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Long-term nutrient variation trends and their potential impact on phytoplankton in the southern Yellow Sea, China

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Abstract

The concentration and composition of nutrients, such as N, P, and Si, respond to biogeochemical processes and in turn, impact the phytoplanktons' community structure and primary production. In this study, historical data was systematically analyzed to identify long-term variations in nutrient trends, red tide frequency, phytoplankton community abundance, and dominant species succession in the southern Yellow Sea (SYS). Results showed that N/P concentration ratios dramatically increased as a function of increasing dissolved inorganic nitrogen concentrations, and Si/N concentration ratios were generally larger than 1, indicating that N limitation morphed to P limitation and potentially to Si limitation, which impacted the phytoplankton community. Furthermore, inter-annual trends over the past 50 years show that phytoplankton community abundance has been higher in spring and summer, relative to autumn and winter. Moreover, with respect to red tide frequency, diatom abundance gradually decreased, while that of dinoflagellates gradually increased. Dominant species succession showed that the phytoplankton community exhibited an evident tendency to transform from diatoms to dinoflagellates. These research results clearly depict the presence of an important correlation between the phytoplankton community and nutrient structure in the SYS.

Key words: southern Yellow Sea, nutrients structure, succession of phytoplankton community, diatom, dinoflagellate

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1 Introduction

Nutrients, including nitrogen (N), phosphorus (P), and silicon (Si), are essential biogenic elements, as they are required for marine phytoplankton reproduction and growth and support their primary productivity (Grasshoff et al., 1999). Phytoplankton are generally regarded as the foundation to maintain and affect the material cycle and energy flow in aquatic systems. Variations in nutrient concentrations and compositions are closely coupled with biogeochemical processes, and heavily impact the phytoplankton community structure and primary production (Jin et al., 2013; Wang et al., 2003). Due to rapid industrial and agricultural development, large amount of N and P has been transported offshore through terrestrial inputs (aquaculture wastewater; industrial, agricultural, and domestic sewage) which transport high concentrations of nutrients to estuary and influence the environment of coastal waters, and atmospheric transport which can impact on remote oceanic regions due to the long-range transport of dust (Chung et al., 1998; Guo et al., 2020; Liu et al., 2000; Seok et al., 2021; Shen et al., 2006; Shi et al., 2013; Wang et al., 2019; Zhang and Liu, 1994; Zheng and Zhai, 2021). In response, the nutrient composition and stoichiometry in coastal areas have been significantly altered, resulting in increased dissolved inorganic nitrogen (DIN) and asymmetric ratios between N, P, and Si (Wei et al., 2015; Wang et al., 2021a). A serious consequence of this imbalance is eutrophication, which facilitates catastrophic marine ecological phenomena, such as red tides, green tides, ocean hypoxia, and acidification (Lund, 1967; Wang et al., 2018; Chen et al., 2021; Xiao et al., 2021).

Because C:N:P=106:16:1 is the main element stoichiometry in phytoplankton, it serves as the foundation for aquatic biogeochemistry studies, and is widely used to evaluate various marine environments (Redfield et al., 1963; Fisher et al., 1992; Sterner et al., 2008). Extensive research has shown that over the past 40 years, nutrient limitation conditions in the Bohai Sea have changed from N control to P and Si control (Xin et al., 2019; Wang et al., 2019). Due to excessive DIN entering via terrestrial runoff and anthropogenic activities, coastal area of the southern Yellow Sea (SYS) showed strong signs of potential P limitation (Guo et al., 2020). Furthermore, in response to global climate change and anthropogenic activities, offshore phytoplankton biomass increased in the Baltic Sea, Black Sea, and Northeast Atlantic (Suikkanen et al., 2007; Mikaelyan, 1997; Edwards et al., 2001). In addition, analysis of phytoplankton community succession showed that abundance of diatoms are decreasing, while that of

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dinoflagellates are increasing, indicating that long-term environmental variation trends favor dinoflagellate growth (Wasmund et al., 1998; Lin et al., 2005; Yang, 2016; Ma, 2019; Yang et al., 2018; Ning et al., 2010; Zhou et al., 2008; Zhang et al., 2020).

The SYS is a semi-closed marginal sea that is located in the Northwest Pacific Ocean between Chinese mainland and Korean Peninsula. It is bounded to the north by the northern Yellow Sea (NYS) and the south by the East China Sea, and is impacted by the Yellow Sea Coastal Current, Korean Coastal Current, and Yellow Sea Warm Current (Hwang et al., 2014). Since 1978, China's economy has undergone expansive changes, which came hand-in-hand with environmental problems (Wang et al., 2014, 2018). Since the 1980s, anthropogenic activity has resulted in consistently increasing concentrations of DIN and phosphate (PO_4^{3-}) being transported to the ocean; while silicate (H_4SiO_4) concentrations remained relatively stable, due to transport being prevented by dams (Liang and Xian, 2018; Wang et al., 2018; Humborg et al., 2008; Guo et al., 2020a). Coastal cities around the SYS are highly developed and densely populated, and thus, have an important impact on marine environment. As such, inter-annual nutrient variation studies in the SYS and their potential impact on marine ecosystems have gained significant attention from the scientific community (Wang et al., 2003; Lin et al., 2005; Fu et al., 2009; Shi et al., 2013; Li et al., 2015; Liu et al., 2015; Wei et al., 2015).

Unlike previous phytoplankton nutrient limitation studies conducted in the SYS, this work mainly focused on local waters, while large scale interannual variations were only minutely considered. In this study, the correlation between nutrients and the phytoplankton community in the Haizhou Bay area was analyzed; and long-term nutrient variations and ecosystem coupling changes in the SYS, including red tides, phytoplankton abundance, and community composition, are discussed (Fig. 1). The results from this analysis shed light on the relationship between nutrient structure and the phytoplankton community, thereby enhancing our understanding of how anthropogenic activities impact the foundation of marine primary productivity.

2 Materials and methods

2.1 *Environmental parameter data*

Nutrient data, including DIN, PO_4^{3-}, and H_4SiO_4 concentrations for the SYS and Changjiang River, were mainly obtained from the following sources: (1) Summer values from 1990 to 2019 were acquired from the marine environmental quality monitoring work of China's State Oceanic Administration (SOA) and Ministry of Ecology and Environment (MEE). To improve the comparability of the nutrient data, DIN and PO_4^{3-} long-term data were classified as nearshore and offshore using the National Marine Functional Zoning (Ministry of Ecology and Environment of the People's Republic of China, 2020). The mean values of nutrients in surface layer and bottom layer in the nearshore (defined by National Marine Functional Zoning) and offshore (adjacent seas of China excluding the nearshore) were calculated to determine the annual variations in the SYS. (2) SOA and MEE H_4SiO_4 data were available only for 2014–2019, and therefore inadequate. Additional H_4SiO_4 data for 1958–1959 and 1997–1999 were obtained from published work by Wei (2015), Wang (2003), and Wang et al. (2003). Because H_4SiO_4 was not sufficiently represented (data of H_4SiO_4 in the offshore is less than 20 per year), the H_4SiO_4 data was averaged and discussed for the SYS as a whole. (3) Nutrient data for the Haizhou Bay for 2003–2017 were obtained from the work of SOA's red tide monitoring. Surface water nutrient mean values were calculated and used to represent monthly concentrations. (4) Changjiang River DIN and PO_4^{3-} data was collected from Wang et al. (2018), while that of H_4SiO_4 was obtained from Ma (2015).

2.2 *Red tide data*

The SYS red-tide event data from 1990–2019 were obtained from the work of red-tide monitoring of SOA and Ministry of Natural Resources. For each red tide event, the occurrence time, location, distribution area, and dominant species were recorded. Cumulative frequency, cumulative area, and the occurrence of diatom and dinoflagellate red tides were calculated respectively.

