NUTRIENT DISTRIBUTIONS IN THE EAST CHINA SEA AND CHANGES OVER THE LAST 25 YEARS

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Abstract. A survey of seasonal variations in the distribution of nutrients along a standard section of the East China Sea was carried out between 1991 and 2015. Here, interannual variations of five key nutrients are analyzed and discussed in the context of possible controlling factors, with special attention given to seasonal variations recorded over the year 2015. Results indicate that concentrations of dissolved inorganic nitrogen (DIN), phosphate (DIP) and silicate (DSi) exhibit gradual increases over this period. Measured concentrations of DIN, DIP and DSi were 5.34–11.87 μmol/L, 0.31–0.53 μmol/L and 11.12-23.15 μmol/L, respectively. Since 1996, concentrations of DIN, DIP and DSi have increased significantly. The annual average concentration of DIN , DIP and DSi in the East China Sea were ~2 times greater in 2015 than in 1991, respectively. The N/P ratio exhibits an increasing trend followed by steady-state asymmetric M-type fluctuations. Conversely, the Si/N ratio is generally stable and exhibits an asymmetric W-type fluctuation. The distribution of nutrients in the East China Sea exhibits clear seasonal variations, with phosphate and inorganic nitrogen contents generally higher in autumn and winter than in spring and summer. Silicate content is higher in summer and winter than in autumn and spring. The main source of nutrients into the East China Sea is the Yangtze River plume, with the Taiwan Warm Current providing an additional, secondary source. The combined effects of the Yangtze River plume and the Taiwan Warm Current enforce a decreasing trend in nutrient concentrations from west to east.

Keywords: *inorganic nitrogen, phosphate, silicate, interannual change, seasonal variation*

Introduction

The East China Sea is bordered by the Yangtze River estuary in the west, the Pacific Ocean in the east, the Taiwan Strait in the south and the Yellow Sea in the north. The boundary between the East China Sea and the Yellow Sea is defined by the line between the northern end of the Yangtze River estuary and the southernmost point of Jeju Island. The eastern boundary of the East China Sea with the Pacific Ocean is defined by the Ryukyu Islands. The East China Sea is fed by more than 40 rivers exceeding 100 kilometers in length (Yanzgi et al., 1993; Chu et al., 2005). These rivers add nutrients to the East China Sea which, in turn, supports one of the most highly productive marine ecosystems in the world (Li and Dag, 2004). As a

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result, the distribution and variations in nutrient concentrations in the East China Sea is a topic of great importance that has inspired numerous scientific investigations.

For example, Wang et al. (2008) studied the three-dimensional distribution and seasonal variation of nutrients in the East China Sea. Wang et al. (2019) then improved on these studies by incorporating a three-dimensional low-trophic ecosystem model that revaluated budgets of nutrients and biogenic particles (phytoplankton and detritus) in the East China Sea and found that export of biogenic particles through the Tsushima Strait is greater than that through the continental shelf break.

Based on surveys of nutrient concentrations along three sections in the East China Sea, Cao et al. (2014) compared profiles collected during the relatively wet period of 1989–2010 to more typical profiles along the same sections to identify long-term changes in the concentrations and distributions of inorganic nitrogen, active phosphate and silicate. To evaluate the contribution of each source of nutrients to the nutrient inventory and primary production over the continental shelf, Zhang et al. (2019) tracked the behavior of all of the state variables incorporated into a low-trophic ecosystem model. Using this technique, Zhang et al. (2019) discovered strong seasonal variations in the spatial distribution of nutrient sources attributable to seawater circulation, mixing, and stratification. The ecological effect of these nutrients is in turn affected by the level of nutrient limitation, light availability and water temperature at the locations of each of these sources (Zhang et al., 2019). Mi et al. (2011) used the survey data collected between April and August, 2011, to analyze and discuss the distribution of nutrients in the East China Sea and adjacent Yellow Sea over the spring and summer seasons, as well as various influencing factors. Based on survey data collected over four voyages conducted in the East China Sea (120°–128°E, 25°–33°N) over the spring, summer, autumn and winter of 2013, Ye et al. (2015) analyzed the spatiotemporal distribution and structure of nutrients in the East China Sea and discussed limitations to the growth of phytoplankton.

Excess nutrients can lead to pollution of water in the East China Sea. Moreover, the concentrations and distributions of these nutrients vary from year to year. Therefore, to promote the rational exploitation and utilization of the marine resources in the East China Sea it is important to generate accurate and timely understandings of the distribution, spatiotemporal variations and influencing factors of nutrients in the East China Sea.

