Earth's Future

RESEARCH ARTICLE
10.1029/2020EF001516

Special Section:
Quantifying Nutrient Budgets for sustainable nutrient management

Spatially Explicit Inventory of Sources of Nitrogen Inputs to the Yellow Sea, East China Sea, and South China Sea for the Period 1970–2010

Junjie Wang[1,2], Arthur H. W. Beusen[2,3], Xiaochen Liu[2], Rita Van Dingenen[4], Frank Dentener[4], Qingzhen Yao[1], Bochao Xu[1], Xiangbin Ran[5], Zhigang Yu[1], and Alexander F. Bouwman[1,2,3]

[1]Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, Qingdao, China, [2]Department of Earth Sciences—Geochemistry, Faculty of Geosciences, Utrecht University, Utrecht, The Netherlands, [3]PBL Netherlands Environmental Assessment Agency, The Hague, The Netherlands, [4]European Commission, Joint Research Centre (JRC), Ispra, Italy, [5]Ministry of Natural Resources, First Institute of Oceanography, Qingdao, China

Key Points:
- Since 1970, N inputs to Chinese seas increased rapidly due to rapidly increasing river export, atmospheric deposition, and mariculture
- River export is the largest source, with a dominant contribution from agriculture and recently growing shares of aquaculture and sewage
- N inputs are spatially concentrated and increase much faster in Chinese coastal waters than in other countries

Supporting Information:
- Supporting Information S1
- Table S1
- Table S2
- Movie S1
- Movie S2

Correspondence to:
A. F. Bouwman,
Lex.Bouwman@pbl.nl

Citation:
Wang, J., Beusen, A. H. W., Liu, X., Van Dingenen, R., Dentener, F., Yao, Q., et al. (2020). Spatially explicit inventory of sources of nitrogen inputs to the Yellow Sea, East China Sea, and South China Sea for the period 1970–2010. *Earth's Future*, *8*, e2020EF001516. https://doi.org/10.1029/2020EF001516

Received 11 FEB 2020
Accepted 11 AUG 2020
Accepted article online 17 AUG 2020

Abstract Symptoms of eutrophication (including biodiversity loss, harmful algal blooms, and hypoxia) are an increasing problem in Chinese seas. Nutrient enrichment is primarily caused by accelerated human activities that cause nutrient pollution of the aquatic environment. In this study, the Integrated Model to Assess the Global Environment–Global Nutrient Model (IMAGE-GNM) was used to estimate nitrogen inputs from river discharge, submarine fresh groundwater discharge, and mariculture, and TM5-FAst Scenario Screening Tool (TM5-FASST) for atmospheric nitrogen deposition to the three Large Marine Ecosystems (LMEs, i.e., Yellow Sea/Bohai Sea, YS/BS; East China Sea, ECS; South China Sea, SCS) bordered by China and several other countries for the period 1970–2010. China's river nitrogen export was the largest nitrogen source in YS/BS and ECS. In SCS, however, China and other countries contributed equally and although decreasing, the proportion of natural sources remain considerable. The total nitrogen inputs to YS/BS (1.0 to 4.1 Tg year^{-1}), ECS (1.3 to 5.5 Tg year^{-1}), and SCS (2.1 to 5.8 Tg year^{-1}) increased rapidly during 1970–2010. River export is dominated by agriculture; nitrogen inputs from atmospheric deposition and mariculture have been increasing rapidly in recent years. Considering only the coastal zone of the three LMEs, our results show that the total nitrogen inputs are strongly concentrated spatially in areas close to river mouths and those confined regions with mariculture production. To sustain the food production and economic growth in the coming decades, nitrogen inputs may increase further, depending on future eutrophication mitigation policies.

Plain Language Summary Nitrogen is a limiting nutrient for plant production. Excessive nitrogen use in agriculture and discharge from wastewater is the primary causes of eutrophication in aquatic environments. Symptoms of eutrophication (including biodiversity loss, harmful algal blooms (HABs), and hypoxia) are an increasing problem in Chinese seas. Here we quantified the nitrogen inputs from river export, atmospheric deposition, submarine fresh groundwater discharge, and mariculture to the Yellow Sea/Bohai Sea (YS/BS), East China Sea (ECS), and South China Sea (SCS) bordered by China and other countries for the period 1970–2010. Nitrogen inputs increased rapidly, mainly due to increasing land-based sources (river export and atmospheric deposition), while nitrogen from mariculture started to increase recently. River export is dominated by agriculture with growing proportions of sewage and freshwater aquaculture. Nitrogen inputs were spatially concentrated and increased faster in Chinese coastal waters than in other countries. China's contribution to nitrogen pollution exceeded that of other countries in YS/BS and ECS, while China and other countries contributed equally in SCS. Nitrogen concentrations and HAB frequency in the seas seem to be correlated as both increased substantially since 1970s. Mitigation of nitrogen pollution is therefore urgent because human activities that trigger nitrogen discharge may increase further in future.

1. Introduction

Nitrogen is an essential element for all organisms. It is a limiting nutrient for primary production in both terrestrial and aquatic ecosystems (Ohyama, 2010). However, excessive nitrogen is one of the primary factors causing eutrophication in both freshwaters (Conley et al., 2009; Han et al., 2014) and marine environments (Strokal et al., 2014; Wang et al., 2018). The global biogeochemical cycle of nitrogen has been accelerated by anthropogenic activities (Fowler et al., 2013; Galloway et al., 2004). Important nitrogen sources in inland surface waters are agriculture (Bouwman et al., 2009; Bouwman, Klein Goldewijk, van der Hoek, et al., 2013; de Wit et al., 2002), aquaculture (Wang et al., 2020), and domestic and industrial wastewater (Morée et al., 2013; van Puijenbroek et al., 2019) due to increasing population and rapid economic development over the past decades. Between 1900 and 2000, the total nitrogen delivery to global freshwater systems increased from 34 to 64 Tg N year^{-1}, and the total nitrogen export from rivers to global coastal marine ecosystems increased from 19 to 37 Tg N year^{-1} (Beusen et al., 2016).

China has the largest population of all countries in the world (Food and Agriculture Organization [FAO], 2019a), reaching 1.46 billion inhabitants in 2018 (Figure S1a) which is 20% of the global population (FAO, 2019a). To feed such a large and rapidly increasing population, the production of crops, livestock, and aquaculture fish have been boosting during the past 50 years (Figure S1b, FAO, 2019a, 2019b). China's national wastewater discharge also increased from 34 to 70 Gt year^{-1} during 1985–2017 (National Bureau of Statistics of China, 1986). Consequently, the nitrogen discharge to aquatic environments increased dramatically due to the intense human activities. The total nitrogen discharge to the Yellow Sea and East China Sea by the Yangtze River increased from 0.3 Tg N year^{-1} in 1900 to 5.9 Tg N year^{-1} in 2010 (Liu, Beusen, et al., 2018). The nitrogen concentration in Chinese coastal seas has been increasing over the past four decades (Jiang et al., 2010; Ning et al., 2009; Wang et al., 2018; Wang et al., 2019; Wei et al., 2015; Yang et al., 2018). Many negative impacts may be associated with nitrogen pollution, such as the frequent occurrence of eutrophication-related harmful algal blooms (HABs) (Liu, Pang, et al., 2013; Wang et al., 2008; Wang et al., 2018; Xiao et al., 2019; Yu et al., 2018), loss of biodiversity (Chang et al., 2012; Li et al., 2010), hypoxia (Chen et al., 2007; Wang et al., 2016), and acidification (Cai et al., 2011). The world's largest green tide occurred in the Yellow Sea every year since 2007 (Liu, Keesing, et al., 2013; Wang et al., 2015). The HAB frequency in Chinese seas increased from almost 0 to over 100 events year^{-1} in the early 2000s and remained high since then (Wang et al., 2018; Xiao et al., 2019).

To develop mitigation strategies for nitrogen pollution and improve the water quality in Chinese seas, it is essential to know the spatial and temporal distribution and long-term changes of all external nitrogen. Consistent long-term quantitative estimates of nitrogen inputs from all external sources to Chinese seas are not available. The long-term changes in nitrogen export are known for a few rivers, such as the Yangtze River (Liu, Du, et al., 2018) and Yellow River (Fan & Huang, 2008; Liao et al., 2013; Ma et al., 2004). However, knowledge is lacking on the long-term changes in nitrogen export by other rivers draining to Chinese coastal seas. Furthermore, rivers are not the only nutrient sources. Re-deposition in China's seas of the rapidly increasing NO_x and NH_3 emission from China (Lü & Tian, 2007; Ohara et al., 2007) is becoming an increasingly important nitrogen source (Luo et al., 2014; Shou et al., 2018; Zhang et al., 2010). There is evidence that locally nitrogen from submarine fresh groundwater discharge is responsible for the high nitrogen concentration observed in various parts of China's coastal seas (Liu, Du, et al., 2017; Liu, Su, et al., 2017). Finally, the rapidly developing mariculture is now comparable to river export in some parts of China's coastal seas (Wang et al., 2020).

