Environmental Science & Technology LETTERS

pubs.acs.org/journal/estlcu

Letter

Harmful Algal Blooms in Chinese Coastal Waters Will Persist Due to Perturbed Nutrient Ratios

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Cite This: *Environ. Sci. Technol. Lett.* 2021, 8, 276−284

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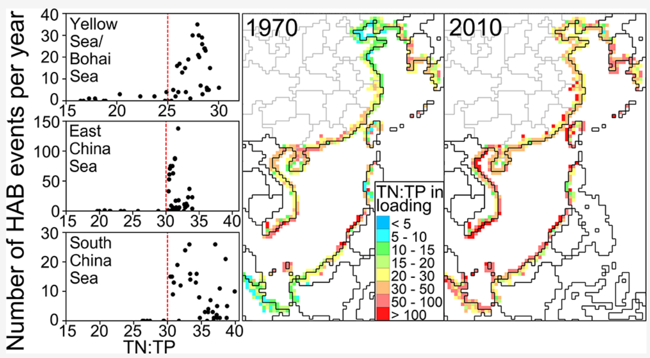
ABSTRACT: The three large marine ecosystems (LMEs) bordering China (Yellow Sea/Bohai Sea, East China Sea, and South China Sea) have received excess nitrogen (N) and phosphorus (P) in the past decades with detrimental consequences for ecosystem functioning, such as increased productivity, loss of biodiversity, and proliferation of harmful algal blooms (HABs). N loading increased much faster than that of P. Here, we show that HABs in the three LMEs started to proliferate after the N:P molar ratio exceeded the threshold value of 25 in the 1980s. The mismatch of N and P inputs is not only related to differences in loads but also inherent to the differences in their biogeochemical cycles which more efficiently filter P than N in land- and waterscapes. Future Shared Socioeconomic Pathways show that high N:P ratios will persist for decades to come, even worsening in a future oriented toward sustainability, and indicate that HABs may be a persisting problem in China's coastal waters. While efforts in agricultural systems are governed by the agronomic crop requirements and are not easy to manage with respect to N:P ratios, the separate collection of urine in urban and rural areas could contribute to decreases in both total nutrient loads and N:P ratios.

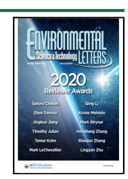


■ INTRODUCTION

The rapidly growing population and production of food and energy, expanding agricultural land, and increasing fertilizer use, discharge of sewage water, and animal waste have perturbed the Earth system by accelerating the global cycles of essential nutrients such as nitrogen (N) and phosphorus (P). Loads and concentrations of N and P are now increasing in nearly every water body across the globe with multiple effects on the ecosystem structure and functioning, such as changes in the community structure (e.g., from macrophytes to fast-growing macro- and microalgae and from diatoms to flagellates) and reduced water clarity due to enhanced algal production. The decay of algal detritus may eventually deplete the oxygen in the water and lead to hypoxia, causing an array of secondary problems such as degradation of marine habitats and massive fish death.[1−6] Under specific conditions, ecosystem changes may lead to the proliferation of toxic algae.[1−6] Hence, harmful algal blooms (HABs) producing harmful effects on fish, shellfish, birds, marine mammals, and humans comprise nontoxic high-biomass and toxic bloom events both with substantial economic losses.[7−9] The proliferation of HABs does not follow a simple dose−response relationship, as HABs may increase disproportionally with eutrophication, depending on which nutrient changes and in what proportion.[10,11] In addition to nutrient availability and ratios, the vulnerability of a particular sea area to HAB proliferation is also determined by the hydrodynamic conditions, particularly stratification and circulation.[12] Climate change may thus play a critical role through warming and changes in stratification and circulation.[8]

In recent decades, HABs have been spreading in China's coastal waters.[7,13] Herein, we explore the changing sources, loads, and element ratios of total nitrogen (TN) to total phosphorus (TP) in China's coastal waters, and where, when, and how these changes are related to the occurrence of HABs. To assess future risk of HAB proliferation in China's coastal waters, we project future nutrient loadings and their cocktail (i.e., ratios) based on the community consensus Shared Socioeconomic Pathways (SSPs). Since projecting future nutrient loading is fraught with uncertainties, we avoid selecting a most probable scenario and present the outcomes

Received: January 6, 2021
Revised: January 18, 2021
Accepted: January 19, 2021
Published: January 22, 2021



https://dx.doi.org/10.1021/acs.estlett.1c00012
Environ. Sci. Technol. Lett. 2021, 8, 276−284

