Coastal eutrophication in China: Trend, sources, and ecological effects

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ABSTRACT

Eutrophication in coastal waters caused by excess nutrient inputs has occurred widely on a global scale. Due to the rapid economic development over the last four decades, most of the Chinese coastal waters have experienced a eutrophic process. Major observed trends of coastal eutrophication include two periods, a slow development from the 1970s to 1990s and a fast development after 2000, with major contributions of increased nitrogen (N) and phosphorus (P) from river inputs, atmospheric deposition, and submarine groundwater discharge (SGD). Nutrient composition and stoichiometry have been significantly changed, including increased ammonium, bioavailable organic N and P, and asymmetric ratios between N, P and silicate (Si). Most of these changes were related to the rapid increases in population density, fertilizer application, sewage discharge, aquaculture and fossil fuel combustion, and have resulted in distinctly increased harmful algal blooms. Coastal eutrophication combined with the effects of climate change is projected to continually grow in coming decades. Targeted research is therefore needed on nitrogen reduction and control, potential adaptation strategies and the consequences for ecosystems and economic sustainability.

1. Introduction

Eutrophication has occurred widely in marine and freshwater systems over the last century and forces a large array of ecological and biogeochemical consequences (Cloern, 2001). It was first presented by Lund in 1967 with the definition being “the process of becoming rich in dissolved nutrients” (Lund, 1967). Because eutrophication is a process rather than a state, the definition is gradually developed from “the enrichment of a given aquatic environment by inorganic or organic nutrients, particularly forms of nitrogen (N) and phosphorus (P), leading to a change in its nutritional state” (Richardson and Jorgensen, 1996; Andersen et al., 2006) to “the increase in the rate of organic matter supply to an ecosystem” (Nixon, 1995; 2009). On a global scale, nutrient addition in estuarine and coastal environments is largely contributed by anthropogenic N and P, particularly from N. The global N export from rivers was estimated to be about 60 Tg N yr⁻¹ (1 Tg = 1 million tons) in the mid-2000s, which increased more than twofold compared to the 1860s (Boyer et al., 2006; Howarth, 2008). Increased N and P loads were largely derived from anthropogenic activities (e.g., agricultural and industrial activities) and related to the population density of the watershed (Nixon, 1995).

China now accounts for about 32% of the world’s consumption of N fertilizers, with the rates of anthropogenic N discharge to freshwater being about 14.5 ± 3.1 Tg N yr⁻¹ (Yu et al., 2019). Approximately 23% of the human population live within the narrow coasts from global average (Nicholls and Small, 2002), the number however in Chinese coast was nearly 44% and they contributed about 60% of the national gross domestic product (Wang et al., 2014a). Consequently, eutrophication caused by N enrichment was observed widely in Chinese coastal waters, over 20% of the coastal waters in China now could be classified as “affected by eutrophication” (MEE, 2020a). According to the Chinese national seawater quality standards (GB 3097-1997), the sea areas with...
water quality worse than Class II reached approximately 55,340 km² in 2019, which is approximately 23.4% of total coastal areas (MEE, 2020a; Fig. 1).

