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Long-Term Nutrient Variations in the Bohai Sea Over the Past 40 Years

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Key Points:

- DIN increased; PO_4-P decreased; SiO_3-Si decreased before 1987 and increased after 1987 in the Bohai Sea; winter concentrations are higher
- N/P increased, Si/N decreased, and Si/P slightly increased in the Bohai Sea; ratios are higher in winter than in summer
- Variations in Si/N ratios may have ecological impacts on the red tide features in the Bohai Sea

Supporting Information:

- Supporting Information S1
- Table S1

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Abstract As China's only continental sea, the Bohai Sea is a relatively closed environment and is vulnerable to natural changes and human activities. In this paper, the long-term variations in nutrients in the Bohai Sea and the potential influencing factors were analyzed based on historical summer and winter data from 1978 to 2016. The results showed that the concentrations of dissolved inorganic nitrogen (DIN) in the Bohai Sea continuously increased from 1990 and rapidly increased after 2002, the phosphate (PO_4-P) concentration exhibited a decreasing trend, and the silicate (SiO_3-Si) concentration decreased from 1978 to 1987 and increased from 1987 to 2008. The nutrient concentrations were lower in summer than in winter, and the bottom concentrations were higher than the surface concentrations in summer, whereas the vertical differences were insignificant in winter. The dominant factor determining the long-term variations in DIN were atmospheric deposition and nonpoint sources due to various human activities; the primary factors affecting PO_4-P were riverine inputs and nonpoint sources related to natural changes and human activities; the primary factors afftecting SiO_3-Si were riverine inputs. The N/P ratio followed the DIN variation, the Si/P ratio followed the SiO_3-Si variation, and the Si/N ratio decreased. The nutrient ratios were lower at the bottom than at the surface and were lower in winter than in summer. The nutrient limitation changed from nitrogen limitation to phosphorus and silicon limitations. The long-term nutrient variations in the Bohai Sea have potential ecological impacts on the local red tide features.

Plain Language Summary Nutrients are important to primary productivity and thus important to marine ecosystems. Nutrient variations in coastal oceans may potentially impact the occurrence of ecological disasters. As the only continental sea in China, the Bohai Sea is a relatively closed environment and is vulnerable to natural and anthropogenic impacts. We found that the concentration of dissolved inorganic nitrogen in the Bohai Sea increased from 1990 to 2006; the phosphate concentration decreased from 1978 to 2016, and the silicate concentration decreased before 1987 and increased after 1987. We also found that N/P increased and Si/N decreased. The nutrient concentrations were higher, whereas the nutrient ratios were lower in winter than in summer. In summer, the bottom nutrient concentrations were higher than the surface concentrations, whereas the bottom ratios were lower than the surface ratios; in winter, the vertical differences were insignificant. The primary factors driving the silicate variations were riverine inputs related to natural changes and human activities; the primary factors in the phosphate variations could be riverine inputs and nonpoint sources related to natural changes and human activities; the dominant factor in the dissolved inorganic nitrogen variations could be atmospheric deposition and nonpoint sources due to various human activities. The red tide features in the Bohai Sea seemed to be potentially related to the long-term nutrient variations.

1. Introduction

Nutrients are very important to primary productivity, which is the basis of marine ecosystems. The addition of nutrients to aquatic environments will stimulate the preferential growth of certain phytoplankton species under suitable conditions (Ferreira et al., 2007) and lead to further changes in their communities (Justić et al., 1995; Yunev et al., 2007). With the accelerated development of agriculture and industry over the past four decades, nutrient inputs from land have undergone significant changes globally, and these changes have resulted in variations in the concentrations and compositions of nutrients in seawater (Danielsson et al., 2008; Jickells, 1998; Paerl, 2006; Yu & Shen, 2011). Due to the potential effects of nutrient variations

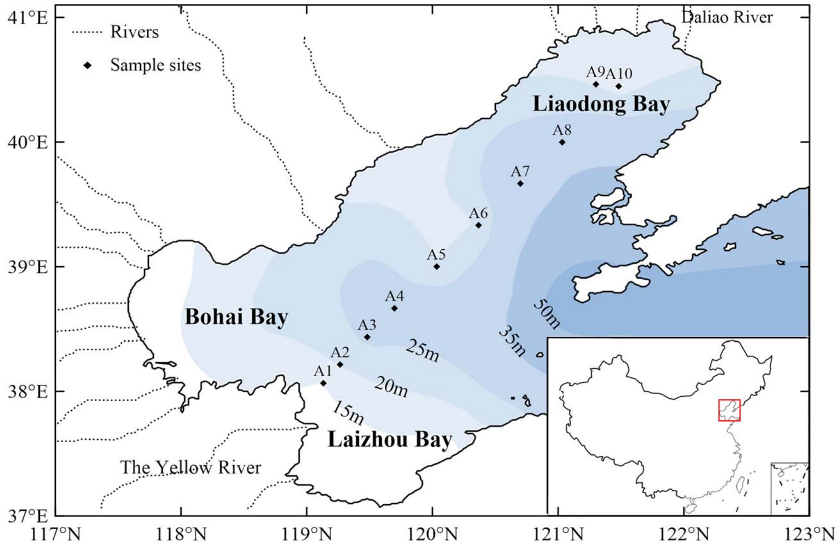

Figure 1. Schematic illustration of the sites where historical data were collected in the Bohai Sea. The dots with "A1–A10" represent the sample sites from 1978 to 2016; the dotted lines represent the rivers.

on marine ecosystems, harmful algal blooms (Anderson et al., 2002; Glibert & Burkholder, 2011; Paerl, 2006; Yu & Shen, 2011; Yunev et al., 2007), jellyfish blooms (Purcell, 2012; Shin-Ichi, 2008; Xian et al., 2005), hypoxia, and ocean acidification may occur in coastal oceans (Cai et al., 2011; Kemp et al., 2009; Rabalais et al., 2002, 2014).

The Bohai Sea is a continental sea in China, which has a total area of 77,284 km^2 and an average depth of 18 m (Zhang et al., 2006). The Bohai Sea is composed of three bays (Liaodong Bay, Bohai Bay, and Laizhou Bay) and a central area, and it is connected to the northern Yellow Sea via the Bohai Strait (Figure 1). Approximately 40 rivers, including four major rivers (the Yellow River, Hai River, Daliao River, and Luan River), flow into the area and transport large amounts of nutrients. With the rapid expansion of industry, agriculture, and mariculture developments, nutrient fluxes from rivers (Zhang, 1996; Zhang et al., 1994), atmospheric deposition (Liu et al., 2008), wastewater discharge (Liu, 2006; Wang et al., 2016), and other sources have substantially increased. As an important spawning ground for fish and shrimps, the Bohai Sea has abundant marine resources and provides very important economic and ecological functions. Shandong Province, Liaoning Province, Hebei Province, and Tianjin municipality are located along the coast of the Bohai Sea, making it one of the most populated areas in China. The Bohai Sea is a relatively closed environment that is vulnerable to anthropogenic stress. Under the influence of dense human activities combined with changes in the natural environment, variations in the concentrations and compositions of nutrients in the Bohai Sea may lead to eutrophication and consequent ecological disasters. In recent decades, the frequency and affected area of red tides have increased significantly in the Bohai Sea (Shen, 2001; Zhao, 2010; Zou et al., 1985). Moreover, the variation in nutrients may boost phytoplankton production and algae blooms in the Bohai Sea, which may further enhance zooplankton production and supply more food to jellyfish, thus resulting in jellyfish blooms in the Bohai Sea (Dong et al., 2010; Purcell, 2012; Shin-Ichi, 2008; Xian et al., 2005). Hypoxia and coastal acidification (Yu & Zhang, 2016; Zhai et al., 2012; Zhang et al., 2016) have also been observed in the Bohai Sea since 2011. These ecological disasters in recent years have severely threatened the health of local marine ecosystems, and the variations in nutrients and the ecological environment of the Bohai Sea have raised increasing attention (Liu et al., 2008; Ning et al., 2010; Wu et al., 2013; Yu et al., 2000).

To examine the nutrient variations in the Bohai Sea, a study by Liu et al. (2008) showed that from 1959 to 1999, the annual mean concentration of dissolved inorganic nitrogen (DIN) and the N/P ratio increased, while the concentration of dissolved silicate (SiO_3-Si) and the Si/N ratio decreased; the concentration of dissolved inorganic phosphate (PO_4-P) slightly increased from 1959 to 1982 and decreased from 1982 to 1999, which was also proven by the results of Cui and Song (1996). Similar increases in the DIN concentration and N/P ratio and similar decreases in the PO_4-P and SiO_3-Si concentrations from the early 1980s to the late 1990s were also found in the studies by Yu et al. (2000) and Ning et al. (2010). However, the variation patterns of the nutrient concentrations in the Bohai Sea over the last 40 years are not the same as those observed in other previous studies, and ambiguity likely exists. In contrast to the continuous decreasing trends of the PO_4-P and SiO_3-Si concentrations from the early 1980s to the late 1990s in the aforementioned studies, Li et al. (2003) and Jiang et al. (2005) observed decreases from 1982 to 1992 and increases from 1992 to 1998 in the SiO_3-Si and PO_4-P concentrations in the Bohai Sea. This result was supported by the results of the study by Zhang et al. (2006) from 1985 to 1998, which showed a decrease before 1990 and an increase after 1990 in the PO_4-P concentration. In addition, the results of the study by Li et al. (2003) showed that the DIN concentration in the Bohai Sea decreased from 1982 to 1992 and then increased from 1992 to 2000, whereas the results of the study by Xu et al. (2010) based on previous studies found that the DIN concentration increased from 1980 to 1995 and decreased from 1995 to 2005. These two distinct patterns of the variations in DIN in the Bohai Sea are not consistent with the aforementioned continuously increasing trend. Evidence has shown that discharge from the Yellow River (Cui et al., 1994; Li et al., 2003; Liu et al., 2012; Xu et al., 2010; Zhao et al., 2002) and atmospheric deposition (Cui, 2008; Liu et al., 2008; Shou et al., 2018; Zhang et al., 2004) are important sources of nutrients in the Bohai Sea, whereas on-land sewage treatment and chemical fertilizers may be other sources of DIN and PO_4-P (Li et al., 2003; Liu et al., 2005; Xu et al., 2010). As the ratios of different nutrients in the Bohai Sea changed with the variations in nutrient concentrations, it was believed that a gradual change from nitrogen limitation to phosphate and silicate limitations occurred during the early 1980s to the late 1990s (Jiang et al., 2005; Yu et al., 2000), and a large variance in the phytoplankton community structure was observed (Liu et al., 2008; Wu et al., 2013; Xu et al., 2010). According to the study by Yu et al. (2000), the increased N/P ratio and the decreased Si/N ratio from 1982 to 1999, which are potentially attributed to the increase in fertilizer and decrease in the flow of the Yellow River into the Bohai Sea, may be the major factors driving the occurrence of red tide dinoflagellates (Pyrrophyta) in the Bohai Sea. These previous studies primarily applied data from discrete years (i.e., discontinuous time series such as 1982, 1992, and 1998) to represent the long-term variations in nutrients in the Bohai Sea during different periods and focused on the variation trends before 2000. To date, the patterns of the long-term variations in the nutrients in the Bohai Sea remain controversial and unclear, especially the continuous variation trends after 2000. In this paper, a retrospective analysis of the long-term variation patterns of nutrients and their compositions in the Bohai Sea was performed based on historical data from 1978 to 2016. Seasonal differences were also considered by comparing the trends in summer and winter. This paper constitutes a considerable expansion of previous work, and we also explored potential influencing factors to reveal the typical regional response of the Bohai Sea to external forcing and its relationship to changes occurring in the ecosystem during this period.

