^- and NO_2^- fluctuated slightly but were generally stable. The composition of DIN substantially changed in 2020–2021, with the highest proportion of NH_4^+ observed in 2020. However, NO_3^- replaced NH_4^+ as the most dominant DIN component in 2021 (Figure 2d). Generally, the nutrients (DIN and SiO_3^{2-}) in Daya Bay showed a clear perturbation with an increasing trend over the last 40 years.

3.2. Long-Term Variations in the Trophic Structure of Daya Bay

The N/P, Si/P, and Si/N ratios in Daya Bay ranged from 1.35 to 122.11 (mean value = 35.45), 30.79 to 769.62 (mean value = 121.74), and 0.66 to 42.60 (mean value = 9.86), respectively. The N/P in Daya Bay was found to be at low levels (1.35–3.03) in 1985–1986 but increased significantly to a higher level of 80.54 in 1987. The N/P ratio in Daya Bay started to decline and returned to the same level as that of 10 years ago, i.e., in 1987–1995. The N/P ratio started to increase every year during 1985–1999 and reached 48.50. Despite slight fluctuations during 2000–2015, the N/P ratio in Daya Bay has been relatively stable at high levels compared to the 1990s. In addition, the N/P began to increase linearly during 2017–2019 and reached a record high (55.15) in 2021. The N/P ratio in Daya Bay showed an overall increasing trend over the last 40 years and was at high levels in recent years.

The Si/P ratio in Daya Bay showed an overall increasing trend from 1985 to the beginning of this century, reaching a record high value in 2019, but decreased to a relatively normal level in 2021 (Figure 3b). The Si/N was slightly higher before 2000 compared to the post-21st century. Although the highest value of Si/N was observed in 2019, it was relatively stable with a slightly declining trend over the last 40 years (Figure 3c). According to the fitting lines in Figure 3, the nutrient limitation of the trophic structure of Daya Bay gradually changed from N-limited conditions at the end of the last century to P-limited conditions, and this trend has become more prominent over the last few years. The N/P values were significantly higher than those in a previous study, indicating that the DIN levels in Daya Bay waters have increased significantly in the last few years, which is consistent with the findings of a previous study [24]. The Chl-a and N/P variations per decade in Daya Bay and the variations in Chl-a during 1985–2021 are shown in Figure 4.

3.3. Long-Term Variations in the Trophic Levels and Eutrophication Degree in Daya Bay

During 1985–2021, the DIN and PO_4^{3-} levels followed the first-class standard of Chinese seawater quality standards for the vast majority of this period (Table 1). However, the DIN levels in 2010–2012 (mean = 280.34 $\mu g \cdot L^{-1}$) were higher than the first-class seawater standard. In particular, the DIN level in 2020 (558.42 $\mu g \cdot L^{-1}$) exceeded the fourth-class seawater standard. Furthermore, the PO_4^{3-} levels in 1985–1986, 2004, and 2010–2012 did not meet the criteria for first-class water quality standards.

Although the long-term trend in overall seawater quality in Daya Bay was following the first-class standard, and the quality also followed the second-class and above standards for certain periods. The NQI was calculated to monitor the nutrient quality status in Daya Bay. The NQI for 1985–2003 was less than two, implying the oligotrophic level of Daya Bay waters during this period. The NQI of Daya Bay started to rise after 2003 and reached 2.58 in 2005 when Daya Bay waters were at a mesotrophic level. The trophic level of the Daya Bay waters entered eutrophication for approximately ten years from 2010 to 2021 when the NQI dropped to the lowest level recorded and then returned to the oligotrophic level (Figure 5a).