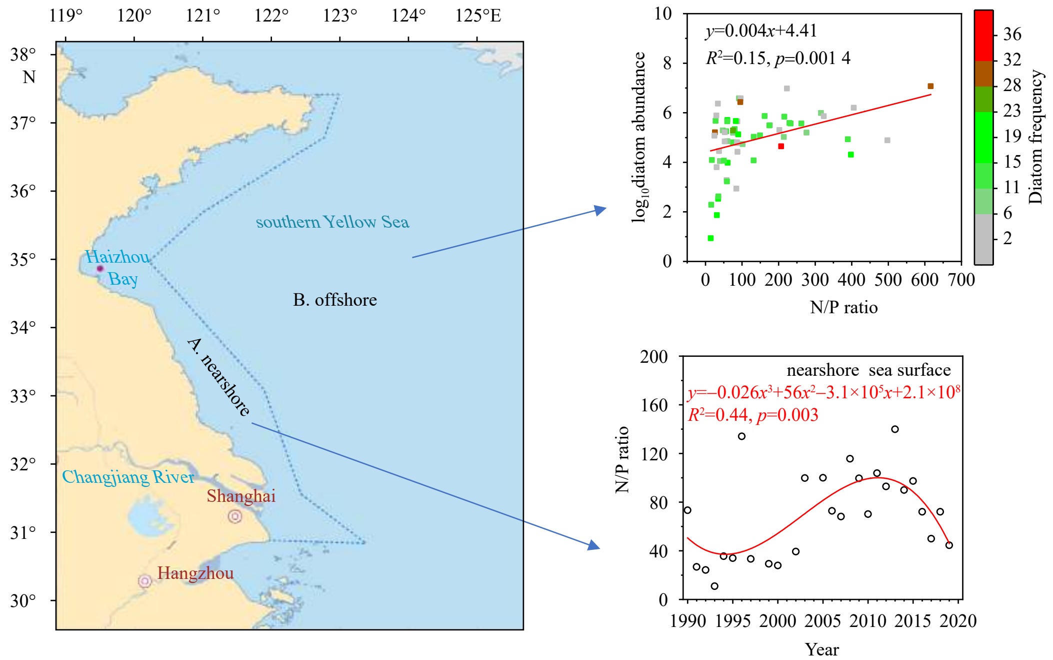

Fig. 1. Research area in the southern Yellow Sea. N/P is the molar ratio between N concentration and P concentration.

2.3 *Phytoplankton community data*

The SYS phytoplankton (net sample) abundance and species data were obtained for the period 1959–2015 from Wang (2001), Fu et al. (2012), Yang (2016), Xu (2007) , Jiang et al. (2020) and Luan et al. (2020) (Table 1). The data were analyzed and discussed in quarterly and annual increments. In addition, the Haizhou Bay phytoplankton data (surface water sample) from 2003 to 2017 were obtained from the work of SOA's red tide monitoring, while frequency and abundance of the phytoplankton community's dinoflagellates and diatoms were characterized monthly and annually.

3 Results

3.1 *Nutrient concentration and structure variations*

3.1.1 *Nutrient concentration*

Examination of long-term DIN variability in the SYS (i.e., 1990 to 2019) showed that the surface and bottom waters both exhibited a similar DIN concentrations trend characterized by an initial increase that was followed by a decrease (Fig. 2). Furthermore, due to intense anthropogenic activity and terrestrial input, the nearshore DIN concentrations were substantially higher than those measured offshore, and the DIN concentrations in surface layer were significantly higher than those in the bottom layer. Specifically, nearshore surface layer concentrations increased significantly from 1990 to 2012, with the highest values generally observed from 2005–2012 (excepting 2010). During the peak, the average DIN concentration was >19.0 μmol/L. Beginning in 2013, the DIN concentration gradually decreased with the lowest value (7.73 μmol/L) recorded in 2019. DIN concentrations in the nearshore bottom layer showed a similar trend to that of the surface layer, although the concentrations were substantially lower. With respect to the offshore environment, DIN surface water trends also showed an increase followed by a decrease, while the bottom water DIN concentrations remained relatively stable throughout the DIN study period. Moreover, because the offshore environment is not directly affected by pollutants delivered from rivers, DIN concentrations in this locale were much lower than those in the nearshore and showed only slight interannual fluctuations.

Table 1. Phytoplankton community data sources (net samples)

Type	Period	Reference
Phytoplankton abundance	1959–1960	Fu et al. (2012)
	1984–1985	Fu et al. (2012)
	1985–1986	Wang (2001)
	1998–2000	Wang (2001)
	2001–2002	Fu et al. (2012)
	2011	Jiang et al. (2020)
	2005	Xu (2007)
	2006–2007	Fu et al. (2012)
	2008	Fu et al. (2012)
Phytoplankton abundance, Diatom abundance, Dinoflagellate abundance	1985–1986	Luan et al. (2020)
	1998–2000	Luan et al. (2020)
	2005–2008	Luan et al. (2020)
	2014–2015	Luan et al. (2020)
	1958–1959	Yang (2016)
	2000–2003	Yang (2016)
	2011–2013	Yang (2016)

Long-term PO_4^{3-} variation trends in the SYS were different from those of DIN (Fig. 3). The nearshore surface and bottom water PO_4^{3-} concentrations showed slightly different variation characteristics. Mainly different variations were observed from 1990 to 1999 and then concentration variation tended to have a similar variation from 2000 to 2019. Specifically, concentrations in the nearshore surface layer gradually increased from 1990 to 1999 and then were maintained between 0.25–0.56 μmol/L (i.e., fluc-

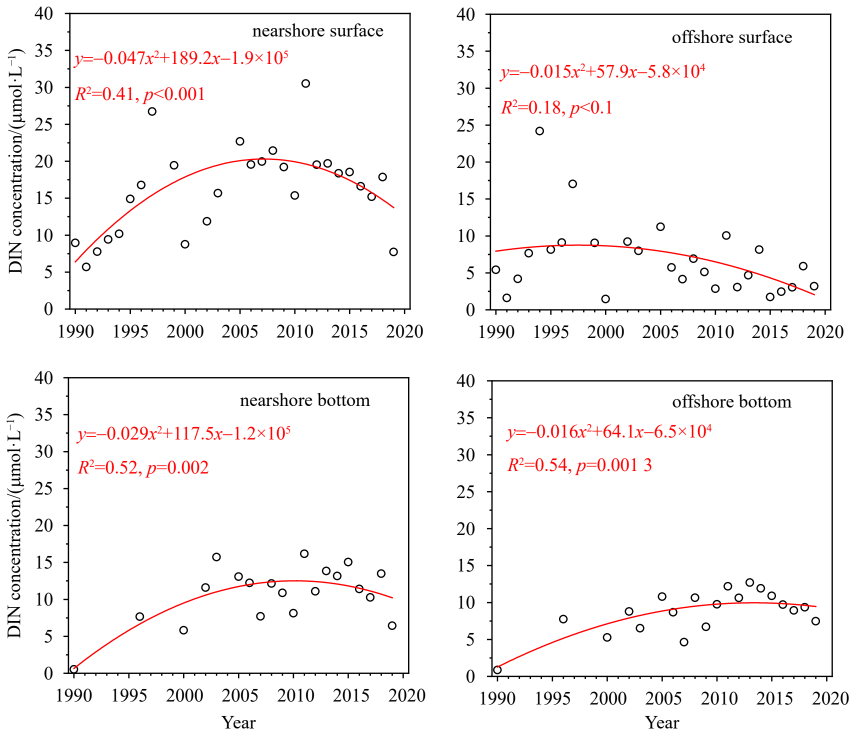

Fig. 2. Temporal variations of DIN concentrations in the southern Yellow Sea (1990–2019).