In this paper we present long-term estimates of the nitrogen inputs to seas from land-based sources including river export, atmospheric deposition, submarine fresh groundwater discharge, and mariculture. The nitrogen dynamics due to water exchange (e.g., ocean currents) and water-sediment exchange in the seas were not considered. We used the Integrated Model to Assess the Global Environment–Global Nutrient Model (IMAGE-GNM) (Beusen et al., 2015, 2016; Bouwman, Beusen, Glibert, et al., 2013; Wang et al., 2020) to estimate the spatial distribution of annual nitrogen inputs from river discharge, submarine fresh groundwater discharge and mariculture to the three Large Marine Ecosystems (LMEs, i.e., Yellow Sea/Bohai Sea, YS/BS; East China Sea, ECS; South China Sea, SCS, Figure 1) that border the coasts of China and several other countries for the period 1970–2010. Estimates of atmospheric nitrogen deposition to the seas were based on TM5-FAst Scenario Screening Tool (TM5-FASST) (Figure 2). Within river nitrogen export,

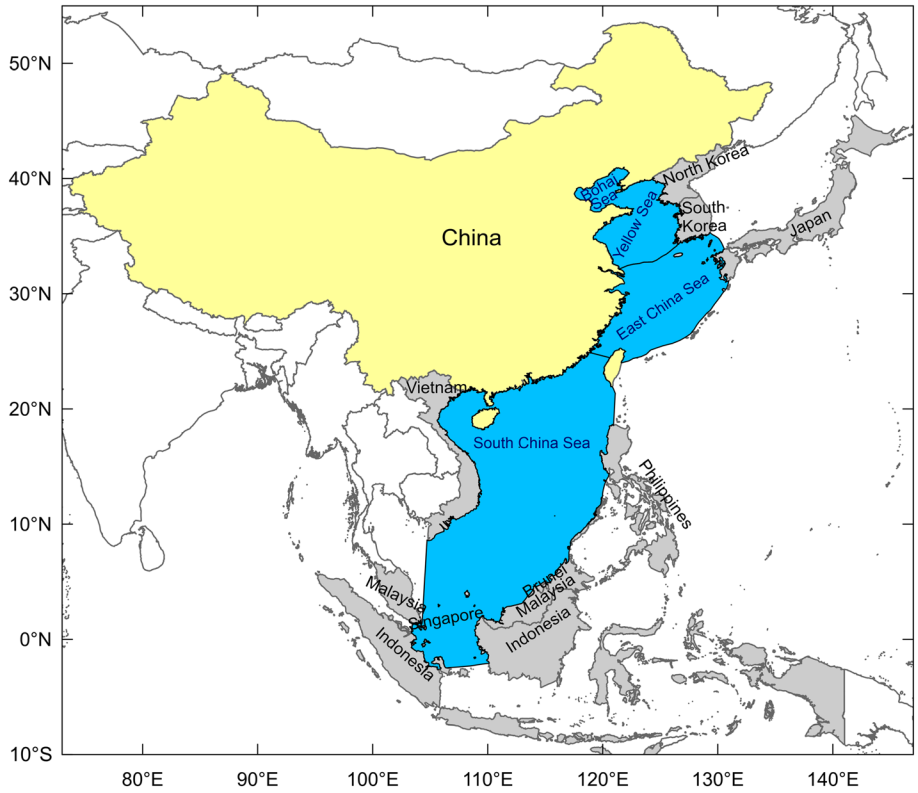

Figure 1. Location of the Yellow Sea/Bohai Sea, East China Sea, and South China Sea. The yellow area represents China, and the gray areas represent all countries that border the three LMEs.

IMAGE-GNM keeps track of the contributions of different natural and anthropogenic sources such as agriculture, aquaculture, and wastewater (Figure 2). This paper has online supporting information.

2. Methods and Data Used

2.1. Nitrogen River Export and Submarine Fresh Groundwater Discharge

The IMAGE-GNM was used to simulate the nitrogen delivery from all known sources to surface water (hereinafter referred to as "delivery") and river nitrogen export (hereinafter referred to as "export") as well as submarine fresh groundwater discharge to the YS/BS, ECS, and SCS. Coupling the integrated assessment model IMAGE (Stehfest et al., 2014) with the global hydrological model PCR-GLOBWB (Van Beek et al., 2011), the IMAGE-GNM model (Beusen et al., 2015) is a spatially explicit, distributed modeling framework for simulating the delivery of nitrogen and phosphorus to surface water from various sources with a spatial resolution of 0.5 by 0.5 degrees (Figure S2). IMAGE-GNM includes diffuse nitrogen delivery from agricultural and natural ecosystems based on nitrogen budgets including input terms (fertilizer use, animal manure, biological N_2 fixation, and atmospheric deposition) and output terms (crop/grass uptake and gaseous emission). Transport pathways include surface runoff, leaching from soils to shallow and deep groundwater, and via riparian zones finally to surface water. IMAGE-GNM calculates direct discharge from point sources (wastewater from sewage systems), freshwater aquaculture, allochthonous organic material from vegetation in floodplains, and atmospheric deposition to streams. Submarine fresh groundwater discharge calculated by IMAGE-GNM (Beusen et al., 2013) is the groundwater and nitrate outflow for all coastal land by shallow and deep aquifers in a narrow zone along the coast.

IMAGE-GNM uses data on crop and livestock production and fertilizer use from Chinese provincial statistics (China Livestock Yearbook Editing Committee, 2014; China Ministry of Agriculture, 2014; National Bureau of Statistics of China, 2014) and national data for the other countries from FAO (2019a) as described

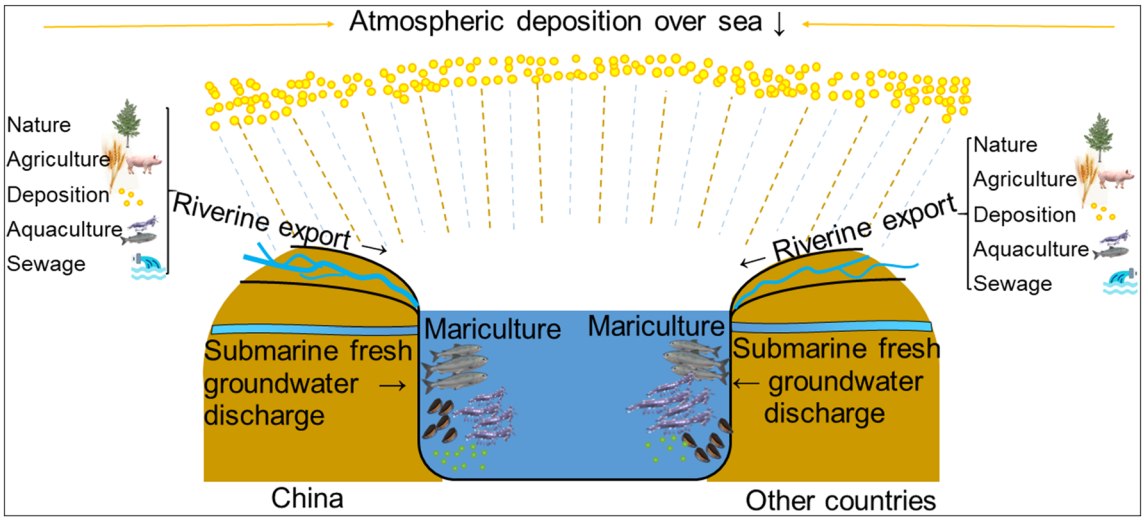

Figure 2. Scheme for nitrogen inputs from all external sources to seas.

by Bouwman et al. (2017) for agriculture. Aquaculture is discussed separately in section 2.2. Wastewater flows are based on national data as described by van Puijenbroek et al. (2019). All other soil, land use, lithological, climate, and environmental data are described by Beusen et al. (2015). Land use distributions are from the IMAGE model (Stehfest et al., 2014). Wastewater discharge is spatially distributed using population densities (van Puijenbroek et al., 2019).

The nutrient spiraling method (Newbold et al., 1981; Wollheim et al., 2008) is employed to calculate the in-stream nitrogen retention. More details on the IMAGE-GNM framework, applications, and sensitivity analyses can be found elsewhere (Beusen et al., 2015, 2016; Liu, Beusen, et al., 2018). Here, we validated the model results of total nitrogen and discharge in the three largest river basins in China, that is, the Yangtze River, Pearl River, and Yellow River (see Table S1 and Figure S3).

The location of YS/BS, ECS, and SCS is shown in Figure 1, and their areas, number of the rivers that drain into them, and other characteristics are in Table S2. We classify rivers draining to an LME into two classes according to the location of the river mouths, that is, rivers from China and "other countries", and we account for the whole river basin even if a country border has been crossed. Since the Yangtze River debouches at the border of YS and ECS, we assume that a proportion of 14% of the nitrogen discharge ends in the YS/BS and 86% in ECS following Liu et al. (2016).

2.2. Nitrogen Release From Aquaculture

The IMAGE-GNM aquaculture nutrient budget model developed by Bouwman et al. (2011), Bouwman, Beusen, Overbeek, et al. (2013), and Bouwman, Beusen, Glibert, et al. (2013) was used to calculate the nitrogen release from freshwater aquaculture in all river basins draining into the three LMEs and mariculture (i.e., aquaculture in brackish and marine environments) within the three LMEs with a 0.5° spatial resolution. The aquaculture systems include finfish (omnivores and carnivores), mollusks (including suspension-feeding bivalves and gastropods), crustaceans, and algae in freshwater, brackish, and marine environments. The International Standard Statistical Classification of Aquatic Animals and Plants is used to group the various species. The nutrient flows described in the system include nutrients in feed inputs and filtered suspended matter, feed conversion, harvested fish, outflows in the form of particulate feces and dissolved nutrients, apparent digestibility, and recycling of pond sediment. Aquaculture production data per country, species group, sea area, and type of environment are available in units of live weight for the period 1970–2010 from the FISHSTAT database (FAO, 2012). Aquaculture is spatially distributed within provinces or countries on the basis of water body type, population density, and temperature for inland and brackish aquaculture and coastal type, coastline length, and temperature as described by Wang et al. (2020). The model scheme is in Figure S4. More details can be found in Bouwman et al. (2011), Bouwman, Beusen, Glibert, et al. (2013), and Bouwman, Beusen, Overbeek, et al. (2013).