2. Materials and Methods

2.1. Data Sources

Ten sampling sites A1-A10 are located on a transect across the Bohai Sea from southwest to northeast within the range of 119.13°E to 121.48°E longitude and 38.07°N to 40.45°N latitude (Figure 1). Site A1 between Bohai Bay and Laizhou Bay in the southwestern Bohai Sea is close to the Yellow River Estuary, whereas site A10 in Liaodong Bay in the northeastern Bohai Sea is close to the Daliao River Estuary. The transect survey data used in this study from 1978 to 2006 and from 2015 to 2016 were obtained from the State Oceanic Administration of China, and the data from 2013 to 2014 were obtained from the Yellow Sea Fisheries Research Institute, Chinese Academy of Fishery Sciences. The surveys were conducted every season (February, May, August, and November) and almost every year from 1978 to 1995 and 2013 to 2016, but only in winter (February/January) and summer (August/July) from 1996 to 2006. We selected the data from the winter and summer seasons to ensure that the data were comparable and the time series was continuous. The survey parameters included dissolved nitrate (NO_3-N), nitrite (NO_2-N), ammonia (NH_4-N), phosphate (PO_4-P), and silicate (SiO_3-Si). DIN was the sum of NO_3-N, NO_2-N, and NH_4-N. However, NO_3-N and NO_2-N measurements began in 1984, and NH_4-N measurements began in 1990; no nitrogen measurements were collected from 1996 to 1999, and SiO_3-Si was not measured from 2007 to 2016. The reference data for nutrients in the summer of 2008 (Liu et al., 2011) and the reference data for PO_4-P in the summers of 2007–2011 (Shi, 2013) that were collected along the same transect were adopted to fill the data gaps during this period. All sampling and analytical methods followed the Chinese national seawater analysis standards, which ensured that the data were comparable throughout the study period.

In addition to the aforementioned survey data, the water discharge and sediment load data from the Yellow River at the Lijin station (obtained from the Yellow River Water Resources Commission and Yuan et al., 2006) were also analyzed to obtain the riverine inputs. The fertilization data for the Bohai Rim, which were obtained from China Statistical Yearbook (the National Bureau of Statistics of the People's Republic of China, 1978–2016), were analyzed to reveal the human impacts on the river inputs. The annual precipitation data from Tianjin (obtained from the National Bureau of Statistics of the People's Republic of China, 1978–2016 and Li et al., 2010) were used to reflect the wet deposition in the Bohai Sea. The annual frequencies of sand and dust events in northern China (obtained from the China Meteorological Administration, National Climate Committee, China Climate Bulletin) were used to reflect the dry deposition in the Bohai Sea. The data on wastewater discharge into the Bohai Sea were obtained from China Marine Statistical Yearbook (the State Oceanic Administration, 1985–2016). The data on mariculture area in the Bohai Sea were obtained from China Fishery Statistical Yearbook (Bureau of Fisheries of the Ministry of Agriculture, 1979-2016). Moreover, the red tide frequency and affected area in the Bohai Sea (obtained from the China Marine Statistical Yearbook and State Oceanic Administration, People's Republic of China, Chinese Marine Disaster Bulletins) were applied to analyze the potential ecological effects.

2.2. Data Analyses

The mean values of the nutrient concentrations at the surface layers and the bottom layers (i.e., 2 m above the seabed) of the 10 sites along the transect were calculated in both the winter and summer seasons, and the mean values of the two seasons were calculated as the annual means. Thus, the interannual variations in the surface and bottom nutrients for each component along the transect were obtained (1978–2016 for PO_4-P, 1978–2006 for SiO_3-Si, 1984–2014 for NO_3-N and NO_2-N, 1990–2014 for NH_4-N, and 1990–2016 for DIN), and linear regression analytical methods (including simple linear regression and binomial regression) were used to evaluate the variation trends. However, during the period from 2007 to 2012, no survey data were obtained from the transect in summer and winter. In the summer of 1993 and the winter of 1984, no parameters were measured; in the winter of 1990, NH_4-N was not measured, which indicated that the NH_4-N time series in winter began in 1991; in 2015 and 2016, the DIN and PO_4-P concentrations were measured only at the surface, and thus, no bottom data were analyzed. Due to the limitations of the different nutrient measurements stated above, the long-term analysis of the variations in N/P ratios was limited to the summers from 1990 to 2016 and winters from 1991 to 2016. For Si/N, the summers from 1990 to 2008 and winters from 1991 to 2006 were analyzed, and for Si/P, the summers from 1978 to 2008 and winters from 1978 to 2006 were analyzed.

3. Results

3.1. Variations in Nutrient Concentrations

In summer, the surface and bottom concentrations of PO_4-P generally exhibited decreasing trends, whereas the concentrations of SiO_3-Si decreased from 1978 to 1987 and showed an increasing trend from 1987 to 2008 (Figures 2a–2f). The surface and bottom concentrations of DIN exhibited an overall increasing trend from 1990 to 2016 with a significant increase from 2002 to 2008 and subsequently maintained high values. The surface and bottom concentrations of NO_3-N showed synchronous increasing trends with the increasing DIN concentrations, whereas the concentrations of NH_4-N and NO_2-N showed moderate increasing trends. For the vertical differences, the bottom concentrations of nutrients in summer were slightly higher than the surface concentrations. The proportions of NO_3-N showed a significant increasing trend, while the proportions of NH_4-N and NO_2-N showed decreasing trends (Figures 2g–2h). In the summers from 1990 to 2000, NH_4-N accounted for the largest proportion of DIN, NO_3-N accounted for the second largest proportion, and NO_2-N contributed the least. However, as the proportion of NO_3-N increased, NO_3-N tended to be the main form of DIN in the Bohai Sea from 2000. The proportion of NO_3-N was slightly larger at the surface than near the bottom; the opposite pattern was observed for the proportion of NH_4-N.

In winter, the surface and bottom concentrations of PO_4-P fluctuated with a slightly increasing trend from 1978 to 2016 (Figures 3a–3f). Sharp decreases were found in 1987 and 1997. The SiO_3-Si concentrations in