From the perspective of the natural hydrodynamic environment, Chinese coastal waters are susceptible to nutrient enrichment. Numerous rivers are flowing into the China Seas, including three world-class large rivers, the Huanghe River (Yellow River), Changjiang River (Yangtze River), and Zhujiang River (Pearl River). These rivers transport a large amount of nutrients into coastal waters and the water export rate from coast to shelf is a pivotal factor determining the residence time of nutrients in coastal waters. Chinese Shelf Seas are broad and shallow which generate a long residence time for matter exchange from coast to shelf and finally to the deep sea, e.g., the modelling calculation found that water residence time in the Bohai, Yellow and East China Seas were 11.6, 4.95 and 0.39 years, respectively (Lin et al., 2020). Large amounts of nutrient input from land combined with relatively weak water exchange capacity of Chinese Shelf Seas are major causes leading to the eutrophic processes of Chinese coastal waters. Besides, the branches of Kuroshio (e.g., Taiwan Warm Current, Yellow Sea Warm Current) also regulate the water exchange and nutrient replenishment of the China Seas. For example, in the East China Sea, water flux from the Kuroshio is 1-2 orders of magnitude larger than that from the Changjiang (Guo et al., 2006; Isobe, 2008), thus, the nutrient flux (especially for P) from the Kuroshio is larger than that from the Changjiang River (Chen and Wang, 1999; Yang et al., 2013; Zhang et al., 2007a), which can promote phytoplankton growth and even lead to dinoflagellate blooms (Tseng et al., 2014; Zhao et al., 2019; Zhou et al., 2019). Xiao et al. (2019a) analyzed the frequency of HABs of the whole Chinese coastal waters over the last 40 years and found it increased at a rate of 40 ± 4% per decade; and moreover, the frequency sped up since 2000 and increased to 35–120 incidents per year (Wang et al., 2018a). The total affected area of HABs reached thousands or even tens of thousands of square kilometers and the duration prolonged from weeks to months (Yu and Liu, 2016). These pieces of evidence indicated a rapid development of eutrophication in Chinese coastal waters. Previous studies showed that the rapid eutrophic process of Chinese coastal waters is mainly caused by the terrigenous inputs (e.g., Strokal et al., 2014; Wang et al., 2018a; Xiao et al., 2019a), thus, the influence of river inputs, atmospheric deposition and submarine groundwater discharge (SGD) was concerned in this study.

Over the last decade, there is emerging and growing evidence that climate change has an interactive effect with nutrients and this could further elevate the eutrophic process and the frequency and scale of HABs in future (Wells et al., 2015). Climate change pressure led to seawater warming and increased precipitation and runoff in many coastal regions. By the year 2050, the global annual average temperature could keep increasing if humanity does not significantly mitigate greenhouse gas emissions IPCC (2018). Ocean warming can strengthen

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**Fig. 1.** The historical status of coastal water quality from 2001 to 2019 which are determined by the sea areas with water quality worse than Class I (a: Bohai Sea; b: Yellow Sea; c: East China Sea; and d: South China Sea) and the spatial pattern of seawater quality matching the coastal population density (e). The classification of water quality is from the Chinese national seawater quality standards (GB 3097-1997), which is mainly based on the concentrations of dissolved inorganic nitrogen (DIN) and dissolved inorganic phosphorus (DIP). Data sources are from the Bulletin of Marine Ecology and Environment Status of China in 2019 (MEE, 2020a, b) and Statistical Communique on the 2019 National Economic and Social Development (NBS, 2020).
seawater stratification and reduce the vertical exchange rate of nutrient cycling while increasing precipitation and runoff enable to promote the nutrient flux from land to sea. These changes combined with biological metabolic processes can produce a series of ecological effects, including the acceleration of eutrophication and HABs (Wells and Karlson, 2018). Since the late 1970s, the sea surface temperature (SST) displayed a rapid increase in China Seas, although there was a slowdown in the warming period (1998–2011). During the warming acceleration period (1979–1998), the mean warming rate was about ~0.31 °C decade⁻¹, and the fastest-warming region reached 0.80–1.30 °C decade⁻¹, far exceeding the mean rate of global ocean surface warming (~0.07 °C decade⁻¹) during the same period (Tang et al., 2020). Xiao et al. (2019a) analyzed the correlation of HABs and environmental conditions during 1970–2015 in Chinese coastal waters and found that HAB frequency was significantly correlated with the warming trend but the most positive relationship appeared in more eutrophic waters. The shift of phytoplankton assemblages between diatoms and non-diatoms over the last six decades was observed in the Changjiang Estuary, and increased non-diatom was regarded as a result of the combining effect between eutrophication and rising SST (Wang et al., 2021).