2.3. Deposition Over Sea

TM5-FASST is a global reduced-form air quality source-receptor model that computes ambient pollutant concentrations with a 1 by 1 degree resolution. (Van Dingenen et al., 2018). TM5-FASST is based on linearized emission-concentration and deposition sensitivities derived with the three-dimensional global atmospheric chemical transport model TM5 that simulates transport, chemical processes, and wet and dry deposition of chemically active atmospheric trace gases (e.g., O_3, SO_2, NO_x, and NH_3) and particulate components (e.g., SO_4^{2-}, NO_3^-, NH_4^+, primary $PM_{2.5}$, and its components black carbon and organic carbon) for a single meteorological year (2001) and emission inventory (RCP year 2000) (Van Dingenen et al., 2018). We used national data on NO_x and NH_3 emissions from EDGAR (2019) and aggregated them to 56 regional levels to drive TM5-FASST. Since NO_x and NH_3 are taken as the main components for total nitrogen from atmospheric deposition (Galloway et al., 2004), we took the sum of the simulated NO_x and NH_3 concentrations as the total nitrogen concentration. Data were converted to 0.5 by 0.5 degree resolution by interpolation.

3. Results

3.1. River Nitrogen Export

During 1970–2010, the nitrogen export by rivers increased rapidly to all LMEs (from 0.8 to 3.3 Tg year^{-1} to YS/BS; 1.0 to 4.6 Tg year^{-1} to ECS; 1.4 to 3.8 Tg year^{-1} to SCS; see Figure 3). Since the natural sources of nitrogen remained almost unchanged during the four decades, their proportions decreased as the total river nitrogen export continuously increased. For YS/BS and ECS, the natural sources were less dominant than other sources; however, for the nitrogen export to SCS from rivers in other countries, the natural sources account for 27–51% with a dominant contribution of vegetation in floodplains (19–37%). Agriculture was the dominant source of nitrogen from river export to the three LMEs (69% for YS/BS, 75% for ECS, and 55% for SCS in 1970). With a stagnating agricultural source of nitrogen since the mid-1990s, its contribution to YS/BS and ECS decreased to 58% and 70%, respectively (Figure 3). Nitrogen discharge from wastewater increased by a factor of 9.5, 10.9, and 10.3 in YS/BS, ECS, and SCS between 1970 and 2010, respectively, while the nitrogen release from freshwater aquaculture increased dramatically by a factor of 54 in YS/BS, 50 in ECS, and 13 in SCS (Figure 3). Nitrogen from freshwater aquaculture in China increased faster than that in other countries for ECS and SCS. The contribution of nitrogen from atmospheric deposition over inland waters was only 0.1–1.1% for all LMEs during 1970–2010. In 2010, the combined contribution from wastewater, aquaculture, and atmospheric deposition to total river nitrogen export reached 38%, 26%, and 21% for YS/BS, ECS, and SCS, respectively, which is comparable to the contribution of agriculture and partly accounts for the decreasing proportion of the agricultural nitrogen source in YS/BS and ECS.

China's contribution to total river export was dominant for ECS (increasing from 89% in 1970 to up to 97% in 2010) and YS/BS (increasing from 64% in 1970 to up to 90% in 2010), while river export from other countries remained stable and their contribution decreased. Since the river export from China to SCS increased faster than that from other countries, the contribution from China (51%) and other countries (49%) were similar in 2010.

Comparison of the nitrogen delivery to surface water within the river basins (see Figure S5) and river export to the LMEs (Figure 3) indicates that the fractions of nitrogen from natural sources were larger in the delivery than in the export to the LMEs. In contrast, the fractions of nitrogen from wastewater and aquaculture were larger in the river export than in the delivery for the three LMEs.

3.2. Other Nitrogen Inputs to the Three LMEs

Atmospheric nitrogen deposition in the YS/BS, ECS, and SCS continuously increased during 1970–2010 (Figures S6a–S6c). Atmospheric nitrogen deposition in YS/BS increased most rapidly by more than a factor of 4 during 1970–2010, while that of ECS and SCS increased by about 3 times.

During 1970–2010, the nitrogen release from mariculture increased rapidly from almost 0 to 0.1 Tg year^{-1}, 0.1 Tg year^{-1}, and 0.2 Tg year^{-1} in YS/BS, ECS, and SCS, respectively, and the increases were very rapid after the mid-1990s (Figures S6d–S6f). China's contribution to the nitrogen release from mariculture was much larger than that from the other countries in all three LMEs in 2010.

10.1029/2020EF001516

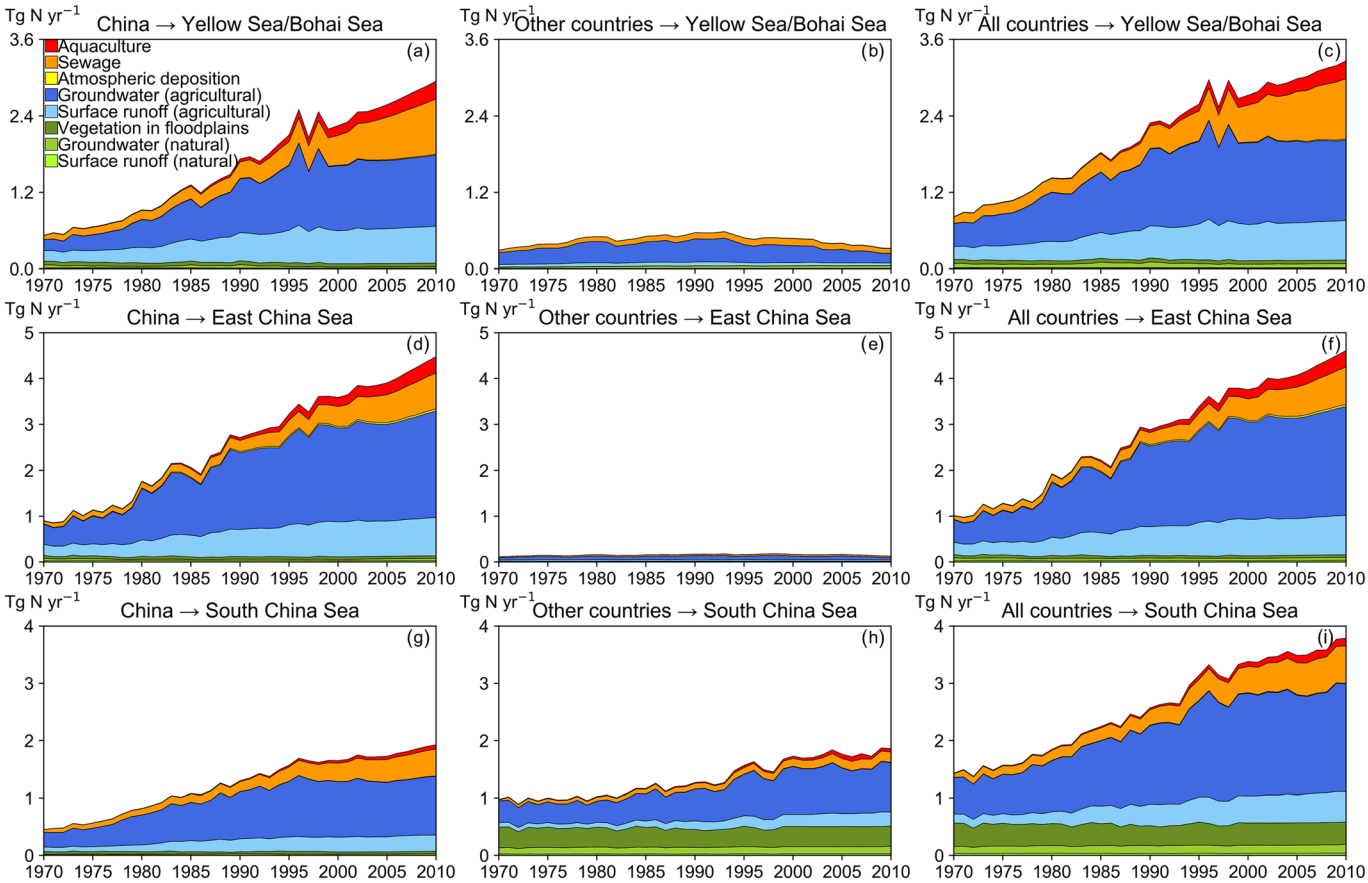

Figure 3. Contribution of land-based sources of nitrogen in the river discharge during 1970–2010 to YS/BS (a–c), ECS (d–f), and SCS (g–i). Left column is data for China, middle column is data for the other countries, and the right column presents data for the total.

Submarine fresh groundwater discharge is a small contribution to the external nitrogen inputs to the three LMEs (Figure 4), with roughly half coming from China, and with an increasing trend in SCS and stable nitrogen discharge in recent years in YS/BS and ECS (Figures S6g–S6i).

The total nitrogen inputs more than doubled from 1.0 to 4.1 Tg year^{-1} (YS/BS), 1.3 to 5.5 Tg year^{-1} (ECS), and 2.1 to 5.8 Tg year^{-1} (SCS) between 1970 and 2010 (Figure 4). River export was the dominant source, with a larger contribution in YS/BS (80–84%) and ECS (77–84%) than in SCS (64–69%) during 1970–2010 (Figure 4). The contribution from atmospheric deposition to total external nitrogen inputs was much more important in SCS (29–32%, fluctuating trend) than in YS/BS (14–16%, increasing) and ECS (15–22%, decreasing). In 1970, submarine fresh groundwater discharge (1–2%) and mariculture (<0.2%) contributed small amounts to the external nitrogen inputs to the three LMEs. However, since the nitrogen from submarine fresh groundwater discharge was relatively stable, while the total nitrogen inputs increased rapidly, the contribution from submarine fresh groundwater discharge decreased. Meanwhile, the contribution from mariculture increased rapidly to 1–3% and exceeded that of submarine fresh groundwater discharge in 2010.