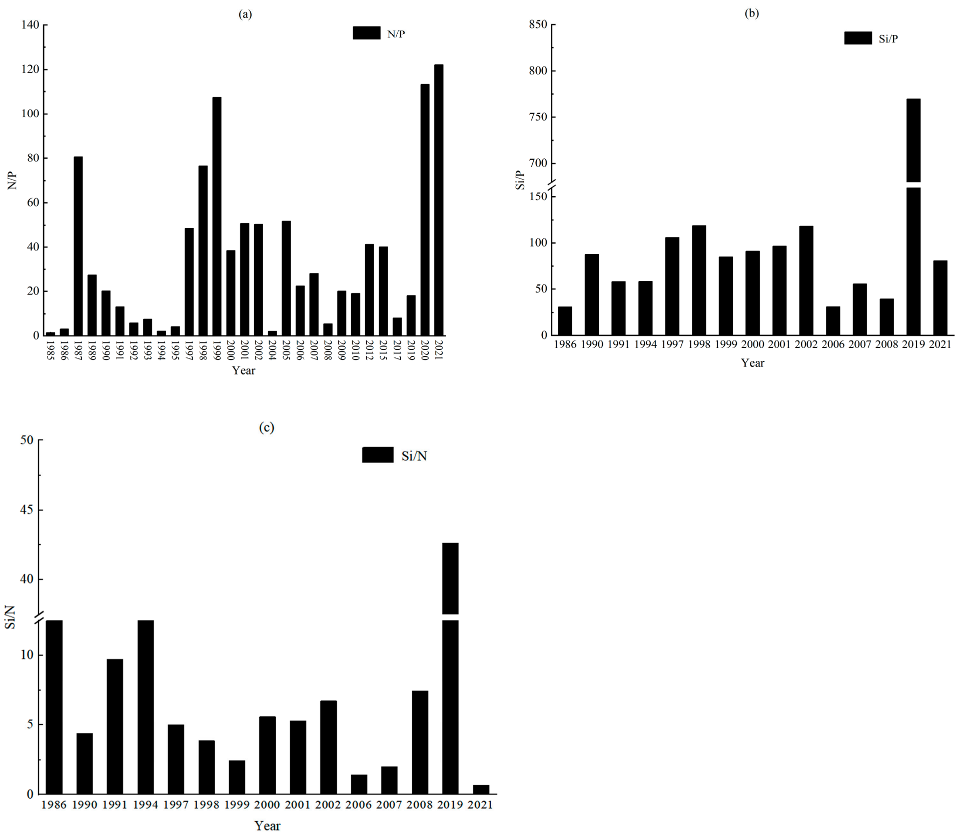

Figure 3. Long-term variations in the trophic structure of Daya Bay during 1985–2021: (**a**) N/P variation in Daya Bay during 1985–2021; (**b**) Si/P variation in Daya Bay during 1985–2021; (**c**) Si/N variation in Daya Bay during 1985–2021.

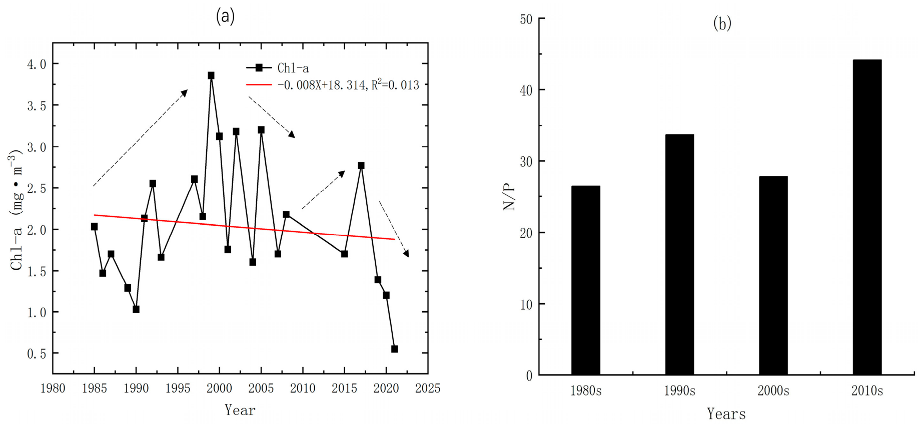

Figure 4. Chl-a and N/P variations per decade in Daya Bay: (**a**) variation in Chl-a in Daya Bay during 1985−2021; (**b**) variations in N/P ratio in Daya Bay from the 1980s to 2010s.