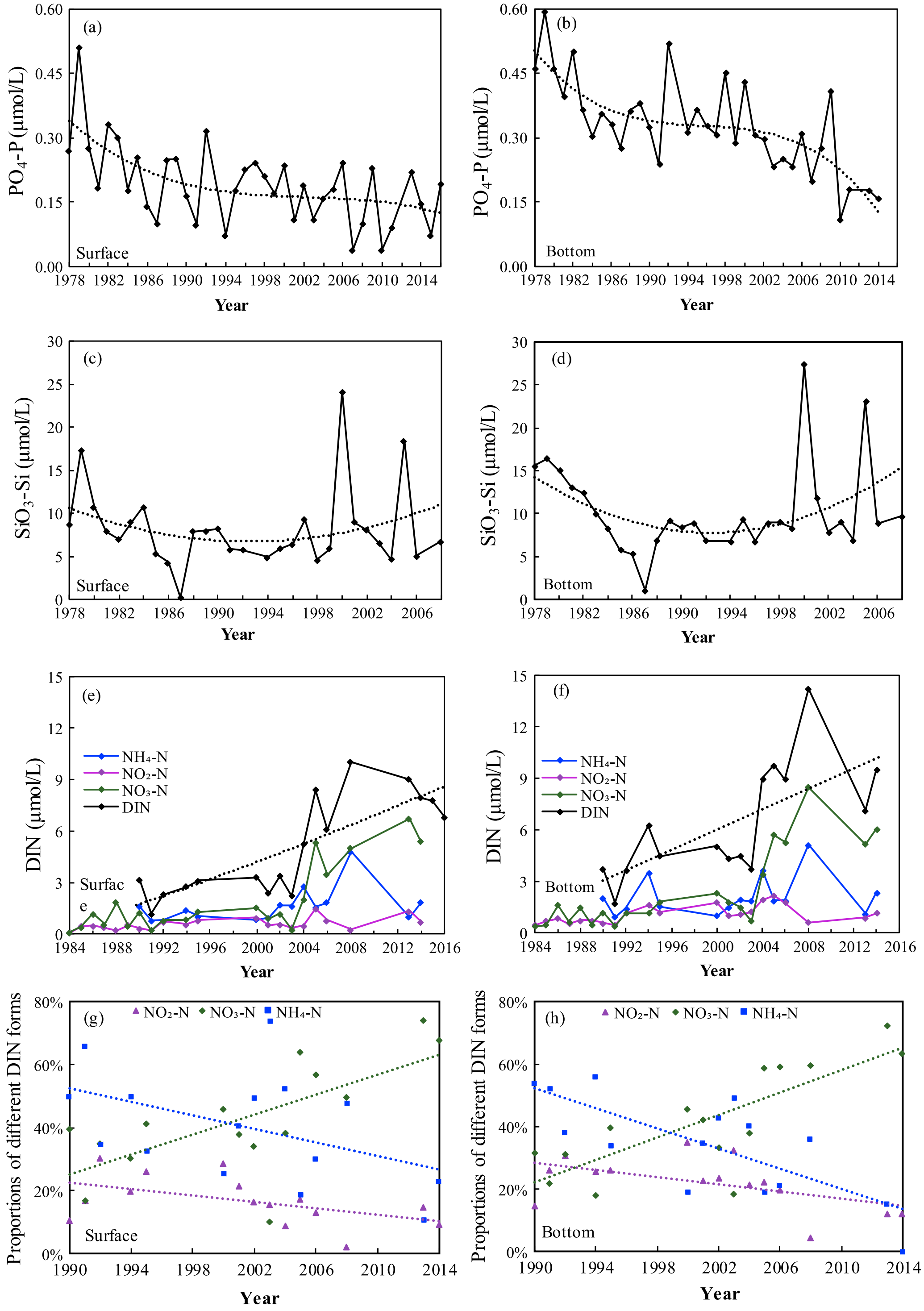

Figure 2. Temporal variations in nutrient concentrations along the A1–A10 transect in the Bohai Sea in summers from 1978–2016: (a) surface PO_4-P concentration, (b) bottom PO_4-P concentration, (c) surface SiO_3-Si concentration, (d) bottom SiO_3-Si concentration, (e) surface dissolved inorganic nitrogen (DIN) concentration, (f) bottom DIN concentration, (g) surface proportions of different DIN forms, and (h) bottom proportions of different DIN forms. The dotted lines represent the variation trend lines determined by linear regression (including simple linear regression and binomial regression).

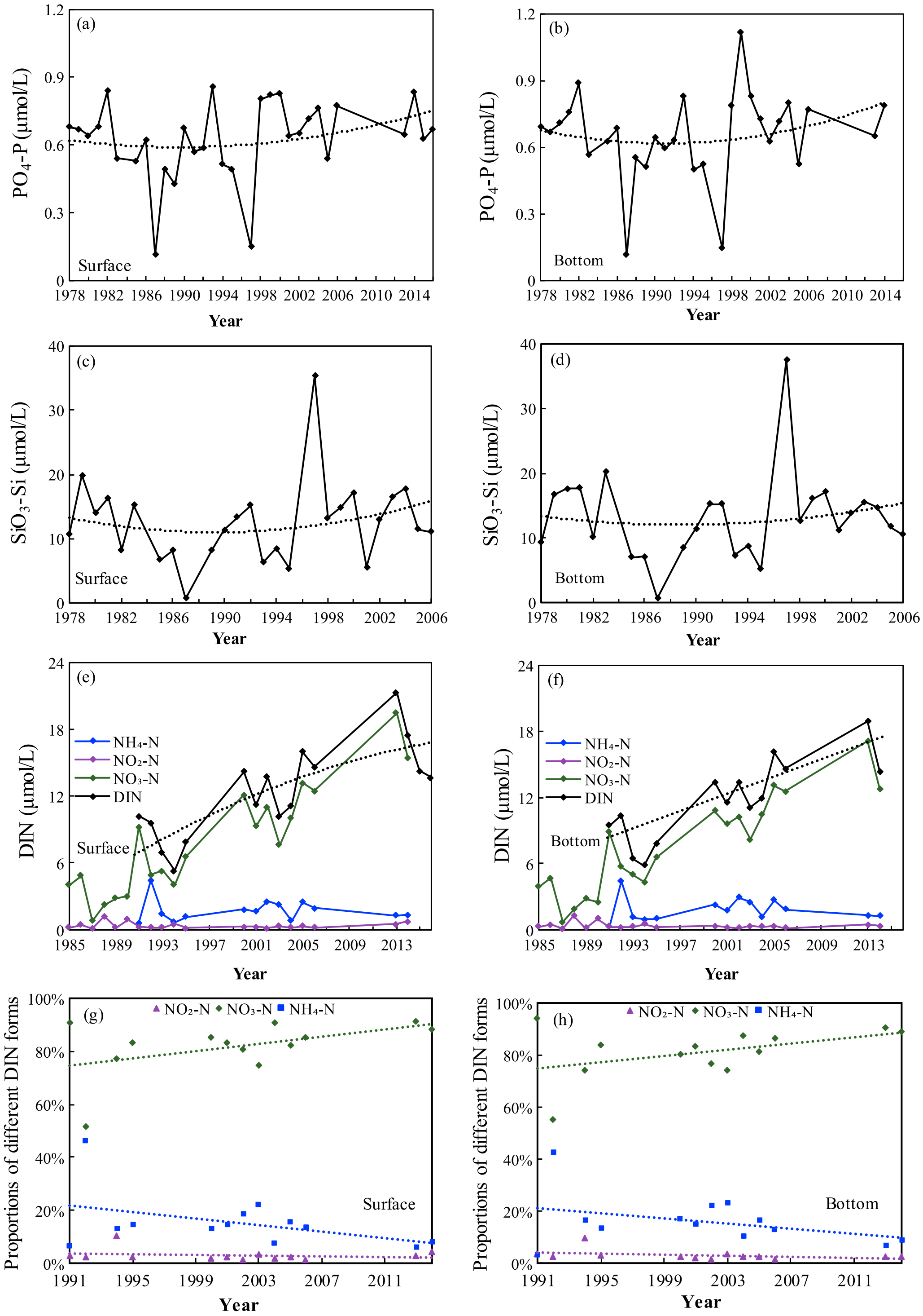

Figure 3. Temporal variations in nutrient concentrations along the A1–A10 transect in the Bohai Sea in winters from 1978–2016: (a) surface PO_4-P concentration, (b) bottom PO_4-P concentration, (c) surface SiO_3-Si concentration, (d) bottom SiO_3-Si concentration, (e) surface dissolved inorganic nitrogen (DIN) concentration, (f) bottom DIN concentration, (g) surface proportions of different DIN forms, and (h) bottom proportions of different DIN forms. The dotted lines represent the variation trend lines determined by linear regression (including simple linear regression and binomial regression).

winter showed a trend that was similar to that observed in summer, decreasing from 1978 to 1987 and increasing from 1987 to 2008. The surface and bottom concentrations of NO_3-N exhibited uniform increasing trends from 1991 to 2016, and the concentrations of DIN showed synchronous increasing trends. The concentrations of NH_4-N and NO_2-N fluctuated over relatively steady ranges. The vertical differences in nutrient concentrations were less pronounced in winter than in summer. Moreover, the nutrient concentrations in winter tended to be higher than those in summer. The variation patterns of the proportions of different DIN forms were almost the same in the surface and bottom layers of the Bohai Sea in the winters from 1991 to 2014 (Figures 3g–3h). In the winters during this period, the proportions of NO_3-N were the largest and showed a continuous increasing trend. The proportions of NH_4-N were the second largest but much smaller than those of NO_3-N. The proportions of NO_2-N were the smallest and remained at approximately 3%.

The annual mean concentrations of PO_4-P generally exhibited a slight decreasing trend, and the variations in the annual mean concentrations of SiO_3-Si and DIN were similar to those in summer and winter (Figures 4a–4f). The bottom concentrations of PO_4-P and SiO_3-Si were slightly higher than the surface concentrations, while the concentrations of DIN exhibited the opposite patterns. In terms of the DIN composition at the surface and bottom of the Bohai Sea during the study period, the average proportions of NO_3-N were the highest, those of NH_4-N were the second highest, and those of NO_2-N were the lowest (Figure 4g). The differences between the proportions of NO_3-N and NH_4-N were 2.43% at the surface and 6.40% at the bottom in summer and 66.49% at the surface and 65.21% at the bottom in winter. The proportions of NO_3-N were higher in winter than in summer as well as throughout the year, while the NH_4-N and NO_2-N proportions were lower in winter. The proportions of NO_3-N were greater at the surface than at the bottom in different seasons and throughout the year.

3.2. Variations in Nutrient Ratios

In summer, the surface and bottom ratios of N/P exhibited increasing trends, with rapid increases after 2002 (Figure 5a), and these changes are similar to the variation patterns of the DIN concentrations. The N/P ratios were also higher than the Redfield ratio of 16:1 after the 2000s. The Si/N ratio showed an overall decreasing trend and tended to be lower than the Redfield ratio of 1:1 from 2004 (Figure 5d). The Si/P ratio showed an overall increasing trend, especially after 1987 (Figure 5g), which is similar to the variation pattern of the SiO_3-Si concentration. The bottom ratios tended to be lower than the ratios at the surface. The patterns of the variations in the nutrient ratios in summer in the Bohai Sea indicated that the nutrient limitation condition changed from a nitrogen limitation to phosphorus and silicon limitations, which may have influenced the uptake and utilization of phytoplankton in the Bohai Sea. The overall trend of the variations in the nutrient ratios in winter generally followed the variation patterns observed in summer (Figures 5b, 5e, and 5h). The nutrient ratios in winter were much lower than those in summer, and the slopes of the variations in winter were much smaller than those in summer. The vertical differences in nutrient ratios were not prominent in winter and were much smaller than those in summer. The variations in the annual mean nutrient ratios generally followed the patterns in summer (Figures 5c, 5f, and 5i). The annual mean nutrient ratios were lower at the bottom than at the surface, while the vertical differences in the N/P and Si/N ratios were small.