No doubt, coastal eutrophication is still a vital prerequisite for the formation of interactive effects between climate and nutrients (Anderson, 2012; Griffith and Gobler, 2020). In the coming decade, ocean eutrophication is likely to increase continually, because the human population will keep increasing and megacities (>10 million people) will be concentrated continually along coastlines (United Nations, 2014; Heffer and Prud’homme, 2016). To support increased population and required food supply, global fertilizer application will continue to rise (Heffer and Prud’homme, 2016) and China Seas might face more pressure in future. In this context, it is necessary to review the eutrophication process and its characteristics in Chinese coastal waters to enhance our understanding and improve the management to cope with future changes. In this review, we traced the evolution processes of Chinese coastal eutrophication on the aspects of nutrients, climate changes and HABs over the last five decades, explored the nutrient sources and forms, analyzed their corresponding relationship with HABs, and discussed the trend of nutrient enrichment correlated with ocean warming and economic development and their ecological effects on HABs.

2. Temporal trends of nutrient enrichment in Chinese coastal waters

The first research article focused on eutrophication and HABs in Chinese coastal waters was published in 1983. Zou et al. (1983) investigated the nutrient levels in the Bohai Bay during 1978–1981, after the bloom of Prorocentrum minimum in the summer of 1977 was reported in local news. In that study, the authors compared the nutrient levels in the Bohai Bay with those in the Tokyo Bay, Baltic Sea and the Seto Inland Sea, and pointed out that the increased N input from rivers enriched the estuarine region of the Bohai Bay and led to algal blooms. The article had a milestone effect, after which more and more scientists realized that increased N and P derived from the land had produced a negative ecological effect on the coastal system (e.g., Yu et al., 2006; Zhang et al., 2004; Song et al., 2016). Subsequent investigations since the 1980s depicted the time trend of N enrichment in the Bohai Sea (Fig. 2). The average dissolved inorganic nitrogen (DIN) in the Bohai Sea was less than 5 µM in the early 1980s, but it increased to 10–25 µM in the 2010s; and increased HABs matched the process of N enrichment (Fig. 2). A large number of studies have shown that throughout most of the world, the rate of N increase is much slower than that of P, leading to high N:P ratios (Jeuven et al., 2015, 2016). A similar pattern was also observed in the Bohai Sea, the average dissolved inorganic phosphorus (DIP) kept a level at 0.3–0.7 µM during the 1980s to the 1990s with a slowly increasing trend, but it decreased to 0.2–0.5 µM in the 2000s (Fig. 2). The decline in DIP over the last two decades was mainly attributed to the government policy of limiting the use of P since 1996 (Liu and Qiu, 2007).

The change of DIN concentrations in the Bohai Sea enables to represent the eutrophication timeline in most Chinese coastal waters. The initiation of N enrichment occurred during 1975–1985 when the fertilizer application increased significantly after agricultural reform in the late 1970s in China; the eutrophic process was accelerated since the 2000s, corresponding to rapid economic development and increased population along the coastline (Liu et al., 2008, 2013a). Here we gave an example of Sishili Bay in the Yellow Sea to show the trajectories of human activity related to N enrichment (Fig. 3). In the second period, N enrichment continued and eutrophic status worsened due to expansion of marine aquaculture, increased domestic sewage discharge and increasing population density from the 1990s to 2000s (Fig. 3). A large expansion of marine aquaculture activity occurred in the 1990s (Fig. 4b), when economic conditions had improved significantly, e.g., suspended scallop aquaculture in the bay carried a standing stock of 30,000 t (Zhou et al., 2006). Meanwhile, increased population and industrial activity during the development of urbanization in the 1990s accelerated the scale of sewage discharge (Fig. 4c, d). Significant positive correlations between TN values of the sediment core b in Fig. 3 and data of the surrounding population density ($R^2 = 0.8695$), sewage discharge ($R^2 = 0.6287$) and fertilizer use ($R^2 = 0.7689$) in Fig. 4 further proved the dominant contribution of human activities to the N enrichment and eutrophic process of the Sishili Bay (Wang et al., 2013). Corresponding to the two stages of N enrichment (Fig. 3), two significant biomass increases in diatom and dinoflagellate occurred in the Sishili Bay after 1975 and 2000, respectively (Fig. 5).