Table 1. DO, DIN, PO_4^{3-}, and COD limits according to the Chinese seawater quality standards.

Seawater Quality Standards	DO ($mg \cdot L^{-1}$)	DIN ($\mu g \cdot L^{-1}$)	PO_4^{3-} ($\mu g \cdot L^{-1}$)	COD ($mg \cdot L^{-1}$)
First class	>6	≤200	≤15	≤2
Second class	5–6	200–300	15–30	2–3
Third class	4–5	300–400	15–30	2–3
Fourth class	3–4	400–500	30–45	4–5

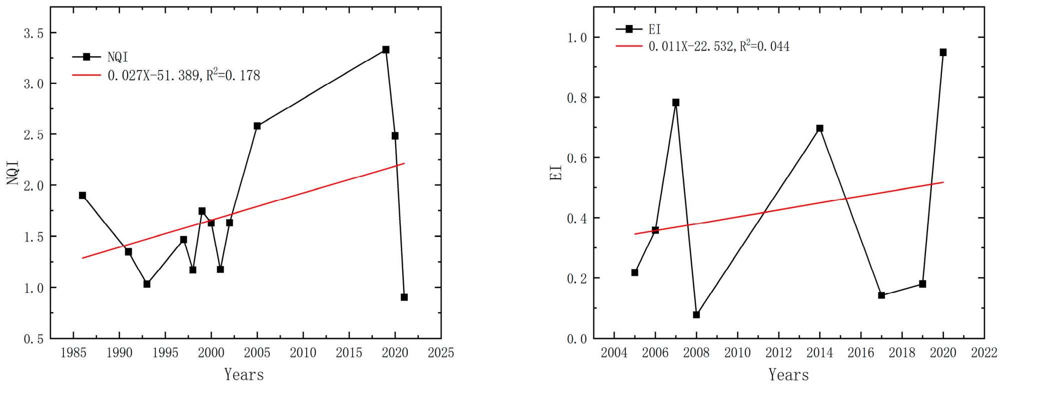

Figure 5. Long-term variations in the trophic levels and eutrophication degree in Daya Bay: (**a**) variation in the NQI in Daya Bay during 1985–2021; (**b**) variation in the EI in Daya Bay during 2005–2021.

The NQI reached a mesotrophic level in 2005 and reached 3.33 (eutrophic level) in 2019. The EI was again used to study the changes in Daya Bay from 2005 to 2020 and to further investigate the trophic levels in Daya Bay. For all 15 years, the EI was less than one, indicating that bay did not reach the eutrophication level. However, a linear fit revealed an increasing trend in the EI from 2005 to 2020, which approached a slight eutrophication threshold of one in some years (Figure 5b). This trend was consistent with that of the NQI during the same period.

4. Discussion

4.1. Analysis of Long-Term Nutrient Levels in Daya Bay

The biogenic elements in Daya Bay, which are directly linked to human activities, showed an overall increasing trend during 1985–2021 [25]. Huizhou and Shenzhen, which are adjacent to Daya Bay, have been undergoing expansion in size and population since the Chinese economic reform and opening up in 1978 [26], resulting in more effluent discharge into the river. According to the *Huizhou Statistical Yearbook 2021*, the population of Huizhou has increased approximately three-fold from 1985 (2,088,800) to 2021 (6,606,000). The growth of the city and the population was found to be closely related to the increase in the DIN in Daya Bay. The urban growth has resulted in impermeable, hardened surfaces along the streets of Daya Bay. In addition, nitrogen compounds from domestic waste and domestic sewage enter Daya Bay along with surface runoff caused by rainfall.