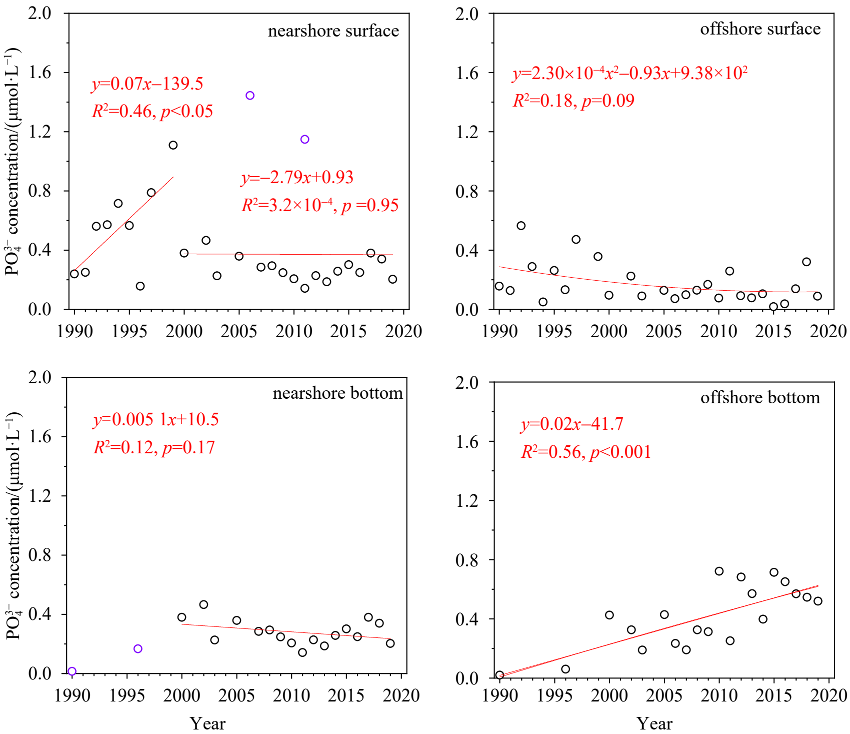

Fig. 3. Temporal variations of PO_4^{3-} concentrations in the southern Yellow Sea (1990–2019).

tuating ~0.40 μmol/L) from 2000 to 2019, excepting 2006 and 2011. In contrast, with the exception of 1990, PO_4^{3-} concentrations in the nearshore bottom fluctuated between 0.14–0.47 μmol/L throughout the PO_4^{3-} study period. Offshore PO_4^{3-} trends differed significantly from those observed nearshore. With respect to offshore surface water, PO_4^{3-} concentrations gradually decreased from 1990 to 2000, then remained stable at ~0.10 μmol/L from 2000 to 2019. In contrast, offshore bottom water PO_4^{3-} concentration has increased since 1990.

Inter-annual variations of H_4SiO_4 in the SYS showed that H_4SiO_4 concentrations initially decreased, then subsequently increased from 1958 to 2019 (Fig. 4). Surface water concentrations decreased from 10.19 μmol/L in 1958 to 4.97 μmol/L in 1990s, and then steadily increased with values generally larger than 10.00 μmol/L in 2010s (excepting 2019). The bottom layer H_4SiO_4 concentration trend parallels that of the surface layer, with values of 13.42 μmol/L in 1958, 8.31 μmol/L in 1990s, and >10.00 μmol/L in 2010s (excepting 2019).

3.1.2 *N/P and Si/N concentration ratios*

N/P concentration ratios results from the surface and bottom layers of the SYS were greater than the Redfield ratio (16), demonstrating the presence of an evident P limitation (Fig. 5). In addition, N/P ratios were greater nearshore than offshore, and surface layer values were higher than those in bottom layer. With respect to long term trends, N/P ratios in the nearshore and offshore surface water, as well as the nearshore bottom water, depicted a similar variation pattern, i.e., an increase followed by a decrease. Higher values were primarily observed from 2003–2015, during which time, the average ratios were 96 (nearshore surface), 76 (nearshore bottom), and 71 (offshore surface). The N/P ratios gradually decreased from 2016 to 2019. The lowest value, i.e., 44, was recorded in 2019, and approximated values were observed in 1990s. This change in N/P ratios is consistent with the DIN fluctuations in the SYS. In contrast, N/P ratios in the offshore bottom layer were relatively stable, and generally fluctuated in the range of 14–48 with an average value of 28 (excepting

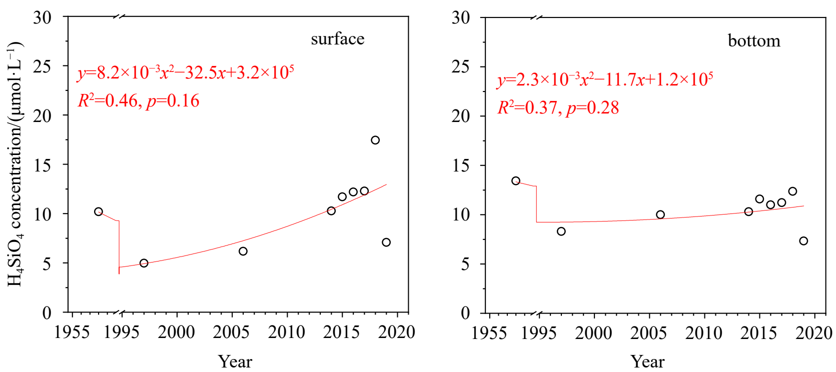

Fig. 4. Temporal variations of H_4SiO_4 concentrations in the southern Yellow Sea (1958–2019) (data of 1958, 1997, and 2006 were obtained from the report of Wei et al. (2015)).

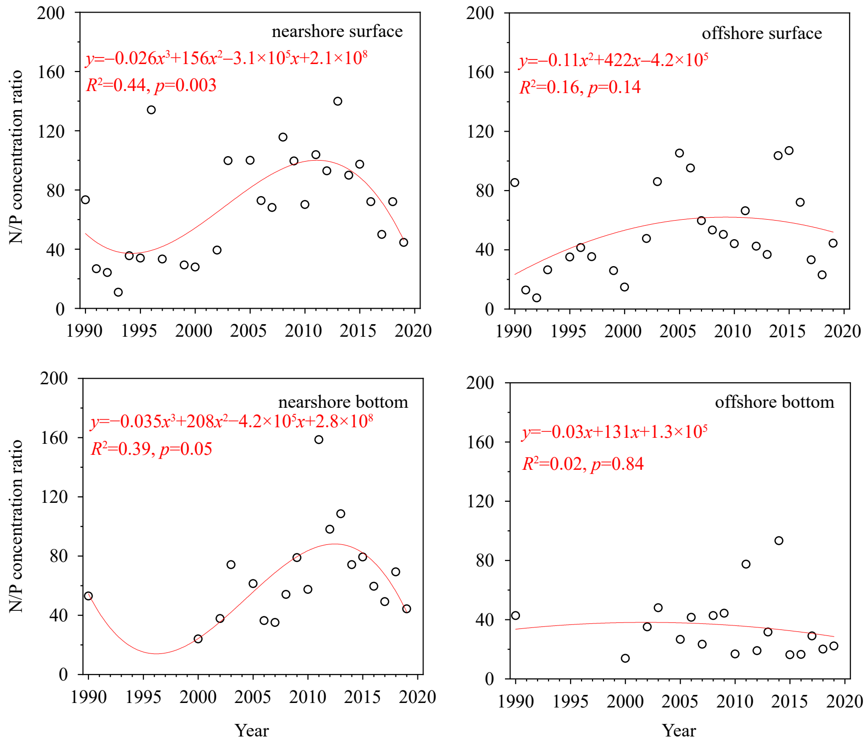

Fig. 5. Temporal variations of N/P concentration ratio in the southern Yellow Sea (1990–2019).

2011 and 2014).

