AGU PUBLICATIONS

Earth's Future

Supporting Information for

Spatially explicit inventory of sources of nitrogen inputs to the Yellow Sea, East China Sea and South China Sea for the period 1970-2010

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Introduction

The Supporting Information provides other details besides the main text to support this study, including statistical data on population and food production, method schemes, model validation results, information on the study area, modelled results of nitrogen delivery to surface waters, nitrogen retention loads in river basins and nitrogen inputs from each external source and their spatial distributions in the coast, and reported eutrophication status and red tide outbreaks in Chinese seas. Movies and modelled output data are in separate files and briefly described in this text.

1. Figure S1. Population and production of crop, livestock and aquaculture in China for 1970–2017

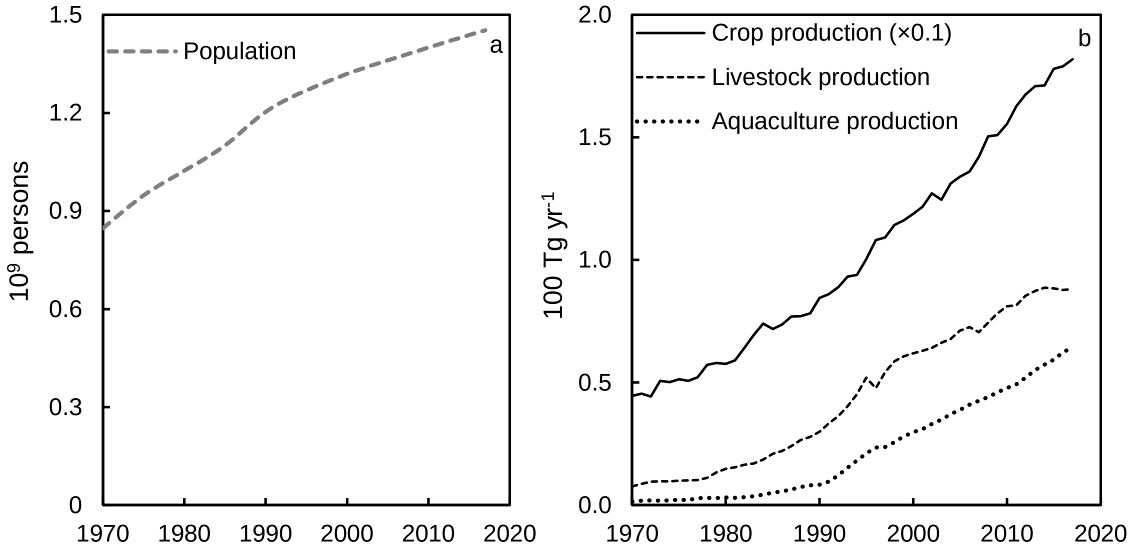

Figure S1. Population and production of crop (cereals in dry weight and others in wet weight), livestock meat (in carcass weight, excluding offal and slaughter fats) and aquaculture (in wet weight) in China for 1970–2017. Data are from FAO (FAO, 2018, 2019).