In addition to the rapid industrial development in Huizhou, the impact of industries along the coast of Daya Bay is also increasingly evident [27]. Industrial emissions showed a positive correlation with the changes in the DIN concentrations (Table 2). In 2019–2020, industrial exhaust gases increased from 40.091 billion m^3 to 1255.49 billion m^3, and the DIN increased from 99 $\mu g \cdot L^{-1}$ to 558.42 $\mu g \cdot L^{-1}$. Industrial fuels react with oxygen and produce nitrogen oxides during combustion. NOx, nitrogen oxides in the exhaust gas, may account for the changes in the DIN concentrations in Daya Bay waters. In addition, the soot generated during industrial combustion was also associated with elevated DIN levels. Industrial soot emissions increased from 190 tons in 2006 to 24,500 tons in 2014 (i.e., an increase of 12.89 times). During this period, the DIN concentrations also increased by 2.37-fold. However, industrial soot emissions decreased from 44,500 tons in 2014 to 14,100 tons in 2017. In addition, the DIN concentrations in Daya Bay also started to decrease, with the DIN concentration in 2017 being only 22.22% of that in 2014. Therefore, it can be stated that the DIN in Daya Bay is influenced by industrial soot emissions, which are associated with the atmospheric deposition of particulate matter from industrial soot. Approximately 20–38% of elemental nitrogen in coastal waters is derived from atmospheric deposition [28]. Nitrogen deposition in the Chinese offshore region was primarily contributed by fossil fuels and industrial emissions, with fossil fuels being the predominant source and accounting for 86% of nitrate nitrogen [29]. In addition, large fluctuations in the DIN concentrations were observed in Daya Bay during 2008–2021, with the concentrations attaining a peak in 2019 and decreasing rapidly after 2020. This trend was similar to those of both domestic and industrial wastewater discharge and industrial emissions, which were observed to be at record highs in 2019 and declined after 2020, consistent with the dynamics of Daya Bay over the last few decades. These factors could explain the overall increase in DIN in Daya Bay for nearly 40 years.

Table 2. The numbers in the table are Pearson correlation coefficients. Due to the lack of data on SiO_3^{2-} in some years, the correlation value of SiO_3^{2-} is abnormal, so it will not be discussed for the time being.

Factor	DIN	PO_4^{3-}	SiO_3^{2-}
Population	0.393	−0.024	0.118
Gross domestic product	0.471	−0.033	0.762
Chemical fertilizer usage	0.219	−0.014	0.001
Mean amount of precipitation	−0.031	0.140	−0.119
Mean sunshine hours	−0.055	−0.051	-
Domestic sewage discharge	−0.093	−0.079	-
Industrial wastewater discharge	−0.079	−0.098	−0.189
Industrial exhaust emission	0.578	−0.076	-
Industrial soot emission	−0.057	−0.109	-

The construction of the Daya Bay Nuclear Power Plant also had an impact on the biogenic elements in this region. The DIN concentrations in the sea area of Daya Bay increased from 1985 to 2005. The DIN concentration in the seawater near the plant exceeded the first-class water quality standard in 1997, and the DIN concentration in the surface layer of the seawater exceeded the first-class water quality standard in 1999. SiO_3^{2-} was found to be at a relatively high level in 1985 and demonstrated an intermittent decrease in the next 15 years, which may be attributed to the increase in the Si content in the seawater after the plant's operation [30].

Extreme changes in Pacific waters due to the fact of global warming have led to a range of impacts on the climate in the northern South China Sea [31]. The coastal areas of the South China Sea have experienced several climate anomalies within these 40 years. According to the precipitation characteristics of China during 1961–2018, a significant increase in extreme precipitation events across the country was recorded, with the increase in extreme precipitation and extreme precipitation days primarily concentrated in the southeastern coastal and western regions, and the national average of continuous extreme precipitation events demonstrating a nonsignificant increasing trend [32]. Both extreme precipitation and extreme drought can lead to certain impacts on the nutrients in Daya Bay and change the composition of the trophic structure. The precipitation affected the PO_4^{3-} concentrations, with more precipitation leading to lower PO_4^{3-} concentrations because of the dilution of PO_4^{3-} in the water by rainfall. The SiO_3^{2-} content in Daya Bay increased due to the increase in precipitation, which was attributed to SiO_3^{2-} from the land contributed by stormwater runoff. In addition, the length of daylight has a significant effect on SiO_3^{2-} as diatoms in seawater photosynthesize under sunlight [33]. The life activity of diatoms is influenced by photosynthesis. Therefore, daylight also determines the SiO_3^{2-} concentration in Daya Bay waters. However, it does not have any significant impact on DIN and PO_4^{3-} concentrations in the Bay.