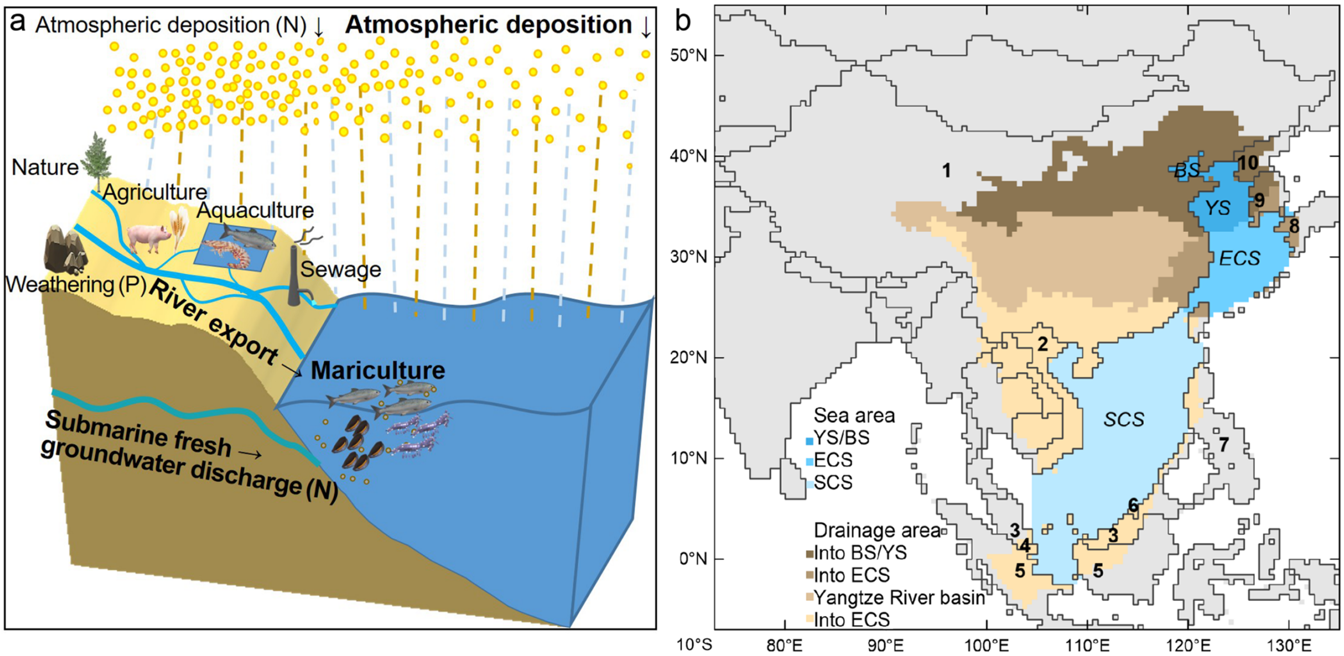


Figure 1. (a) Our conceptual model of nitrogen and phosphorus loadings to coastal waters. (b) Location and drainage area into the three large marine ecosystems (LMEs) bordering China, i.e., Yellow Sea/Bohai Sea (YS/BS, in darkest colors), East China Sea (ECS), and South China Sea (SCS, in lightest colors). Brown or gray colors represent drainage areas and blue or white colors represent sea areas. The numbers in the figure represent different countries: (1) China, (2) Vietnam, (3) Malaysia, (4) Singapore, (5) Indonesia, (6) Brunei, (7) Philippines, (8) Japan, (9) South Korea, and (10) North Korea. Data on land area, agricultural area, cropland area, population, and river discharge for the years 1970 and 2000 are listed in Table S1 in the Supporting Information.

for five different SSPs that represent a wide range of future scenarios.

Excess nutrients in China's coastal systems originate from agriculture, aquaculture, industry, and wastewater inputs through rivers and groundwater (Figure 1a).[14] Since the 1970s, in China, most food is produced in a nutrient-inefficient agricultural system;[15] its aquaculture system makes up for more than 60% of the global production,[16] with freshwater aquaculture contributing to river export and mariculture directly discharging nutrients in coastal waters.[16] Urban and rural wastewater is an important and rapidly growing source of nutrients in Chinese inland and coastal waters.[17] Submarine fresh groundwater discharge may locally be an important source, particularly of N.[18] Finally, atmospheric deposition driven by N gas emissions from agriculture, fossil-fuel combustion (both stationary and mobile), and P-containing dust from soil erosion may form an important nutrient input to the coastal and open oceans.[19]