The correlations between N enrichment and increased phytoplankton biomass or HABs in the Bohai Sea and Sishili Bay illustrated the eutrophic history typical in most Chinese coastal waters. For example, similar trends were also observed in the Changjiang Estuary (Feng et al., 2008; Zhu et al., 2014), coastal waters in the East China Sea (Gao and Wang, 2008; Xing et al., 2016) and Zhujiang Estuary (Jia et al., 2002, 2013). Therefore, coastal eutrophic processes in China can be divided into two periods, a slow development from the 1970s to 1990s and faster development after 2000. Since 2010, the Chinese government significantly enhanced N management to control N pollution in aquatic
systems. The policy significantly reduced the rates of eutrophication, the sea areas with eutrophic status showed a decreasing trend during 2012–2019 (Fig. 6). One example is in the Laizhou Bay, Bohai Sea, where reduced ammonium loads from rivers led to the decrease of HAB frequency on a decadal scale (Jiang et al., 2018). Although it will take a long time to alleviate eutrophication, the reduction in both N and P loads is still an important way to achieve long-term and large-scale ecological restoration.

3. Sources of nutrients to coastal ecosystems

3.1. River input

Riverine nutrient loads to coastal waters play an important role in coastal eutrophication, fertilizer application, wastewater disposal and aquaculture are the major sources of N and P in rivers and estuaries (Liu et al., 2009; Wang et al., 2018a). For example, approximately 25% of the riverine DIN to the Bohai Sea was from sewage in 2000, and approximately 40% was from agriculture (Strokal et al., 2014), Chinese rivers deliver about 5–10% of global freshwater input and 15–20% of the global continental sediment to the world ocean Zhang (2002). Among them, freshwater discharge from the three largest rivers (Huanghe, Changjiang and Zhujiang Rivers) represents 73% of the total river discharge into the China Seas (Wang et al., 2018a). The important impact of large rivers is also reflected in the map of coastal water quality, the most eutrophic seawaters are mainly distributed in these three largest estuaries (Fig. 1).

The N pollution in the Chinese rivers is higher than or comparable to the levels of eutrophic rivers in Europe and North America (Liu et al., 2003; Zhang, 1996). According to the classification of pollution index based on riverine DIN concentration devised by Smith et al. (2003), 41% of the rivers emptying into the Chinese Seas are at the levels between the average global conditions (52 μM) and polluted waters (110 μM), and the other 59% are at the levels between polluted and extremely polluted waters (347 μM) (Liu et al., 2009). Moreover, N and P components in the rivers displayed two distinct characteristics. One is that the proportion of ammonium to the DIN load is relatively high compared to other rivers in the world (Turner et al., 2003), e.g., ammonium represented >20% of DIN in the Changjiang and Zhujiang Rivers during the winter time (Liu et al., 2009). The other is that the proportions of dissolved organic nitrogen (DON) and phosphorus (DOP) in N and P pools significantly increased, e.g., the proportion of DON to total dissolved nitrogen (TDN) was approximately 13% in the Huanghe Estuary (Liu et al., 2009), but it exceeded 50% in the Changjiang Estuary (Zhang et al., 2015; Xin et al., 2019) and 80% in the Zhujiang Estuary (Wang et al., 2003; Zhang et al., 2006; Qiao, 2016); the proportion of DOP to total dissolved phosphorus (TDP) accounted for 41% in the Huanghe Estuary (Liu et al., 2009). These data indicated that DON and DOP might play more important roles in coastal eutrophication than expected.

The decadal changes of DIN concentrations in the three largest rivers illustrate the contribution of riverine nutrients to coastal eutrophication (Fig. 7a-c). In general, DIN concentrations in the three largest rivers displayed a distinct uptrend during the 1970s to the 2010s, the first increase initiated in the 1980s with a relatively slow rate, then it reached to peak after the 2000s at a faster rate (Fig. 7a-c). Here we took the Changjiang River as an example to show its spatial linkage with coastal eutrophication (Fig. 7a-c). In general, riverine DIN concentrations increased from 27 μM in 1978 to 118 μM in 2016 (Fig. 7b). Consequently, riverine DIN loads in the East China Sea increased from 4.0 × 10^5 to 10^6 μ g yr^−1 in the early 1980s to 6.2 × 10^6 to 10^7 μ g yr^−1 in the mid-1980s, then increased to 18 × 10^6 to 10^7 μ g yr^−1 in the early 2000s (Wu et al., 2012). Historical DIN concentration in the Changjiang Estuary was less than 10 μ M in the early 1960s (Zhou et al., 2008) but reached 20–77 μ M in the 2000s and was predicted to increase continually in the coming years (Wang and Cao, 2012; Zhou et al., 2008).