We also calculated the external inputs from river export, atmospheric deposition, mariculture, and submarine fresh groundwater discharge for the coastal cells in the three LMEs where the inputs occur (Figure 5). Between 1970 and 2010, the total nitrogen inputs to coastal waters increased dramatically in YS/BS, ECS, and SCS from 0.9 to 3.8 Tg year^{-1}, 1.1 to 5.0 Tg year^{-1}, and 1.7 to 4.7 Tg year^{-1}, respectively. River export constituted >80% of the total inputs to coastal waters (87–90% for YS/BS, 90–93% for ECS, and 80–87% for SCS). Atmospheric deposition in the coastal waters more than tripled between 1970 and 2010 and accounted for 8–9%, 6–9%, and 11–15% of the total nitrogen inputs to YS/BS, ECS, and SCS, respectively. As the total nitrogen inputs increased, the fractions of mariculture increased from insignificant to an amount

10.1029/2020EF001516

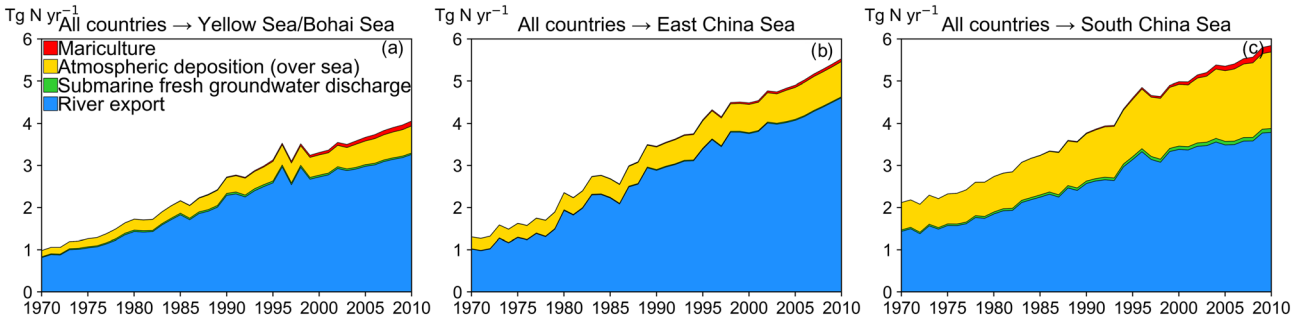

Figure 4. Total nitrogen inputs to (a) YS/BS, (b) ECS, and (c) SCS from river export, atmospheric deposition, mariculture, and submarine fresh groundwater discharge during 1970–2010.

comparable to that of atmospheric deposition. The increase in the total nitrogen inputs to the Chinese coastal waters was much faster than that in the other countries.

4. Discussion

4.1. River Nitrogen Delivery and Export

By comparing the total nitrogen delivered by rivers (Figure S5) and their export to LMEs (Figure 3), we see major differences in the in-stream retention (22–27% to YS/BS, 28–38% to ECS, and 24–31% to SCS). During 1970–2010, the in-stream retention increased for YS/BS but decreased for ECS and SCS. With larger retention load (defined as the sum of nitrogen load in rivers, lakes, and reservoirs which is removed from the water column due to removal processes [i.e., retention]; Beusen et al., 2015) in lakes and reservoirs, the in-stream retention load in river basins draining into ECS is larger than in those draining into YS/BS and SCS (Figure S7).

Another factor determining the overall retention is the location of the nitrogen sources such as discharge from wastewater and freshwater aquaculture, which are larger in river export than in the delivery for the three LMEs. This is very much related to the location of these discharges. Many large Chinese cities are located in coastal regions, where the distance to the sea is much smaller than for inland population centers. As a consequence, retention fractions are smaller, and the contribution of wastewater to river export is larger. Aquaculture activities are very much concentrated in a few provinces in the eastern part of China, so transport distances are relatively small and retention therefore limited.

4.2. Nitrogen Inputs to Coastal Waters

The nitrogen inputs to the coastal waters are strongly concentrated to areas close to river mouths and areas with intense mariculture (Figures 6 and S8), which is consistent with the spatial distribution of eutrophication status and seawater quality status of inorganic nitrogen in Chinese seas in 2018 (Figure S9). Between 1970 and 2010, nitrogen inputs to the coastal areas of the three LMEs from riverine export, atmospheric deposition, mariculture, and submarine fresh groundwater discharge showed prominent increases both for the total (Figure 5) and the spatial distributions (Figures 6 and S8), and the increases were faster in China's coastal waters than in those of the other countries bordering the three LMEs. The YS/BS shows a high intensity and concentration of nitrogen inputs in coastal waters of all countries (Figures 6 and S8). Riverine export and atmospheric deposition are responsible for most of the nitrogen inputs in most coastal areas (Figures 6 and S8). However, with the rapid increase in fish and shellfish production in marine waters particularly in China (Wang et al., 2020), mariculture is now locally as important as riverine export and atmospheric deposition (Figure S8).

4.3. Comparison With Observations and Literature

Several estimates of dissolved inorganic nitrogen (DIN) export by rivers to the LMEs were based on observations (Table 1), including 0.4 Tg N year^{-1} for the YS in late 1990s (Liu et al., 2003), 0.1 Tg N year^{-1} for the BS in the late 2000s (Liu et al., 2011), and 0.5 Tg N year^{-1} for the YS/BS in the early 1990s (Wang et al., 2002). Using the ratio of DIN concentration:total nitrogen (TN) concentration of 0.8 in the Yellow River (Tan, 2002)

10.1029/2020EF001516

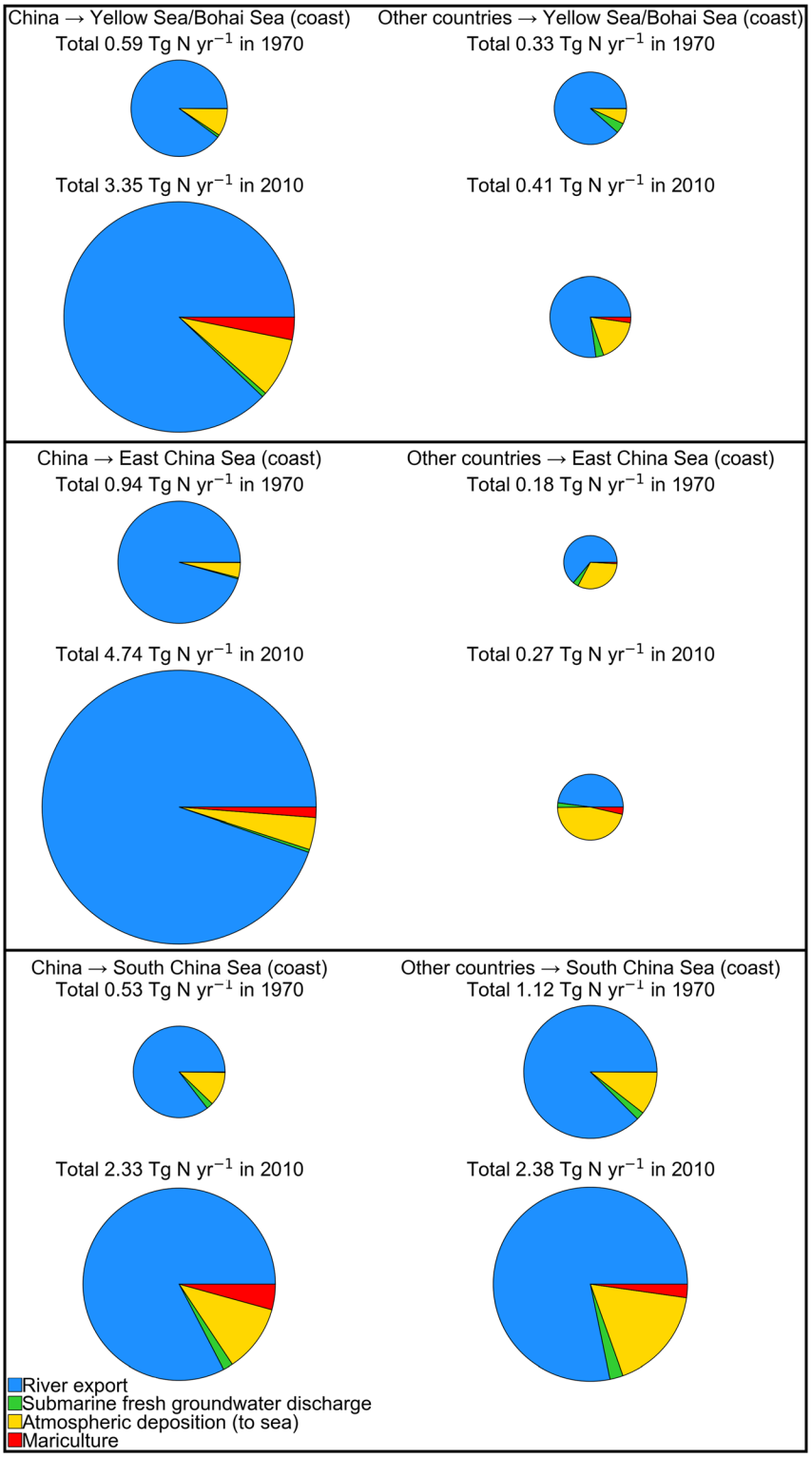

Figure 5. Total nitrogen inputs to the coastal waters of (a) YS/BS, (b) ECS, and (c) SCS from river export, atmospheric deposition, mariculture, and submarine fresh groundwater discharge in 1970 and 2010.