We consider the scale of large marine ecosystems (LMEs).[20] There are three LMEs bordering China's coastline and those of neighboring Asian countries, i.e., Yellow Sea/Bohai Sea (YS/BS), East China Sea (ECS), and South China Sea (SCS) (Figure 1b). These LMEs span a range of climates from temperate to subtropical to tropical zones, have different geological settings, drainage patterns, and freshwater discharges, and differ in their watershed population density, nature and intensity of economic and agricultural activities, and changes therein during past decades (Table S1a). The BS is a shallow continental sea bordered by the Chinese provinces of Liaoning, Hebei, and Shandong and municipality of Tianjin and connected to the YS via the Bohai Strait, with an area of 77,000 km^2 and an average water depth of 18 m. The YS is a semiclosed continental shelf sea between the Chinese mainland (Liaoning, Hebei, Shandong, and Jiangsu) and the Korean Peninsula (Table S1b), with an area of 380,000 km^2 and an average water depth of 44 m. The total area draining the 381 km^3 of freshwater into the YS/BS is 1.95 Mkm^2, accommodating 551 million inhabitants with an increase of over 60% for the past four decades (Table S1a). The ECS has an area of 760,000 km^2 and consists primarily of continental shelves with depths less than 200 m bordered by China (Shanghai, Zhejiang, Fujian, and Taiwan), South Korea, and Japan. The land area draining the 939 km^3 of freshwater into the ECS is 0.9 Mkm^2, with a population that increased by 57% to 556 million inhabitants between 1970 and 2010. The SCS covers an area of ∼3.5 Mkm^2, has an average water depth of ∼1200 m, and is more open to the ocean than the YS and ECS. Watersheds in the land area surrounding the SCS cover 2.3 Mkm^2, deliver 2450 km^3 of freshwater, and host 426 million inhabitants with a nearly 2-fold increase since 1970 (Table S1a).

■ MATERIALS AND METHODS

This section presents a summary of methods and data used. Details are presented in Text S1 in the Supporting Information.

Modeling Nutrient Loadings to Coasts. The Integrated Model to Assess the Global Environment−Global Nutrient Model (IMAGE−GNM) (0.5° × 0.5° resolution)[14,21] was used to simulate various nutrient sources to rivers, riverine nutrient export, and submarine fresh groundwater N discharge to the three LMEs for the period 1970−2010, based on grid-based land-use and climate data from the Integrated Model to Assess the Global Environment (IMAGE)[22] and hydrological data from PCRaster Global Water Balance (PCR-GLOBWB).[23] The IMAGE-GNM aquaculture nutrient budget