In this study, we analyze the results of nutrient monitoring surveys conducted between 1991 and 2015 and evaluate interannual trends and variations in nutrient concentrations and distributions. Additional survey data collected from 30 stations in the East China Sea in the spring (May), summer (August), autumn (November) and winter (February) of 2015 to determine the distribution of nutrients in East China Sea and possible influencing factors are likewise discussed and analyzed. The goal of this study is to systematically investigate the distribution of nutrients in the East China Sea and to identify seasonal, interannual and long-term variations in the distributions of these nutrients over the past 25 years, thereby providing fundamental data by which to advance scientific study of the East China Sea and to promote rational development and utilization of marine resources in this region.

Materials and methods

Sample collection

Samples were collected during a series of four oceanographic research cruises conducted by the Ningbo Marine Environmental Monitoring Center along standard sections of the East China Sea (27–33°N, 121–128°E), China, between 1991 and 2015. The total of 30 survey stations are arranged into 3 oceanographic sections supplemented by four auxiliary stations. A map of the relevant survey stations is presented in *Fig. 1*.

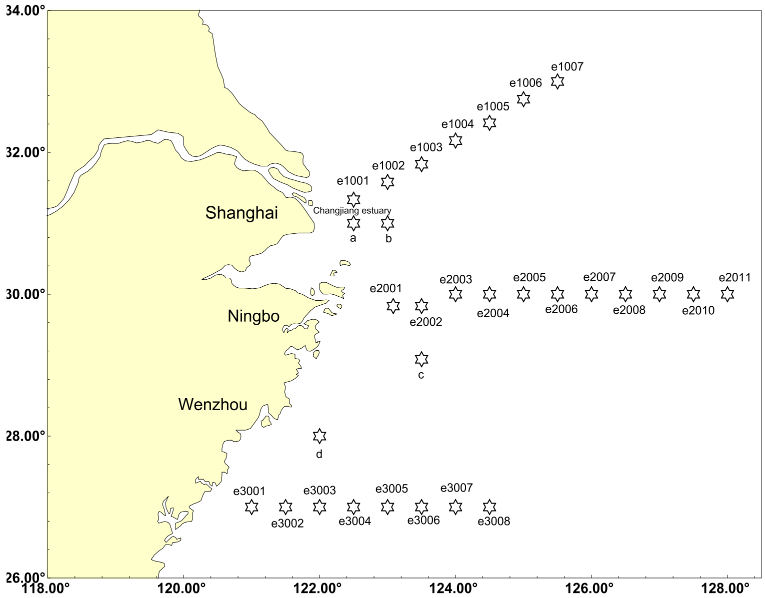

***Figure 1.** Sampling sites in the East China Sea*

Sampling and analysis

Full water-depth sampling of seawater was conducted using a CTD self-capacitive sampler (Seabird, USA). Samples were filtered using a HCl acid-washed neutral pH cellulose acetate fiber filtration membrane (0.45 μm). After filtration, nutrient assays were conducted using the methods outlined in the fourth volume of the marine survey code (GB17378.4-2007): the investigation of the chemical elements in sea water (Xu et al., 2005). The analytical methods and detection limits for dissolved inorganic phosphate (DIP), nitrite (NO_2-N), nitrate (NO_3-N), ammonium salt (NH_4-N) and silicate (DSi) are presented in *Table 1*. Dissolved inorganic nitrogen is the sum of the nitrate, nitrite and ammonia nitrogen components (*i.e.*, DIN =NO_3-N+NO_2-N+NH_4-N). Data interpolations between sampling locations, statistical data processing and visualization were carried out using the Surfer software (Golden Software, USA).

***Table 1.** Analytical methods by component*

Project	NO_2-N	NO_3-N	NH_4-N	DIP	DSi
Analysis method	Naphthalene ethylenediamine spectrophotometric method	Zinc and cadmium reduction	Hypobromate oxidation	Phospho molybdenum blue method	Silicon molybdenum yellow method
Instruments	723C visible spectrophotometer				

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Data processing

Long-term trends in nutrient concentrations were evaluated by performing a linear regression on monitoring results obtained in August and calculating the Pearson correlation coefficient, R. (*Eq. 1*)

This coefficient was defined as the correlation coefficient between the time series {X} and the integers {n}, i=1,2,3,...,n. In this study, n is the total span of years of the data. The coefficient was computed from the following equation:

$$R = \frac{\sum (x-\bar{x})\cdot(y-\bar{y})}{\sqrt{\sum (x-\bar{x})^2 \cdot \sum (y-\bar{y})^2}} \quad \text{(Eq.1)}$$

where y=(n+1)/2. Its significance level is determined from the Student's t-test. A positive (negative) value of *R* indicates that the time series {X} has a linear positive (negative) trend (Liu et al., 2005).

