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PAPER

Historical trends of riverine nitrogen loading from land to the East China Sea: a model-based evaluation

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Supplementary material for this article is available online

Abstract

East Asia is the one of the hotspot regions with too much reactive nitrogen (N) inputs from anthropogenic sources. Here, we evaluated historical total inorganic N (TIN) load from land to sea through the rivers surrounding the East China sea using biogeochemical model 'VISIT' combined with a newly developed VISIT Off-line River Nitrogen scheme (VISIToRN). VISIT calculated N cycling in both natural and agricultural ecosystems and VISIToRN calculated inorganic N transport and riverine denitrification through the river channels at half degree spatial resolution. Between 1961 and 2010, the estimated TIN load from land to the sea surrounding the East China Sea increased from 2.7 Tg-N Year^{-1} to 5.5 Tg-N Year^{-1}, a twofold increase, while the anthropogenic N input to the East China Sea basin (N deposition, N fertilizer, manure, and human sewage) increased from 12.9 Tg-N Year^{-1} to 36.9 Tg-N Year^{-1}, an increase of about 3 times. This difference in the rate of increase is due in large part to the terrestrial nitrogen budget, and the results of the model balance indicate that TIN load to rivers has been suppressed by improvements in fertilizer application rates, harvesting on agricultural land, and nitrogen accumulation in forests. The results of the model balance showed that the increase rate of nitrogen runoff from Chinese rivers has been declining since 2000. In our estimation by VISIToRN, the amount of nitrogen removed by river denitrification in the river channel before the mouth is not negligible, ranging from 1.6 Tg-N Year^{-1} to 2.16 Tg-N Year^{-1}. The N load from agricultural sources is still significant and needs to be further reduced. TIN load tended to increase in years with high precipitation. In order to effectively reduce TIN load, it is necessary to consider climate change-adaptive agricultural N management.

1. Introduction

Anthropogenic activities have greatly altered global nitrogen (N) cycling in terrestrial and ocean environments (Gruber and Galloway 2008) and it become above the planetary boundary for N, that is capacity of stabilizing the state of the Earth System, in current era (Steffen *et al* 2015). In addition, by 2100, it is plausible that the amount of synthetic N fertilizer use will continue to increase, possibly up to twice the current level (Winiwarter *et al* 2013). Against this backdrop of high N load by human, N load from land to sea seriously cause in coastal eutrophication (Seitzinger *et al* 2005, Howarth 2008). Eutrophication is thought to have a negative impact on ecosystems by causing deoxygenation in vulnerable areas including coastal areas and some in coastal waters and some semi-enclosed seas (Breitburg *et al* 2018). Reducing the nitrogen load from land is one of the most important measures (Breitburg *et al* 2018).

East-Asia is known to be a hotspot of reactive N (Nr) pollution. In the last half century, the use of N fertilizer in this region was rapidly growing (Nishina *et al* 2017). Due to the high consumption of chemical fertilizers in the area, East-Asia is a biggest source of atmospheric N_2O among the global regions (Tian *et al* 2016, Shang *et al* 2019, Tian *et al* 2020). The amount of nitrogen fertilizer consumed in East Asia has increased more than 20 times from the 1960s to 2000, and is currently about 43.6 Tg N Year^{-1}, or 53% of global consumption (Nishina *et al* 2017). In addition to N fertilizer, various non-point source of N (N deposition (Liu *et al* 2013), manure (Zhang *et al* 2017)) have also increased during the same period. Furthermore, East Asia is fast urbanising particularly in recent decades, which also contributed increase of reactive N as a point source to a river via wastewater release (Shindo *et al* 2003, Gu *et al* 2012).