From 1997 to 2019, Si/N ratios were increasing, while in general, the values were >1 (Fig. 6). These results indicate that Si was not the main limiting factor in the SYS. However, there may be Si limitation in some areas in response to massive dam construction and the subsequent freshwater inflow decrease.

3.2 *Phytoplankton community variation*

3.2.1 *Phytoplankton abundance*

Interannual changes of phytoplankton abundance in the SYS are shown in Fig. 7. Since 1958, phytoplankton abundance has fluctuated significantly, in some cases by up to 8 orders of magnitude. The highest phytoplankton abundance value, i.e., >600× 10^6 cells/m^3, was observed in the summer of 2006–2007. With respect to seasonal changes, the values in spring and summer were generally higher than those in autumn and winter. Maximum seasonal values for spring, summer, fall, and winter were approximately 8×10^6 cells/m^3, 600×10^6 cells/m^3, 2×10^6 cells/m^3, and 6×10^6 cells/m^3, respectively. From 1958 to 2015, long-term phytoplankton abundance variations depicted an increasing trend in spring and summer, but this trend was not evident in autumn and winter.

3.2.2 *Phytoplankton community composition*

Figure 8 and Table 2 show the percentage of total phytoplankton abundance represented by diatoms and dinoflagellates. The summer diatom percentage contribution in the SYS dropped from 70.8%–85.1% in 1959 to 18.8%–27.4% in 2012. Spring (May) diatom abundances also showed a similar decrease (Table 2), dropping from 88.9% in 1986, to 69.5% in 1998, and 50.9% in 2005. In contrast, the spring (May) dinoflagellate abundance in the SYS increased from 11.1% in 1986, to 30.5% in 1998, and 49.1% in 2005 (Wang, 2001). The gradually decreasing diatom

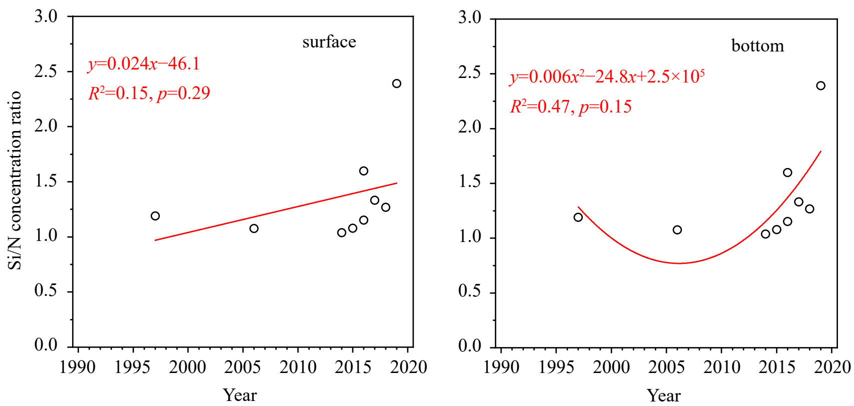

Fig. 6. Temporal variations of Si/N concentration ratio in the southern Yellow Sea (1997–2019).

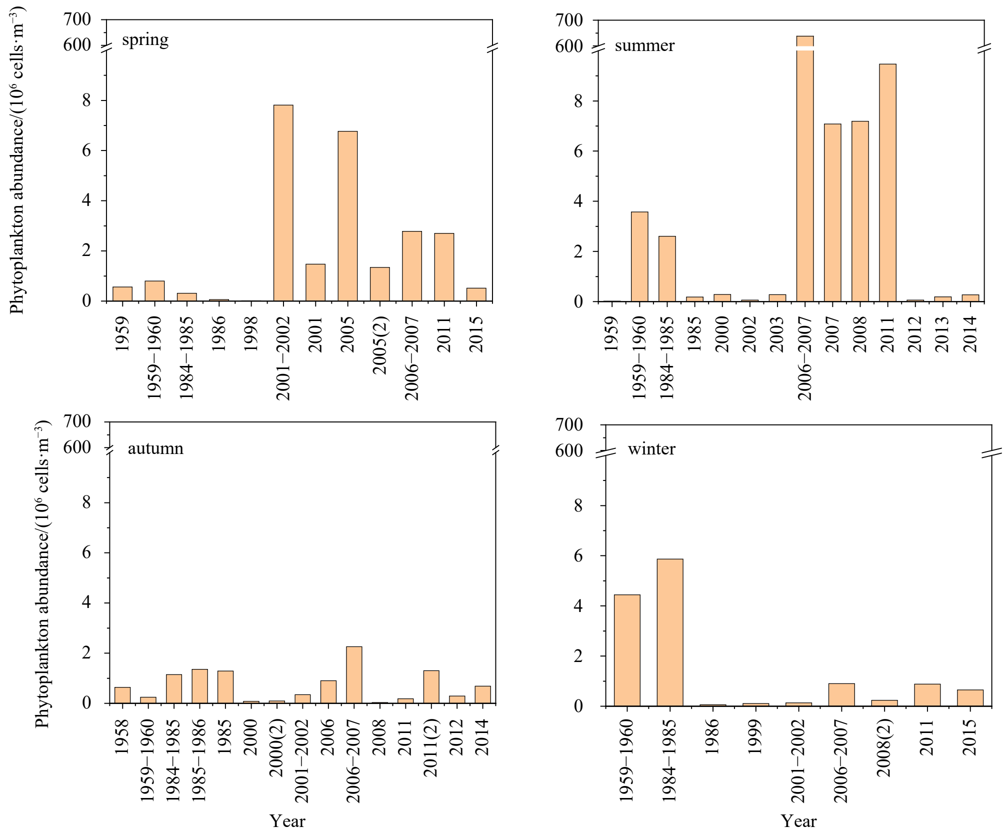

Fig. 7. Seasonal variations of phytoplankton abundance in the southern Yellow Sea (1958–2015). Data of 1959–1960, 1984–1985, 1985–1986, 2001–2002, 2006–2007, 2008 were redrawn from Fu et al. (2012); Data of 1985, 1986, 1998, 1999, 2000, 2005(2), 2006, 2007, 2008(2), 2014, 2015 were redrawn from Luan et al. (2020); Data of 1958, 1959, 2000(2), 2001, 2002, 2003, 2011(2), 2012, 2013 were redrawn from Yang (2016); Data of 2005 were redrawn from Xu (2007); Data of 2011 were redrawn from Jiang et al. (2020). Detailed data of this figure are shown in Table S1.

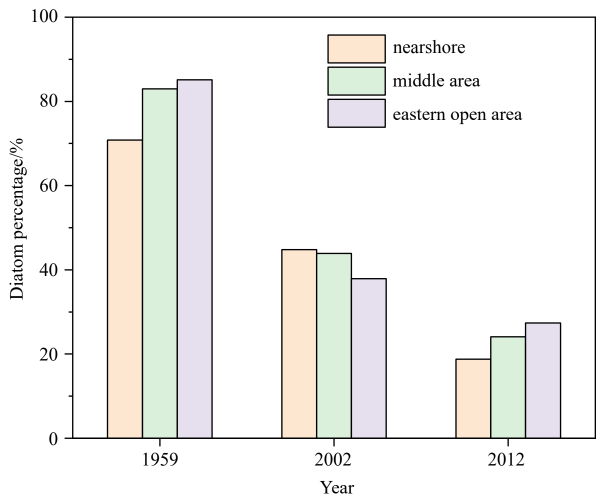

Fig. 8. Percentage of summer diatoms contributing to total phytoplankton abundance in the southern Yellow Sea (data from Yang (2016)).

and increasing dinoflagellate trends demonstrate that, over the last 50 years, the phytoplankton community exhibited an evident tendency to shift from diatoms to dinoflagellates.