The decadal changes of DIP concentrations in the three largest rivers are different (Fig. 7). DIP concentration in the Changjiang and Zhujiang Rivers also displayed an increasing trend (Fig. 7b, c), but it has decreased in the Huanghe River in recent decades (Fig. 7a). The Huanghe River is the largest river connecting to the Bohai Sea, the decreased DIP in Huanghe River was consistent with the trend of DIN concentrations in the Bohai Sea. Before the 1990s, the Bohai Sea was characterized by lower DIN concentrations (< 2.5 μ M) and relatively high DIP concentrations (> 0.50 μ M), but the pattern shifted to relatively high DIN concentrations (> 5 μ M) and low DIP concentrations (> 0.35 μ M) during the 1990s to 2000s (Xin et al., 2019). Consequently, the nutrient regime in the Bohai Sea changed from N-limitation before the 1990s to P-limitation in the 2000s.

It is hard to fully explain the reason for P decrease, but the similar variation trend of DIN and DSI in the 1990s might be related to the increased nutrient retention within the river systems because of the construction of dams, which occurred simultaneously with the reduction in annual water discharge and sediment load in the lower reaches of the Huanghe River during this period (Liu et al., 2012a). Further, the government’s policy of limiting the use of P after 1997 and the local river pollution control policy implemented by the Shandong provincial government since 2000 should also have played a role in P decrease (Liu et al., 2009).
et al., 2009; Liao et al., 2013).

3.2. Atmospheric deposition

N and P released by the combustion process in the industry and agriculture exist in gaseous, particulate and aqueous phases, they can be delivered to coastal waters via wind and became an important external source for coastal eutrophication (Paerl et al., 2002). Atmospheric-sourced N includes inorganic and organic species, inorganic nitrogen (nitrate and ammonium) was the dominant species deposited into the sea (Zhang et al., 2004; Zhang et al., 2007b), in contrast, the contribution of organic nitrogen in total nitrogen of atmospheric deposition occupied about 15–30% (e.g., Shi et al., 2010; Zhang et al., 2012; Xing et al., 2018). Over the past few decades, atmospheric deposition of N and P displayed an uptrend in China, high atmospheric deposition in China Seas is not only related to the increase in energy consumption during economic development, but also to the increase of sandstorm events under the background of climate change (Zhang et al., 2010).

The relative contribution of atmospheric deposition to the N and P pool of coastal ecosystem greatly depends on the basin area, human activity intensity and weather conditions (e.g., wind and precipitation) (Liu et al., 2013b; Luo et al., 2014a). The limited observational data in China Seas showed that N deposition flux had significant temporal and spatial variations (Table 1). In general, wet deposition fluxes of DIN are 2–10 times higher than the dry deposition fluxes over Chinese coastal waters. In the Bohai Sea, for instance, wet deposition fluxes of DIN were 71.7 ± 47 mmol m−2 yr−1 in 2018 and 73.3 ± 36.2 mmol m−2 yr−1 in 2019, the levels being comparable with that observed in 1995–1996 (80.1 ± 38.6 mmol m−2 yr−1) (Table 1). It indicates that the value of total DIN deposition flux in the Bohai Sea (124.2 ± 54.8 mmol m−2 yr−1) estimated by Zhang et al. (2004) in 1995–1996 was reasonable.