The economic development of Guangdong Province and the increased demand for seafood have led to an incremental increase in the scale and volume of mariculture in Daya Bay [34]. The high concentrations of the biogenic elements N and P in sediments near typical aquacultural areas in Daya Bay can make the cultured waters a potential source of endogenous pollution. The raft culture area in Dapengao was only 540 m^2 in 1994, while it expanded to more than 1300 m^2 in 1998. According to a previous study [35], the NH_4^+ and PO_4^{3-} in Aotou waters reached 13.5 $t.a^{-1}$ and 0.34 $t.a^{-1}$, respectively, indicating the large contribution of the aquatic sediments in Daya Bay to the N and P contents in seawater. These nutrients exhibit a band distribution with the direction of the tide, indicating different degrees of eutrophication. The increase in the scale of cage aquaculture in Daya Bay can inevitably lead to more severe environmental issues related to water [36]. The feed used in aquaculture is rich in protein, which increases the N content in the water body. In addition, the warm drainage from the plant in Daya Bay, the oil spills from the petrochemical zone, the yearly increase in fertilizer usage in the upper reaches of the rivers along Daya Bay, domestic wastewater, and the oil spills that accompany the mooring of a large number of ships all affect the concentrations of nutrients in the waters [37]. As a complex and dynamic system, the nutrient concentration in the water body will also be affected by external and internal factors, similar to the decomposition of organic matter, and the release of substances in sediment, bottom-up and top-down mechanisms in the water body, and other biochemical factors will also affect its concentration changes.

4.2. Long-Term Nutrient Limitation of Long-Term Water Bodies in Daya Bay

The standing stock of phytoplankton, the most important producer in marine ecosystems [38], can generally be characterized by *Chl-a* concentration. The N/P ratio in marine waters is an important factor for determining whether the waters are N limited or P limited and whether the trophic structure can limit phytoplankton growth [39]. Redfield proposed a threshold value of 16:1 for the N/P ratio, and a ratio close to this threshold is the optimal ratio. A study indicated that the water bodies with an N/P ratio close to this threshold were most suitable for phytoplankton growth, water bodies with a ratio above this threshold were P limited, and water bodies below this threshold were N limited [40]. According to the 10-year average N/P ratio (Figure 4b), Daya Bay waters were primarily P limited from the 1980s to 2000s, and from the 1980s to 2000s they were close to N limited. Daya Bay was closest to Redfield's N/P threshold in the 2000s and rose above this threshold in the 2010s, which coincided with the increase followed by a decreasing trend in *Chl-a* from 1985 to 2010 (Figure 4a). The results of this study further validate Redfield's threshold theory that phytoplankton growth is the least restricted and the fastest in water bodies with an N/P ratio close to 16:1. The economic reform and opening up policy led to an increase in the population size as well as industrial and agricultural growth. Consequently, these nutrients from the Daya Bay coastline were carried into the Bay with increasing human activities, resulting in higher N/P ratios and phytoplankton populations during the 1980s and 1990s. However, China's national policy on scientific development, formulated in 2003 [41], focused on the harmonious development of humans and nature and considered the association of economic construction and population growth with resource utilization and ecological environmental protection. As a result, the N/P ratio in Daya Bay started to decline, which, in turn, affected the change in the nutrient limitations in waters from N limited to P limited.