Tables 3–5 depict the phytoplankton community's dominant genera and dominance index. From 1958 to 1959, diatoms were the dominant species in the SYS. While dinoflagellates were presented during this time, they did not occupy a dominant position in the phytoplankton community. From 2011 to 2013, dinoflagellates such as *Ceratium* and *Noctiluca* gradually became the dominant genera in the phytoplankton community, creating an environment in which both dinoflagellates and diatoms were dominant species. The shift shows that the dominant position of dinoflagellates in phytoplankton community has gradually prominent in recent years.

Table 2. Percentage of spring diatoms and dinoflagellates contributing to the total phytoplankton abundance in the Yellow Sea (YS)

Area	Survey time	Diatom/%	Dinoflagellate/%	Reference
YS	May 1986	88.90	11.10	Wang et al. (2001)
YS	May 1998	69.50	30.50	Wang et al. (2001)
SYS	March 2005	99.72	0.28	Fu et al. (2012)
SYS	May 2005	50.89	49.11	Fu et al. (2012)
SYS	April 2006	99.63	0.37	Fu et al. (2012)
SYS	April 2007	99.80	1.20	Fu et al. (2012)

Note: SYS is the abbreviation of southern Yellow Sea.

3.2.3 *Red tide frequency*

Figure 9 shows the cumulative frequency and cumulative area of red tide occurrences in the SYS, from 1990 to 2019. During this period, red tides occurred 73 times and covered a total

Table 3. Dominant spring phytoplankton genera in the southern Yellow Sea from 1959 to 2011 (net sampling, data from Yang (2016))

Dominant genera		1959		2011	
		Abundance/(10^4 cells·m^{-3})	Dominance index	Abundance/(10^4 cells·m^{-3})	Dominance index
Diatom	*Ditylum*	–	–	80.04±238.51	0.274
	Navicula	–	–	15.35±53.33	0.024
	Chaetoceros	–	–	73.92±317.98	0.269
	Rhizosolena	53.60±243.3	0.949	4.16±7.10	0.018
	Thalassiosia	–	–	20.18±62.94	0.046
Dinoflagellate	*Ceratium*	–	–	4.20±9.49	0.017

Note: – represents no data.

Table 4. Dominant summer phytoplankton genera in the southern Yellow Sea from 1959 to 2013 (net sampling, data from Yang (2016))

Dominant genera		1959		2012		2013	
		Abundance /(10^4 cells·m^{-3})	Dominance index	Abundance /(10^4 cells·m^{-3})	Dominance index	Abundance /(10^4 cells·m^{-3})	Dominance index
Diatom	*Coscinodiscus*	0.158±0.244	0.07	0.481±0.568	0.078	0.887±1.495	0.042
	Rhizosolenia	0.258±0.518	0.071	–	–	10.613±29.963	0.441
	Hemiaulus	0.262±0.520	0.043	–	–	–	–
	Chaetoceros	–	–	2.947±5.367	0.213	1.859±5.156	0.033
	Paralia	1.046±2.051	0.172	–	–	–	–
	Nitzschia	0.188±0.492	0.041	–	–	–	–
	Pseudonitzschia	–	–	–	–	3.386±9.874	0.060
Dinoflagellate	*Ceratium*	–	–	1.287±1.912	0.186	1.572±4.223	0.075
	Noctiluca	–	–	0.935±1.080	0.135	–	–

Note: – represents no data.

Table 5. Dominant autumn phytoplankton genera in the southern Yellow Sea from 1958 to 2012 (net sampling, data from Yang (2016))

Dominant genera		1958		2011		2012	
		Abundance /(10^4 cells·m^{-3})	Dominance index	Abundance /(10^4 cells·m^{-3})	Dominance index	Abundance/ /(10^4 cells·m^{-3})	Dominance index
Diatom	*Biddulphia*	1.61±3.9	0.017	–	–	–	–
	Thalassiosira	1.34±2.95	0.015	–	–	–	–
	Rhizosolenia	3.51±5.95	0.040	–	–	2.01±6.28	0.035
	Paralia	16.34±43.29	0.153	–	–	–	–
	Nitzschia	33.7±84.78	0.246	–	–	–	–
	Chaetoceros	–	–	16.29±59.01	0.386	3.57±8.73	0.069
	Coscinodiscus	–	–	0.29±0.43	0.011	–	–
	Pseudonitzschia	–	–	–	–	19.86±73.48	0.215
Dinoflagellate	*Ceratium*	–	–	0.71±1.07	0.034	0.41±0.70	0.013

Note: – represents no data.

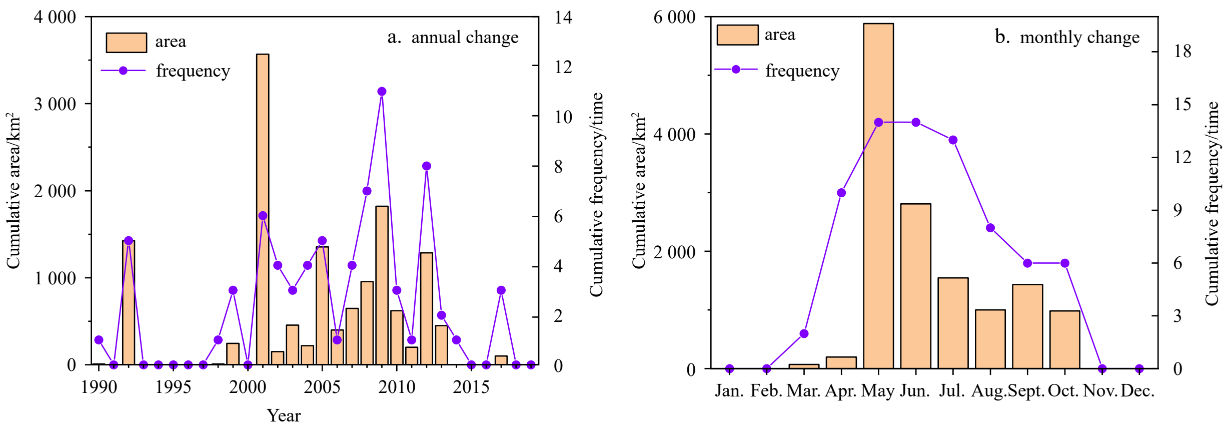

Fig. 9. Variations in red tide area and frequency in the southern Yellow Sea (1990–2019).

area of 13 918 km^2. Red tide events were mainly recorded in coastal regions, with hotspots located in the Jiaozhou Bay, Haizhou Bay, and Rudong coastal waters (Fig. S1). Red tide occurrences corresponded with sites characterized by high pollution and eutrophication.

Red tides were relatively rare from 1990 to 2000, but large events were frequent from 2001 to 2013. The maximum annual frequency occurred in 2009, when 11 red tide events were recorded; while 2001 holds the record for the maximum affected area (3 500 km^2) (Fig. 9a). After 2013, the frequency and scale of red tides in the SYS showed a clear decreasing trend. Because these events are seasonal, the red tides occurred from March to October, with most taking place between May and October. In fact, 83.6% of the total red tide events and 98.1% of the total affected area occurred during these six months (Fig. 9b).

Changes in algal species over time are also observable in the red tides. The frequency and area of dinoflagellate red tides in the SYS increased significantly from 1990 to 2019. Furthermore, dinoflagellate red tides were significantly more frequent than diatom red tides, although both types covered a similar size area (Fig. 10). Specifically, dinoflagellate red tides accounted for 47.9% of the red tide events in the SYS and covered a cumulative area up to 26.6%; while diatom red tides were responsible for 17.8% of the red tide events and covered a cumulative area up to 16.5%. Studies in the Changjiang River Estuary and Yellow Sea show that dinoflagellate abundance and dinoflagellate red tides have both increased significantly (Zhou et al., 2008; Jiang et al., 2014).