In contrast, DIN deposition fluxes in the Yellow and East China Seas were variable (Table 1). It ranged from 34.7 mmol m−2 yr−1 in 1999–2003 to 208.1 mmol m−2 yr−1 in 2015–2016 in the Yellow Sea (Qi et al., 2013; Xing et al., 2017, 2018). The annual input of DIN fluxes was estimated as 380–2277 kt, which accounts for 0.3–6.7% of the new productivity in the Yellow Sea (Qi et al., 2013). DIN deposition fluxes in the East China Sea ranged from 68.4 mmol m−2 yr−1 in 1999–2003 to 201 mmol m−2 yr−1 in 2004 (Chen et al., 2011; Zhang et al., 2007b; Zhu et al., 2009; Liao et al., 2013).

![Fig. 4. The trajectories of human activity related to nitrogen (N) enrichment around the Sishili Bay, Yellow Sea (Data source: revised from Fig. 8 in Liu et al., 2013a).](image-url)
et al., 2013), which was estimated as 790–2323 kt of annual input, accounting for 1.1–3.9% of the new productivity (Zhang et al., 2010). DIN deposition fluxes in the South China Sea displayed a significant difference in space, e.g., it was 92.3 mmol m$^{-2}$ yr$^{-1}$ during 2015–2017 at Daya Bay near the city but decreased to 26.9 mmol m$^{-2}$ yr$^{-1}$ in the Yongxing Island, far away from the city. The DIN deposition fluxes observed in China Seas were generally higher than those reported in other large-scale water regions around the world. For instance, the deposition fluxes ranged from 18.1 to 47.7 mmol m$^{-2}$ yr$^{-1}$ around the Mediterranean during June 2001-May 2002 (Markaki et al., 2010), and 46.2 ± 4.93 mmol m$^{-2}$ yr$^{-1}$ around the Black Sea during 2004–2010 (Medinets and Medinets, 2012). Compared to the coastal waters, lower deposition levels were observed over the marginal seas and the open oceans (Qi et al., 2020).

In addition, fluxes of gaseous reactive nitrogen, such as HNO$_3$ and NH$_3$, were not measured in these studies and this might result in the underestimation of N deposition over the Chinese coastal waters, especially in spring and summer. Except for DIN fluxes from atmospheric deposition, the ratios of N:P (119 and 832 for wet deposition, and 418 and 729 for dry deposition at Qianliyan and Shengsi, respectively, and
167 for dry deposition at Daya Bay) in both wet and dry deposition were measured and they were much higher than those in surface seawater and the uptake ratios of phytoplankton (Zhang et al., 2007b; Wu et al., 2018a). In contrast, the studies on atmospheric P deposition were relatively less than N and it is generally considered that the flux of P deposition was very low (Baker et al., 2003; Mahowald et al., 2008). In any case, atmospheric deposition of nutrients may change the nutrient structure in coastal waters (Yau et al., 2020; Zhang et al., 2007b), especially during the periods of dust and haze events (Qi et al., 2018; Zhang et al., 2019).

### 3.3. Submarine groundwater discharge (SGD)

SGD, an important component of the global water and biogenic element sources, has been considered as a significant pathway for material exchange at the land-sea interface of coastal ecosystems Moore (2010). SGD-driven biogenic elements could alter the nutrient environment in marine ecosystems via changing their components and ratios (Slomp and van Cappellen, 2004; Lee et al., 2010; Chen, 2019). Investigations of SGD rate along the Chinese Coastline have been carried out over the last decade (Table 2). SGD rate displayed distinctly spatial variations, e.g., the highest SGD rate was 127 cm d⁻¹, which found in the Zhujiajian Bay, East China Sea (Ji et al., 2012), while the lowest one (0.08 cm d⁻¹) appeared in the Minjiang Estuary, East China Sea (Liu et al., 2016). In estuarine regions, the SGD rates at large rivers (Huanghe Estuary: 10⁻¹⁰ cm d⁻¹; Changjiang Estuary 18⁻⁴⁵ cm d⁻¹; Zhujiang Estuary: 6⁻⁵⁰ cm d⁻¹) are much higher than at a smaller river (Wanquan River Estuary: 0.49 cm d⁻¹) (Su et al., 2011; Xu et al., 2013; Liu et al., 2018a).