The conditions in Daya Bay have changed from N limited during the 1980s–2000s to P limited in the 2010s. In addition, the N/P ratio increased to 44.09 during this period, which was 1.59 times as high as that in the 2000s and was consistent with the variations in the *Chl-a* concentrations. However, despite the significantly higher N/P ratio in the 2010s relative to the N/P ratio in the 1990s, the concentration of *Chl-a* in the 2010s was lower than that in the 1990s. Therefore, the phytoplankton population did not increase significantly with the rapid increase in the N/P ratio in the 2010s. This phenomenon was caused by multiple factors. The growth rate of algae is only significantly limited by the N/P ratio at low N and P concentrations, and the optimal N/P ratio required for the growth of algae varies depending on the species of algae. For example, the competitive inhibition parameter (α) of *Microcystis aeruginosa* against *Oscillatoria* was greater than that (β) of *Oscillatoria* against *Microcystis aeruginosa* at medium and high N/P ratios, whereas the opposite phenomenon was observed at low N/P ratios [42]. Japanese scholar Seung HoBaek proposed the luxury consumption theory that excessive cell storage is not related to the growth rate [43]. Daya Bay may have been in a state of luxury consumption in the 2010s, with certain phytoplankton such as *Ceratium furca* having low nutrient uptake half-saturation constants (Ks) and specific nutrient uptake characteristics. The change in the phytoplankton community structure may be one of the primary reasons for the limitation in the trophic structure. According to a previous long-term study on algae in Daya Bay [44], the phytoplankton community structure in Daya Bay was determined to be influenced by the environmental factors during the major years of the 2010s, including low temperature and high salinity in the water, high DIN concentration, and high N/P ratio. Under high DIN and high N/P conditions, some pollutant-tolerant species' (e.g., *Pseudo nitzschia* and *Skeletonema costatum*, among the major species in Daya Bay) interspecific competitive advantage was significantly higher than that of other species, such as *Chaetoceros* and *Thalassionema nitzschioides*. In addition, these species belong to eurythermal and euryhaline organisms and can adapt to the variations in temperature and salinity in water caused by the invasion of external seawater. Therefore, their abundance and abundance percentage demonstrated a significantly increasing trend. In addition, they can adapt to variations in temperature and salinity. Therefore, they have a stronger competitive advantage relative to other dinoflagellate species, resulting in a significantly increasing trend in their abundance between 1999 and 2017 and becoming the absolute dominant species among the dinoflagellate species in Daya Bay. As a result, the growth of these species dominated in the 2010s. The conditions in Daya Bay have shifted from N limited to the current P limited over the last decade. Briefly, the limitation of the trophic structure affects the phytoplankton growth, whereas the change in the phytoplankton community structure also affects the change in the trophic structure.

4.3. Analysis of Eutrophication in Daya Bay over the Past 40 Years

Daya Bay experienced the Zhimazhou Blast and the Mabianzhou Blast in 1993 and 1994, respectively. Based on the variations in the NQI values before and after the initiation of the South China Sea Petrochemical Project and the Zhimazhou Blast, it was observed that the waters of Daya Bay were at a clean level before the start of the Project and the Blast. The water quality was affected during the construction process, and slight water pollution was observed in the construction area. Although blasting led to significant changes in the water quality, it returned to pre-blasting levels within a short period [45]. The South China Sea Fisheries Research Institute of the Chinese Academy of Fisheries Sciences [46] monitored the blasting site at Mabianzhou and evaluated the blasting effects. The institute observed that the blasting had a high transient impact on the waters around Mabianzhou. The effects of blasting were large for some time immediately after blasting but returned to normal levels during the post-blasting period, indicating that the blasting had no significant impact on the quality of the surrounding waters. Therefore, the blasting project did not have a significant impact on the variations in the water quality parameters in Daya Bay.