Results and discussion

Interannual variations in nutrient concentrations

Analyses of dissolved nutrient concentrations indicate that concentrations of DIN in the East China Sea gradually increased over the study period, from 5.34 μmol/L in 1991 to 11.87 μmol/L in 2015 (*Fig. 2*). Concentrations of DIP likewise gradually increased, from 0.31 μmol/L in 1991 to 0.49 μmol/L in 2015 (*Fig. 3*). Concentrations of DSi exhibit high levels of interannual variability (*Fig. 4*). However, with the exception of a large increase in 1999, the trend is generally increasing, from 11.12 μmol/L in 1991 to 23.15 μmol/L in 2015. Since 1996, the average concentration of DIN, DIP and DSi in the East China Sea have increased significantly. The annual average concentration of DIN in the East China Sea in 2015 was ~2 times greater than that measured in 1991, the annual average concentration of DIP increased ~1.6 times, and the annual average concentration of DSi increased ~2 times.

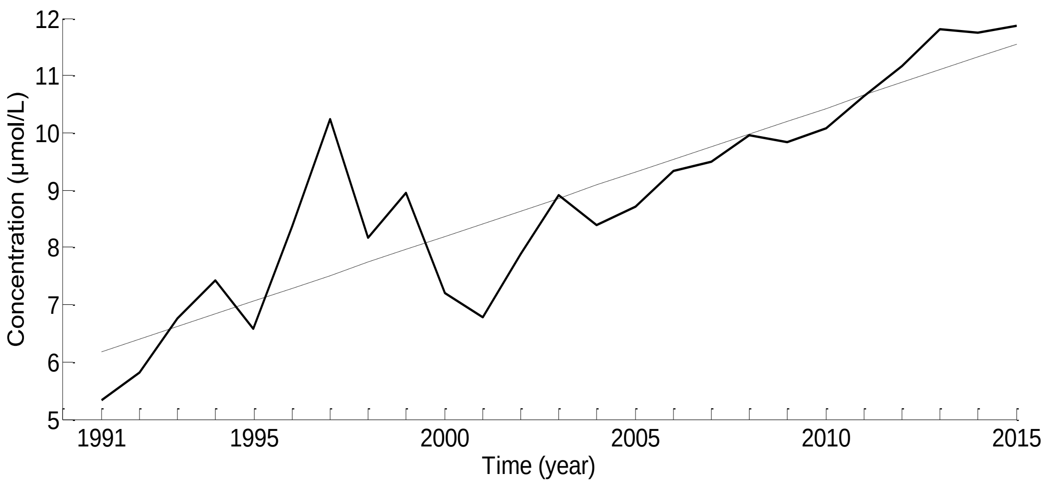

Figure 2. *Interannual variations and long-term trends in DIN concentrations in the East China Sea*

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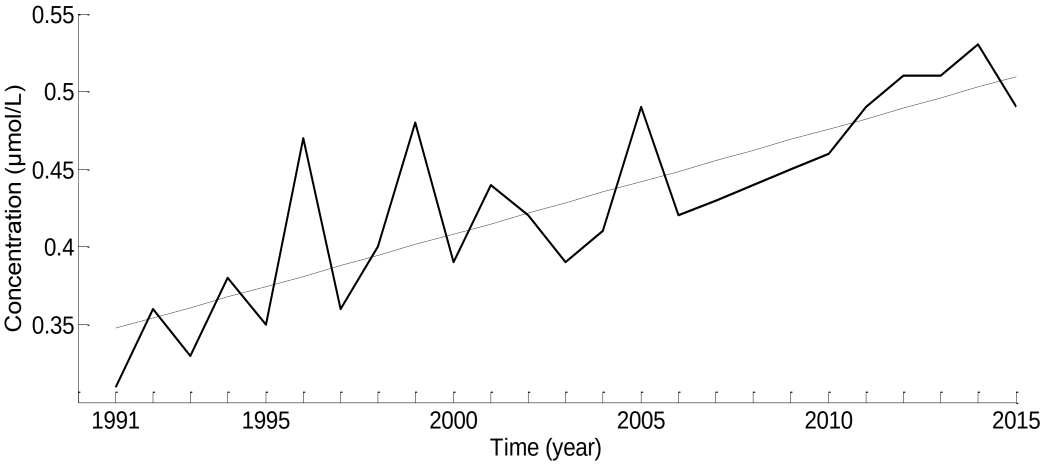

Figure 3. *Interannual variations and long-term trends in DIP concentrations in the East China Sea*