10.1029/2020EF001516

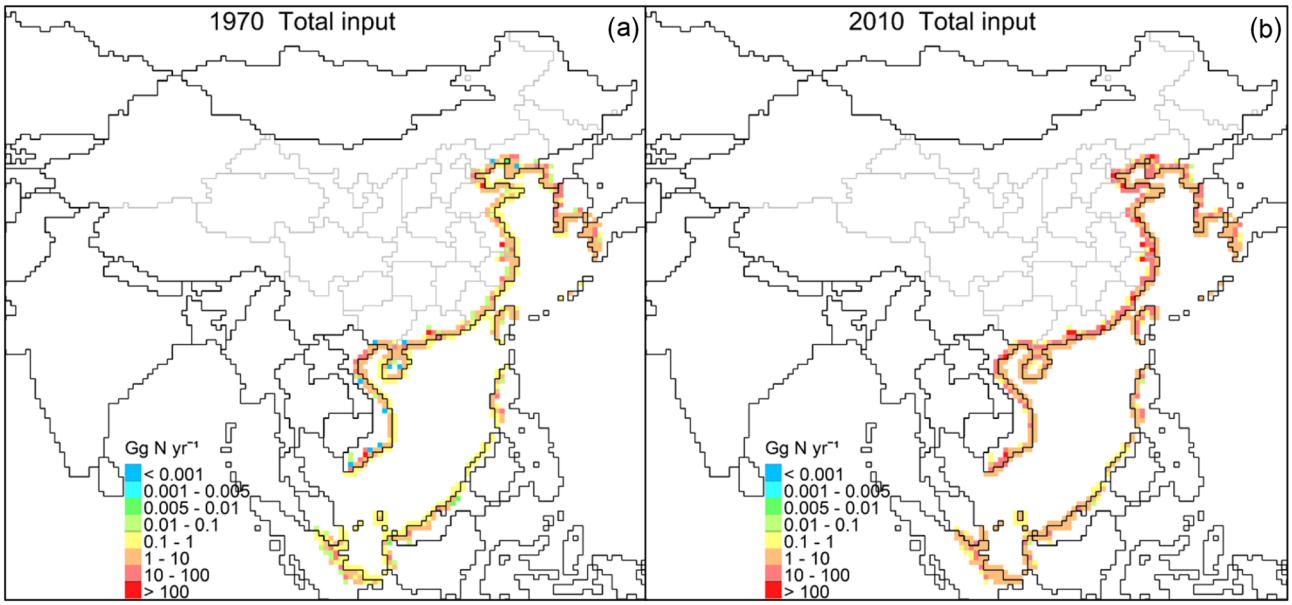

Figure 6. Spatial distribution of nitrogen inputs from all external sources (including river export, atmospheric deposition, submarine fresh groundwater discharge, and mariculture) to the coastal areas of YS/BS, ECS, and SCS in (a) 1970 and (b) 2010.

and of 0.5 for the Yangtze River (Yan et al., 2001; Zhang, 1990), the observation-based estimate of total nitrogen from riverine export to YS/BS was 0.6–1.0 Tg N year^{-1} during the early 1990s to late 2000s, which is lower than our modeled 2–3 Tg year^{-1} during the same period. Li et al. (2014) estimated that the DIN export by the Yangtze River increased from 0.8 to 3.0 Tg N year^{-1} during the years 1980–2006. Using the DIN:TN ratio of 0.5, the increase of total nitrogen export would be from 1.6 to 6.0 Tg N year^{-1}, which is close to our modeled increase of nitrogen export from Chinese rivers to YS/BS and ECS together from 3 to 7 Tg N year^{-1} during the same period. The underestimation of measurement-based results is mainly because they only accounted for a few rivers while the nitrogen export by other rivers was not included.

The estimated nitrogen input from submarine fresh groundwater discharge in the Pearl River Estuary is 0.02 Tg year^{-1} for 2014–2015 (Liu, Du, et al., 2018), which is comparable to our simulated total nitrogen from China's total submarine fresh groundwater discharge of 0.04 Tg year^{-1} to SCS for 2010 (Table 1). Our estimation of nitrogen release from Chinese mariculture of 0.27 Tg year^{-1} in 2010 is close to 0.18 Tg year^{-1} by X. Zhang et al. (2015), and the difference lies in the incomplete list of species and the use of aggregated nutrient use efficiencies adopted by X. Zhang et al. (2015) as discussed in Wang et al. (2020).

The difference between our model results and measurement-based estimates of nitrogen from atmospheric deposition may be the result of scale issues (e.g., spatial and temporal variability that is not captured when the number of sampling sites or the period covered by measurements is limited). Despite this, the measurement-based estimation of nitrogen from atmospheric deposition of 0.4 Tg year^{-1} to YS/BS during 1992–1998 (Wang et al., 2002) almost equals our model-based result of 0.4–0.5 Tg year^{-1} for the same years, while the estimate of 1.0 Tg year^{-1} during 1994–1997 (Bashkin et al., 2002) is also comparable (Table 1). The measurement-based estimation of 0.21 Tg year^{-1} to YS/BS and 0.43 Tg year^{-1} to ECS during 1999–2003 are comparable to our modeled 0.48–0.51 Tg year^{-1} and 0.66–0.70 Tg year^{-1} during the same period (Zhang et al., 2007). Our modeled result of 0.7 Tg year^{-1} to ECS in 2000–2001 is close to the reported 0.9 Tg year^{-1} (Wan et al., 2002). The 0.52 Tg year^{-1} to YS/BS during 2008–2011 measured by Qi et al. (2018) is close to our modeled ~0.6 Tg year^{-1} during the same period. Our modeled 0.65 Tg year^{-1} to YS/BS and 0.83 Tg year^{-1} to ECS in 2010 respectively match the ranges of 0.37–2.1 Tg year^{-1} and 0.64–3.7 Tg year^{-1} during 2014–2015 measured by Luo et al. (2018).

Table 1
Comparison of the Measurement-Based and Modeled Nitrogen Inputs From River Export, Submarine Fresh Groundwater Discharge, Mariculture, and Atmospheric Deposition to YS/BS, ECS, and SCS to YS/BS, ECS, and SCS

Source of nitrogen inputs	Large Marine Ecosystems	Measured period	Measurement-based nitrogen inputs (Tg year^{-1})	Modeled period	Modeled nitrogen inputs (Tg year^{-1})
River export	YS/BS	Early 1990s to late 2000s	0.6–1.0 (Liu et al., 2003, 2011; Wang et al., 2002)	1990–2009	2–3
	YS/BS + ECS	1980–2006	1.6–6.0 (Li et al., 2014)	1980–2006	3–7
Submarine fresh groundwater discharge	SCS	2014–2015	0.02 (Liu, Du, et al., 2018)	2010	0.04
Mariculture	YS/BS + ECS + SCS (Chinese coast)	2010	0.18 (X. Zhang et al., 2015)	2010	0.27
Atmospheric deposition	YS/BS	1992–1998	0.4 (Wang et al., 2002)	1992–1998	0.4–0.5
	YS/BS	1994–1997	1.0 (Bashkin et al., 2002)	1994–1997	0.4–0.5
	YS/BS	1999–2003	0.21 (Zhang et al., 2007)	1999–2003	0.48–0.51
	ECS	1999–2003	0.43 (Zhang et al., 2007)	1999–2003	0.66–0.70
	ECS	2000–2001	0.9 (Wan et al., 2002)	2000–2001	0.7
	YS/BS	2008–2011	0.52 (Qi et al., 2018)	2008–2010	0.62–0.65
	YS/BS	2014–2015	0.37–2.1 (Luo et al., 2018)	2010	0.65
	ECS	2014–2015	0.64–3.7 (Luo et al., 2018)	2010	0.83

4.4. Implications for Coastal Waters With Increasing Eutrophication

Since the nitrogen inputs from all the external sources increased by a factor of 4.1, 4.2, and 2.8 in YS/BS, ECS, and SCS during 1970–2010, respectively, the nitrogen concentrations in the LMEs would rise correspondingly, assuming the water volume and nitrogen retention in Chinese seas remain stable. With the increase of nitrogen input to coastal waters by a factor of 4.1 for YS/BS, 4.5 for ECS, and 2.9 for SCS, the increases in nitrogen concentrations in the coastal waters may be even faster than in the average concentrations of the whole LMEs. If we focus on nitrogen input to the coastal grid cells bordering China, the problem is even more severe (an increase by a factor of 5.7 for YS/BS, 5.0 for ECS, and 4.4 for SCS). This corresponds to the long-term changes in the DIN concentration measured in these seas: increases by >4 times in the southern YS during 1984–2006 (Wei et al., 2015), ~3 times in the northern YS during 1991–2006 (Yang et al., 2018), >2 times in the BS during 1990–2016 (Wang et al., 2019), >2 times in the Yangtze River Estuary (ECS) during 1981–2016 (Wang et al., 2018), and >5 times in the northern SCS during 1989–2004 (Ning et al., 2009).

Higher nitrogen concentrations, changed nutrient stoichiometry and increased exports of certain forms of nitrogen (e.g., ammonium and urea) can promote preferential growth of certain phytoplankton species and cause HABs if other conditions are suitable (Anderson et al., 2002; Heisler et al., 2008; Wang et al., 2019). Over the recent decades, the frequency and affected area of red tides (Figure S10) (Wang et al., 2008; Wang et al., 2019; Xie & Yu, 2007) and hypoxia (Yu & Zhang, 2016; Zhai et al., 2012; Zhang et al., 2016) in Chinese seas have been increasing rapidly. The annual frequency of HABs in China's coastal area has increased from almost 0 in 1970 to >30 year^{-1} in YS/BS, >120 year^{-1} in ECS, and >25 year^{-1} in SCS in the mid-2000s and remained high afterwards (Figure 7).