The East China Sea is located in trans-boundary region among East-Asian countries, and nutrient loading from multiple nations. The Yangtze River basin, the largest river flowing into the East China Sea, is the area with the highest increase rate in anthropogenic nitrogen in China in recent decades (Ti *et al* 2012). Thus, heavy N load from land to rivers and eventually to the seas has occurred and resulted in eutrophication of coastal areas in this region (e.g., Wang *et al* 2018). Due to the anthropogenic nutrients inputs from land, this region has been suffered from hypoxia for several decades (e.g., Daoji and Daler 2004). The East China Sea is mostly a shallow continental shelf with a depth of about 200m, which is susceptible to anthropogenic nutrient loading. The river plumes of large Chinese rivers with high NO_3^- concentration extend towards the east and propagate across the shelf in the East China sea (Chen 2008). So, not just in the coastal areas, but throughout the East China Sea, eutrophication effects are evident. For example, Jellyfish blooms have been on the rise due to the eutrophication after the 1990's (Jiang *et al* 2008), which could cause the decline of fishery resources (Purcell and Arai 2001).

In this study, we focused on the TIN load to the East China Sea and we evaluated historical trends from land to sea in order to effectively manage Nr impacts in this region. To this end, it is essential to quantitatively comprehend each Nr source and Nr flows in watersheds surrounding the East China sea in East-Asian countries. Previous modeling studies have already been done on the rivers flowing into the East China Sea (e.g., Yan *et al* 2010, Strokal *et al* 2014, Wang *et al* 2014, Chen *et al* 2019), however, the studies on the entire East China Sea as a trans-boundary area are still limited (Wang *et al* 2020a). Considering the nitrogen cascade (Galloway *et al* 2003), that is the flow of reactive nitrogen from one environment to another (e.g., air, soil, water, sea and human sectors), is important for understanding the nitrogen balance of the basin, but this perspective is lacking. It is known that meteorological factors are important for nitrogen runoff (e.g., Ballard *et al* 2019), but this has not been fully investigated for TIN load to the East China Sea. So, this study aimed to evaluate historical N load to sea by the rivers and the contribution of major point sources as well as non-point sources to the N load to seas in East-Asia from the view to the N cascade concept (Galloway *et al* 2003). Previous studies of nitrogen loading to the ocean based on the assessment of the nitrogen loading of each source, but have not focused on the dynamics of nitrogen on land and during river transportation. We have already evaluated the amount of direct N_2O emission and N cycling in the land ecosystems in this region using biogeochemical model 'VISIT' (Ito *et al* 2018). In this study, we further assessed the N load from land to sea by developing a simple off-line riverine N scheme for biogeochemical models. This model, which we named VISIToRN (VISIT Off-line River Nitrogen scheme), is an independent off-line model from VISIT, which uses the output of VISIT (nitrogen load and flow) as input data to calculate the nitrogen load in rivers. Using this scheme, we assessed the historical dynamics of nitrogen in the cascade from the land to the ocean and used the simulation results to address the relationship between meteorological factors and TIN loading.

2. Materials and methods

2.1. Calculation of nitrogen flows in terrestrial ecosystems

We used a process-based terrestrial ecosystem model Vegetation Integrative SImulator for Trace gases (VISIT) (Inatomi *et al* 2010, Ito and Inatomi 2012) to estimate N loadings from natural and agriculture ecosystems. This model can simulate the energy budget, hydrology, carbon cycle, and N cycle of natural vegetation and agricultural land separately at each grid, , using meteorological variables and various environmental data as input variables. The hydrological scheme simulates surface radiation and water budgets for two (near-surface and below) soil water pools and snow layer, which outputs are evapotranspiration rate from the soil surface and vegetation canopy surface, infiltration, and runoff discharge. For snow layer, snow accumulation and melting are also simulated. The carbon cycle scheme consists of three (leaf, stem, and root) plant and two (litter and humus) soil carbon pools. This scheme calculates photosynthetic assimilation, respiratory emission, allocation to C pools, litterfall, and microbial soil decomposition. Carbon budgets has been validated by net primary production (Inatomi *et al* 2010) and satellite-based estimates of photosynthetic parameters (Ito *et al* 2017). The N cycle scheme can calculate major N budget, which are based on the C flows in the model, and simulate soil N processes such as mineralization, nitrification, denitrification, nitrate leaching. Fertilizer is considered as an