4 Discussion

4.1 *Primary nutrient source*

Terrestrial inputs in general, and river input specifically, are the most important external source of nutrients affecting coastal waters in the adjacent seas of China (Qu and Kroeze, 2010). The Changjiang River, which is characterized by an annual runoff of 9.2×10^{11} m^3/a, is the main nutrient input source for the SYS, and the discharge is carried into the Yellow Sea by Changjiang River effluent waters (Le, 1984; Tian et al., 1993). Considering Yellow Sea riverine nutrient inputs from China and Korea, NO_3^- mainly originates from the Changjiang (40%) and Yalu (50%) rivers; NH_4^+ and PO_4^{3-} are primarily delivered by the Han (50%) and Changjiang (20%–30%) rivers, while H_4SiO_4 is mainly contributed by Changjiang (57%) and Yalu (30%) rivers (Liu et al., 2003).

Increased use of fertilizers, exceptionally high sewage discharge, and the exacerbation of soil erosion led to an increase in the concentration of N-based species entering the ocean. Chemical fertilizer used in the Changjiang River basin increased threefold from 1980 (3.02×10^6 t) to 1996 (9.37×10^6 t) (Yan et al., 2003; Gao and Li, 2009). Inter-annual changes in the DIN concentration throughout the Changjiang River significantly increased from the 1970s to 2016. Notably, an exceptionally rapid increase began in the mid-1980s. Since that time, the concentration has fluctuated, but has remained at high levels (Fig. 11a) (Wang et al., 2018). The increasing DIN concentration carried by the Changjiang River to the sea is consistent with the increasing DIN measured in the Yellow Sea from 1990 to 2016. Due to national management policies that aimed at reducing land-based emissions and curbing marine environment deterioration, the DIN concentration in the Yellow Sea decreased from 2017 to 2019.

Approximately 75% of the PO_4^{3-} in the Yellow Sea is inputted from external sources (Liu et al., 2003). From the 1970s to 2016, the PO_4^{3-} concentration in the Changjiang River has steadily increased, with the most significant increase being observed since

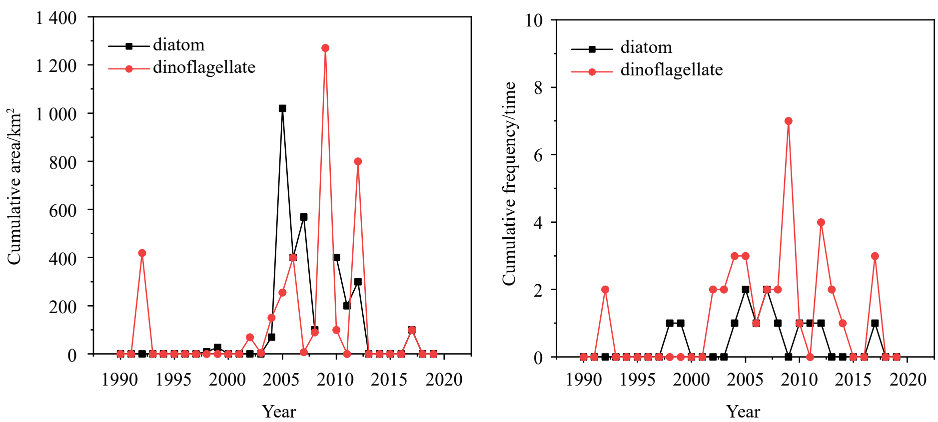

Fig. 10. Variations in area and frequency of diatom and dinoflagellate red tides in the southern Yellow Sea (1990–2019).

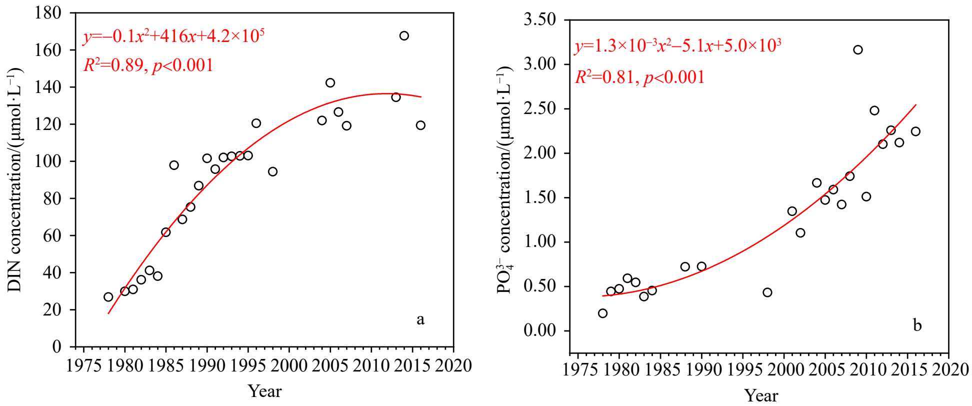

Fig. 11. Long-term variations in riverine nutrient concentrations in the Changjiang River (data were obtained from Wang et al. (2018)).

the end of the 1990s. Since 2012, the PO_4^{3-} concentration has fluctuated, but has remained at high levels (Wang et al., 2018) (Fig. 11b). Interestingly, the interannual variation of PO_4^{3-} in the SYS differs significantly from that in the Changjiang River. This inconsistency may be attributed to two factors: First, the increase rate of PO_4^{3-} input from the rivers is lower than that of DIN. DIN input promotes phytoplankton absorption of PO_4^{3-} indirectly, resulting in oceanic PO_4^{3-} concentrations to depict slightly increasing or decreasing trends. Second, oceanic PO_4^{3-} has endogenous cycling characteristics, and is therefore closely related to both terrestrial input and a series of additional interactions. Examples of the latter include, but are not limited to: absorption and utilization of marine phytoplankton and adsorption/desorption of particulate matter in the sea.

The "artificial lake effect" produced by anthropogenic activities like dam building, dry flow, and farmland irrigation has resulted in dissolved H_4SiO_4 depicting a notable decreasing trend in rivers all over the world (Wang et al., 2018). The H_4SiO_4 concentration in the Changjiang River Estuary decreased by a factor of three from 1960 to 1985 and has basically remained stable (Ma, 2015) (Fig. 12). Lin et al. (2005) found that the Yellow Sea H_4SiO_4 concentration showed a gradual decrease from 1976 to 2000.

Atmospheric deposition is considered an important external nutrient source for the upper ocean and has been shown to impact phytoplankton growth and community structure (Jickells et al., 2005; Shi et al., 2012; Chien et al., 2016; Zhang et al., 2019). On the global scale, atmospheric N input to the coastal waters is generally equivalent to riverine N input (Qi et al., 2013). DIN and PO_4^{3-} from atmospheric deposition in the western Yellow Sea account for 58% and 75% of terrigenous input, respectively (Zhang et al., 1999; Gao and Zhang, 2019). Studies have shown that for external sources, i.e., atmospheric and riverine inputs, the former provides 94% NH_4^+ and 68% PO_4^{3-} to the Yellow Sea, while the latter provides 95% H_4SiO_4 and 66% NO_3^- (Liu et al., 2003). Song et al. (2019) conducted a study from 2009 to 2010 and demonstrated that when considering the entire Yellow Sea, the nutrient contribution from dry deposition was equivalent to that of wet deposition. Furthermore, annual NH_4^+, NO_2^- + NO_3^-, H_4SiO_4, and PO_4^{3-} dry atmospheric deposition input to the Yellow Sea accounts for 87%, 53%, 3%, and 50%, respectively, of the total input from atmospheric deposition and rivers combined. Shi et al. (2013) analyzed the impact of sandstorms on the Yellow Sea in 2007 and showed that the dry deposition nutrient flux during sandstorms is five times higher than that during normal periods. Atmospheric deposition significantly influences marine ecosystems, and total atmospheric N deposition can support 0.3%–6.7% of the new productivity in the Yellow Sea (Qi et al., 2013). What's more, on-board microcosm experiments showed that dust additions supplied a considerable amount of N and negligible P to the seawater. The primary beneficiary was the nano-sized phytoplankton (primarily consisting of dinoflagellates) in the euphotic zone (Zhang et al., 2019), and the atmospheric deposition's fertilization effect was evident in the SYS during spring (Liu, 2014).