4. Discussion

4.1. Comparison With Previous Studies

During the period from the early 1980s to the late 1990s, an increase in the DIN concentration and a decrease in the PO_4-P concentration were observed in the Bohai Sea in three studies using 1982-1992-1998 time series (Cui & Song, 1996; Liu et al., 2008; Yu et al., 2000) and one study using a continuous time series from 1978 to 1996 (Ning et al., 2010). Our study revealed the same variation patterns of the annual mean concentrations of DIN and PO_4-P during the same period. The decreasing trend of the PO_4-P concentration was also observed by Shi (2013), who analyzed the PO_4-P variation in the Bohai Sea in the summers from 1978 to 2012. Moreover, according to the continuous increase in the DIN concentration from 1985 to 1998 in the study by Zhang et al. (2006) and the similar increase starting from 1959 in the studies by Cui and Song (1996) and Liu et al. (2008), the increasing trend of the DIN concentration can be traced back to 1959; based on the results of this study, particularly the DIN variation after 2000, we suggest an increasing trend of the

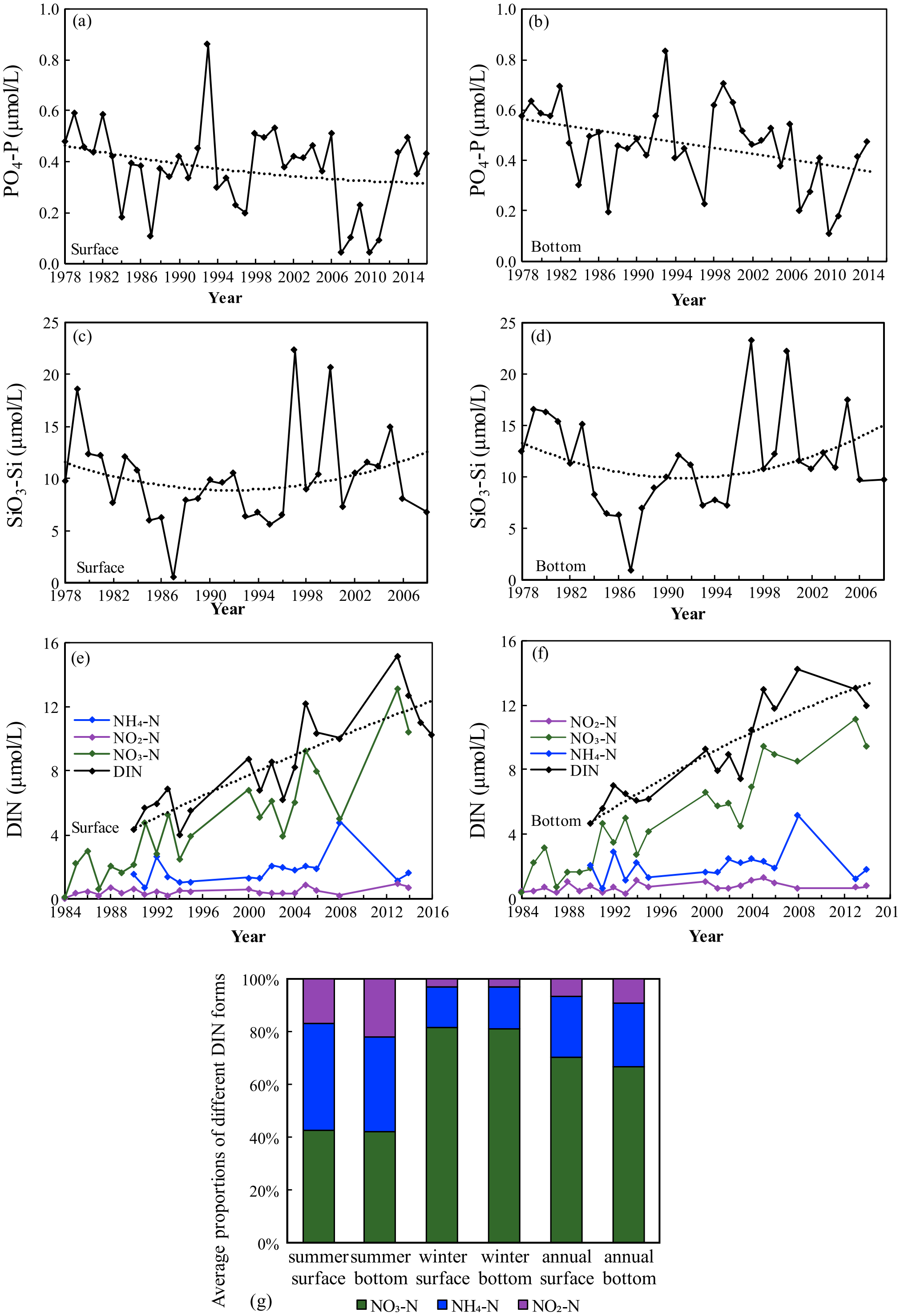

Figure 4. Temporal variations in annual mean nutrient concentrations along the A1–A10 transect in the Bohai Sea from 1978–2016: (a) surface PO_4-P concentration, (b) bottom PO_4-P concentration, (c) surface SiO_3-Si concentration, (d) bottom SiO_3-Si concentration, (e) surface dissolved inorganic nitrogen (DIN) concentration, (f) bottom DIN concentration, and (g) comparison of the average proportions of different DIN forms from 1990 to 2014. The dotted lines represent the variation trend lines determined by linear regression (including simple linear regression and binomial regression).

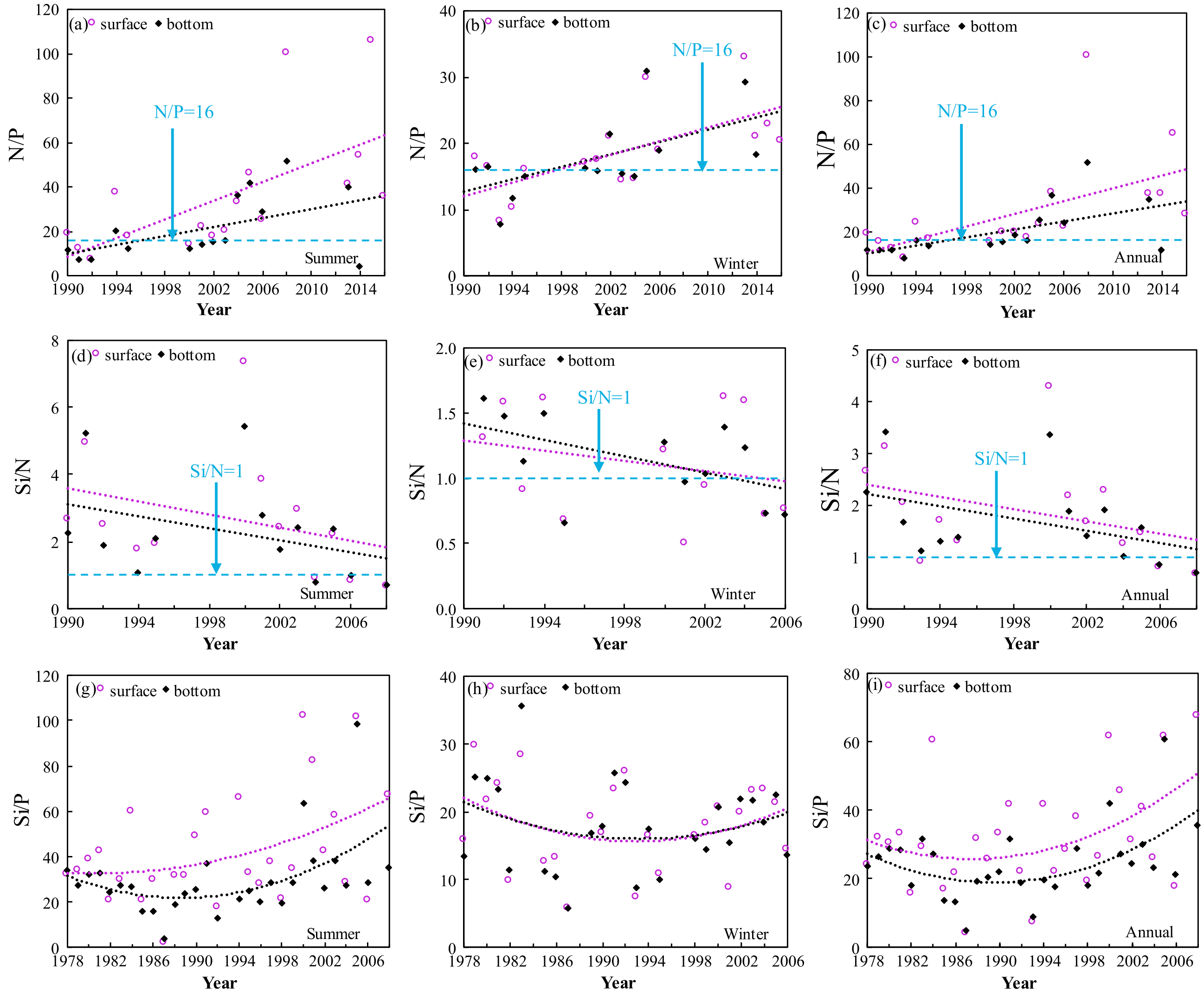

Figure 5. Temporal variations in nutrient ratios along the A1–A10 transect in the Bohai Sea in summers, winters, and throughout the years from 1978–2016: (a) N/P ratio in summer, (b) N/P ratio in winter, (c) annual mean N/P ratio, (d) Si/N ratio in summer, (e) Si/N ratio in winter; (f) annual mean Si/N ratio, (g) Si/P ratio in summer, (h) Si/P ratio in winter, and (i) annual mean Si/P ratio. The dotted lines represent the variation trend lines determined by linear regression (including simple linear regression and binomial regression).

DIN concentration in the Bohai Sea from 1959 to 2016, with a rapid increase after 2002. Similarly, according to the results of by Cui and Song (1996) and Liu et al. (2008), the PO_4-P concentration slightly increased and maintained a high level from 1959 to the early 1980s; based on our results of the PO_4-P variation from 1978 to 2016, we suggest an overall decreasing pattern of the PO_4-P concentration in the Bohai Sea from 1959 to 2016, with a slight increase during the period from 1959 to 1982.