2. Figure S2. Scheme of Integrated Model to Assess the Global Environment–Global Nutrient Model (IMAGE–GNM)

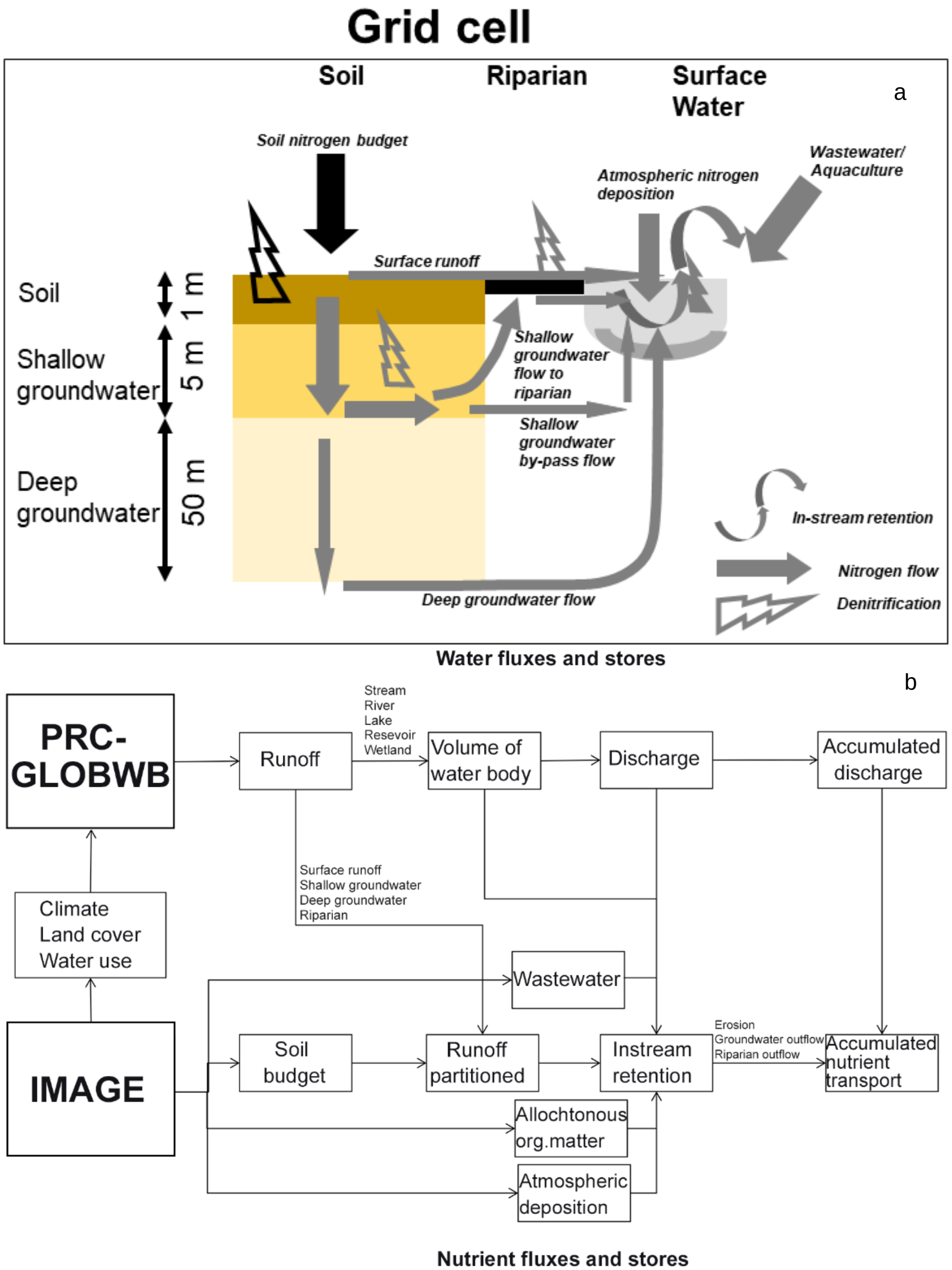

Figure S2(a). Scheme of Integrated Model to Assess the Global Environment–Global Nutrient Model (IMAGE–GNM), adapted from Beusen et al. (2015); **(b).** scheme of the model framework with PCR-GLOBWB and IMAGE and data flows between the models, adapted from Beusen et al. (2015) and Liu et al. (2018).

IMAGE-GNM uses the 0.5°-grid-based global hydrological model PCRaster Global Water Balance (PCR-GLOBWB) (Van Beek et al., 2011) to quantify the hydrological properties in river basins, including water stores and fluxes, volume, surface area, and thus depth of water bodies, and water travel time (Beusen et al., 2015). According to the IMAGE-GNM model framework in the publications (Beusen et al., 2015), a) each grid cell in the basin receives water and dissolved and suspended nitrogen from upstream grid cells; b) inside the grid cell, nitrogen is delivered to water bodies via diffuse sources (surface runoff, shallow and deep groundwater, riparian zones; litterfall in floodplains; atmospheric deposition) and wastewater; c) nitrogen retention in a water body is calculated based on water residence time and nutrient uptake velocity; d) water and nitrogen are transported to downstream grid cells. In-stream retention process in the river basin included in IMAGE-GNM comprises of denitrification, sedimentation, and uptake by aquatic plants.

For more details on IMAGE-GNM, please go to the descriptions in previous publications (Beusen et al., 2015; Beusen et al., 2016; Liu et al., 2018).

3. Model validation and measurement data sources

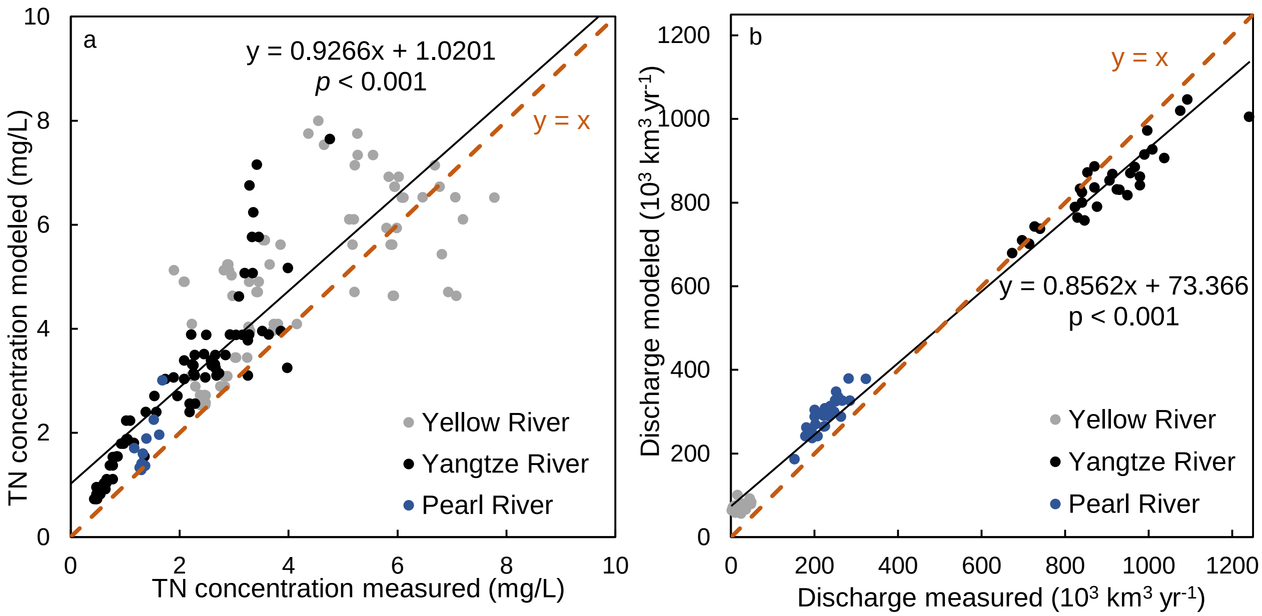

Figure S3. (a) TN concentration comparison and (b) discharge comparison: measured versus modelled using IMAGE-GNM at river mouths of China's 3 major rivers (i.e. the Yellow River, Yangtze River and Pearl River). The red dashed line is the 1:1 line, and the black line is the linear fitting line.

Table S1. Measurement data sources for model validation.

River	LMEs drained into	Station	Parameter	Year and sources	Observed nitrogen forms	Transfer ratio and reference
Yellow River	YS/BS	Lijin	TN concentration	1980-2010 (Fan and Huang, 2008; Gu, 2018; Li, 2010; Liao et al., 2013; Ma et al., 2015; Tan, 2002)	DIN, TN, TDN, DON, TON, NO_3-N, NO_2-N, NH_4-N, TPN	DIN/TN = 80% (Tan, 2002)
Yangtze River	YS and ECS	Datong	TN concentration	1970-1986, 1997-2010 (Dai et al., 2011; Liu et al., 2003a; Shen et al., 2003; Xu, 2013)	DIN, TN, TDN, DON, TON, NO_3-N, NO_2-N, NH_4-N, TPN	DIN/TN = 50% (Yan et al., 2001; Zhang, 1990)
Pearl River	SCS	Gaoyao	TN concentration	1980-1989 (Duan et al., 2000)	DIN	DIN/TN = 50% (Yan et al., 2001; Zhang, 1990)
Yellow River	YS/BS	Lijin	Discharge	1970-2000 (Ministry of Water Resource of China, 2003)	—	—

Yangtze River	YS and ECS	Datong	Discharge	1970-2000 (Ministry of Water Resource of China, 2003)	—	—
Pearl River	SCS	Gaoyao	Discharge	1970-2000 (Ministry of Water Resource of China, 2003)	—	—

DIN: dissolved inorganic nitrogen; TN: total nitrogen; TDN: total dissolved nitrogen; DON: dissolved organic nitrogen; TON: total organic nitrogen; NO_3-N: nitrate; NO_2-N: nitrite; NH_4-N: ammonium; TPN: total particulate nitrogen.