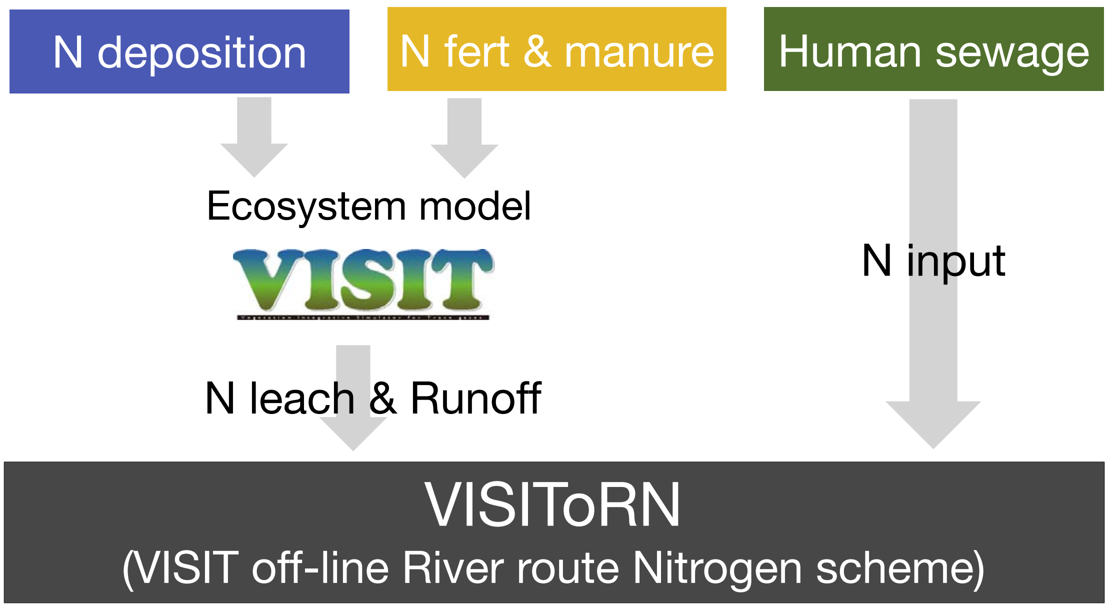

Figure 1. Scheme of calculation in this study. VISIT is biogeochemical model (Inatomi *et al* 2010) and VISIToRN is a scheme for river routing and N reactions in river developed in this study.

input to the ammonium and nitrate pools and manure as an input into the litter organic nitrogen pool. N_2O emissions through nitrification and denitrification are estimated using the scheme developed by Parton *et al* (1996).

In this study, the same protocol was used to calculate N cycling of terrestrial ecosystems as in the study published by (Ito *et al* 2018), The simulation protocol of this study is exactly the same as that of previous studies, except that it replaces the latest nitrogen fertilizer data which is based on sub-national scale statistics (i.e., county, municipal, provincial or state levels) (Wang *et al* 2020b). Spatial resolution is half degree in latitude and longitude and the simulation periods was during 1961 to 2010 in this study. The list of major inputs dataset are as follows; climate variables are obtained from the CRU TS3.25 dataset (Harris *et al* 2014) for each grid. For Land-use and its transition, we used a harmonization of land-use data for Earth System models (Hurtt *et al* 2011). N deposition were derived from the results of atmospheric chemistry model simulation studies (Sudo *et al* 2002, Dentener *et al* 2006),

The synthetic N fertilizer and manure inputs were used a new dataset developed by Wang *et al* (2020b). This dataset distinguishes N application rate via manure or synthetic N for rice and for all other crops. So, according to global crop distribution maps (Monfreda *et al* 2008), we divided the agriculture ecosystems into rice and other crops in order to calculate weighted N fertilizer inputs in each grid. Prior to using this dataset, all grids with values greater than 2000 kg-N ha^{-1} $year^{-1}$ were replaced with 2000 kg-N ha^{-1} $year^{-1}$ for each grid in order to remove outliers in both synthetic N fertilizer and manure dataset.

2.2. Calculation of nitrogen transport in river channels

We developed a river routing module namely 'VISIToRN' to compute daily discharge volume and N loading in each half grid cell, which is a kind of simple linear reservoir model (Nash 1957). We used this model combined with VISIT output. Since the calculation step of VISIToRN is daily, the monthly output from VISIT was simply divided by the number of days and used as daily step input data. Similarly, the nitrogen in wastewater estimated for the year was divided by the number of days in the year and used as the daily nitrogen input. Figure 1 summarize the simulation protocol in this study. In this section, we described VISIToRN in detail.