The NQI of Daya Bay waters was at the oligotrophic level from 1985 to 2005. The human activities during this period led to fluctuations in the NQI of Daya Bay waters. From 2005 onwards, the NQI of Daya Bay waters exceeded 25 (Figure 4), indicating that the trophic level of Daya Bay shifted from oligotrophic to mesotrophic, and an increasing trend was observed during 2005–2019. During 2005–2020, Daya Bay waters experienced a rich and significantly increasing trophic level for three years (2007, 2014, and 2020), indicating a remarkable increase in the nutrient input into the waters of the bay and an increase in the COD levels, which may be attributed to the industrial and agricultural changes in Daya Bay. During this period, the increasing scale of the petrochemical industry, nuclear power, shipping industry, and cage aquaculture in Daya Bay [47], along with the increase in different types of effluent discharges, led to an increase in the nutrient input load and elevated nitrogen and phosphorus concentrations in Daya Bay. If the nutrient load into the seawater cannot be precisely and effectively controlled, the EI in Daya Bay can be every 6–7 years, and the waters of Daya Bay will probably reach the eutrophic level around midcentury, based on the prediction from the trend observed during 2005–2020. In this study, the relevant data were collected for specific correlation analyses to investigate the causes of eutrophication in Daya Bay (Figure 6). The EI and DIN in Daya Bay demonstrated a strong correlation (correlation coefficient = 0.8), indicating that DIN is the primary factor affecting eutrophication in Daya Bay. In addition, PO_4^{3-} is the secondary factor showing a general correlation with EI, indicating that PO_4^{3-} can exert a certain influence on eutrophication in Daya Bay. However, *Chl-a* and COD in Daya Bay waters have little effect on the EI. After the operation of the nuclear power plant, the COD in Daya Bay waters showed a decreasing trend every year, especially in the western part.

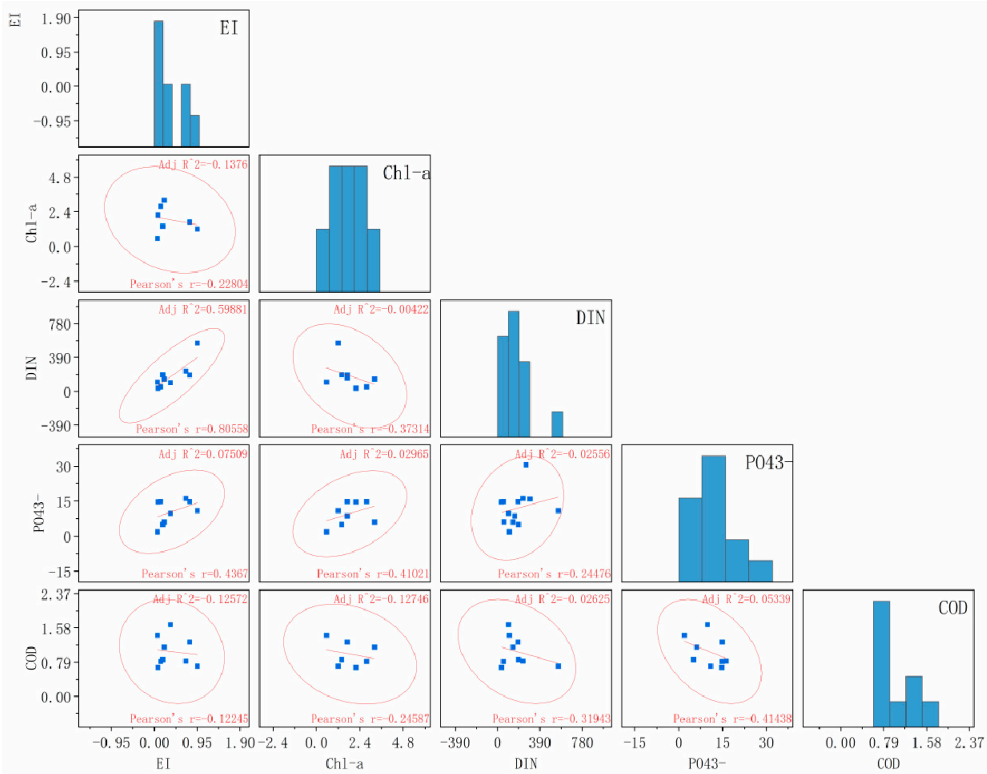

Figure 6. Pearson correlation among the eutrophication factors of the Daya Bay waters.