SGD processes are complex with different kinds of hydrological processes such as precipitation, geomorphic condition, tidal/wave height, etc. (Santos et al., 2012). Although uncertainty exists in SGD-derived fluxes of biogenic elements, especially at coastal zones with high anthropogenic activities, some studies have indicated the importance of SGD for coastal nutrient budgets (Liu et al., 2017a; Chen et al., 2018). For example, SGD-derived alkalinity was considered as an important factor to cause seawater acidification in the coral system of the Sanya Bay, South China Sea (Wang et al., 2014b). In the Yellow Sea, SGD-derived nutrients were estimated as a dominant source to support macroalgal blooms (Liu et al., 2017b). In the Changjiang Estuary, the oxygen level can be impacted by anoxic groundwater (Guo et al., 2020). In future, the contribution of SGD-derived nutrient fluxes in Chinese coastal waters needs to be further assessed.

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**Fig. 7.** The historical changes of nutrient concentrations in the Lijin Station of the Huanghe River (a), Datong Station of the Changjiang River (b) and Badakoumen Station of the Zhujiang River (c) (Data source from Wang et al., 2018a). The spatial pattern of seawater quality (d) matching the coastal population density (Data source: Statistical Communique on the 2019 National Economic and Social Development) was modified from the Bulletin of Ecology and Environment Status of China in 2019 (MEE, 2020a).
amino acids, amino sugar, glucose-6-P, adenosine-5'-monophosphate (AMP) and ADP. Bioavailable forms of DON and DOP are diverse, including urea, 

"4. Ecological effects of nutrient enrichment"

The excessive N and P transported from land to sea not only increased the concentrations of these elements in coastal waters but also changed the composition of these nutrients in the water body. In terms of nutrient forms, the proportions of DON and DOP in the TDN and TDP increased the concentrations of these elements in coastal waters but also changed the composition of these nutrients in the water body. In terms of nutrient forms, the proportions of DON and DOP in the TDN and TDP have been associated with compositional shifts in phytoplankton assemblages, away from diatoms to many HAB species previously suspected. Although there are some controversial opinions, some flagellate species (P. micans, Alexandrium tamarense, Chattonella marina and H. akashiwo) grew well under various DON and DOP regimes (Wang et al., 2011; Ou et al., 2014).

In general, nitrate is the dominant component of DIN in the ocean. Ammonium however was significantly elevated in many Chinese coastal waters over the last four decades, due to the expansion of marine aquaculture and sewage discharge. Ammonium is the reduced state of nitrogen and can be synthesized to amino acids under the action of enzymes without changing the valence state of nitrogen, whereas nitrate is the oxidized state of nitrogen and needs to be reduced to ammonium before biosynthesis. Therefore, ammonium uptake enables reduced energy consumption which leads to the priority of ammonium utilization by phytoplankton. Excessive ammonium however can impact the structure of phytoplankton assemblages (Gilbert et al., 2014). More and more evidence indicated that diatoms were more susceptible to the negative effects of ammonium than many other HAB species, especially cyanobacteria that can thrive on high ammonium concentration (Glibert et al., 2016; Glibert et al., 2018).

"4.2. The ecological effects of changed nutrient stoichiometry"

During the process of eutrophication, the increasing rate of N in coastal waters was much higher than that of P. In contrast to increases in N and P, dissolved silicate (DSi) decreased in many coastal waters, mainly due to retention behind dams (e.g. Vorosmarty et al., 2003; Beusen et al., 2009). Such asymmetric changes between N, P and Si have resulted in a gradual shift from N-limitation to P- and Si-limitation in many coastal systems (Conley and Malone, 1992; Ragueneau et al., 2008). Long-term increases in the ratios of DIN:DSi and DIN:DIP have been associated with the phenomenon also occurred in many Chinese coastal waters, and
Table 2
The estimated SGD rate (cm d⁻¹) in Chinese coastal waters