According to the results of the SiO_3-Si variation in the Bohai Sea from Cui and Song (1996), Yu et al. (2000), and Liu et al. (2008) using discontinuous (1959-)1982-1992(-1998) time series and the results from Ning et al. (2010) analyzing the continuous variation from 1978 to 1996, the SiO_3-Si concentration tended to decrease from 1982 to 1998. However, the overall decreasing pattern of SiO_3-Si from 1978 to 1996 observed by Ning et al. (2010) included a decreasing period from 1978 to 1987 and an increasing period from 1978 to 1996, which is roughly consistent with the variation patterns observed by Li et al. (2003) and Jiang et al. (2005) using a 1982-1992-1998 time series and by Zhang et al. (2006) using a continuous time series from 1985 to 1998. In accordance with the decrease in SiO_3-Si from 1959 to 1982 (Liu et al., 2008), the decreasing trend of the SiO_3-Si concentration can be traced back to 1959; based on our results, we suggest that the SiO_3-Si concentration in the Bohai Sea decreased from 1959 to 1987 and increased from 1987 to 2008.

It is likely that the adoption of a discontinuous time series (e.g., the 1982-1992-1998 series in Yu et al., 2000; Li et al., 2003; and Jiang et al., 2005) and the trend over different seasons (e.g., May and October in Yu et al., 2000, and May, August, and October in Jiang et al., 2005) may have resulted in the differences and inaccuracies in some previous studies that analyzed the long-term variations in nutrients in the Bohai Sea. The seasonal differences are also proven in this study. We find that the nutrient concentrations in the Bohai Sea are much lower in summer than in winter (Figures 2 and 3): the PO_4-P concentrations in summer range from [0.040, 0.595] (μmol/L), whereas the concentrations in winter range from [0.114, 1.119] (μmol/L); the SiO_3-Si concentrations in summer range from [0.207, 27.387] (μmol/L), whereas the concentrations in winter range from [0.658, 37.482] (μmol/L); and the DIN concentrations in summer range from [1.183, 14.217] (μmol/L), whereas the concentrations in winter range from [5.275, 21.320] (μmol/L). The variation patterns of nutrients also show seasonal differences (Figures 2 and 3): the PO_4-P concentration in summer decreases from 1978 to 2016, whereas the concentration in winter shows an overall increasing trend; the DIN and NO_3-N concentrations in summer show significant rapid increases after 2002, whereas the increasing patterns in winter tend to be uniform; the increasing trends of the NH_4-N and NO_2-N concentrations in summer are also different from the relatively steady concentration levels in winter. Moreover, the proportions of different DIN forms differ in the different seasons (Figures 2 and 3). The proportion of NH_4-N tends to be higher in summer than in winter, especially from 1990 to 2000, when NH_4-N accounted for the largest proportion of DIN. The proportion of NO_3-N was much higher in winter than in summer and contributed an average of more than 80% of the total DIN during the study period. Therefore, it is strongly recommended that the nutrient conditions should be measured in more than one season in a year, and the data that are measured in different seasons should be averaged to eliminate the seasonal differences to obtain the long-term variation patterns of the nutrients in the Bohai Sea.

Consistent with the results of Yu et al. (2000), Jiang et al. (2005), and Ning et al. (2010), the increasing N/P ratio and decreasing Si/N ratio in our results indicate an evolutionary trend from an earlier nitrogen limitation to the potential phosphorus and silicon limitations from 1978 to 2016 in the Bohai Sea. The Si/P variation from our study roughly corresponds to the interdecadal results of Jiang et al. (2005), who observed a decrease from 1982 to 1992 and a substantial increase from 1992 to 1998. In addition, the increasing trend of the Si/P ratio after 1990, which is similar to the variation in the SiO_3-Si concentration, and the overall decreasing trend of the Si/N ratio are also consistent with the trends observed in the southern Yellow Sea (Wei et al., 2015).

4.2. Main Nutrient Sources

In this section, the long-term variations in riverine inputs, atmospheric deposition, and nonpoint sources under natural changes and anthropogenic impacts are discussed as the main factors (Li, 2010; Mackas & Harrison, 1997) that influence the long-term variations in the nutrients in the Bohai Sea.

4.2.1. The Variations in Riverine Inputs

In coastal and estuarine areas, riverine inputs are likely to account for a majority of the total nutrient sources. Studies have shown that changes in riverine nutrient loads may shift the distribution and composition of nutrients in coastal continental shelf areas worldwide, thus exerting effects on coastal ecosystems (Humborg et al., 1997; Liu et al., 2005; Rabalais et al., 1996; Zhang, 1996).

As the main water body that receives flow from more than 40 rivers, the environmental conditions of the Bohai Sea may be greatly impacted by the discharge from the surrounding rivers. Rivers may play an important role in nutrient transport from land to the semiclosed Bohai Sea. The Yellow River is the second longest river in China and contributes 64.3% of the freshwater discharge from all rivers flowing into the Bohai Sea (Liu, 2006). Thus, the Yellow River was analyzed as the main riverine source of nutrients to the Bohai Sea in this study. In recent decades, there has been a decreasing trend of water and sediment fluxes from rivers into the Bohai Sea due to the impacts of natural changes and human activities, such as the construction of dams and reservoirs and diversion irrigation from mainstreams (Wang, Yang, et al., 2006, 2007). The water discharge and sediment load of the Yellow River from 1978 to 2002 also showed an overall decreasing trend that was probably due to the construction of hydropower stations during this period because the shrinkage in water discharge was particularly sharp after the river closure at Longyangxia at the end of 1979, the reservoir impoundment of the Longyangxia hydropower station in October 1986, and the river closure at the Xiaolangdi hydropower station in October 1997 (Figure 6a). According to the statistics, the drying up days

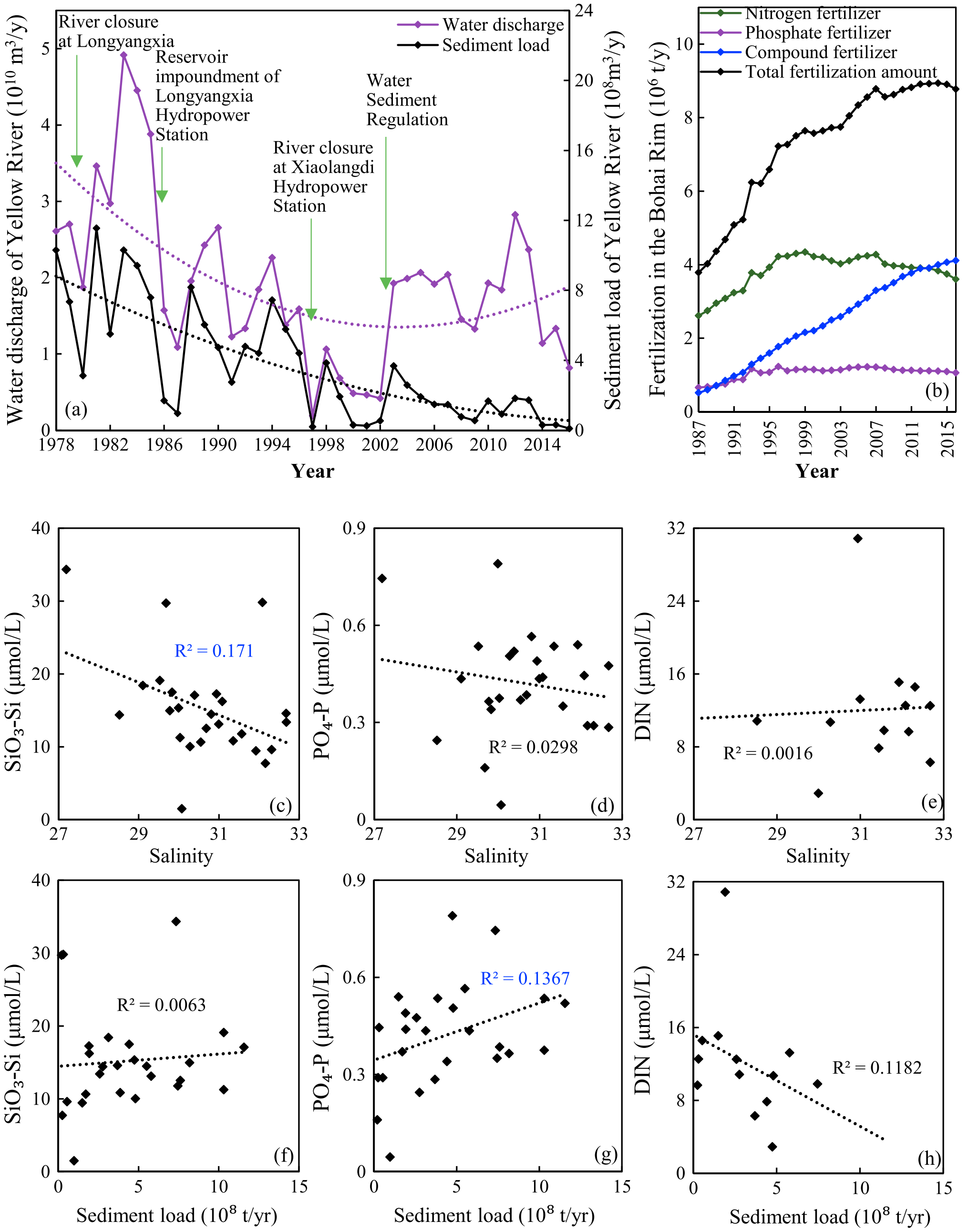

Figure 6. (a). Long-term variations in water discharge and sediment load of the Yellow River at the Lijin station. The data were obtained from the Yellow River Water Resources Commission and Yuan et al. (2006). (b) Long-term variations in fertilization in the Bohai Rim. The data were obtained from the National Bureau of Statistics of the People's Republic of China. (c–e). Plots of the annual mean nutrient concentrations versus salinity at the surface at site A1 near the Yellow River Estuary: (c) SiO_3-Si, (d) PO_4-P, and (e) DIN. (f–h). Plots of the annual mean nutrient concentrations at site A1 versus the annual sediment load of the Yellow River at the Lijin station: (f) SiO_3-Si, (g) PO_4-P, and (h) DIN.

of the Yellow River at the Lijin station reached a maximum of 226 days in 1997 compared to 8 days in 1980 (Yellow River Water Resources Commission). The water discharge and sediment load increased significantly in 2002 when the water sediment regulations came into operation (Figure 6a), and drying up has not occurred in the Yellow River since 2002 (Yellow River Water Resources Commission). From 2003 to 2013, the water discharge of the Yellow River showed an increasing trend, while the sediment load continued to decrease (Figure 6a).