Each grid cell is considered to have a surface water storage pool 'S' (m^3). The change with time t of S is represented as a following equation;

$$\frac{dS_{riv}}{dt} = Q_{in} - Q_{out} + R_{VISIT} \tag{1}$$

Q_{in} is water discharge from upper-stream cells and Q_{out} is the discharge to a down-stream cell. R_{VISIT} indicates the runoff of each grid cell calculated by VISIT model (m^3 d^{-1}). For the transport directions, we used the river routing map TRIP (Oki and Sud 1998), which defined the river network topology under the assumption that each grid cell can drain into one of the eight next-neighbor cells. Q_{in} is calculated as the sum of the upper stream Q_{out} according to the routing map. Q_{out} is defined as follow;

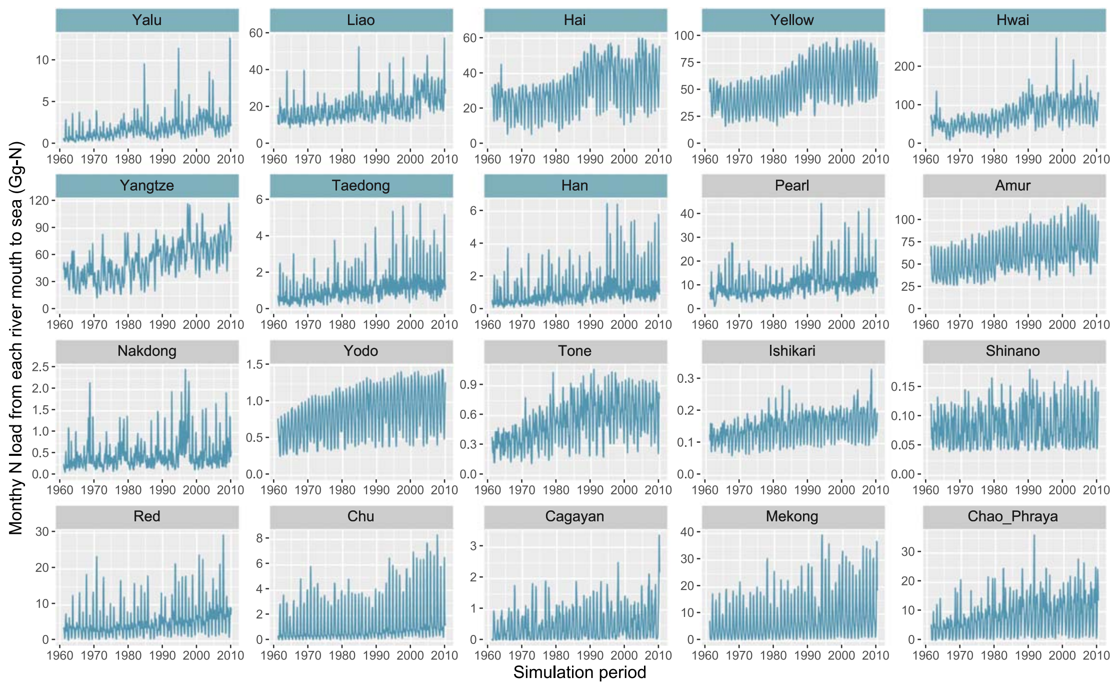

Figure 2. Monthly inorganic N loading to the ocean at major river mouths in the East China Sea (9 rivers, blue) and adjacent East Asian (10 rivers, gray) watersheds. The locations of the river mouths are shown in figure S2.

$$Q_{out} = S_{riv} \times \min\left(\frac{\nu}{d}, 1.0\right) \tag{2}$$

where ν is the flow rates and d is the river length in the grid cell. We set ν to be 0.5 m s^{-1} according to the global hydrological model H08 (Hanasaki *et al* 2006).