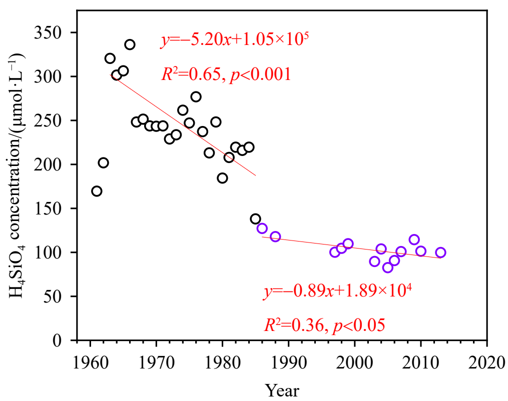

Fig. 12. Interannual dissolved H_4SiO_4 variations in the Changjiang River (data from Ma et al. (2015)).

4.2 *The relationship between nutrient and phytoplankton community*

4.2.1 *Phytoplankton community succession*

In recent years, the significant increasing dinoflagellate abundance trend in the SYS has been observed and confirmed by many scholars. The escalating dominance of dinoflagellate red tides with respect to frequency and scale was recorded in the Yellow Sea, and is closely related to eutrophication and the development of mariculture industry (Guo et al., 2015; Li et al., 2021). Yang (2016) found that dinoflagellate abundance in the SYS has increased significantly compared to that in 1959 and the percent carbon content from dinoflagellates contributing to the total carbon content has also increased. The increasing proportion of dinoflagellate phytoplankton abundance depicts an evident evolution in the phytoplankton community in China's offshore waters (Yang, 2016). According to the observation from 1985 to 2015, diatom abundance had been supplanted by dinoflagellate abundance in the Yellow Sea was confirmed. The average diatom abundance in the Yellow Sea accounted for 80.3% of the total average abundance for the period from 1985 to 2015, and only 67.5% in 2010. In contrast, the average dinoflagellate abundance accounted for 19.7% of the total average abundance for the period from 1985 to 2015, and 32.5% in 2010 (Fig. 13) (Luan et al., 2020). Xiao (2018) forecasted that by the end of the 21st century, diatom biomass in 60% of the East China Sea will have decreased and the dinoflagellate biomass in 70% of the East China Sea will have increased. These results imply that dinoflagellate blooms will become more frequent and intense, and will impact coastal ecosystem functions (Xiao et al., 2018; Huang et al., 2021).

4.2.2 *Nutrient structure and stoichiometry ecological impacts*

Previous studies have established that changes in nutrient concentrations and structure have dramatic impacts on marine phytoplankton. With respect to nutrient structure, some phytoplankton species can absorb and utilize low-molecular-weight dissolved organic nitrogen (DON) or dissolved organic phosphorus (DOP) directly (Huang et al., 2005; Xiao et al., 2019; Wang et al., 2021a, 2021b). Compared with nitrate, ammonium can be preferentially up-taken by phytoplankton species due to the unchanged valence state of N (Glibert et al., 2014). Nutrient stoichiometry is one of the most important factors impacting changes in phytoplankton species composition and dominant species shifts (Mathew et al., 2021). Long-term N/P ratios in the SYS showed a rapidly increasing trend from 1990 to 2015 and a decreasing trend from 2016 to 2019 (Fig. 5). Si/N ratios depicted a slightly increasing trend from 1958 to 2019, with ratio values that were generally larger than 1 (Fig. 6). Asymmetric changes in nutrient structure indicated the presence of obvious P-limitation and potential Si limitation in some areas, which impacted the SYS phytoplankton community. Nutrient limitation studies have shown that the phytoplankton's growth threshold is a key factor dictating their community characteristics. Under normal conditions, when the phytoplankton has not reached the maximum

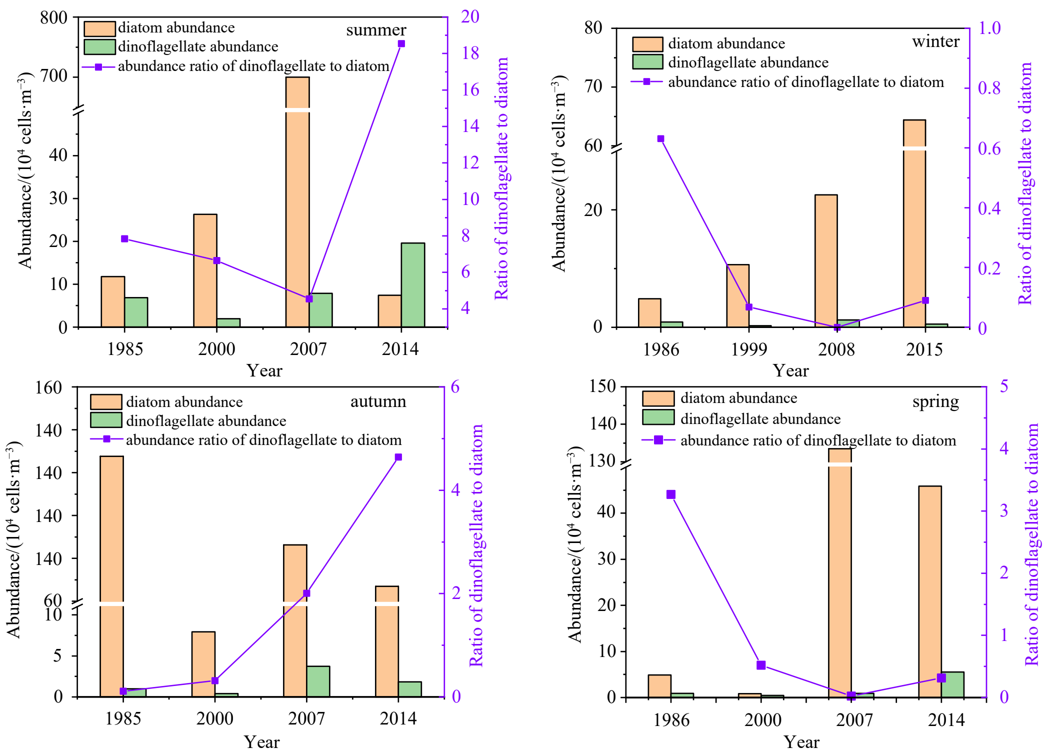

Fig. 13. Interannual variation of phytoplankton abundance in the southern Yellow Sea (data from Luan et al. (2020)).

growth rate, the phytoplankton N/P concentration demand ratio is positively correlated with the N/P supply ratio in the surrounding environment (Klausmeier et al., 2004; Zhang et al., 2018). This relationship reflects the phytoplanktons' ability to adapt to the surrounding environment by efficiently adjusting their physiological state and modifying the phytoplankton community structure (Gao and Zhang, 2019). Li (2008) and Xiao et al. (2018) studied the phytoplankton community changes in the Changjiang River Estuary and showed that the higher N/P ratio or higher N concentration was beneficial to dinoflagellate growth. The reduction of nutrients caused by climate warming (layers) and the increase of nutrients imported from land sources facilitated dinoflagellates to supplant diatoms as the dominant species (Li, 2008; Xiao et al., 2018; Huang et al., 2021). Lin et al. (2005) reported that when the northern Yellow Sea was in a P-limited and N-sufficient state, the main phytoplankton community species transformed from diatoms to dinoflagellates. In this work, the significant summertime increase in the SYS phytoplankton abundance observed after 2000 is attributed to the DIN and N/P ratio fluctuations in the SYS surface water, which maintained a high concentration from 2000 to 2015. This indicates that these nutrients are an important control on phytoplankton growth (Figs 2, 5 and 7).