<table>
<thead>
<tr>
<th>Sea regions</th>
<th>Study areas</th>
<th>Methods</th>
<th>SGD rate</th>
<th>References</th>
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<td>Ji et al., 2012</td>
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here examples in the Bohai and Yellow Seas are given (Fig. 8). DIN:DIP and DIN:DSi in the Bohai and Yellow Seas showed a rapid increase over the last four decades; the range of DIN:DIP in the Bohai Sea increased from 2–45 during the 1980s and 1990s to 20–87 during the 2000s and DIN:DSi increased from 0.1–1.26 during the 1980s and 1990s to 0.68–2.89 during the 2000s (Fig. 8a). In the Yellow Sea, DIN:DIP kept increasing since the 1990s, with a range of 40-80 after the 2010s, DIN: DSI increased a lot from 1990 to 2000, but declined to a reasonable level (close to 1:1) during 2000 and 2010 (Fig. 8b). Asymmetric changes in nutrient structure have resulted in the shift of phytoplankton assemblages in Chinese coastal waters. The phytoplankton biomarkers (brassicasterol, dinosterol and alklenones) in the sediment cores from some coastal bays revealed the increase of phytoplankton productivity during the 1960s–2010s, accompanying with a change from diatom dominance to dinoflagellate dominance (Liu et al., 2013a; Chen et al., 2019).

Xiao et al. (2018) quantified the functional relationships between diatoms and dinoflagellates and environmental factors in coastal waters of the East China Sea. The results indicated a combined effect between temperature and DIN:DIP, with higher temperature and DIN:DIP ratio seemed to favor the dinoflagellates (Fig. 9). This might help to explain some extremely large HABs resulted from dinoflagellates in this region, e.g., a 10,000 km² bloom caused by P. donghaiense in May 2004, and a 10,000 km² bloom caused by K. mikimotoi in the summer of 2005 (Chen et al., 2006; Zhou and Yu, 2007). Xiao et al. (2018a) further analyzed the long-term (45 years) correlation between HABs and environmental factors in 11 coastal regions of China, spanning from 18° to 43° N. This data set provides a solid foundation to understand the impact of climate change in a rich waterbody, including a seasonal delay of coastal HABs along this latitudinal gradient, a shift of HABs towards an advanced seasonality, and increased HABs frequency with warming. These studies suggest the need for better management of nutrient enrichment under future climate change.

5. Conclusion and perspective

The historical evolution of coastal eutrophication in China exhibited two periods driven by the speed of economic and social development: one was a relatively slow eutrophic process from the 1970s to 1990s and the other was a faster eutrophic process after 2000. Nutrients from rivers, marine aquaculture, atmospheric deposition and SGD are the major external sources, which have changed nutrient composition and stoichiometry in the Chinese coastal waters. Increased proportions of DON and DOP and asymmetric changes between N, P and Si in coastal waters caused a fundamental impact on coastal ecosystems, e.g., increased HABs. Clearly, more work is necessary to understand the ecological influence brought by these significant changes, e.g., the physiological responses of different phytoplankton taxa to excessive ammonium, DON and DOP, and the threshold for phytoplankton to utilize different nutrient forms.

In future, eutrophication in coastal zone combined with the complexity of climate change is likely to continually increase the pressure on coastal ecosystems. While the challenges of managing eutrophication and HABs will continue to be great, what gives people hope is that the Chinese government has strengthened the treatment and management of coastal eutrophication over the last two decades. Defining standard values for seawater quality is an essential first step (Fig. 1), and reducing both terrigenous N discharge and emissions of fossil fuel burning are important policies to slow down coastal eutrophication. High efficiencies to remove ammonium from the treatment of municipal wastewater have developed rapidly in China’s coastal cities since 2005, continuous decrease in ammonium concentration in total municipal wastewater discharge was observed from 2006 to 2015 (Zhang et al., 2020).

Scientists have gradually deepened their understanding of the relationship between the eutrophication and ecological consequences. One is that integrated monitoring (especially for SGD) efforts should be paired with laboratory and field process studies to better understand the chemical and biological effects of changing nutrient composition and stoichiometry at organism, population and ecosystem levels. Moreover, we need to pay more attention to the effects of ocean acidification, HABs
and hypoxia amplified by eutrophication and warming.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Harmful Algae xxx (xxxx) xxx


Y. Wang et al.
Y. Wang et al.

Harmful Algae xxx (xxxx) xxx