For the N transport, we formulated in the same manner as water discharge as follows;

$$\frac{dSn_{riv}}{dt} = Qn_{in} - Qn_{out} + Ln + Hn - U_{den} \tag{3}$$

Sn_{riv} indicates Rriver N storage (kg-N) in each grid. Qn_{in} and Qn_{out} are inflows from upper stream and an outflow to a down-stream for N loading (kg-N d^{-1}), respectively. Ln is leaching N as to be NO_3^- from both natural and agricultural ecosystems, which are the outputs of VISIT model in this study. Hn is the input of total inorganic N (TIN) derived from human sewage in each grid. Qn_{out} is defined as follow;

$$Qn_{out} = Sn_{riv} \times \min\left(\frac{\nu}{d}, 1.0\right). \tag{4}$$

Ln in equation (3) is N inputs in each grid via N leaching as nitrate from the ecosystem (kg-N d^{-1} in each grid^{-1}), which is estimated by VISIT model. We assumed H_n to be dissolved inorganic nitrogen forms (i.e. mainly NH_4^+ or NO_3^-) and oxidized to nitrate by nitrifier by the time it entered the main channel. Hence, TIN was treated as to be equivalent to NO_3^- in the river storage in the model and can be denitrified as following the equation;

$$U_{den} = \frac{V_{\max}[NO_3]}{k + [NO_3]} R_{\text{area}} \tag{5}$$

U_{den} is denitrification rates (kg m^{-2} day^{-1}). [NO_3] is a the NO_3^- concentration (mg-N L^{-1}) in the river storage, which is defined as Sn_{riv}/S_{riv} in this study. These variables are calculated on a daily basis and are endogenously calculated in the model. Here, denitrification rate are defined as Michaelis–Menten kinetics, which V_{max} and k represent the maximum rate and the substrate concentration at half V_{max} value. In this study, these parameter values are obtained from Mulholland *et al* (2009). V_{max} and k are 93.6 mg-N m^{-2} day^{-1} and 0.422 mg-N L^{-1}, respectively. R_{area} indicates river surface area in each grid. For R_{area}, we used a dataset based on open street map (Yamazaki *et al* 2019).

To estimate human sewage input Hn_{total}, we utilized the following country-specific data set provided by Bouwman *et al* (2005). They are: total nitrogen discharge into wastewater per capita (Cn; kg-N capita^{-1} year^{-1}), nitrogen removal efficiency of wastewater treatment facilities (R_{wwt}; %), percentage of the population with access to sanitation in urban areas (A_U; %), and percentage of the population with access to sanitation in non-urban areas (A_R; %). These data were smoothed with a spline function for three different years (1970, 1990, and 2030) to create a data set for the period 1960-2010 (with negative values and those exceeding 100% rounded up

Table 1. Summary of input data used in this study.

Input data	Spatial & Time resolution	Reference
Fertilizer & Manure	half degree & yearly	Wang *et al* (2020b)
Manure	half degree & yearly	Wang *et al* (2020b)
N deposition	half degree & monthly	Sudo *et al* (2002), Dentener *et al* (2006)
Sewage	country & yearly (interpolated)	Bouwman *et al* (2005)
Population	half degree & yearly	Klein Goldewijk *et al* (2017)
Climate	half degree & monthly	CRU TS3.25 dataset in Harris *et al* (2014)
Land-use map	half degree & yearly	Monfreda *et al* (2008)

and down, respectively) to provide time series data. We used historical gridded population (P) data at half degree resolutions in Hyde3.2 database (Klein Goldewijk *et al* 2017) at this period. The urbanization fraction for each grid (U_{frac}; %) used was the year 2000 data (Gao and O'Neill 2020) for the entire simulation period. Using these data, the total wastewater nitrogen loadings to rivers were calculated using the following equation;

$$Hn_{total} = CnP - CnPU_{frac}\,U_{frac}A_U R_{wwt} - CnP(1 - U_{frac})A_R R_{wwt} \tag{6}$$

We took only total inorganic nitrogen for Hn, therefore, we estimated DIN in sewage effluents using the same calculation method adopted by Dumont *et al* (2004) and He *et al* (2011) as follow;

$$Hn = Hn_{total}[0.485 + 0.255T_N / max(T_N)] \tag{7}$$

T_N is a country by country fraction of TN removed by wastewater treatment compiled by Bouwman *et al* (2005). A detailed explanation is in Dumont *et al* (2004).