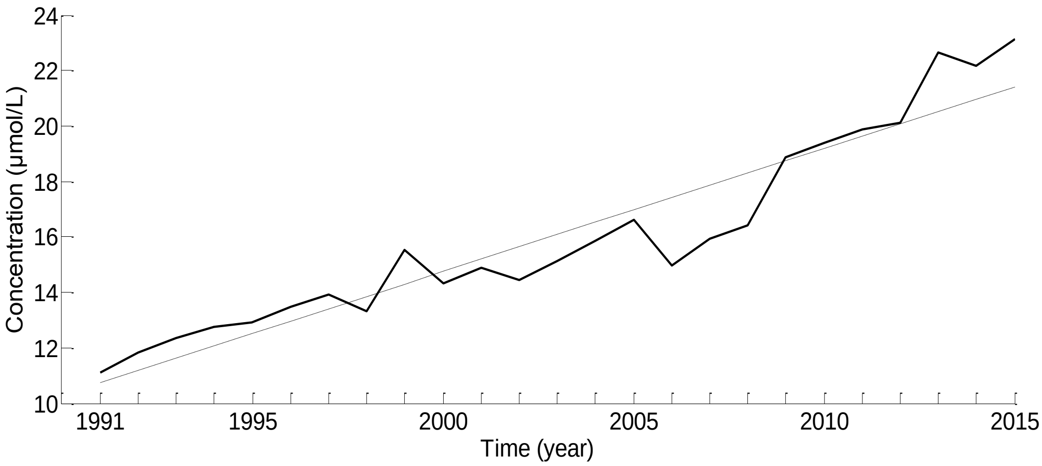

Figure 4. *Interannual variations and long-term trends in DSi concentrations in the East China Sea*

The nutrient content of samples collected at Datong Station on the Yangtze River exhibit a similar historical change to that of the East China Sea (Li et al., 2007), suggesting that annual variations in DIN and DIP in the East China Sea are synchronized with those of the Yangtze River estuary (Chen et al., 2011). As a result, we propose that the increase of DIN and DIP in the East China Sea is primarily attributable to runoff from the Yangtze River. Since 1996, the concentration of DIN and DIP in the East China Sea has increased significantly (Cao et al., 2014). Bing et al. (2019) analyzed velocity and nutrient data collected from satellite images and *in situ* measurements between 1993 and 2014 to study spatiotemporal variations in cross-shelf nutrient exchanges at the 200 m isobath. Average integrated nitrate transport (FQ) over this 22-year period is 22.0±12.4 kmol/s (Bing et al., 2019). Furthermore, the on- and off-shelf components of FQ are roughly twice that of FQ itself, such that variations in FQ over time are primarily determined by the on-shelf component (Bing et al., 2019).

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While interannual variations in FQ are greater than seasonal variations, no persistent interannual signal was identified (Bing et al., 2019). Our results show that this increase is also in line with a longer term gradual trend, which would suggest that these increases in nutrient concentrations are caused by long-term environmental changes. For example, the development of agriculture and the establishment of chemical industrial facilities along the coast and along the Yangtze River would tend to increase the input of nutrients from land sources, including runoff from the Yangtze River. Acting in contrast to this trend, the expansion of sewage processing facilities would tend to decrease the amount of coastal industrial pollutants and related inorganic nutrient directly discharged into the sea. Processes within the ocean, such as physical dilution by oceanic seawater and the absorption of nutrients by marine phytoplankton would also serve to modify nutrient concentrations in the East China Sea (Luo, 2006).

As concentrations of DIN, DIP and DSi change, derivative ratios such as N/P and Si/N also change (Li et al., 2007). It is generally considered that the proportion of C: Si: N: P suitable for diatom growth is 106: 16: 16: 1 (Xia et al., 2007), and that changes in nutrient concentrations affect the cell size and community composition of phytoplankton (Pätsch et al., 1997; Egge et al., 1998). Between 1991 and 2015, the N/P ratio of nutrients in the East China Sea increased gradually and then stabilized after 2009, exhibiting an asymmetric M-type pattern (*Fig. 5*). The maximum and minimum values of N/P appeared in 1997 and 2001, respectively, and their values were 28.44 and 15.41. Between 1991 and 2009, the average Si/N ratio in the East China Sea exhibited an asymmetric W-type fluctuation (*Fig. 6*). The maximum and minimum values of Si/N appeared in 2001 and 1997, respectively, and their values were 2.20 and 1.36. After 2009, the Si/N ratio gradually stabilized between 1.80 and 1.95.