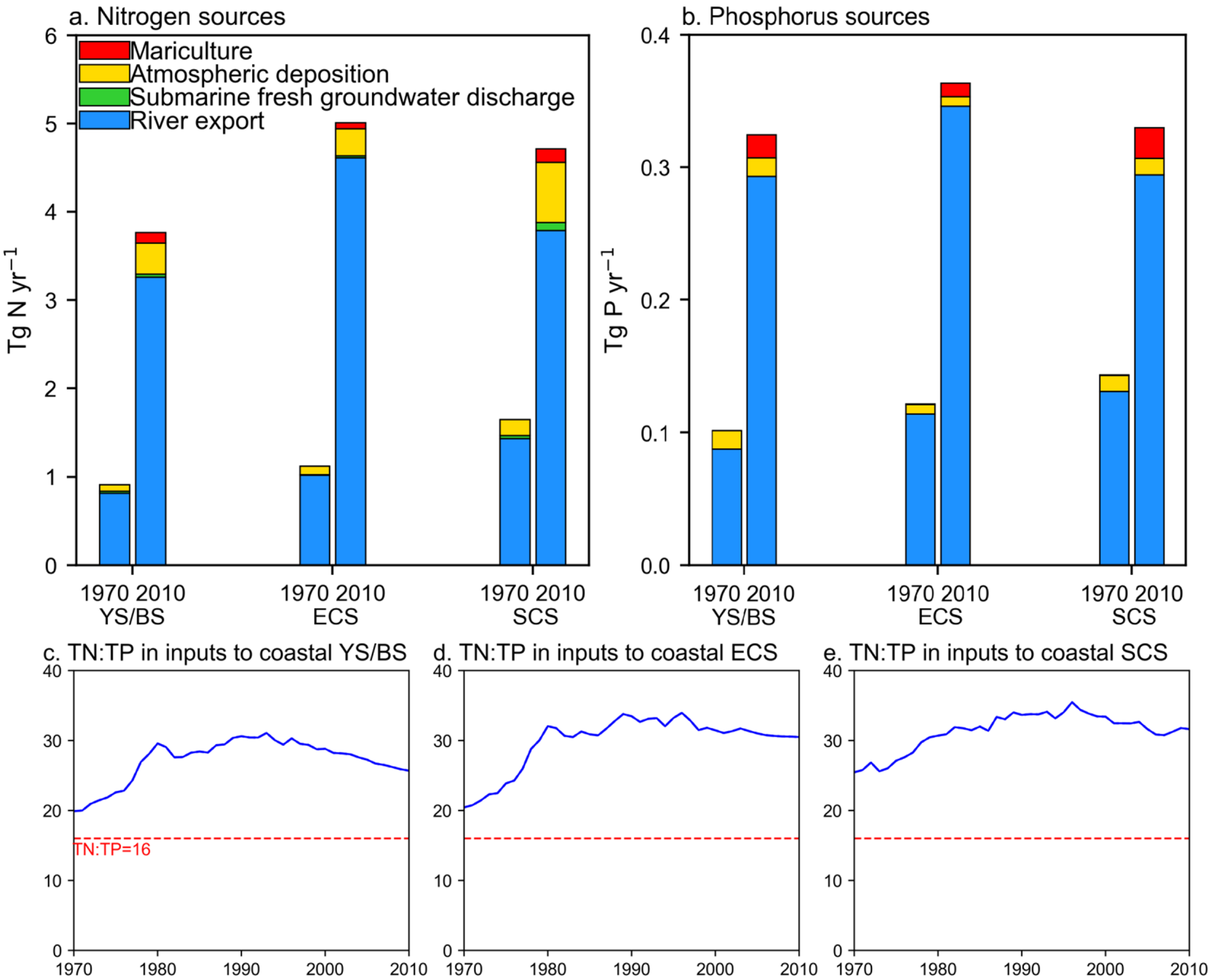


Figure 2. Sources of nitrogen (a) and phosphorus (b) inputs to the coastal waters of the Yellow Sea/Bohai Sea (YS/BS), East China Sea (ECS), and South China Sea (SCS) for 1970 and 2010 and molar TN:TP ratios in inputs to coastal BS/YS (c), ECS (d), and SCS (e) between 1970 and 2010. Data represent the aggregated inputs to all coastal 0.5° × 0.5° grid cells.

model[24−26] (0.5° × 0.5° resolution) was used to calculate nutrient flows in mariculture systems based on FISHSTAT data.[27] The TM5-FAst Scenario Screening Tool (TM5-FASST) model (1° × 1° resolution)[28] and Community Atmospheric Model (CAM4) (2° × 2° resolution)[29] were used to calculate the atmospheric nutrient deposition onto coastal grid cells and the results were converted to 0.5° × 0.5° resolution by interpolation. Extensive sensitivity/uncertainty analyses and model validation are presented in several previous studies.[14,21,24,26,28,30,31]

SSP Scenarios. The PCR-GLOBWB hydrology model was run until 2099 using anomalies from the RCP simulations from the HadCM3 Global Circulation Model[32] bias-corrected with observed weather over the historical period 1960−1999.[33] On the basis of the projected global change in radiative forcing from IMAGE, combinations of SSP1-RCP4.5, SSP2-SSP4 with RCP6.0, and SSP5-RCP8.5 were implemented. In the SSPs, P fertilizer projections used projected food demand, trade, and production from IMAGE in a soil P dynamic model included in IMAGE-GNM.[34] N fertilizer projections are based on nitrogen use efficiency (NUE). Scenarios for wastewater N and P discharge to surface water are based on relationships between income and human emissions, degree of connection to sewerage systems, presence of wastewater treatment plants, and their level of nutrient removal efficiency.

Data Sources of Chlorophyll-*a* and HABs. Annual average chlorophyll-*a* (Chl-*a*) concentrations in the coastal areas of the LMEs for the period 1978−2010 (9 km × 9 km resolution) were obtained from SeaWiFS and CZCS datasets (https://oceancolor.gsfc.nasa.gov) and rescaled to 0.5° × 0.5° resolution. The time, location, frequency, area, and dominant species of reported HABs in Chinese coastal waters during 1970−2010 were obtained from the Investigation and evaluation of red tide disasters in China.[35]