In this study, we focused on and analyzed the N budget of East China Sea (including Yellow sea) because this sea is trans-boundary region of East-Asian countries. Table 1 summarize input data. We set the boundary as in figure S1 (in supplemental material available online at stacks.iop.org/ERC/3/085005/mmedia). The assumed total area of watersheds surrounding East-Asia sea and Yellow sea is 4 330 837 km^2 in the model .

3. Results and discussions

3.1. N loading in simulation results and previous literatures

The almost all rivers in East-Asian region showed the increase trends during 1961 to 2010, however, the extent of TIN load to seas largely varied among the rivers (figure 2). Yangtze river, that is the largest river in East-Asia, had the highest TIN load among the watersheds of the East China sea, which ranged from 0.46 Tg-N year^{-1} to 1.39 Tg-N year^{-1} during 1961–2010 in our estimation. For example, compared to previous modeling evaluation Ti *et al* (2012), the estimates of annual TIN load to the ocean in the Yangtze River ranged from 0.74 Tg-N Year^{-1} to 1.01 Tg-N Year^{-1} between 1985 and 2007, whereas the present study's estimates for the same period ranged from 0.65 Tg-N Year^{-1} to 1.14 Tg-N Year^{-1}. In assessing the TIN loadings of the Yangtze River by observations, (He *et al* 2011) reported 0.58 Tg-N Year^{-1} in 1995, Müller *et al* (2012) reported 1.0 Tg-N Year^{-1} in 2010. On the other hand, the TIN loading in the 1960s was about 0.1 Tg-N Year^{-1} in Müller *et al* (2012), which is considerably lower than our study. It was determined as a product of annual average nitrate concentration and annual flow rate, and although it is an actual measurement-based estimate, there is a high degree of uncertainty in the estimate. Thus, we have genenally similar TIN estimates for the Yangzte River with previous estimates, while our estimates for the Yellow River in this period (from 0.65 Tg-N Year^{-1} in 1985 to 0.85 Tg-N Year^{-1} in 2007) are larger than the Ti's estimates (from 0.36 Tg-N Year^{-1} in 1985 to 0.59 Tg-N Year^{-1} in 2007) (Ti *et al* 2012). Estimates of the annual total nitrogen loading of the Yangtze River have been studied relatively more and the estimates are more available. Liu *et al* (2018) summarized the previous estimations on the total N load on the mouth of Yangtze and noted that the estimates of annual total nitrogen loading in 2000 considerably varied depending on the method of estimation and calibration (e.g., 1.1 Tg-N in NEWS model (Mayorga *et al* 2010), 1.1 Tg-N in Global NEWS-2 model (Strokal *et al* 2016), and 3.5 Tg-N (Liu *et al* 2018) in IMAGE-GNM model). Liu *et al* (2018) pointed out that previous studies have not validated for N load at the Yangtze River estuary, but at the monitoring station more than 600km upstream from the mouth of the river. Our simulation shows that the nitrogen load in the Hwai River exceeded the Yangtze and isthe largest in the region, at 1.11 Tg-N Year^{-1} in 2000. The river channel model used in this study Oki and Sud (1998) didn't take into account the tributaries of the Hwai River, which split and partially join the Yangtze River at the downstream near the mouth of the Hwai river. Therefore, we thought that the Hwai river has the maximum nitrogen load, which is an artifact. If this branching and confluence of the Hwai River were taken into account, the nitrogen load in the Yangtze River