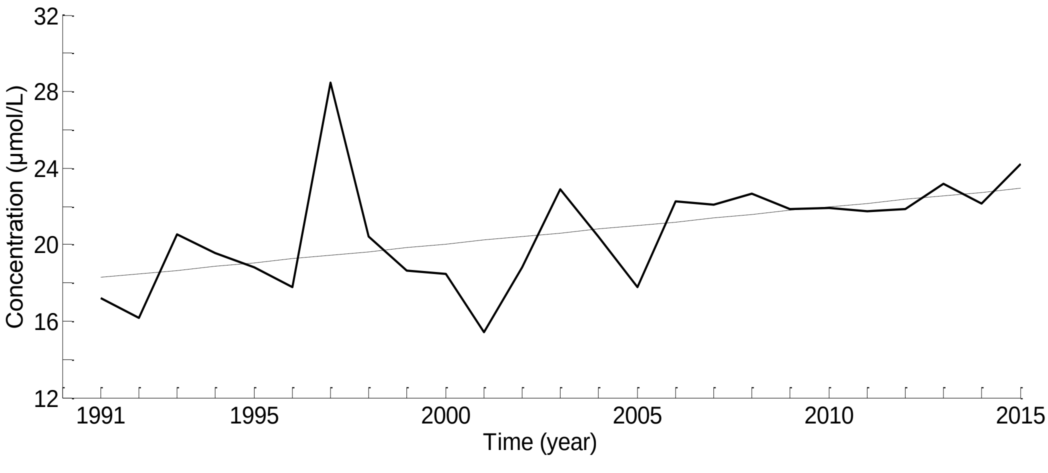

Figure 5. *Interannual variations and long-term trends in the N/P ratio in the East China Sea*

A large number of studies have shown that the value of N/P is closely related to the growth of phytoplankton (e.g., Chen et al., 2011). When plankton masses proliferate, although nitrogen and phosphorus are consumed in large quantities, inorganic nitrogen still maintains a high concentration. However, the proportion of N/P in the East China Sea was basically maintained at 21.93 ± 6.51. This ratio is only slightly higher than that reported by Redfield (1958) and is largely attributable to the high concentration of inorganic nitrogen.

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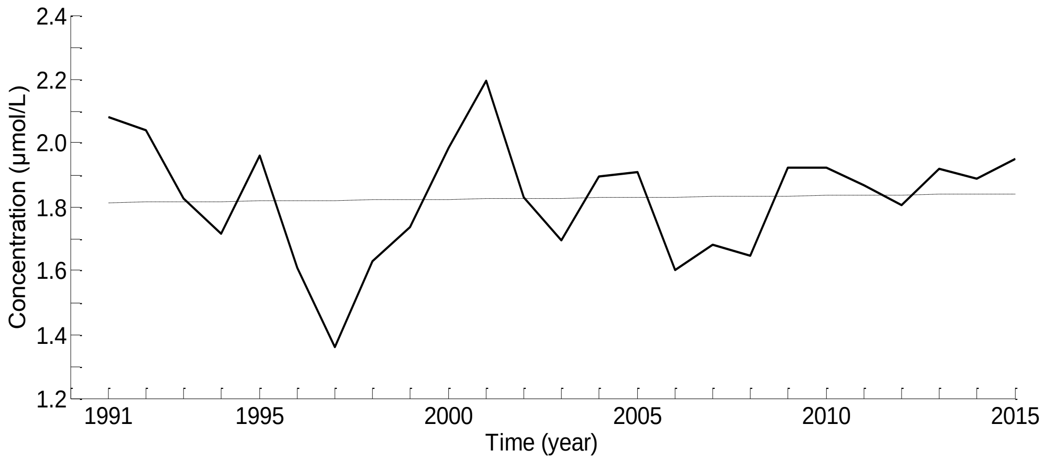

Figure 6. *Interannual variations and long-term trends in the Si/N ratio in the East China Sea*

The Si/N ratio in East China Sea is greater than 1. Therefore, DSi is not a limiting factor in the growth of diatoms. In recent years, changes in sediment transport have had a direct effect on the flux of dissolved silicate to the ocean (Humborg et al., 1997). However, our data show that despite high levels of interannual variation, the Si/N ratio of the East China Sea has been generally stable. Over the past twenty-five years, the N/P and Si/N ratios of seawater in the East China Sea show a trend of roundabout rise, and all tend to be stable. The interannual variation of nutrients in the East China Sea is becoming much more stable, which may be related to changes the runoff, sediment transport and the structure of N and P use in chemical fertilizers (Chen et al., 2011). This apparent stabilization is closely related to the relevant policies of the total pollution control of the marine environmental and the protection of the marine environment.