RESULTS AND DISCUSSION

Changes in the Coastal N and P Cocktail and HABs. The total nutrient inputs to the three LMEs, accounting for river export, submarine fresh groundwater discharge, atmospheric deposition, and mariculture release, increased rapidly during the period 1970−2010 (Figure 2 and Figure S1). If only coastal waters bordering China are considered, nutrient

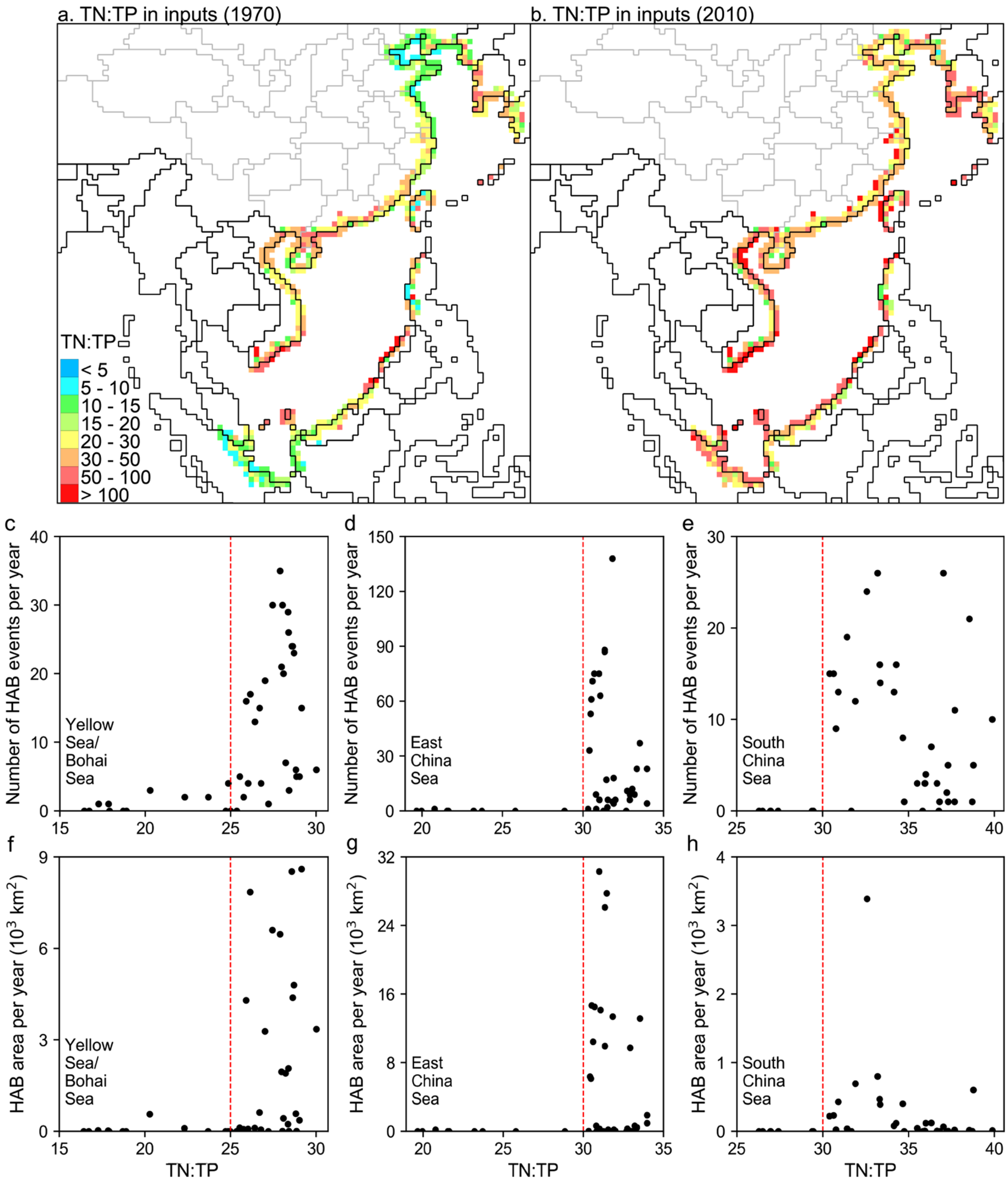


Figure 3. Molar TN:TP ratio in inputs to the coastal grid cells in the Yellow Sea/Bohai Sea, East China Sea, and South China Sea for 1970 (a) and 2010 (b) and relationship of the aggregated molar TN:TP ratio and the number and area of reported HABs per year in Chinese coastal waters of the Yellow Sea/Bohai Sea (c, f), East China Sea (d, g), and South China Sea (e, h) for the period 1970–2010. See Figure S2 for specification of the HAB species involved.