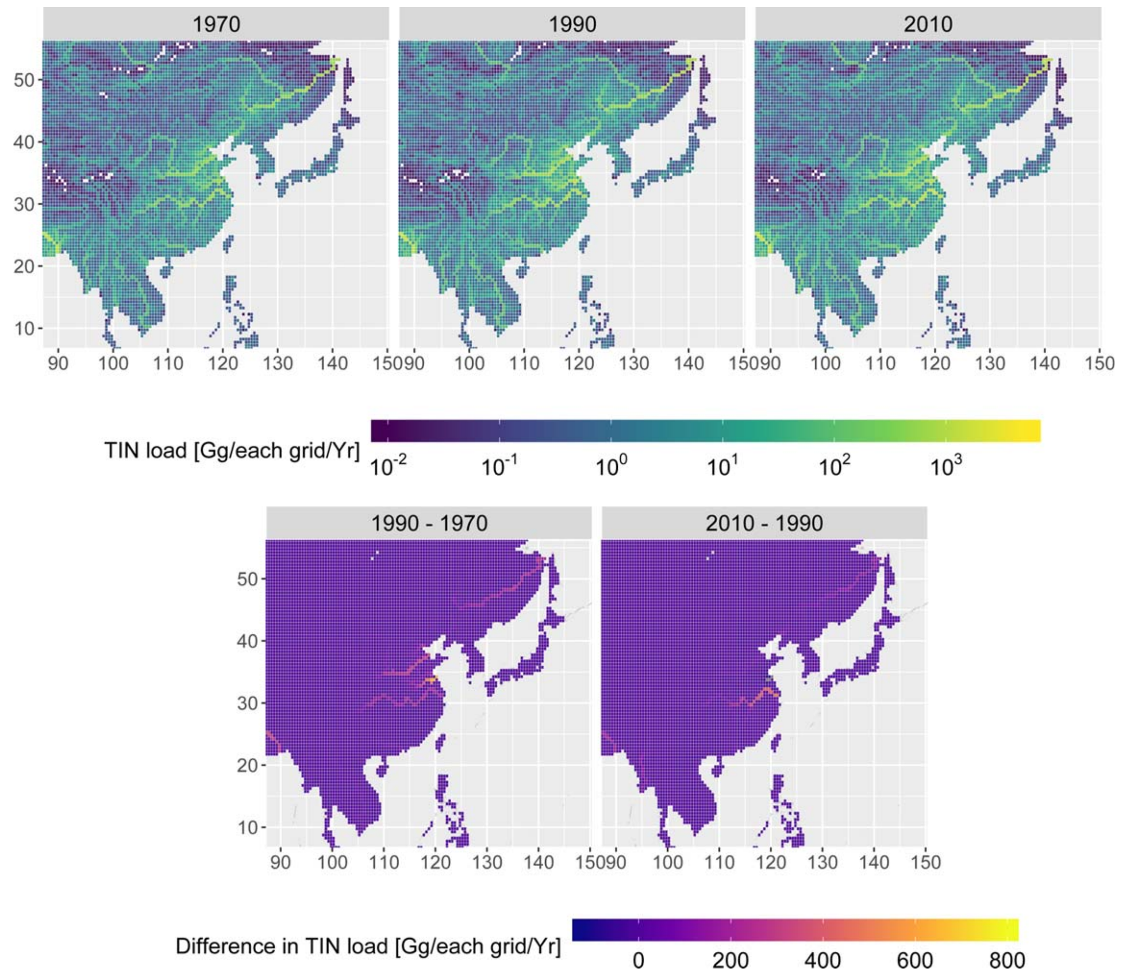

Figure 3. Spatial distribution of inorganic N load in rivers (Gg-N/cell/year). Lower plots indicate the difference in spatial distribution N load between 1970 and 1990 and between 1990 and 2010 (Gg-N/cell/year). The area of a cell is about 2500km^2 at the equator.