Distribution of nutrients in East China Sea District surface waters

Figure 7 shows the planar distribution of phosphate, inorganic nitrogen, and silicate in the surface waters based on samples collected during the four research cruises. It can be seen from the figure that high surface concentrations of phosphate, inorganic nitrogen, and silicate in the East China Sea in the spring (May), summer (August), autumn (November) and winter (February) of 2015 appear in the waters of the Yangtze River estuary, and expand to form dense isolines. As the importance of oceanic waters increases with distance offshore, surface concentrations of phosphate, inorganic nitrogen, silicate gradually decrease. Controlling for location, surface concentrations of phosphate are higher in winter and autumn than in summer and spring. This pattern may be related to the gradual growth of surface water phytoplankton in the in the spring and the associated large amount of phosphate absorption. Surface concentrations of inorganic nitrogen were likewise higher in winter and autumn than in spring and summer. Surface concentrations of inorganic nitrogen increased in autumn and reached the highest value in winter before decreasing in spring, and reaching the lowest value in summer. Concentrations of DIN may be affected by the expansion of runoff from the Yangtze River. Areas of high DIN concentrations are centered around the Yangtze River estuary, whereas DIN concentrations generally decrease with distance from the estuary. The main reason for this structure of nutrients is the segregations of surface

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waters from nutrient-rich deep waters over the summer and subsequent depletion of biogenic nutrients in surface waters (Ning et al., 1998). Surface concentrations of DSi are higher in summer and winter than in autumn and spring. The distribution of silicate in the waters of the Yangtze River estuary is basically the same as that of the Yangtze River, which extends as two tongues, one to the northeast and a second to the southeast, with a wide range of influence (Zhu et al., 2005).

Analysis of changes in nutrients

Changes in the distribution of nutrients in the East China Sea are closely related to seasonal variations in marine biological activities and internal biogeochemical nutrient cycling mechanisms. Nutrient concentrations are additionally controlled by complex circulation patterns in the East China Sea, which are strongly affected by seasonal variations in Yangtze River discharge and associated nutrient transport (Gao et al., 2004; Ye et al., 2017). Previous studies of nutrients in the East China Sea have indicated that the Kuroshio, Taiwan warm current, the Yellow Sea warm current and the Yangtze River estuary plume with its associated freshwater input greatly affect the distribution of nutrients and that substantial exchanges occur between these contrasting water masses (Wang et al., 2002; Chen et al., 2007).

The East China Sea is located in the East Asian monsoon region, which has exhibited a high rate of global environmental change. This leads to challenges in researching the dynamics of nutrient concentrations and related biogeochemical processes and marine ecosystems, which are strongly affected by complex monsoon-driven physical and chemical conditions (Liu et al., 2003). The outer sea area of the East China Sea is strongly influenced by the western side of the Taiwan warm current. Hence, contrasting concentrations of nutrients occur in the surface water and the bottom water. The invasion path of the Taiwan warm current into the East China Sea is characterized by a high level of nutrients in the bottom water (Wang et al., 2008). Nutrient concentrations in East China Sea surface water are primarily influenced by the input of freshwater from the Yangtze River, and high concentrations of nutrients occur in coastal surface waters of the Yangtze River estuary, as well as off the coasts of Zhejiang and Fujian provinces. Surface waters in the open ocean are greatly affected by the influence of the Kuroshio Current, where nutrient concentrations are generally low. Deeper waters are affected by the decomposition of organic matter, and nutrient concentrations generally increase with depth (Mi et al., 2012). The concentration of nitrate in the freshwater of the Yangtze River is high (the highest concentration is up to 100 μmol/L), but the concentration of phosphate is relatively low (Liu et al., 2008). As a result, the concentrations of nutrients in the sea area near the Yangtze River estuary is significantly larger than that of the middle and bottom layers. This further indicates that the main source of the nutrients into the East China Sea is the Yangtze River, and that phosphate concentrations in the East China Sea are primarily controlled by concentrations in the Yangtze River and the Taiwan warm current (Shi et al., 2003). Nutrient concentrations exhibit a decreasing trend from west to east or, alternatively, with distance offshore. The nutrient concentrations of DIN, DIP and DSi in the Kuroshio Subsurface Water and Kuroshio Intermediate Water are additionally quite high, indicating that these water masses, together with Changjiang Diluted Water (CDW), are probably important contributors to the nutrients inventories of the northeastern section of the East China Sea (Liu et al., 2017).