With the expected population and economic growth, increasing food production and changing human diets (increasing fish and meat consumption) in the coming decades, nitrogen inputs to the YS/BS, ECS, and SCS may increase further. This is an extremely difficult problem to solve, as a large part of the sources of nitrogen are land-based with important contributions from agriculture, urban wastewater, and aquaculture. Wastewater treatment is an effective option, but modern treatment plants with high nutrient removal efficiency are costly, and it may take years or decades to build. However, our results indicate that much of the nitrogen load is from population centers close to the coast, and an initial policy focus on these regions may be more effective than on regions more upstream. Only 24% of the nitrogen in feed are retained by the cultured species and 66% are released from China's mariculture system to coastal waters (Wang et al., 2020), which means that the feed efficiency needs to be improved by adopting formulated feed (Cao et al., 2007; Wu, 1995). The policy of zero increase in fertilizer use is a first step toward reducing nitrogen losses from agriculture (Yu et al., 2019; Y. Zhang , 2015), but more stringent reductions may be needed in view of the legacy of the nitrogen that is temporarily stored in aquifers and that may add to future river

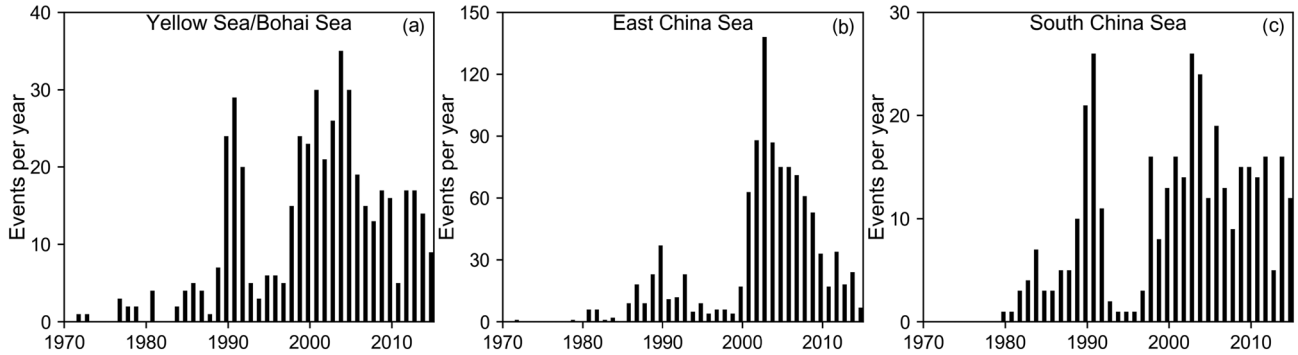

Figure 7. Annual frequency of harmful algal blooms in China's coastal waters in (a) YS/BS, (b) ECS, and (c) SCS during 1970–2015. Data were obtained from *Investigation and evaluation of red tide disasters in China (1933–2009)* (Liang, 2012; Xiao et al., 2019).

export (Bouwman, Bierkens, Griffioen, et al., 2013). Building a new system of recycling livestock and domestic waste as fertilizer and gradually increasing the nutrient-recycling rate should be considered (Yu et al., 2019). However, the problem is extremely complex, since many sources are involved with different retention fractions and location within the river basins that drain into the YS/BS, ECS, and SCS. Therefore, for evaluating the effectiveness of policy strategies to reduce nitrogen pollution, monitoring of the water quality in rivers and coastal marine environments is urgently needed.

4.5. Limitations of the Approach Used

Our model-based estimates are in good agreement with estimates based on observed nutrient concentrations and loads for the three LMEs. However, we also recognize that our approach has limitations. Sensitivity analysis revealed the importance of model parameters or input data such as runoff which is a major factor for nitrogen delivery and export by rivers (Beusen et al., 2015), and feed conversion efficiencies and apparent digestibility of nitrogen in compound feed in aquaculture production (Bouwman et al., 2011; Bouwman, Beusen, Overbeek, et al., 2013). The interannual variability in atmospheric exported nitrogen from source regions and the uncertainty of the aggregated annual deposition at regional scales from TM5-FASST is around 5%. However, the uncertainty of deposition for individual 1 by 1 grid cells is much larger, depending on local meteorological conditions. The most important limitation is in the temporal scale of our data, which is 1 year. HABs develop at much shorter time scale due to coincident specific growth conditions, such as availability of nutrients, nutrient ratios and physical conditions such as stratification and temperature (Glibert, 2017; Glibert et al., 2014).

5. Conclusions

Between 1970 and 2010, nitrogen inputs to the YS/BS, ECS, and SCS LMEs increased substantially mainly due to rapidly increasing river export. Agriculture was the dominant source in river delivery and export, while the proportions of aquaculture and sewage have recently started to increase rapidly, especially in river export because important aquaculture regions and particularly population centers are located in coastal provinces of China. Although decreasing, the proportion of natural sources is considerable in rivers draining into the SCS.

Nitrogen inputs to LMEs increased much faster in Chinese coastal waters than in other countries. Nitrogen inputs from mariculture have been increasing rapidly in recent years, with a strong concentration spatially in areas close to river mouths and confined sea areas. China's contribution to nitrogen pollution exceeds that of the other countries that border YS/BS and ECS, while in the SCS China and all the other countries contribute equally.

Nitrogen concentrations in the three LMEs have been increasing during the past 40 years. The dramatic increase of the HAB frequency in the three LMEs suggests that the assimilative capacity of China's coastal seas may already be exceeded. Our results show that a large part of the increased nitrogen loading is from land-based sources, both via the rivers and indirectly via atmospheric deposition, and an increasing part is from direct nitrogen release from fish and shellfish production. One mitigation strategy is to focus

nutrient reduction strategies initially on population centers and agricultural and aquaculture production regions close to the coast where in-stream retention is small due to short travel times of river water. This requires mitigation strategies for all sources, including advanced treatment systems for urban wastewater and aquaculture, while increasing agricultural nutrient use efficiencies by avoiding excess nutrient applications in crop fields and integrating crop and livestock production systems to increase nutrient-recycling rates. Environmental and agricultural management to reduce nitrogen pollution is urgent, because nitrogen inputs may increase further to sustain future food production and economic growth.

Data Availability Statement

The data supporting the conclusions can be obtained in supporting information for this paper and from 4TU. Centre for Research Data (http://doi.org/10.4121/uuid:3a178393-7c80-4caf-a29e-c6a731f7029c).

Acknowledgments

This work was supported by 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.), and support from PBL Netherlands Environmental Assessment Agency through in-kind contributions to The New Delta 2014 ALW projects no. 869.15.015 and 869.15.014 (A. F. B. and A. H. W. B.).

References

Anderson, D., Gilbert, P. M., & Burkholder, J. M. (2002). Harmful algal blooms and eutrophication: Nutrient sources, composition, and consequences. *Estuaries*, *25*, 704–726.

Bashkin, V. N., Park, S. U., Choi, M. S., & Lee, C. B. (2002). Nitrogen budgets for the Republic of Korea and the Yellow Sea region. *Biogeochemistry*, *57–58*, 387–403.

Beusen, A. H. W., Bouwman, A. F., Van Beek, L. P. H., Mogollón, J. M., & Middelburg, J. J. (2016). Global riverine N and P transport to ocean increased during the 20th century despite increased retention along the aquatic continuum. *Biogeosciences*, *13*, 2441–2451.

Beusen, A. H. W., Slomp, C. P., & Bouwman, A. F. (2013). Global land-ocean linkage: Direct inputs of nitrogen to coastal waters via submarine groundwater discharge. *Environmental Research Letters*, *8*(3), 034035. https://doi.org/10.1088/1748-9326/8/3/034035

Beusen, A. H. W., Van Beek, L. P. H., Bouwman, A. F., Mogollón, J. M., & Middelburg, J. J. (2015). Coupling global models for hydrology and nutrient loading to simulate nitrogen and phosphorus retention in surface water. Description of IMAGE-GNM and analysis of performance. *Geoscientific Model Development*, *8*(4), 4045–4067. https://doi.org/10.5194/gmd-8-4045-2015

Bouwman, A. F., Beusen, A. H. W., & Billen, G. (2009). Human alteration of the global nitrogen and phosphorus soil balances for the period 1970–2050. *Global Biogeochemical Cycles*, *23*, GB0A04. https://doi.org/10.1029/2009GB003576

Bouwman, A. F., Beusen, A. H. W., Glibert, P. M., Overbeek, C. C., Pawlowski, M., Herrera, J., et al. (2013). Mariculture: Significant and expanding cause of coastal nutrient enrichment. *Environmental Research Letters*, *8*(4), 044026. https://doi.org/10.1088/1748-9326/8/4/044026

Bouwman, A. F., Beusen, A. H. W., Lassaletta, L., van Apeldoorn, D. F., van Grinsven, H. J. M., Zhang, J., & van Ittersum, M. K. (2017). Lessons from temporal and spatial patterns in global use of N and P fertilizer on cropland. *Scientific Reports*, *7*(1), 40366. https://doi.org/10.1038/srep40366

Bouwman, A. F., Beusen, A. H. W., Overbeek, C. C., Bureau, D. P., Pawlowski, M., & Glibert, P. M. (2013). Hindcasts and future projections of global inland and coastal nitrogen and phosphorus loads due to finfish aquaculture. *Reviews in Fisheries Science*, *21*(2), 112–156.

Bouwman, A. F., Bierkens, M. F. P., Griffioen, J., Hefting, M. M., Middelburg, J. J., Middelkoop, H., & Slomp, C. P. (2013). Nutrient dynamics, transfer and retention along the aquatic continuum from land to ocean: Towards integration of ecological and biogeochemical models. *Biogeosciences*, *10*, 1–23. https://doi.org/10.5194/bg-10-1-2013

Bouwman, A. F., Goldewijk, K. K., Van Der Hoek, K. W., Beusen, A. H. W., Van Vuuren, D. P., Willems, J., et al. (2013). Exploring global changes in nitrogen and phosphorus cycles in agriculture induced by livestock production over the 1900–2050 period. *Proceedings of the National Academy of Sciences of the United States of America*, *110*(52), 20882–20887. https://doi.org/10.1073/pnas.1012878108

Bouwman, A. F., Pawłowski, M., Liu, C., Beusen, A. H. W., Shumway, S. E., Glibert, P. M., & Overbeek, C. C. (2011). Global hindcasts and future projections of coastal nitrogen and phosphorus loads due to shellfish and seaweed aquaculture. *Reviews in Fisheries Science*, *19*(4), 331–357.