For the period from 1970 to 2010, the model-based TN concentrations generally agree with the measured TN concentrations at the monitoring stations located at the mouths of China's three main rivers (i.e. the Yellow River, Yangtze River and Pearl River)(Figure S3a). Although the model slightly overestimates the TN concentrations, the linear fitting curve with a slope of 0.9266 (p<0.001) in Figure S3a is very close to the curve of y = x. The discrepancy between simulations and observations can be partly attributed to the fixed DIN/TN ratios (Tan, 2002; Yan et al., 2001; Zhang, 1990) used to estimate TN for the entire period, which may be different for different years. However, studies on the measured nitrogen forms and their ratios for different years are very limited for the Yangtze River and Yellow River. Moreover, since nitrogen forms and their ratios have not been measured in the Pearl River, the DIN/TN ratio measured in the Yangtze River was adopted to estimate the measured TN in the Pearl River. Furthermore, we calculate on an annual basis, which may not appropriately capture short-term (i.e. monthly or seasonal) observations while some measurements were only conducted in several months instead of the whole year.

According to the comparison of modelled and measured discharge at the mouths of China's three main rivers (Figure S3b), the discharge of these rivers is well represented in IMAGE-GNM. The discharge of Yangtze River is the largest, that of the Pearl River second, and that of the Yellow River smallest, which also well fits the magnitudes of these rivers. Although the model slightly underestimates the discharge for the "large" Yangtze River and slightly overestimates the discharge for the "small" Pearl River and Yellow River, the linear fitting curve with a slope of 0.8562 (p<0.001) in Figure S3b is very close to the curve of y = x. Since the time step of IMAGE-GNM is yearly, short-term (i.e. monthly or seasonal) variations in discharge may not be appropriately captured.

Overall, the discharge and nutrient transport in China's major river basins are well simulated by IMAGE-GNM. Considering the good validation results for China's three main rivers in this study and other rivers in a previous study (Beusen et al., 2015), the modelled river nitrogen export to the three Large Marine Ecosystems (YS/BS, ECS, and SCS) using IMAGE-GNM is expected to be close to reality.

4. Table S2. Information of the Large Marine Ecosystems that border the coasts of China and other countries.

Table S2. Information of the Large Marine Ecosystems.

Large Marine Ecosystems	Marginal or continental seas	Area (10^3 km^2)	Average depth (m)	Number of River basins from IMAGE-GNM with water discharge	Surrounding countries
Yellow Sea/Bohai Sea (YS/BS)	Yellow Sea and Bohai Sea	444	18 for Bohai Sea (Zhang et al., 2006) and 44 for Yellow Sea (Liu et al., 2003b)	82	China, North Korea and South Korea
East China Sea (ECS)	East China Sea	781	370 (Guan and Mao, 1982)	65	China, South Korea and Japan
South China Sea (SCS)	South China Sea	3232	1350 (Chen et al., 2001)	195	China, Vietnam, Malaysia, Singapore, Indonesia, Brunei and Philippines

5. Figure S4. Scheme of IMAGE–GNM aquaculture nutrient budget model

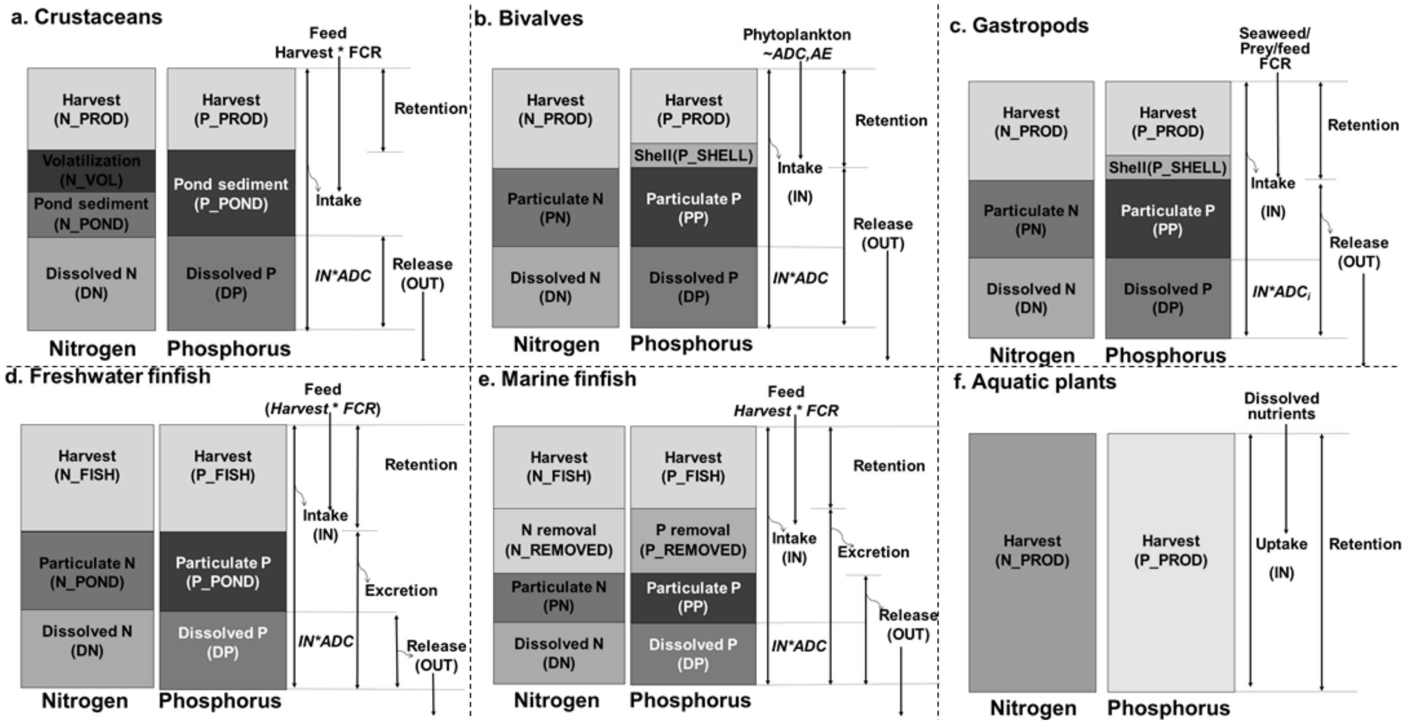

Figure S4. Scheme of IMAGE–GNM aquaculture nutrient budget model, adapted from (Wang et al., 2019).