The N pollution sources leading to the eutrophication of Daya Bay waters over the past 40 years are land-based sources of pollution, sea-based sources of pollution, and atmospheric sources of pollution. According to a database (www.dayawan.gov.cn, Three-Year Action Plan for Marine Environmental Protection in Daya Bay Economic Development Zone), there are 63 land-based sources of discharge into the sea along the coast of the Daya Bay area, including 20 land-based sources of discharge into the sea close to the coast of the petrochemical area, as well as 10 discharge ports and 33 discharge ports along the coastal towns. The sources of marine pollution consist of two sources: mariculture [48] and pollution. The discharge of liquid waste during aquaculture leads to higher than normal levels of pollutants in adjacent waters, which has a significant impact on the ecological functions of water bodies. Daya Bay is a major aquaculture area in Guangdong Province, where aquaculture, especially cage aquaculture, has been highly developed. The impact of cage aquaculture in Daya Bay on the water environment is mainly due to the artificial feeding process in which a large number of nutrients directly enter the water bodies in both dissolved and nondissolved forms. The dissolved nutrients lead to an increase in the content of certain environmental factors in the water body, and the nondissolved nutrients are deposited on the seabed of the cage aquaculture area. These deposits become a potential source of pollution [49]. Cage aquaculture mainly leads to elevated levels of NH_4^+ and TP in the marine environment. Huizhou Daya Bay banned cultured fishing rafts in the jurisdictional waters in 2021. The reduction in the farming area of Daya Bay rapidly decreased the NQI in 2021 (this was the lowest level observed in the past 40 years). Shipping pollution sources primarily include cabin water, ballast water, tank washings, and domestic sewage from ships. Out of these sources, the first three are relatively large in volume. The major pollutants in sewage include petroleum compounds in ballast water and phosphorus in detergents used in the washing process, which are the primary sources of environmental pollution in the port area. Atmospheric pollution is mainly manifested as atmospheric deposition. A large amount of particulate matter from industrial combustion such as waste gas from industries along Daya Bay and the islands enters the atmosphere and settles on the water with precipitation and other means. In addition, NH_4^+ from chemical fertilizers used in nearby agricultural areas is also a considerable source of atmospheric deposition [50]. It is worth affirming that hydrological changes, such as precipitation, evaporation, and exchange with water bodies outside the bay, will also affect water quality, and because no hydrological change data were collected, they were not added as a quantitative assessment.

5. Conclusions

In this study, the dynamic changes in the DIN, PO_4^{3-}, and SiO_3^{2-} levels in Daya Bay waters from 1985 to 2021 and their interrelationships were investigated. The DIN and SiO_3^{2-} levels indicated an increasing trend over the last four decades, whereas PO_4^{3-} showed a stable trend. In addition, the major reasons that may lead to the changes in biogenic elements were also analyzed. The changes in DIN were strongly related to the growth of the population, as well as industrial and agricultural development in Daya Bay. The PO_4^{3-} and SiO_3^{2-} levels, especially SiO_3^{2-}, were less affected by anthropogenic influences but were significantly affected by climate change. In addition, more attention should be paid to the influence of balanced and fixed monitoring stations on long-term water quality changes.

The changes in *Chl-a* in Daya Bay over the past 40 years were generally consistent with the changes in the N/P ratio. The primary productivity of Daya Bay may be influenced by luxury consumption. In addition, the overall N/P ratio in Daya Bay indicated an increasing trend. The limitation conditions of the trophic structure of Daya Bay waters gradually changed from N limited at the end of the last century to P limited, and this trend could become more prominent in the future.

The trophic state of Daya Bay from 1985 to 2021 was analyzed using the NQI method. The NQI of Daya Bay waters in this period showed an overall increasing trend every year. The trophic state increased from the oligotrophic level before 2005 to the mesotrophic level but returned to the oligotrophic level in 2019, indicating an overall increasing trend. In addition, an EI study of the water bodies was conducted after 2005. The NQI method was used to determine the trophic level of the Daya Bay waters. The EI revealed that although the waters did not reach the eutrophication level, they were on the verge of eutrophication. If the nutrient input into Daya Bay is not effectively controlled, Daya Bay could reach eutrophic levels around the middle of this century, leading to further deterioration of the water quality. Therefore, immediate attention should be paid to the eutrophication of Daya Bay.

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