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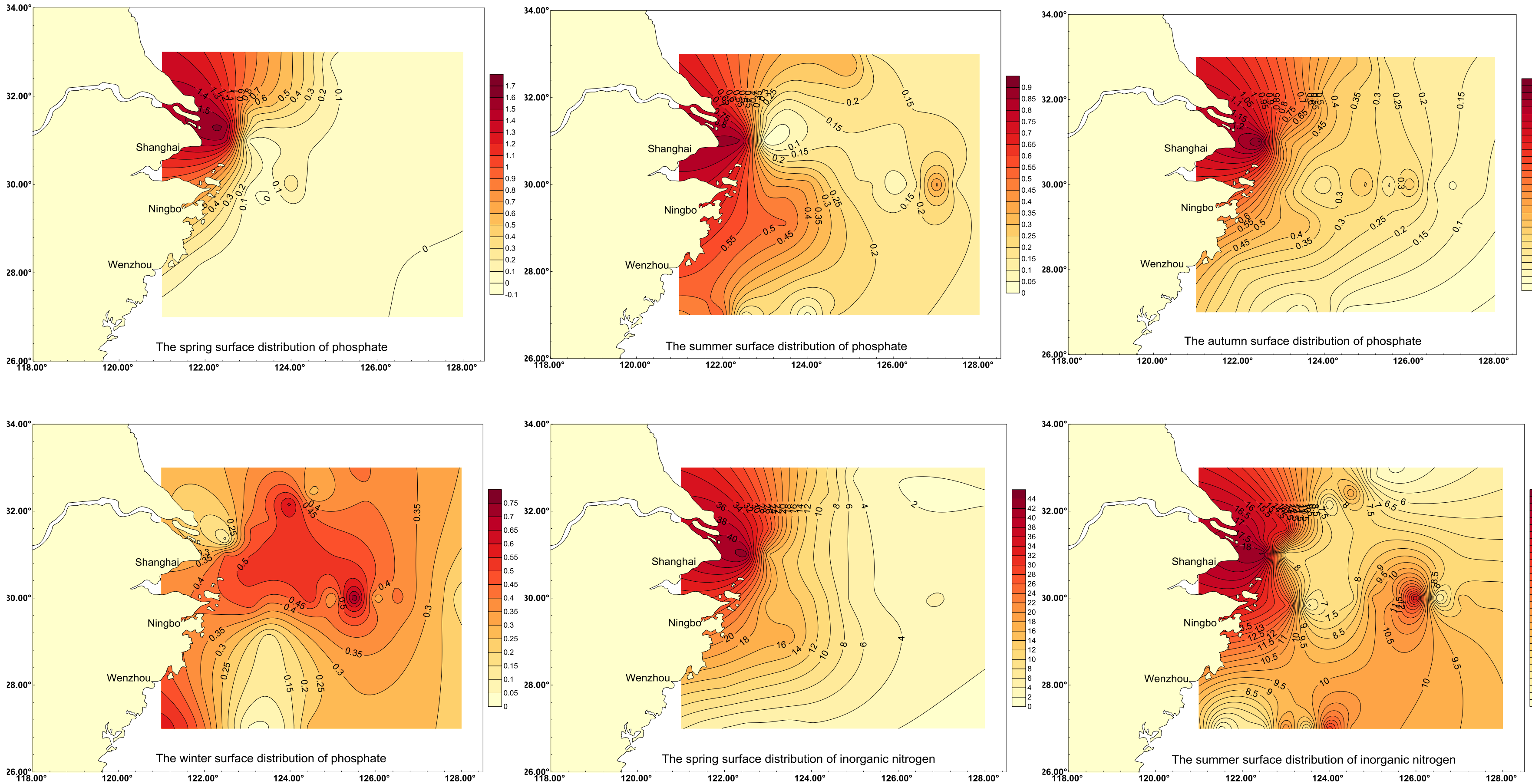