6. Figure S5. Nitrogen delivery to surface water from diffuse and point sources during 1970-2010 for rivers draining to YS/BS, ECS and SCS.

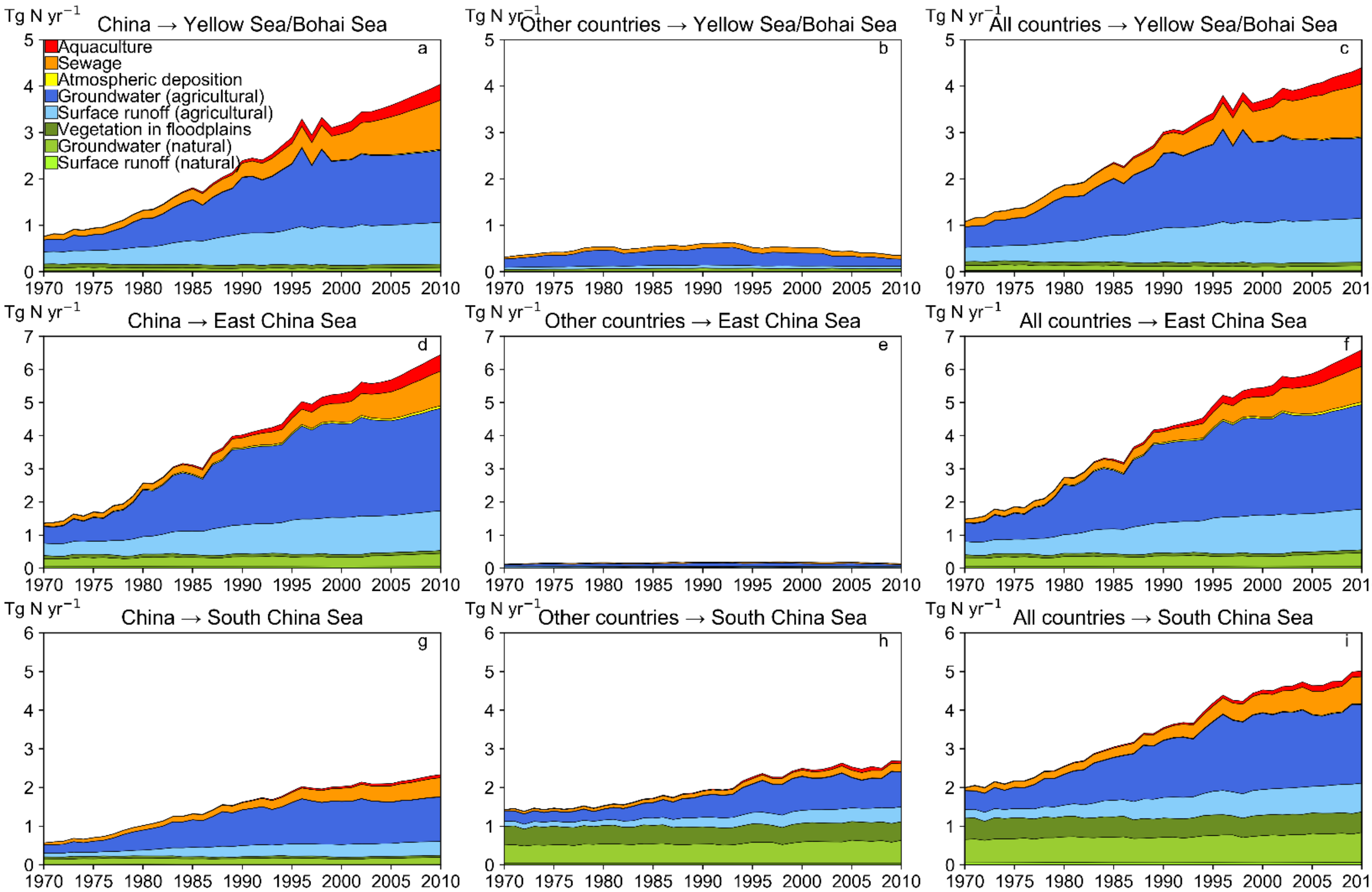

Figure S5. Nitrogen delivery to surface water from diffuse and point sources during 1970-2010 for rivers draining to YS/BS (a-c), ECS (d-f) and SCS (g-i). Left column is data for China, middle column is data for the other countries, and the right column presents data for the total.

7. Figure S6. Nitrogen inputs to YS/BS, ECS and SCS from atmospheric deposition, mariculture and submarine fresh groundwater discharge during 1970-2010.