Table 1
Relative Contributions of the Main Nutrient Sources to the Bohai Sea in 2011

	Riverine input			Atmospheric deposition (dry + wet)	Nonpoint sources	
%	Yellow River (excluding particulate nutrients)	Yellow River (particulate nutrients)	Total riverine inputs (excluding particulate nutrients)		Wastewater discharge	Mariculture
DIN	3.93		4.76	25.81	61.23	8.20
PO_4-P	0.12	20.76	0.48	0.22	71.34	7.20
SiO_3-Si	55.17		95.39	4.61		

Note. The relative contributions were approximated as the proportions of the nutrient fluxes from a nutrient source to the nutrient fluxes from all nutrient sources listed. The riverine nutrient fluxes were estimated using the water discharge data of the Yellow River at the Lijin station in 2011, which were obtained from the Yellow River Water Resources Commission, and the nutrient concentrations of the Yellow River at the Lijin station, which were obtained from Chen et al. (2013). The total particulate phosphorus flux was calculated based on the sediment load at the Lijin station from the Yellow River Water Resources Commission and the particulate phosphorus content from He et al. (2010). The total riverine nutrient fluxes (excluding particulate nutrients) were derived from Liu et al. (2011). The DIN flux from atmospheric deposition was calculated from Shou (2017), while the PO_4-P and SiO_3-Si fluxes were calculated from Zhang et al. (2004). The nitrogen and phosphorus fluxes from the wastewater discharge into the Bohai Sea were estimated from the National Bureau of Statistics of the People's Republic of China. The nitrogen and phosphorus fluxes from mariculture emissions were calculated based on the mariculture area in the Bohai Rim from the National Bureau of Statistics of the People's Republic of China and the unit nutrient emissions from Liu (2006).

As the fresh water of the Yellow River flows into the Bohai Sea, it also carries large amounts of sediments and nutrients that significantly change the nutrient concentrations in the estuarine and adjacent sea areas (Liu, 2015; Liu et al., 2012). The Yellow River transports approximately 410×10^9 mol/year of dissolved ions, while its chemical erosion rate is close to the world average (Zhang et al., 1995). Weathering and erosion accelerated by anthropogenic activities, such as farming in recent decades, may be the main reason for the increases in many chemical element fluxes, including silicates (Hu et al., 1982; Tréguer et al., 1995; Zhang et al., 1995, 2015). The cumulative effects of continued global warming and acidification also favor weathering and erosion in drainage basins (Wei et al., 2015). Previous studies have shown that the significant decreases in the water flux and sediment content of the Yellow River before 2000 resulted in a decreasing trend of the SiO_3-Si concentration in the Bohai Sea (Ning et al., 2010; Yu et al., 2000), and the SiO_3-Si from stream discharge may account for up to 80% of the total SiO_3-Si inputs (Liu et al., 2005). In this study, the variation trend of the SiO_3-Si concentration in the Bohai Sea roughly corresponded to the variation in the riverine water discharge from the Yellow River to the Bohai Sea from 1978 to 2008 (Figures 4c, 4d and 6a). In particular, the sharp decreases in the SiO_3-Si concentration in 1987 and 1997 corresponded to the river closure and reservoir impoundment of hydropower stations. Based on the correlation analyses between the nutrient concentrations and salinity near the Yellow River Estuary during our study period (Figures 6c–6e), the SiO_3-Si concentration was observed to have a fine negative correlation coefficient, which indicated that the fresh water from the Yellow River may have an important impact on the SiO_3-Si concentration. Moreover, the relative contribution of SiO_3-Si from the Yellow River was estimated to be more than 50% of the total sources into the Bohai Sea, and the total riverine input was estimated to be more than 95% (Table 1). Therefore, riverine input is suggested to be an important factor for the long-term SiO_3-Si variations in the Bohai Sea.

Compared to other rivers worldwide, the ratio of sediment load to water discharge is very high in the Yellow River (Zhang et al., 1995). Evidence has shown that phosphorus is transported to the ocean primarily in particulate phases via riverine flux (Conley et al., 1995; Guo et al., 2004; Meng et al., 2015). Dissolved PO_4-P adsorbs to particles such as amorphous ferric hydroxides in freshwater and is then released as salinity increases during the process of freshwater–seawater mixing (Carritt & Goodgal, 1954; Lebo, 1990). Evidence has shown that the DIN and PO_4-P fluxes into the Bohai Sea through rivers tended to increase from 1979 to 1990 and decrease from 1990 to the early 2000s (Liu, 2006), which does not entirely correspond to the nutrient variation patterns in the Bohai Sea. In addition, we found the annual mean concentration of PO_4-P in the Bohai Sea roughly matched the overall decreasing trend in the sediment loads of the Yellow River, and the sharp decreases in 1987 and 1997 corresponded to the decreases in the sediment loads of the Yellow River owing to the river closures (Figures 4a and 4b). Moreover, it was observed that the PO_4-P concentration near the Yellow River Estuary and the sediment load were positively correlated (Figure 6g), and the contribution of PO_4-P from the Yellow River increased from 0.12% to 20.88% of the total sources into the Bohai Sea if the particulate contribution was considered (Table 1). Thus, the difference in the estimated riverine PO_4-P flux and the PO_4-P concentration in the Bohai Sea may arise from the substantial release of particulate phosphorus from sediment, which is underestimated; the riverine inputs, especially the internal mechanisms involving sediment transport, could be at least somewhat important to the long-term variations in PO_4-P in the Bohai Sea. However, the PO_4-P fluxes from the Yellow River into the Bohai Sea that were estimated by Liao et al. (2013) showed an increasing trend from 2002 to 2012, which is contrary to the decreasing trend of the sediment load of the Yellow River during the same period. This phenomenon was also observed in the southern Yellow Sea (Wei et al., 2015), and was attributed to the increasingly extensive fertilizer use. The DIN variation in the Bohai Sea did not match the variations in the water discharge and sediment load of the Yellow River (Figures 4e, 4f and 6a), and the DIN concentration near the Yellow River Estuary was also observed to be uncorrelated with salinity (Figure 6e). According to Cui (2008), improper fertilization led to annual losses of 2.41×10^4 t of nitrogen and 0.61×10^4 t of phosphorus in the three provinces and one municipality in the Bohai Rim. Moreover, nutrient losses due to natural soil losses in the amounts of 0.1×10^4 t of nitrogen and 0.08×10^4 t of phosphorus should be added. A significant increasing trend in the compound fertilizer (mainly including nitrogen, phosphorus, and potassium) used in the Bohai Rim was found, while the amounts of nitrogen fertilizer and phosphate fertilizer remained steadily high from 1987 to 2016 (Figure 6b). The large inputs of fertilizer into the drainage basins as well as the increasing trend of total fertilization in recent decades might partially compensate for the deficits of DIN and PO_4-P caused by the decreased river discharge and sediment load as well as the trap effect (i.e., the absorption of nutrients by phytoplankton in the reservoir and sediment precipitation; Wei et al., 2015) and might even promote increases in the DIN and PO_4-P fluxes from rivers.

4.2.2. The Variations in Atmospheric Deposition

In addition to riverine inputs, atmospheric deposition is also proven to be an important pathway of nutrient inputs to seas and may provide from 10% to more than 40% of the nitrogen in N-limited estuarine and coastal waters (Okin et al., 2011; Paerl et al., 2002; Zhang et al., 1999). Globally, nitrogen from atmospheric deposition to the ocean reached 3.5×10^8 t/year (Wollast, 1991). Evidence has shown that atmospheric deposition is likely an important source of DIN to the Bohai Sea (Liu et al., 2008; Zhang et al., 2004). In 1998, 26180 t of nitrogen was input into the Bohai Sea via atmospheric deposition (Cui, 2008). The relative contribution of DIN from atmospheric deposition was estimated to be larger than that from riverine input (Shou et al., 2018). The contribution of DIN from atmospheric deposition was estimated to be 25.81% of the total DIN input into the Bohai Sea in 2011 (Table 1), which indicated that the variation in atmospheric deposition may be somewhat important to the long-term DIN variations in the Bohai Sea.