loadings increased by a factor of 5.7 for TN and 3.6 for TP in the YS/BS, a factor of 5.0 for TN and 3.2 for TP for the ECS, and a factor of 4.3 for TN and 3.8 for TP in the SCS. Hence, TN loadings increased much faster than those of TP. There is uncertainty in the simulated yearly nutrient loadings. For example, the root-mean-square error of data covering 1960–2010 at Datong station in the Yangtze River (~500 km upstream Shanghai) is 40% for TN,[30] but the modeled trend in concentrations and river export agrees very well with observations for the Yangtze[30] and a range of global rivers.[14,21,31]

Molar TN:TP ratios in the total coastal nutrient inputs steadily increased until the 1990s and afterward remained much higher than the Redfield N:P ratio of 16 (Figure 2). The change in the nutrient ratios has shifted the BS, northern YS, and southern YS ecosystems from an N-limited oligotrophic state before the 1990s to a potentially P-limited eutrophic state as suggested by their observed ratios of the biologically available dissolved inorganic nitrogen (DIN) to dissolved inorganic phosphorus (DIP).[36−38] Meanwhile, the number and area of HAB events in the Chinese coastal waters substantially increased (Figure S2). Our data indicate that HABs started to occur from the 1980s onwards when the TN:TP ratio in inputs to Chinese coastal waters exceeded a value of 25 in the YS/BS and 30 in the ECS and SCS (Figures 2 and 3). When the TN:TP ratio in inputs exceeded these critical values, the HAB frequency and area substantially increased (Figure 3), with more dinoflagellates-dominant HABs (Figures S2 and S3) and more frequently reported fish mortality, shellfish poisoning, economic loss, and even human death.[7,9] The findings from our analysis clearly point to the need for more in-depth analysis of causal factors for individual HAB species.

The molar ratio of DIN:DIP can be much larger than that of TN:TP in the coastal nutrient inputs because a large part of TP consists of inactive P bound to soil particles.[30] Furthermore, the DIN:DIP ratio in the coastal waters has increased further because of the increased DIN concentration[36−40] and DIP buffer mechanism; i.e., the sorption and desorption of DIP by suspended particles (Figure S4) in the coastal waters will maintain the DIP concentration relatively unchanged.[41,42] Our data show that the increases in the DIN:DIP ratios in the coastal waters of the BS, YS, ECS, and SCS since the 1980s−1990s (Figure S5) are similar to those in the TN:TP ratios in their coastal inputs.[36−40]

Causes of the N to P Mismatch. Ratios of TN:TP have increased as a result of many simultaneous changes. The TN:TP ratio of atmospheric deposition showed a rapid increase (Figure S6), as N deposition is primarily from anthropogenic sources that have increased rapidly in recent decades,[43] while P deposition primarily consists of soil particles mobilized by wind erosion that remained relatively stable in recent decades.[44] The TN:TP ratio in river export shows an overall increasing trend, while that from mariculture is more stable, particularly in recent years (Figure S6). River export is the primary source of coastal nutrient inputs (Figure 2).

The causes of changing TN:TP ratios in nutrients exported by rivers are many and influenced by changes in the agricultural and sewage systems (Figure S7). The nutrient ratio in the river export is the result of the element ratios in human food, inputs in agricultural and natural ecosystems, and a series of biogeochemical filters during the transport from land to sea. In landscapes, there is the soil−plant filter, with uptake by plants and biogeochemical processing in soils. After this first landscape filter, water containing nutrients percolates through the soil and moves to groundwater and riparian zones. During this transport, there is further biogeochemical filtering and temporary storage before the water and nutrients are discharged to surface water bodies where further differential processing takes place in streams, lakes, and reservoirs.[14]

The biogeochemistry and mobility of N and P in the environment differ. First, N has many gaseous forms, which can escape to the atmosphere via denitrification, anammox, and ammonia volatilization[45] part of which ends in aquatic ecosystems after redeposition. P lacks a significant gaseous form. Second, N is mobile in soils, and a significant part is lost to aquatic environments via surface runoff, erosion, and leaching to groundwater and surface water. The N entering the groundwater system may be released to surface waters after a long delay, as travel times in groundwater systems vary from years to decades to centuries.[46] P is much less mobile in the soil/sediment system than N, because P interacts with particles in soil[47,48] and sediment;[49] part of the P is retained, and another part is directly available to plants.[48]