Cai, W., Hu, X., Huang, W., Murrell, M. C., Lehrter, J. C., Lohrenz, S. E., et al. (2011). Acidification of subsurface coastal waters enhanced by eutrophication. *Nature Geoscience*, *4*, 766–770.

Cao, L., Wang, W., Yang, Y., Yang, C., Yuan, Z., Xiong, S., & Diana, J. (2007). Environmental impact of aquaculture and countermeasures to aquaculture pollution in China. *Environmental Science and Pollution Research International*, *14*(7), 452–462.

Chang, N., Shiao, J., & Gong, G. (2012). Diversity of demersal fish in the East China Sea: Implication of eutrophication and fishery. *Continental Shelf Research*, *47*, 42–54.

Chen, C., Gong, G., & Shiah, F. (2007). Hypoxia in the East China Sea: One of the largest coastal low-oxygen areas in the world. *Marine Environmental Research*, *64*, 399–408.

China Livestock Yearbook Editing Committee (2014). *China livestock yearbook. Data covering 1999–2011 retrieved 8 October 2014*. Beijing, China: China Agriculture Press (in Chinese).

China Ministry of Agriculture (2014). *The Chinese agricultural statistical report. Data covering 1980–2011 retrieved 8 October 2014*. Beijing, China: China Agriculture Press (in Chinese).

Conley, D. J., Paerl, H. W., Howarth, R. W., Boesch, D. F., Seitzinger, S. P., Havens, K. E., et al. (2009). Controlling eutrophication: nitrogen and phosphorus. *Science*, *323*(5917), 1014–1015.

de Wit, M., Behrendt, H., Bendoricchio, G., Bleuten, W., & van Gaans, P. (2002). The contribution of agriculture to nutrient pollution in three European rivers, with reference to the European nitrates directive. In *European Water Management Online EWA 2002*. Hennef, Germany/Brussels, Belgium.

EDGAR (2019). Emission database for global atmospheric research, 1970–2010 (https://edgar.jrc.ec.europa.eu/overview.php?v = 431). Vol. 2019, Ispra.

Fan, H., & Huang, H. (2008). Response of coastal marine eco-environment to river fluxes into the sea: A case study of the Huanghe (Yellow) River mouth and adjacent waters. *Marine Environmental Research*, *65*, 378–387.

Food and Agriculture Organization (2012). Aquaculture production 1950–2010. FISHSTAT Plus - Universal software for fishery statistical time series by Fisheries and Aquaculture Information and Statistics Service. Food and Agriculture Organization of the United Nations. Rome. https://www.fao.org/fishery/statistics/software/fishstat/en

Food and Agriculture Organization (2019a). FAOSTAT database collections. Data retrieved February 26, 2019. Rome. Food and Agriculture Organization of the United Nations. https://www.fao.org/faostat/en/#data

Food and Agriculture Organization (2019b). Fishery and Aquaculture Statistics. Global aquaculture and capture production 1950–2017 (FishstatJ). In: FAO Fisheries and Aquaculture Department [online]. Rome. Updated 2019. www.fao.org/fishery/statistics/software/fishstatj/en

Fowler, D., Coyle, M., Skiba, U., Sutton, M. A., Cape, J. N., Reis, S., et al. (2013). The global nitrogen cycle in the twenty-first century. *Philosophical Transactions of the Royal Society, B: Biological Sciences*, *368*, 20130164.

Galloway, J. N., Dentener, F. J., Capone, D. G., Boyer, E. W., Howarth, R. W., Seitzinger, S. P., et al. (2004). Nitrogen cycles: Past, present, and future. *Biogeochemistry*, *70*, 153–226.

Glibert, P. M. (2017). Eutrophication, harmful algae and biodiversity—Challenging paradigms in a world of complex nutrient changes. *Marine Pollution Bulletin*, *124*, 591–606.

Glibert, P. M., Icarus Allen, J., Artioli, Y., Beusen, A., Bouwman, L., Harle, J., et al. (2014). Vulnerability of coastal ecosystems to changes in harmful algal bloom distribution in response to climate change: Projections based on model analysis. *Global Change Biology*, *20*, 3,845–3,858.

Han, Y., Fan, Y., Yang, P., Wang, X., Wang, Y., Tian, J., et al. (2014). Net anthropogenic nitrogen inputs (NANI) index application in Mainland China. *Geoderma*, *213*, 87–94.

Heisler, J., Glibert, P. M., Burkholder, J. M., Anderson, D. M., Cochlan, W., Dennison, W. C., et al. (2008). Eutrophication and harmful algal blooms: A scientific consensus. *Harmful Algae*, *8*(1), 3–13.

Jiang, T., Yu, Z., Song, X., Cao, X., & Yuan, Y. (2010). Long-term ecological interactions between nutrient and phytoplankton community in the Changjiang estuary. *Chinese Journal of Oceanology and Limnology*, *28*(4), 887–898.

Li, H., Tang, H., Shi, X., Zhang, C., & Wang, X. (2014). Increased nutrient loads from the Changjiang (Yangtze) River have led to increased Harmful Algal Blooms. *Harmful Algae*, *39*, 92–101.

Li, Y., Li, D., Tang, J., Wang, Y., Liu, Z., & He, S. (2010). Long-term changes in the Changjiang Estuary plankton community related to anthropogenic eutrophication. *Aquatic Ecosystem Health & Management*, *13*(1), 66–72.

Liang, Y. (2012). *Investigation and evaluation of red tide disasters in China (1933–2009)*. Beijing: Ocean Press (in Chinese).

Liao, W., Zhang, L., Chen, H., Xiao, C., & Zhang, X. (2013). Nutrients variations and fluxes estimation in the Yellow River Estuary from 2001 to 2011. *Periodical of Ocean University of China*, *43*, 81–86 (in Chinese with English abstract).

Liu, D., Keesing, J. K., He, P., Wang, Z., Shi, Y., & Wang, Y. (2013). The world's largest macroalgal bloom in the Yellow Sea, China: Formation and implications. *Estuarine, Coastal and Shelf Science*, *129*, 2–10.

Liu, F., Pang, S., Chopin, T., Gao, S., Shan, T., Zhao, X., & Li, J. (2013). Understanding the recurrent large-scale green tide in the Yellow Sea: Temporal and spatial correlations between multiple geographical, aquacultural and biological factors. *Marine Environmental Research*, *83*, 38–47.

Liu, J., Du, J., Wu, Y., & Liu, S. (2018). Nutrient input through submarine groundwater discharge in two major Chinese estuaries: The Pearl River Estuary and the Changjiang River Estuary. *Estuarine, Coastal and Shelf Science*, *203*, 17–28.

Liu, J., Du, J., & Yi, L. (2017). Ra tracer-based study of submarine groundwater discharge and associated nutrient fluxes into the Bohai Sea, China: A highly human-affected marginal sea. *Journal of Geophysical Research: Oceans*, *122*, 8646–8660. https://doi.org/10.1002/2017JC013095

Liu, J., Su, N., Wang, X., & Du, J. (2017). Submarine groundwater discharge and associated nutrient fluxes into the Southern Yellow Sea: A case study for semi-enclosed and oligotrophic seas-implication for green tide bloom. *Journal of Geophysical Research: Oceans*, *122*, 139–152. https://doi.org/10.1002/2016JC012282

Liu, J., Zang, J., Bouwman, L., Liu, S., Yu, Z., & Ran, X. (2016). Distribution and budget of dissolved and biogenic silica in the Bohai Sea and Yellow Sea. *Biogeochemistry*, *130*, 85–101.

Liu, S., Li, L., & Zhang, Z. (2011). Inventory of nutrients in the Bohai. *Continental Shelf Research*, *31*, 1790–1797.

Liu, S., Zhang, J., Chen, S., Chen, H., Hong, G., Wei, H., & Wu, Q. (2003). Inventory of nutrient compounds in the Yellow Sea. *Continental Shelf Research*, *23*, 1161–1174.

Liu, X., Beusen, A. H. W., Van Beek, L. P. H., Mogollón, J. M., Ran, X., & Bouwman, A. F. (2018). Exploring spatiotemporal changes of the Yangtze River (Changjiang) nitrogen and phosphorus sources, retention and export to the East China Sea and Yellow Sea. *Water Research*, *142*, 246–255.

Lü, C., & Tian, H. (2007). Spatial and temporal patterns of nitrogen deposition in China: Synthesis of observational data. *Journal of Geophysical Research*, *112*, D22S05. https://doi.org/10.1029/2006JD007990

Luo, L., Kao, S., Bao, H., Xiao, H., Xiao, H., Yao, X., et al. (2018). Sources of reactive nitrogen in marine aerosol over the Northwest Pacific Ocean in spring. *Atmospheric Chemistry and Physics*, *18*, 6207–6222.

Luo, X., Tang, A., Shi, K., Wu, L., Li, W., Shi, W., et al. (2014). Chinese coastal seas are facing heavy atmospheric nitrogen deposition. *Environmental Research Letters*, *9*, 095007. https://doi.org/10.1088/1748-9326/9/9/095007

Ma, S., Xin, F., Cui, Y., & Qiao, X. (2004). Assessment of main pollution matter volume into the sea from Yellow River and Xiaoqing River. *Marine Fisheries Research*, *25*, 47–51 (in Chinese with English abstract).