Windblown dusts play an important role in marine ecosystems once the dusts are deposited and dissolved (Tu et al., 2015). Approximately 63.9% of sand and dust events likely affect seas in China (Zhang et al., 2005), and the deposition fluxes of particulate phosphorus during the dust storm events associated with precipitation were approximately 500–1,000 times greater than the daily average flux during nondust days in the Yellow Sea (Shi et al., 2013). Dusts moving from the Loess Plateau and Mongolian Plateau were observed to be deposited into the Bohai Sea (Zhang et al., 2005). In 1998, 770 t of phosphorus was input into the Bohai Sea via atmospheric deposition (Cui, 2008). Tianjin is one of the major cities on the periphery of the Bohai Sea. Therefore, the long-term variations in the trends of dust and precipitation in this region can partially reflect the effects of dust events and wet deposition on the Bohai Sea. A study on the dust in this region indicated that the number of days with dust weather in Tianjin decreased from the mid-1960s to 1999, with an abrupt increase in the early 2000s (Duan et al., 2005). The annual frequencies of sand and dust weather in northern China showed a decreasing trend from 2000 to 2017 (Figure 7a), which was consistent with the decreasing trend of the PO_4-P concentration in the Bohai Sea during the same period (Figure 4). However, the contribution of PO_4-P from atmospheric deposition was very limited compared with the contributions from rivers and nonpoint sources (Table 1), which indicated that atmospheric deposition may not be the main factor determining the long-term variation pattern of the PO_4-P in the Bohai Sea. Observational data showed that the overall precipitation in the Tianjin region exhibited a decreasing trend before the late 1990s and a gradually increasing trend from the late 1990s to 2015 (Figure 7a), which roughly corresponded to the long-term variations in the SiO_3-Si concentration in the Bohai Sea (Figure 4). Although the SiO_3-Si flux directly from atmospheric deposition to the Bohai Sea was estimated to be only 4.61% of the total input flux (Table 1), the long-term variations in the annual precipitation affect the Earth surface processes around the sea that are related to wet deposition, and large-scale precipitation may promote water and soil losses

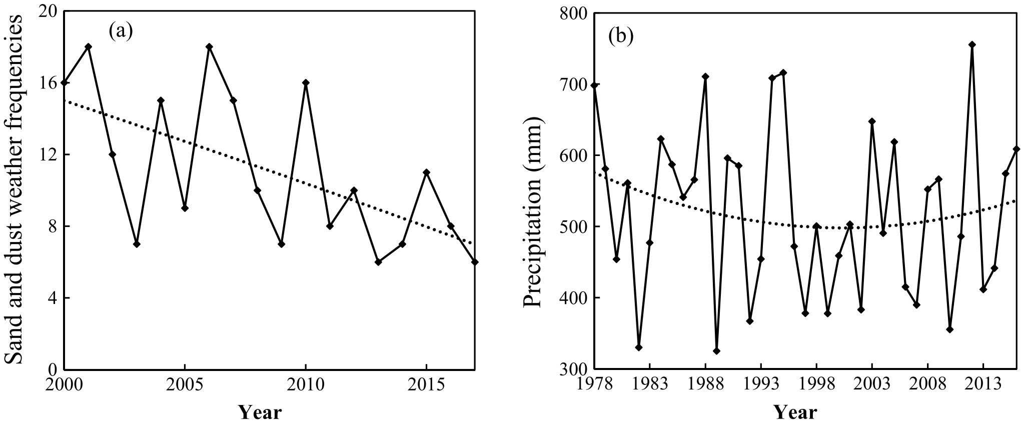

Figure 7. (a) Long-term variations in the annual sand and dust weather frequencies in northern China. The data were obtained from the China Meteorological Administration, National Climate Committee, China Climate Bulletin. (b) Long-term variations in the annual precipitation in Tianjin. The data were obtained from the National Bureau of Statistics of the People's Republic of China, China Statistical Yearbook and Li et al. (2010).

(Wei et al., 2015). Terrestrial SiO_3-Si induced by the scouring and erosion effects of rainfall on the land surface may enter the ocean, thereby increasing the nutrient concentrations in the ocean (Wei et al., 2015).

4.2.3. The Variations in Nonpoint Sources

In addition to the above sources, the variations in nonpoint nutrient sources should also be considered, such as the increasing discharge of industrial, agricultural, and domestic wastewater and the expanding area of mariculture. With the intensification of human activities, a large amount of nutrients have been input into the sea, and the anthropogenic inputs of nutrients will alter the concentrations and composition ratios of nutrients in the sea. These changes will greatly impact the absorption and utilization of nutrients by different phytoplankton species in the local ecosystem.

According to the investigation of nonpoint sources of pollution from land to sea in 2017, 9,600 nonpoint pollution sources (drain outlets) were found along the coast of China (http://www.soa.gov.cn/xw/ztbd/ztbd_2017/2017wthzxdc/mtbd/201801/t20180118_60021.html), which indicates that there was a source every 2 km along the coastline on average. The number of pollution sources along the coastline of the Bohai Sea was estimated to be 640. From 1985 to 2015, the total wastewater discharge continuously increased, the domestic sewage discharge showed the same increasing trend, and the industrial wastewater discharge was maintained at approximately 4×10^9 t with no significant variations (Figure 8a). The study by Xu et al. (2010) indicated the significant influences of sewage disposal on the nutrients in the Bohai Sea. Evidence has shown that wastewater transport leads to increased concentrations of ammonium and phosphate in the surface waters of the Bohai Sea, and the phosphate from wastewater accounts for 31% of the total inputs, whereas the ammonium from wastewater accounts for 84% of the total inputs (Liu et al., 2005). The variations in the trend of the total wastewater discharge induced by increased domestic sewage corresponded to the long-term trend of the DIN concentration in the Bohai Sea (Figures 8a and 4), and the contribution of nitrogen from wastewater was estimated to be 61.23% of the total sources into the Bohai Sea (Table 1). Therefore, it is indicated that the dense population and high volumes of domestic sewage discharge are likely important factors accounting for the rapidly increased DIN concentration in the Bohai Sea.

Expansion of the aquaculture industry has resulted in increasing nutrients in its effluents (Boyd et al., 2007). Fish or other cultured species retain only approximately 20–30% of the nitrogen from their feed and a smaller fraction of phosphorus, while the remaining nutrients are released into the water as dissolved and solid waste (Krom & Neori, 1989; Lupatsch & Kissil, 1998). The contributions of nitrogen and phosphorus from mariculture were estimated to be 8.20% and 7.20% of the total sources into the Bohai Sea, respectively (Table 1). The mariculture area in the Bohai Sea increased from 1979 to 2016 (Figure 8b), which also corresponded to the long-term variations in the DIN concentration in the Bohai Sea. Evidence has shown that aquaculture has a greater influence on nitrogen nutrients than on phosphate nutrients (Kang & Xu, 2016). Therefore, the continuously expanded area used for mariculture in the Bohai Sea could contribute to the increase in the inputs of DIN nutrients.

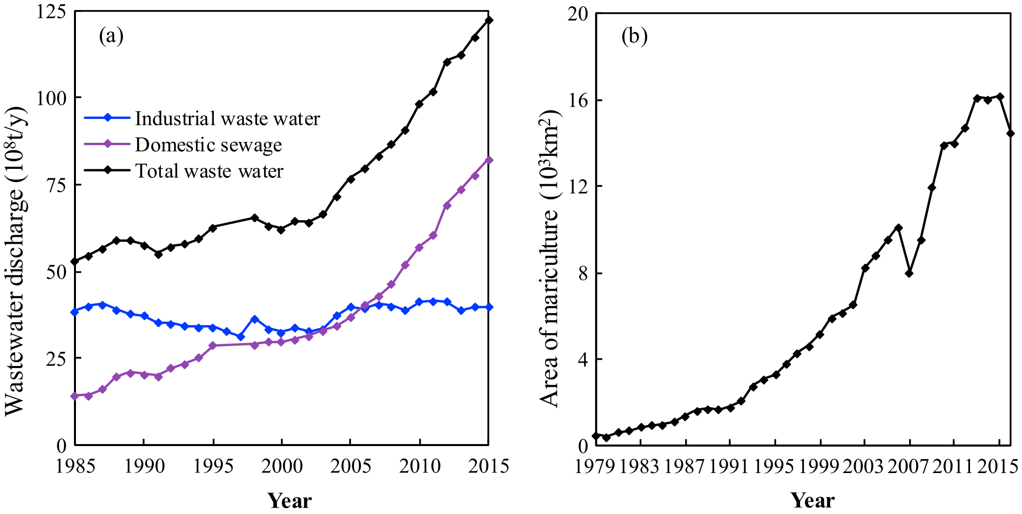

Figure 8. (a). Long-term variations in wastewater discharge into the Bohai Sea. The data were obtained from the State Oceanic Administration, China Marine Statistical Yearbook. (b). Long-term variations in the mariculture area in the Bohai Sea. The data were obtained from the Bureau of Fisheries of the Ministry of Agriculture, China Fishery Statistical Yearbook.

4.3. Potential Effects on Ecological Disasters

The reasons for the occurrence of ecological disasters in the Bohai Sea may be complex. An increasing trend of chlorophyll *a* content from 2002 to 2006 was believed to be caused by increased nutrient concentrations (Xu et al., 2010). As mentioned in section 3.1, the surface nutrient concentrations were significantly lower than the bottom concentrations in summer (Figure 2). During this period, nutrients at the surface of the Bohai Sea were substantially utilized by phytoplankton that were in vigorous growth stages (Cui et al., 1994; Zhou et al., 2017). However, in winter, this phenomenon was mitigated, and there were much smaller differences between the surface and bottom concentrations of nutrients (Figure 3).