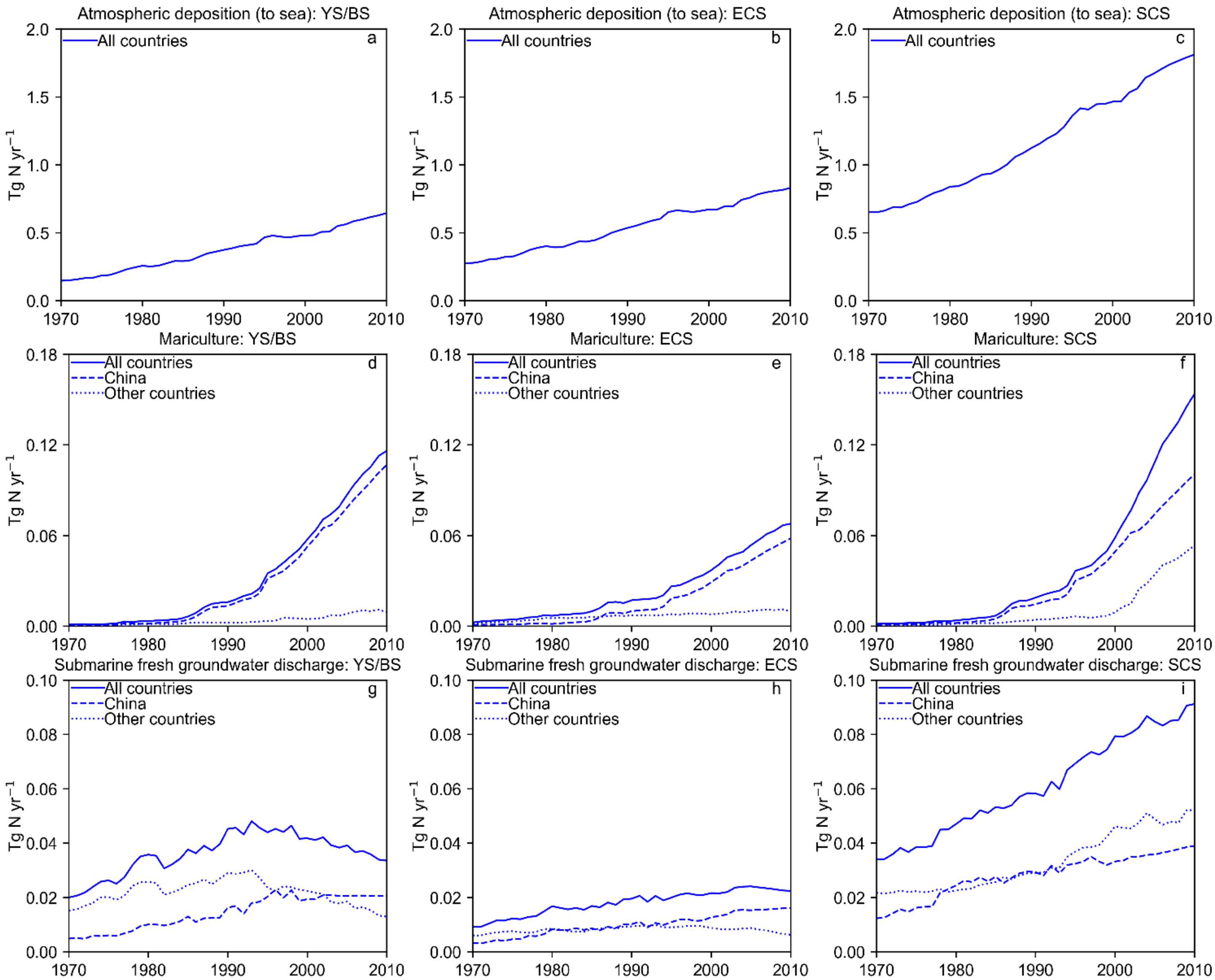

Figure S6. Nitrogen inputs to YS/BS (right column), ECS (middle column) and SCS (right column) from atmospheric deposition (a-c), mariculture (d-f) and submarine fresh groundwater discharge (g-i) during 1970-2010.

8. Figure S7. Nitrogen retention loads from rivers, lakes and reservoirs in river basins draining to YS/BS, ECS and SCS during 1970-2010.