Morée, A. L., Beusen, A. H. W., Bouwman, A. F., & Willems, W. J. (2013). Exploring global nitrogen and phosphorus flows in urban wastes during the twentieth century. *Global Biogeochemical Cycles*, *27*, 836–846. https://doi.org/10.1002/gbc.20072

National Bureau of Statistics of China (1986–2018). *China statistical yearbook. Data covered 1985–2017 retrieved 30 December 2019*. Beijing, China: China Statistic Press (in Chinese).

National Bureau of Statistics of China (2014). *China statistical yearbook. Data covering 1981–2011 retrieved 8 October 2014*. Beijing, China: China Statistic Press (in Chinese).

Newbold, J. D., Elwood, J. W., O'Neill, R. V., & Van Winkle, W. (1981). Measuring nutrient spiraling in streams. *Canadian Journal of Fisheries and Aquatic Sciences*, *38*, 860–863.

Ning, X., Lin, C., Hao, Q., Liu, C., Le, F., & Shi, J. (2009). Long term changes in the ecosystem in the northern South China Sea during 1976–2004. *Biogeosciences*, *6*, 2227–2243.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., & Hayasaka, T. (2007). An Asian emission inventory of anthropogenic emission sources for the period 1980–2020. *Atmospheric Chemistry and Physics*, *7*, 4419–4444. https://doi.org/10.5194/acp-7-4419-2007

Ohyama, T. (2010). Nitrogen as a major essential element of plants. In T. Ohyama, & K. Sueyoshi (Eds.), *Nitrogen assimilation in plants*. India: Signpost, Trivandrum, Kerala.

Qi, J., Liu, X., Yao, X., Zhang, R., Chen, X., Lin, X., et al. (2018). The concentration, source and deposition flux of ammonium and nitrate in atmospheric particles during dust events at a coastal site in northern China. *Atmospheric Chemistry and Physics*, *18*, 571–586.

Shou, W., Zong, H., Ding, P., & Hou, L. (2018). A modelling approach to assess the effects of atmospheric nitrogen deposition on the marine ecosystem in the Bohai Sea, China. *Estuarine, Coastal and Shelf Science*, *208*, 36–48.

Stehfest, E., van Vuuren, D. P., Kram, T., & Bouwman, A. F. (2014). Integrated assessment of global environmental change with IMAGE 3.0. In *Model description and policy applications*. The Hague: PBL Netherlands Environmental Assessment Agency. Retrieved from http://themasites.pbl.nl/models/image/index.php/Main_Page

Strokal, M., Yang, H., Zhang, Y., Kroeze, C., Li, L., Luan, S., et al. (2014). Increasing eutrophication in the coastal seas of China from 1970 to 2050. *Marine Pollution Bulletin*, *85*, 123–140.

Tan, J. (2002). *Study on nutrients of Huanghe River and the transport flux through Lijin hydrological station* (PhD dissertation). Ocean University of China (in Chinese with English abstract).

Van Beek, L. P. H., Wada, Y., & Bierkens, M. F. P. (2011). Global monthly water stress: 1. Water balance and water availability. *Water Resources Research*, *47*, W07517. https://doi.org/10.1029/2010WR009791

Van Dingenen, R., Dentener, F., Crippa, M., Leitao, J., Marmer, E., Rao, S., et al. (2018). TM5-FASST: a global atmospheric source–receptor model for rapid impact analysis of emission changes on air quality and short-lived climate pollutants. *Atmospheric Chemistry and Physics*, *18*, 16,173–16,211.

van Puijenbroek, P. J. T. M., Beusen, A. H. W., & Bouwman, A. F. (2019). Global nitrogen and phosphorus in urban waste water based on the shared socio-economic pathways. *Global Environmental Change*, *231*, 446–456.

Wan, X., Wu, Z., Chang, Z., & Zhang, X. (2002). Reanalysis of atmospheric flux of nutrients to the south Yellow Sea and the East China Sea. *Marine Environmental Science*, *21*, 14–18 (in Chinese with English abstract).

Wang, B., Shan, B., Zhan, R., & Zang, J. (2002). Budget model of inorganic nitrogen in the Bohai and Yellow Sea. *Marine Sciences*, *26*, 33–36 (in Chinese with English abstract).

Wang, B., Xin, M., Wei, Q., & Xie, L. (2018). A historical overview of coastal eutrophication in the China Seas. *Marine Pollution Bulletin*, *136*, 394–400.

Wang, H., Dai, M., Liu, J., Kao, S., Zhang, C., Cai, W., et al. (2016). Eutrophication-driven hypoxia in the East China Sea off the Changjiang Estuary. *Environmental Science and Technology*, *50*(5), 2255–2263.

Wang, J., Beusen, A. H. W., Liu, X., & Bouwman, A. F. (2020). Aquaculture production is a large, spatially concentrated source of nutrients in Chinese freshwater and coastal seas. *Environmental Science & Technology*.*54*(3), 1464–1474. https://doi.org/10.1021/acs.est.9b03340

Wang, J., Yu, Z., Wei, Q., & Yao, Q. (2019). Long-term nutrient variations in the Bohai Sea over the past 40 years. *Journal of Geophysical Research: Oceans*, *124*, 703–722. https://doi.org/10.1029/2018JC014765

Wang, S., Tang, D., He, F., Fukuyo, Y., & Azanza, R. V. (2008). Occurrences of harmful algal blooms (HABs) associated with ocean environments in the South China Sea. *Hydrobiologia*, *596*, 79–93.

Wang, Z., Xiao, J., Fan, S., Li, Y., Liu, X., & Liu, D. (2015). Who made the world's largest green tide in China? An integrated study on the initiation and early development of the green tide in Yellow Sea. *Limnology and Oceanography*, *60*, 1105–1117.

Wei, Q., Yao, Q., Wang, B., Wang, H., & Yu, Z. (2015). Long-term variation of nutrients in the southern Yellow Sea. *Continental Shelf Research*, *111*, 184–196.

Wollheim, W., Vörösmarty, C. J., Bouwman, A. F., Green, P., Harrison, J., Linder, E., et al. (2008). Global N removal by freshwater aquatic systems using a spatially distributed, within-basin approach. *Global Biogeochemical Cycles*, *22*, GB2026. https://doi.org/10.1029/2007GB002963

Wu, R. S. S. (1995). The environmental impact of marine fish culture: Towards a sustainable future. *Marine Pollution Bulletin*, *31*, 159–166.

Xiao, X., Agustí, S., Pan, Y., Yu, Y., Li, K., Wu, J., & Duarte, C. M. (2019). Warming amplifies the frequency of harmful algal blooms with eutrophication in Chinese coastal waters. *Environmental Science & Technology*, *53*(22), 13,031–13,041.

Xie, B., & Yu, K. (2007). Shrimp farming in China: Operating characteristics, environmental impact and perspectives. *Ocean and Coastal Management*, *50*, 538–550.

Yan, W., Zhang, S., & Wang, J. (2001). Nitrogen biogeochemical cycling in the Changjiang drainage basin and its effect on Changjiang river dissolved inorganic nitrogen: Temporal trend for the period 1968–1997. *Acta Geographica Sinica*, *505–514* (in Chinese with English abstract).

Yang, F., Wei, Q., Chen, H., & Yao, Q. (2018). Long-term variations and influence factors of nutrients in the western North Yellow Sea, China. *Marine Pollution Bulletin*, *135*, 1026–1034.

Yu, C., Huang, X., Chen, H., Godfray, H. C. J., Wright, J. S., Hall, J. W., et al. (2019). Managing nitrogen to restore water quality in China. *Nature*, *567*, 516–520.

Yu, J., & Zhang, H. (2016). *Spatial distribution and isotopic signature of organic carbon and nitrogen in surface sediments from Bohai Sea*. Paper presented at Ocean Sciences Meeting, American Geophysical Union.

Yu, R., Lü, S., & Liang, Y. (2018). Harmful algal blooms in the coastal waters of China. In P. M. Glibert, E. Berdalet, M. A. Burford, G. C. Pitcher, & M. E. Zhou (Eds.), *Global ecology and oceanagraphy of harmful algal blooms*, *Ecological Studies (Analysis and Synthesis)* (pp. 309–316). Cham: Springer.

Zhai, W., Zhao, H., Zheng, N., & Xu, Y. (2012). Coastal acidification in summer bottom oxygen-depleted waters in northwestern-northern Bohai Sea from June to August in 2011. *Chinese Science Bulletin*, *57*, 1062–1068.

Zhang, G., Zhang, J., & Liu, S. (2007). Characterization of nutrients in the atmospheric wet and dry deposition observed at the two monitoring sites over Yellow Sea and East China Sea. *Journal of Atmospheric Chemistry*, *57*, 41–57.

Zhang, H., Li, Y., Tang, C., Zou, T., Yu, J., & Guo, K. (2016). Spatial characteristics and formation mechanisms of bottom hypoxia zone in the Bohai Sea during summer. *Chinese Science Bulletin*, *61*, 1612–1620 (in Chinese with English abstract).

Zhang, S. (1990). A study of the mass transport rate of carbon, nitrogen, phosphorus and sulphur of the Changjiang River. *Study on the Background Values of Chemical Elements in Aquatic Environment*.

Zhang, X., Davidson, E. A., Mauzerall, D. L., Searchinger, T. D., Dumas, P., & Shen, Y. (2015). Managing nitrogen for sustainable development. *Nature*, *528*, 51–59.

Zhang, Y., Bleeker, A., & Liu, J. (2015). Nutrient discharge from China's aquaculture industry and associated environmental impacts. *Environmental Research Letters*, *10*, 045002. https://doi.org/10.1088/1748-9326/10/4/045002

Zhang, Y., Yu, Q., Ma, W., & Chen, L. (2010). Atmospheric deposition of inorganic nitrogen to the eastern China seas and its implications to marine biogeochemistry. *Journal of Geophysical Research*, *115*, D00K10. https://doi.org/10.1029/2009JD012814