Similarly, the increase in red tides is likely a response to the change in nutrients. The frequency and area of red tide events in the Bohai Sea tended to increase after 1989 and reached an explosive phase after 1998 when the affected area significantly expanded and the frequency greatly increased (Figure 9a). From 1990 to 1995, the low number of red tide outbreaks in the Bohai Sea matched the low DIN concentrations and N/P ratios during the same period (Figure 4). In addition, during the explosive phase of red tides in the Bohai Sea, the dominant species also changed from single Pyrrophyta species to a more diverse composition, including Pyrrophyta, Bacillariophyta, Rhaphidophyta, Chrysophyta, Cryptophyta, and Cyanophyta (Figure 9a), and the percentage of toxic algal blooms also increased (State Oceanic Administration, People's Republic of China, Chinese Marine Disaster Bulletins). During this phase, the DIN concentration increased significantly, and the SiO_3-Si concentration increased slightly, while the PO_4-P concentration decreased (Figure 4). Notably, the variations in the Si/N ratio during the period from 1990 to 2008 roughly matched the variations in the frequency, area, and species diversity of red tides during this period (Figures 5f and 9a): the decreasing Si/N ratio in the early 1990s corresponded to lower red tide frequencies, and the decreasing Si/N ratio in the 2000s also corresponded to lower red tide frequencies and lower dominant species diversities accompanied by lower ranks of Bacillariophyta as well as larger red tide areas. In addition, in the years when red tide occurred, the DIN concentrations and the N/P and Si/N ratios tended to be higher than those in the years without red tide occurrences; for the SiO_3-Si and PO_4-P concentrations, the differences were not prominent (Figures 9b and 9c). Analysis of the potential relationships between the red tides and nutrients in the Bohai Sea (Figures 9d–9m) revealed that the red tide area and frequency tended to increase when the DIN and PO_4-P concentrations increased. When the SiO_3-Si concentration was lower than approximately 10 μmol/L, the red tide area and frequency tended to increase as SiO_3-Si increased; when the concentration was greater than 10 μmol/L, decreasing trends of the red tide area and frequency were observed. When the N/P ratio was lower than approximately 30, the red tide area and frequency tended to increase as the N/P

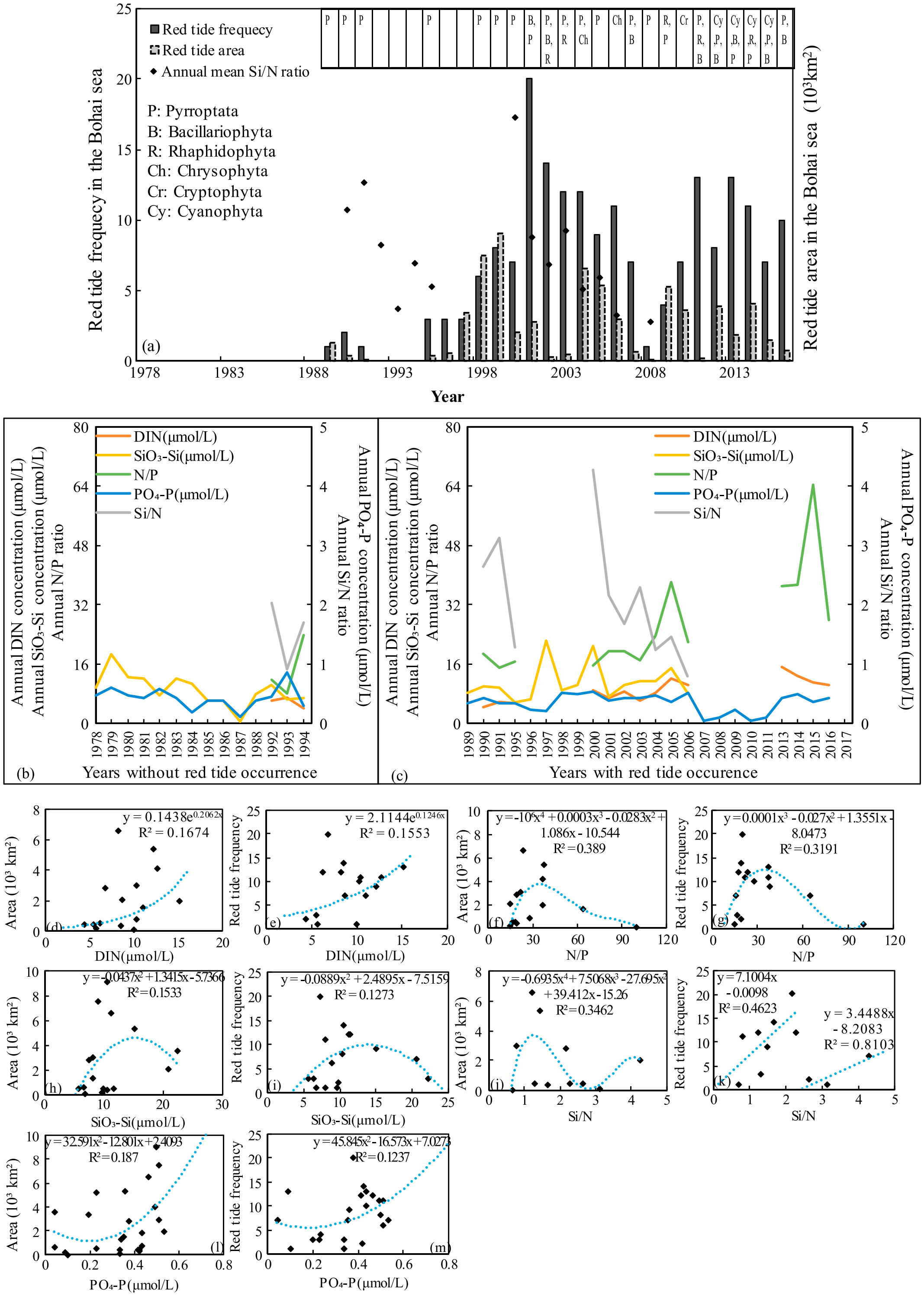

Figure 9. (a). Long-term variations in the red tide frequency and area in the Bohai Sea. The data were obtained from the China Marine Statistical Yearbook and State Oceanic Administration, People's Republic of China, Chinese Marine Disaster Bulletins. The Si/N ratios in the figure are 4 times the real values to provide a better comparison. The dominant species listed in the figure are listed in the order of their affected areas in a year. (b and c). Nutrient concentrations and ratios in years (b) without red tide occurrence and (c) with red tide occurrence in the Bohai Sea. (d-m). The relationships between red tides and nutrients in the Bohai Sea: (d) red tide area versus DIN concentration, (e) red tide frequency versus DIN concentration, (f) red tide area versus N/P ratio, (g) red tide frequency versus N/P ratio, (h) red tide area versus SiO_3-Si concentration, (i) red tide frequency versus SiO_3-Si concentration, (j) red tide area versus Si/N ratio, (k) red tide frequency versus Si/N ratio, (l) red tide area versus PO_4-P concentration, and (m) red tide frequency versus PO_4-P concentration. The dotted blue lines in the figures represent fitting curves.

ratio increased; when the ratio was greater than 30, the red tide area and frequency decreased as the N/P ratio increased. The red tide frequency showed increasing trends as the Si/N ratio increased in the ranges of (0, 2.5) and (2.5, 5), and the area showed a potential decreasing trend as the Si/N ratio increased. Generally, under nonlimiting conditions, diatoms are fast-growing species and have a higher growth rate than flagellates (Banse, 1982). High nutrient levels and sufficient silicon supplies will promote the preferential growth of diatoms (Wang, Qi, et al., 2006), and Pyrrophyta such as *Noctiluca scintillans* will then feed on the enhanced diatoms; during these blooms, substantial amounts of nutrients are consumed (Feng, 2004; Yin et al., 2013). As the nitrogen concentration was generally in excess due to increasing anthropogenic impacts in recent decades, the N/P ratio tended to increase and phosphorus limitation was likely to occur, especially after the consumption of phosphorus by phytoplankton, and during this phase, the proportion of diatoms may decrease by up to 50% (Fu et al., 2012). The increasing N/P ratio in the Bohai Sea during our study period, which was higher than the Redfield ratio after the 2000s, and the decreasing Si/N ratio, which was lower than the Redfield ratio after 2003 (Figure 5), as well as the variations in nutrient concentrations (Figure 4), indicated an evolutionary trend from an early nitrogen limitation to the current potential phosphorus and silicon limitations in the Bohai Sea. This trend may limit the growth of diatoms and promote the development of Pyrrophyta in the Bohai Sea if other conditions are suitable (Yu et al., 2000). It is therefore suggested that the changes in nutrient concentrations and nutrient limitations may be a potential factor accounting for the increasing occurrence of red tides (in terms of area, frequency, and diversity of dominant species) in the Bohai Sea.

5. Conclusions

Nutrients are very important to marine ecosystems as they support the growth of phytoplankton. Due to the accelerated development of agriculture and industry over the past four decades, nutrient inputs from land have experienced significant changes in China, which has resulted in variations in the concentrations and compositions of nutrients in marginal seas. As the only enclosed sea in China, the Bohai Sea is a relatively closed environment that is particularly vulnerable to human activities. In this paper, the long-term variation patterns of nutrients and their compositions in the Bohai Sea, as well as their potential influencing factors, were analyzed based on historical summer and winter data from 1978 to 2016.

The results showed that in summer, the DIN concentration in the Bohai Sea continuously increased from 1990 and showed a more rapid increase after 2002. The PO_4-P concentration exhibited a decreasing trend from 1978 to 2016. The SiO_3-Si concentration decreased from 1978 to 1987 and increased from 1987 to 2008. In winter, the PO_4-P concentration fluctuated with a slight increasing trend, while the concentrations of SiO_3-Si and DIN exhibited increasing trends. The variations in the NO_3-N concentrations were consistent with the variations in the DIN concentrations in summer and winter, while the NH_4-N and NO_2-N concentrations increased in summer but fluctuated over relatively steady ranges in winter. For the vertical difference, the nutrient concentrations were higher at the bottom than at the surface in summer, whereas the difference was not significant in winter. In summer and winter, the N/P ratio increased with a rapid increase after 2002, the Si/N ratio decreased, and the Si/P ratio showed an overall slightly increasing trend. The bottom ratios tended to be lower than the surface ratios in summer, whereas the vertical differences were not prominent in winter. The nutrient ratios were much lower in winter than in summer. The nutrient limitation changed from nitrogen limitation to phosphorus and silicon limitations, which might influence the uptake and utilization of nutrients by phytoplankton in the Bohai Sea. Based on the results of previous studies from 1959 and the results of the present study from 1978 to 2016, there was an overall increase in the annual mean concentration of DIN from 1959 to 2016, an overall decrease in the annual mean concentration of PO_4-P from 1978 to 2016, a decrease in the annual mean concentration of SiO_3-Si from 1959 to 1987, and an increase in the SiO_3-Si concentrations from 1987 to 2008. The dominant factors determining the long-term variations in the DIN concentrations in the Bohai Sea could be atmospheric deposition and nonpoint sources due to various human activities; the primary factors determining the variations in the PO_4-P concentration could be the variations in riverine inputs and nonpoint sources related to natural changes and human activities; riverine inputs have the greatest influence on the variations in the SiO_3-Si concentration. The long-term nutrient variations in the Bohai Sea have potential ecological impacts on the frequencies, affected areas, and diversity of the dominant species in the local red tide blooms.

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