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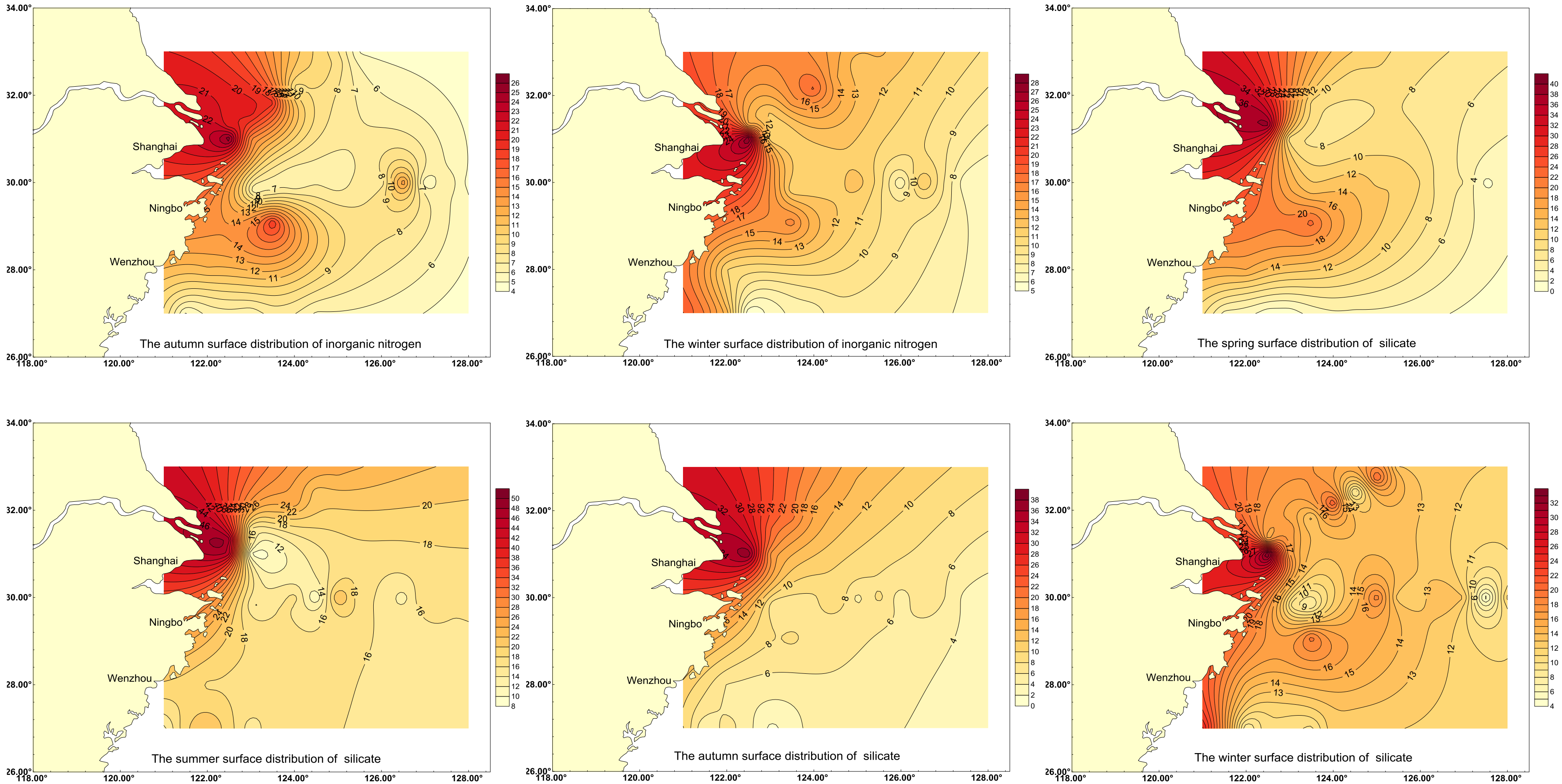

***Figure 7.** Seasonal distribution of phosphate, inorganic nitrogen and silicate in the East China Sea during the spring, summer, autumn and winter, 2015*

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Conclusion

Based on the analysis of nutrient concentrations in seawater samples taken along standard oceanographic sections in the East China Sea for nearly 25 years, we conclude that concentrations of DIN, DIP and DSi exhibit a gradual increase characterized by interannual variability. Concentrations of DIP, DIN and DSi range from 5.34-11.87 μmol/L, 0.31–0.53 μmol/L and 11.12–23.15 μmol/L, respectively. From 1996, the concentrations of DIN, DIP and DSi increased significantly. In 2015, the annual average concentration of DIN in the East China Sea was ~2 times greater than in 1991, the annual average concentration of DIP was ~1.6 times greater, and the annual average DSi concentration was ~2 times greater. This increase in nutrient concentration primarily reflects increases in land-source emissions, physical dilution by ocean water and absorption of nutrients by phytoplankton. The N/P ratio of the East China Sea is generally stable and exhibits an asymmetric M-type fluctuation. The Si/N ratio is likewise generally stable and exhibits an asymmetric W-type fluctuation. This may be related to changes in runoff and sediment transport as well as changes in the N and P structure of chemical fertilizer over recent years. Certain policy changes, such as China's recent efforts to control China's total marine pollution and protect marine environments may also play a significant role.

The distribution of nutrients in the East China Sea exhibits clear seasonal variations. Concentrations of phosphate and inorganic nitrogen are higher in winter and autumn than that in summer and spring. Silicate concentrations are higher in summer and winter than in autumn and spring. The major source of nutrients into the East China Sea is the Yangtze River estuary plume. Consequently, nutrient concentrations decrease from west to east.

Despite the scale of this study, we advocate for the adoption of continued annual marine environmental monitoring, which will provide reliable data support for decision-making by China's marine environmental management departments. We additionally propose strengthening marine environmental monitoring and early warning systems and adherence to the strategy of sea and land pooling. Recognizing that data alone will not solve any environmental issues, we additionally advocate for the strengthening marine protection efforts and promoting these efforts through more widespread awareness and environmental education.

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