A man-made filter is the nutrient removal from wastewater in treatment plants. Wastewater discharge is an important source of N and P, since in most countries bordering the YS/BS, ECS, and SCS, the degree of sewage connection is still relatively low. Moreover, wastewater treatment in most countries bordering the three LMEs (except for Japan and the Republic of Korea) is dominated by primary and secondary systems that remove P more efficiently than N.[17] Recent improvements in sanitation of urban and rural wastewater have led to significantly declining lake P concentrations in China.[50]

The imbalance between N and P is articulated by the in-stream filter. The retention efficiency in river basins draining into the three LMEs is lower for N than that for P (Table S2). River nutrient retention was fairly constant during 1970−2010 (Table S2) as a result of simultaneous saturation due to increasing concentrations and increasing water travel time due to growing reservoir volume.[14] The slight decline of retention in rivers draining into the SCS may be related to the growth of urban centers close to the coast, where the travel time is short and the natural in-stream nutrient retention efficiency is low relative to the middle or upper reaches of the rivers.

A special case is formed by river basins with large water volumes (e.g., lakes and reservoirs) such as the Yangtze River, where P accumulated in sediment may be released when water-pollution control measures successfully reduce P concentrations,[49] when algal blooms may lead to alkalinization of the water through intensive photosynthesis,[51,52] or when hypoxic conditions are established in the bottom waters of reservoirs.[53]

Future HAB Risk in Chinese Coastal Waters. The clear TN:TP threshold value above which HABs start to develop shows that human-induced nutrient flows are essential determinants of the ecology in these coastal marine waters. For the whole of China, the TN:TP ratio of nutrients exported by Chinese rivers increased from 18 in 1970 to 29 in 2010 (Figure 4). Projections for the "business-as-usual" scenario SSP2 implemented with IMAGE-GNM indicate that the river export TN:TP ratio will not decline between 2010 and 2050 (Figure 4), with nearly constant TN and TP loadings. Generally, increasing the efficiency in agriculture to reduce nutrient losses and nutrient removal in wastewater treatment installations are obvious ways to reduce nutrient flows, as illustrated by the contrasting "sustainable" scenario SSP1. In this latter scenario, fertilizer use efficiencies are rapidly improved, and N and P fertilizer uses decline. Moreover, nutrient removal in wastewater treatment is enhanced. Although implementation of these sustainability policies leads to declining TN and TP loads, the trajectory of high TN:TP ratios will persist in the coming decades (Figure 4). Apart from the imbalance created by nutrient management in agricultural systems, the different filters in landscapes may further change the TN:TP ratio in water draining into the coastal ocean. Wastewater treatment is generally more efficient for P than for N. Furthermore, temporary accumulation of N in soils and aquifers and P sorption by sediment may cause a legacy when nutrient-reducing strategies to mitigate eutrophication will be implemented, with unpredictable outcomes for the TN:TP ratio. The TN:TP ratios will continue to increase in SSP3−SSP5 and most rapidly in the "traditional development" scenario SSP5 (Figure S8).

The high ambition of China's government to improve coastal water quality is reflected by the 13th Five-Year Plan for

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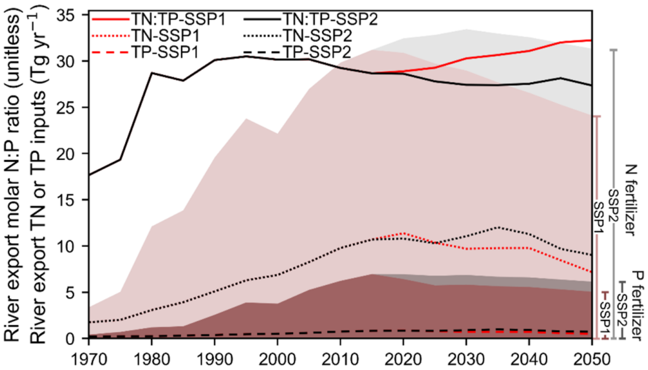

Figure 4. Projected molar TN:TP ratios and TN and TP loadings in river export from China for the period 2010−2050 according to the Shared Socioeconomic Pathways (SSPs), the business-as-usual scenario SSP2 and sustainable scenario SSP1[54] implemented with IMAGE-GNM. The average molar TN:TP ratio in river export increased from 18 in the year 1970 to 30 in 2000 and since then showed a slight decline to 29 in 2010. A further slight decline is projected for the period 2010−2050 towards a value of 27 in SSP2 and an increase to 32 in SSP1. Fertilizer use projections (in Tg N or P yr^{-1}) reflect nutrient use efficiencies that increase from 33% in 2015 to 38% (SSP2) and 46% (SSP1) in 2050. P use is based on a model that accounts for the soil residual P stock and the required production in a specific year. The outcome of the SSP1 scenario is more ambitious than the current Chinese policy to allow no increase in fertilizer use beyond 2020. For SSP3−SSP5, see Figure S8 in the Supporting Information.