4.2.3 *The relationship between nutrient and phytoplankton community in the Haizhou Bay*

Figure 14 depicts the relationship between nutrients and phytoplankton communities in the Haizhou Bay from 2003 to 2017. Asymmetric changes were observed between N/P and Si/N ratios, demonstrating evident P-limitation and potential Si-limitation. Specifically, N/P ratios ranged primarily from 16 to 100 with the maximum value larger than 600, while Si/N ratios ranged mainly from 0.2 to 1.2 with the maximum value larger than 3. Results from the phytoplankton community study showed that diatoms were the dominant species. However, other species, such as dinoflagellates, frequently appeared in the bay, indicating that diatoms and dinoflagellates coexisted in the Haizhou Bay. Furthermore, results from a correlation analysis between diatom/dinoflagellate abundance/frequency and nutrients are presented in Table 6. As shown, diatom abundance is significantly correlated with DIN concentration and N/P ratio, while frequency is correlated with Si/N ratio. Contrarily, dinoflagellate abundance is significantly correlated with Si/N ratio and frequency is correlated with DIN and H_4SiO_4. This phenomenon indicates that although the diatom abundance/frequency correlated slightly differently with the nutrients as compared to that of dinoflagellates, DIN and H_4SiO_4 were the main factors impacting phytoplankton growth.

5 Conclusions

Herein, long-term variations in nutrient concentration and structure, inter-annual phytoplankton community trends, and the relationship between nutrients and phytoplankton community were systematically analyzed in the SYS. Over the past 30 years, due to influences from both anthropogenic activities and terrestrial inputs, nearshore nutrient concentrations have been significantly higher and the structures more diverse than those

Table 6. Pearson correlation between nutrients and the phytoplankton community in the Haizhou Bay

Parameter	DIN	PO_4^{3-}	H_4SiO_4	N/P	Si/N
Diatom abundance	0.328**	–0.036	0.040	0.428**	–0.006
Diatom frequency	0.113	0.167	–0.069	0.076	–0.248*
Dinoflagellate abundance	–0.148	–0.285	–0.069	–0.054	0.494**
Dinoflagellate frequency	–0.318*	–0.138	–0.334*	–0.283	0.066

Note: **Correlation is significant at the p=0.01 level (2-tailed); *Correlation is significant at the p=0.05 level (2-tailed). N/P is the molar ratio between N concentration and P concentration; Si/N, the molar ratio between Si concentration and N concentration.

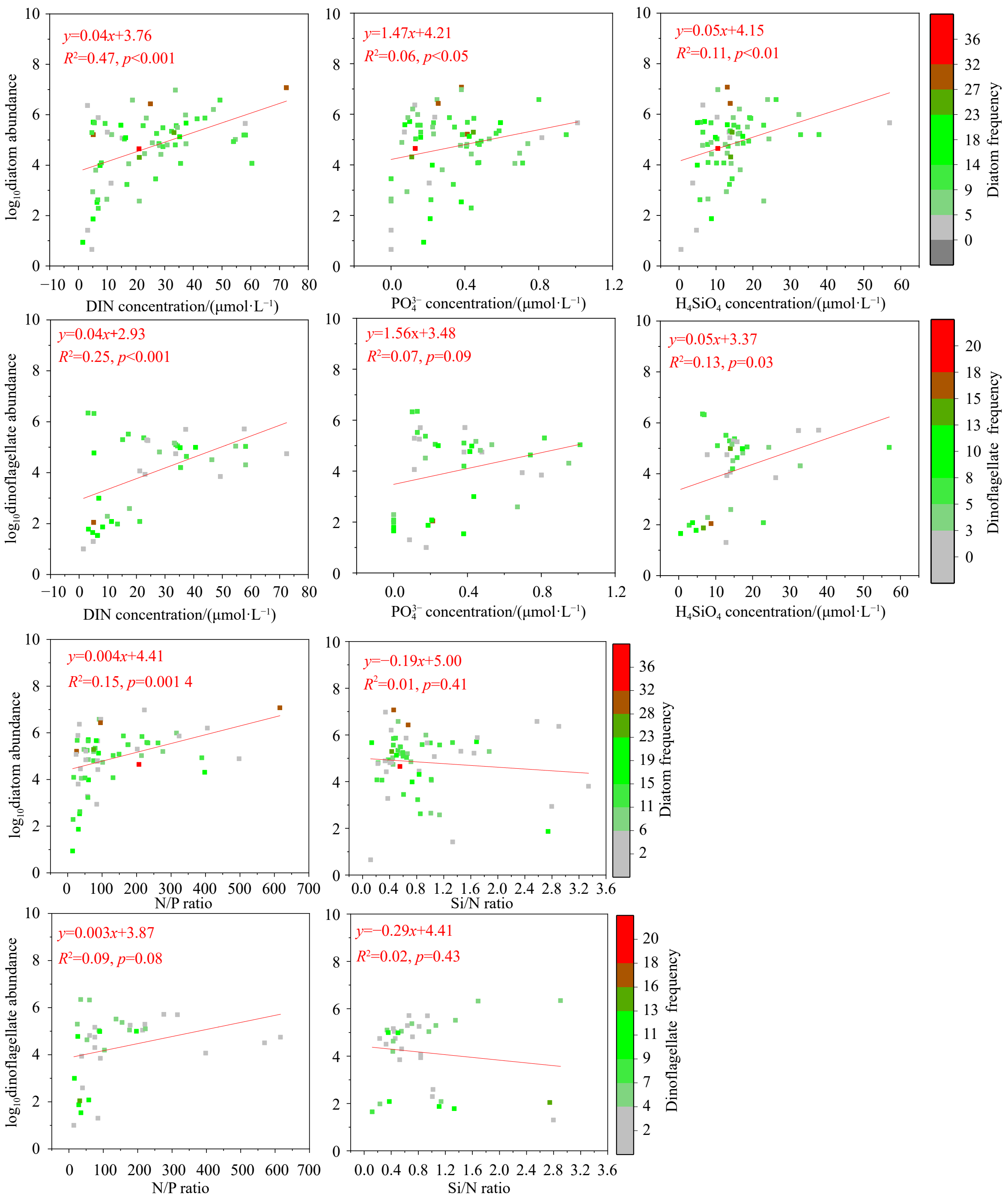

Fig. 14. Correlation between nutrients and phytoplankton community in the Haizhou Bay.

observed in the offshore. Nutrient limitation conditions of the SYS changed from N limitation to P limitation and potentially to Si limitation, which impacted the phytoplankton community. The phytoplankton species composition change and dominant species shift from diatoms to dinoflagellates have been validated by annual changes in red tide frequency and composition. Although diatoms still are the dominant species in phytoplankton community, an evident succession from diatoms to dinoflagellates has exhibited over the last 50 years.

Because the nutrient, red tide, and phytoplankton data discussed herein are relatively independent, it is impossible to quantitatively assess the impact of nutrients on the phytoplankton community. Thus, the relationship between these factors was only qualitatively analyzed. Moreover, although nutrients are the foundation of phytoplankton growth, it is a complex process that is also impacted by factors like light, temperature, and salinity, etc. Thus, the phytoplankton community is not static, and often changes with the marine environment, and this ongoing transition requires further systematic exploration. Therefore, long-term comprehensive investigations that integrate environmental factors with phytoplankton community changes are needed to better understand ecosystem response in the SYS.

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Supplementary information:

Fig. S1. Map of red tide events in the southern Yellow Sea from 1990–2019.

Table. S1. Data of phytoplankton abundance of Fig. 7.

The supplementary information is available online at https://doi.org/10.1007/s13131-022-2031-3 and www.aosocean.com. The supplementary information is published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.