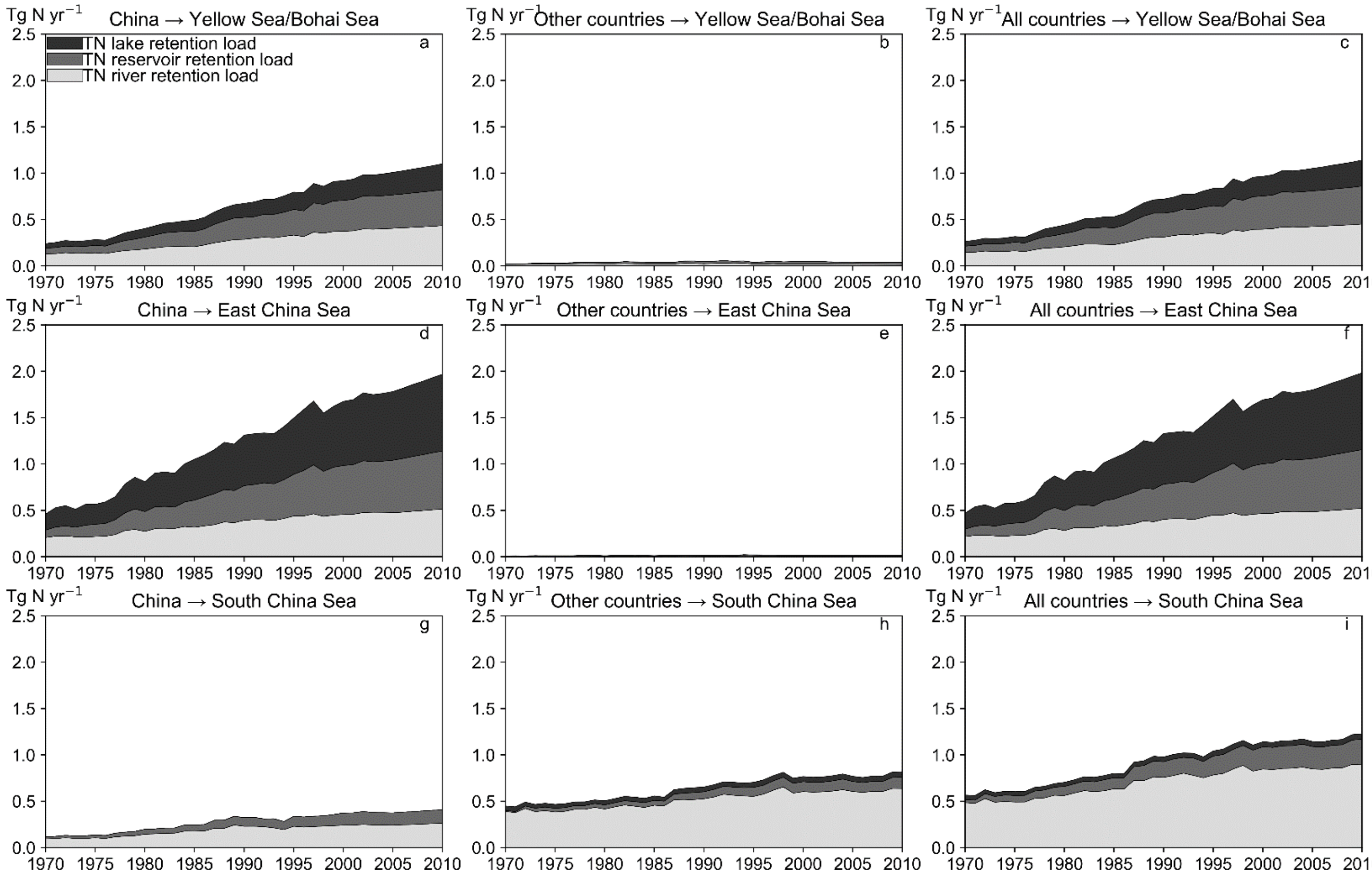

Figure S7. Nitrogen retention loads from rivers, lakes and reservoirs in river basins draining to YS/BS, ECS and SCS during 1970-2010. Total nitrogen retention load = TN lake retention load + TN reservoir retention load + TN river retention load (excluding the lake and reservoir shares). "Nitrogen retention load" is the nitrogen load which is removed from the water column due to removal processes (retention) in a waterbody (Beusen et al., 2015).

9. Figure S8. Spatial distribution of nitrogen inputs from river export, atmospheric deposition, mariculture and submarine fresh groundwater discharge to the coastal areas of YS/BS, ECS and SCS in 1970 and 2010.

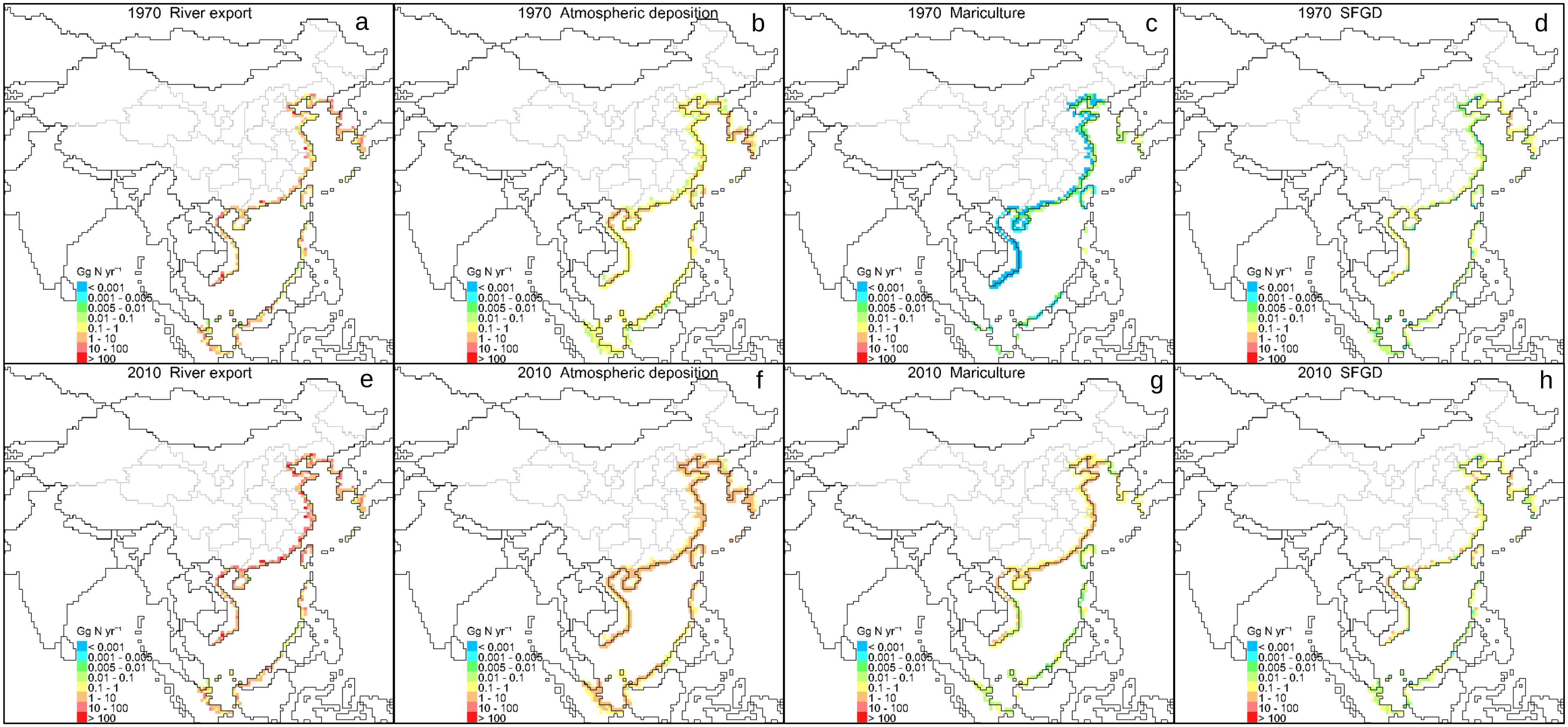

Figure S8. Spatial distribution of nitrogen inputs from all sources to the coastal areas of YS/BS, ECS and SCS in 1970 and 2010: (a, e) river export, (b, f) atmospheric deposition, (c, g) mariculture, and (d, h) submarine fresh groundwater discharge (SFGD).