Ecological & Environmental Protection, in which the goals are to maintain the area of China's coastal waters with status "excellent" (Classes I and II) at around 70% and improve all regions with status worse than Class V to a lower class.[55,56] However, it is not clear whether nutrient ratios are considered in these policy goals. The mismatch of N (high mobility in soils, groundwater memory, atmospheric transport pathway, and low retention in river basins) and P (low mobility in soils, no gaseous forms, more efficient removal of P than N in wastewater treatment, and higher P than N retention in river basins) is primarily related to the difference in their biogeochemical cycles. This turns the HAB problem into a very complex one for environmental managers and policy makers, since the task involves reductions in nutrient loads while maintaining also a healthy balance between the nutrients N and P from all sources combined. A multidisciplinary approach is required that can generate solutions that differ from the middle of the road technological options such as traditional sewage systems and wastewater treatment. Collection of human urine, which contains most N and a large part of P, and its reuse in agriculture could revitalize the traditional recycling of human waste in agriculture.[57] In any case, the reduction of N in waste flows becomes ever more pressing.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.1c00012.

Material and methods used for this study; spatial distribution of changes in TN and TP inputs to the coastal waters of the three LMEs between 1970 and 2010; HAB characteristics in Chinese coastal waters between 1970 and 2010; relationship of the aggregated molar TN:TP ratio and number of reported HABs of per dominant group per year in Chinese coastal waters for 1970−2010; geographical and hydrodynamic conditions of the three LMEs; molar DIN:DIP ratio measured in the surface water in Chinese seas during the 1980s-1990s and 2004−2007; molar TN:TP ratio in inputs from atmospheric deposition, mariculture, and river export to the coastal waters of the three LMEs for 1970−2010; coastal sources of TN and TP for the three LMEs between 1970 and 2010; TN:TP in river export from China until 2050 projected in the five SSPs; relationship between aggregated TN and TP inputs to coastal waters and number of reported HAB events in Chinese coastal waters for 1970−2010; relationship between aggregated TN and TP inputs to coastal waters and observed annual Chl-*a* concentrations in the coastal waters of the three LMEs for 1978−2010; spatial distribution of pearson correlation coefficient for the relationship between TN and TP inputs and observed annual Chl-*a* concentrations during 1978−2010; relationship between TN:TP molar ratio and HABs and Chl-*a*; representative rivers draining into the three LMEs bordering the coast of China and neighboring Asian countries; and changes in TN and TP retention in river basins draining to the three LMEs (PDF)

Data of TN, TP, and TN:TP in inputs to the coastal waters of the three LMEs; HAB frequency and area in Chinese coastal waters for the period 1970−2010; and projected river TN, TP, and TN:TP export in SSPs for the period 1970−2050. (ZIP)

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https://dx.doi.org/10.1021/acs.estlett.1c00012
Environ. Sci. Technol. Lett. 2021, 8, 276−284

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was supported by funding from the China Scholarship Council Grant (No. 201806330024) and Earth and life sciences (ALW) Open Programme 2016 project (No. 476 ALWOP.230) financed by The Netherlands Organization for Scientific Research (NWO) (J.W.), Green Card Talents project (No. 841912031) financed by Ocean University of China (A.F.B.), PBL Netherlands Environmental Assessment Agency through in-kind contributions to The New Delta 2014 ALW projects (No. 869.15.015 and No. 869.15.014) (A.F.B. and A.H.W.B.), Horizon 2020 Framework Programme (VERIFY, No. 776810) (F.D.), China Scholarship Council Grant (No. 201806190149) (Y.Y.), The Netherlands Earth System Science Center (J.J.M.), U.S. National Science Foundation (No. DEB 1911349) (P.M.G.), National Natural Science Foundation of China projects (No. 41776089 and No. 41930862) (X.R.), and Strategic Priority Research Program of Chinese Academy of Sciences (CAS, No. XDA19060203) (R.Y.). We thank the reviewers for constructive comments and suggestions that have helped to improve this paper.

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https://dx.doi.org/10.1021/acs.estlett.1c00012
Environ. Sci. Technol. Lett. 2021, 8, 276−284