10. Figure S9. Eutrophication status and seawater quality status of inorganic nitrogen in Chinese seas in 2018.

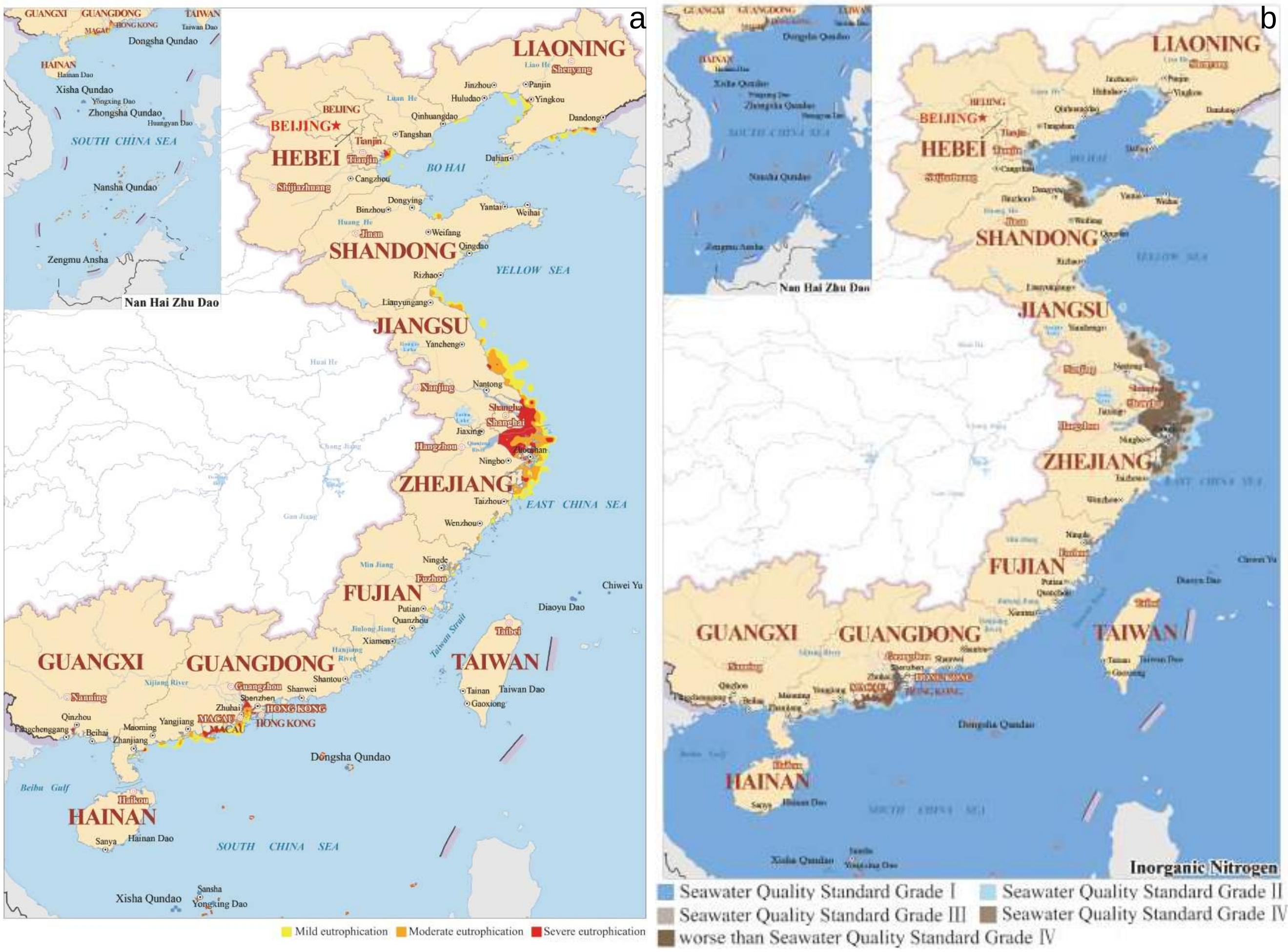

Figure S9. (a) Eutrophication status and (b) seawater quality status of inorganic nitrogen in Chinese seas in 2018 (Ministry of Ecology and Environment of the People's Republic of China, 2019).

11. Figure S10. The distribution of annual frequency of red tide outbreaks in Chinese seas during 1990-1999 and 2000-2009.

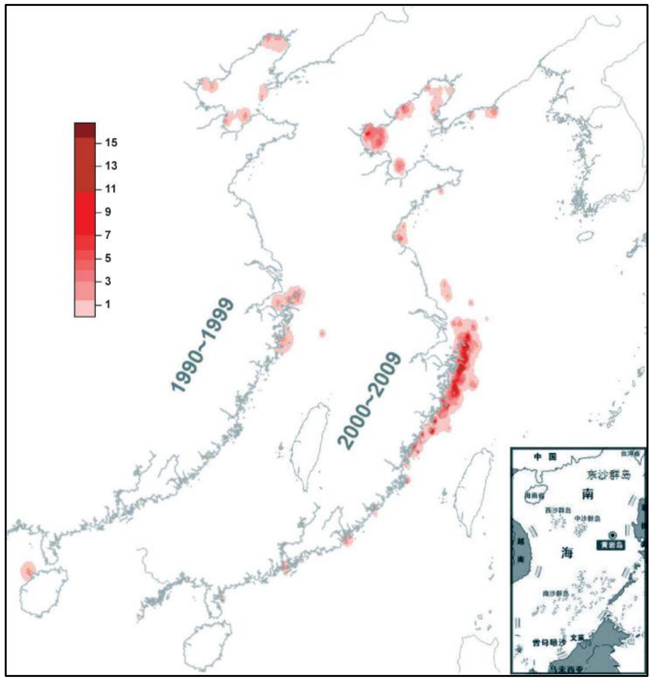

Figure S10. The distribution of annual frequency of red tide outbreaks in Chinese seas during 1990-1999 and 2000-2009 (State Oceanic Administration, 2010).

12. Description on Movies S1. Long-term variations in the detailed sources of river nitrogen export to the LMEs.

The directory "MoviesS1_Sources_of_river_N_export" contains the results of the long-term variations in the detailed sources of nitrogen inputs via river export to the YS/BS, ECS and SCS LMEs for the period 1970-2010 modelled from IMAGE-GNM. The movies show how the total river nitrogen export and contributions of surface water (natural), groundwater (natural), surface water (agricultural), groundwater (agricultural), vegetation in floodplains, atmospheric deposition (to inland waters), sewage and freshwater aquaculture change during 1970-2010 in the rivers draining to the YS/BS, ECS and SCS in China, other countries and all countries.

13. Description on Movies S2. Long-term variations in the sources of total nitrogen inputs to the LMEs.

The directory "MoviesS2_Sources_of_total_N_inputs" contains the results of the long-term variations in the sources of total nitrogen inputs to the YS/BS, ECS and SCS LMEs for the period 1970-2010. The movies show how the total nitrogen inputs to the LMEs and contributions of river export, submarine fresh groundwater discharge, atmospheric deposition (to sea) and mariculture change during 1970-2010.

14. Description on Modelled output data used in this study (separate csv files):

1) Description of Table S3. Detailed sources of river export to the LMEs.

See file Table S3. Detailed sources of river export to the LMEs.csv. Each line contains the fractions of total nitrogen from one detailed source to river export to one LME during 1970-2010.

The names of detailed sources in the second column are explained here:

sro_nat: nitrogen in river export from surface runoff in natural area

sro_agri: nitrogen in river export from surface runoff in agricultural area

grw_nat: nitrogen in river export from groundwater outflow in natural area

grw_agri: nitrogen in river export from groundwater outflow in agricultural area

depo_water: nitrogen in river export from atmospheric deposition in river basins

pointsources: nitrogen in river export from urban wastewater

aquaculture: nitrogen in river export from freshwater aquaculture

veg_flooding: nitrogen in river export from vegetation in floodplains

2) Description of Table S4. Sources of total nitrogen inputs to the LMEs.

See file Table S4. Sources of total nitrogen inputs to the LMEs.csv. Each line contains the fractions of nitrogen input from one source to one LME during 1970-2010.

The names of detailed sources in the second column are explained here:

nload_mouth: nitrogen inputs from river export to the LMEs

sfgd: nitrogen inputs from submarine fresh groundwater discharge to the LMEs

mariculture: nitrogen release from mariculture in the LMEs

deposition_over_sea: nitrogen inputs from atmospheric deposition over the LMEs

deposition_over_coast: nitrogen inputs from atmospheric deposition over the coast of the LMEs

ntotal_sources: total nitrogen inputs from river export, submarine fresh groundwater discharge, mariculture and atmospheric deposition over the LMEs

ncoastal_sources: total nitrogen inputs from river export, submarine fresh groundwater discharge, mariculture and atmospheric deposition over the coast of the LMEs

Reference

Beusen, A. H. W., Bouwman, A. F., Van Beek, L. P. H., Mogollón, J. M., and 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., Van Beek, L. P. H., Bouwman, A. F., Mogollón, J. M., and 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**, 4045–4067, doi:10.5194/gmd-8-4045-2015 (http://www.geosci-model-dev.net/8/4045/2015/).

Chen, C. T. A., Wang, S., Wang, B., and Pai, S. (2001). Nutrient budgets for the South China Sea basin. *Marine Chemistry* **75**.

Dai, Z., Du, J., Zhang, X., Su, N., and Li, J. (2011). Variation of riverine material loads and environmental consequences on the Changjiang (Yangtze) Estuary in recent decades (1955−2008). *Environmental Science & Technology* **45**, 223-227.

Duan, S., Zhang, S., and Huang, H. (2000). Transport of dissolved inorganic nitrogen from the major rivers to estuaries in China. *Nutrient Cycling in Agroecosystems* **57**, 13-22.

Fan, H., and 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.

FAO (2018). Fishery and Aquaculture Statistics. Global aquaculture and capture production 1950-2016 (FishstatJ). In: FAO Fisheries and Aquaculture Department [online]. Rome. Updated 2018. www.fao.org/fishery/statistics/software/fishstatj/en.

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

Gu, J. (2018). Review on the nutrients transportation of Yellow River and its ecological impacts in the Estuary. *Yellow River* **40(2)**, 85-91 (in Chinese with English abstract).

Guan, B., and Mao, H. (1982). A note on circulation of the East China Sea. *Chinese Journal of Oceanology and Limnology* **1**, 5-16.

Li, L. (2010). Effects of exchange fluxes of nutrients at the sediment and water interface and the Huanghe input on nutrient dynamics of the Bohai Sea., Master dissertation, Ocean University of China (in Chinese with English abstract).

Liao, W., Zhang, L., Chen, H., Xiao, C., and 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, S., Zhang, J., Chen, H., Wu, Y., Xiong, H., and Zhang, Z. (2003a). Nutrients in the Changjiang and its tributaries. *Biogeochemistry* **62(1)**, 1-18.

Liu, S., Zhang, J., Chen, S., Chen, H., Hong, G., Wei, H., and Wu, Q. (2003b). 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., and 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.

Ma, Y., Zang, J., Che, H., Zheng, L., Zhang, B., and Ran, X. (2015). Trend and distributions of nutrient elements in the Huanghe (Yellow) River. *Oceanologia et Limnologia Sinica* **46(1)**, 140-147 (in Chinese with English abstract).

Ministry of Ecology and Environment of the People's Republic of China (2019). Bulletin of Marine Ecology and Environment Status of China in 2018.
Ministry of Water Resource of China (2003). China River Sediment Bulletin 2002.
Shen, Z., Liu, Q., and Zhang, S. (2003). Distribution, variation and removal patterns of inorganic nitrogen in the Changjiang River. *Oceanologia et Limnologia Sinica* **34(4)**, 355-363 (in Chinese).
State Oceanic Administration, People's Republic of China. (2010). China Marine Environmental Quality Bulletin 2009. *Ocean Development and Management* **4**, (in Chinese).
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., and Bierkens, M. F. P. (2011). Global monthly water stress: 1. Water balance and water availability. *Water Resource Research* **47**, W07517.
Wang, J., Beusen, A. H. W., Liu, X., and Bouwman, A. F. (2019). Aquaculture Production is a Large, Spatially Concentrated Source of Nutrients in Chinese Freshwater and Coastal Seas. *Environmental Science and Technology* **54(3)**, 1464–1474. https://doi.org/10.1021/acs.est.9b03340.
Xu, H. (2013). The Yangtze Estuary: Nutrients budget and transport response to hunman activities in the river basin., PhD dissertation, East China Normal University (in Chinese with English abstract).
Yan, W., Zhang, S., and 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).
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*, 1212131.
Zhang, Z., Zhu, M., Wang, Z., and Wang, J. (2006). Monitoring and managing pollution load in Bohai Sea, PR China. *Ocean & Coastal Management* **49**, 706-716.