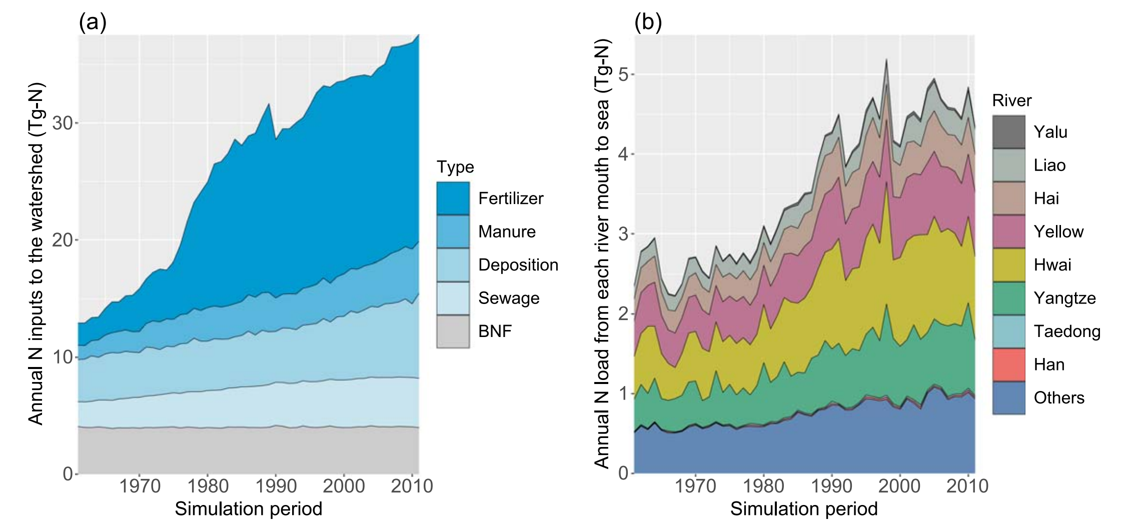

Figure 4. Annual rN inputs to the watershed (a) and riverine TIN load to the East China sea from the river mouths.

would be higher than the current calculations, but still smaller than the estimates of Liu *et al.*'s study (Liu *et al* 2018).

Chinese big rivers (Yellow, Pearl, Hwai, Liao, Yalu, Hai and Amur) showed a clear increase trends in N load and almost its doubling or even increasing in this period. While, as pointed out in the previous study for Chinese N_2O emission (Shang *et al* 2019), the rate of increase of the emission is weakened in China after 2000 (figure 3).

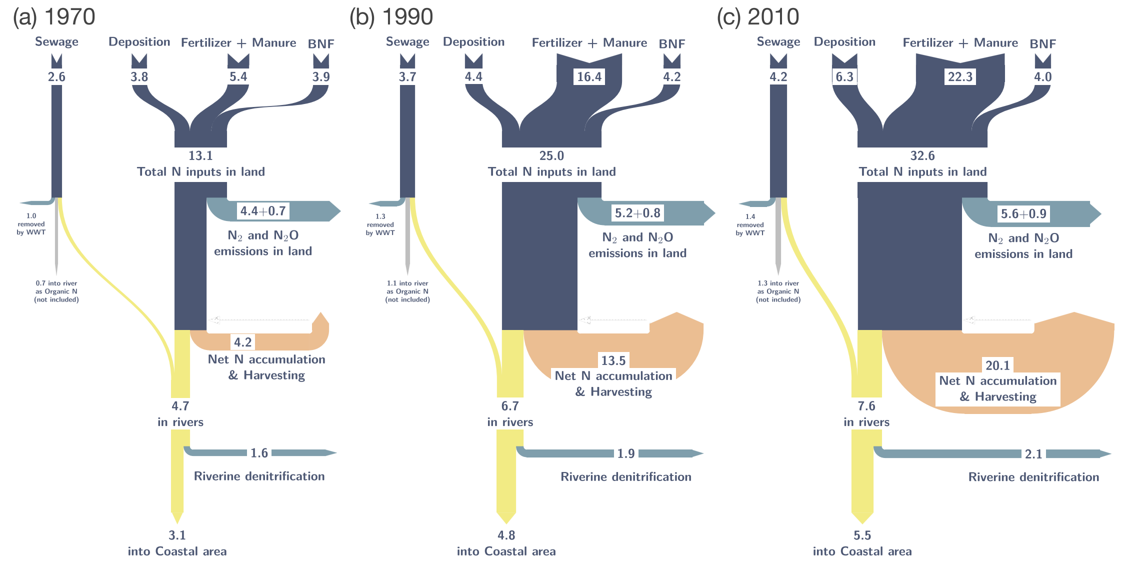

Figure 5. Annual N flows from land to sea in the watersheds surrounding East-China sea (and Yellow sea) in 1970 (a), 1990 (b), and 2010 (c). Width indicates the amount of each N flow. The unit of value is Tg-N. Yellow color indicates inorganic N (TIN) flows in river channels (N leaching, sewage, and N load in rivers). Blue color indicates the reactive N flows. Light blue indicates N_2 (and N_2O) gas emission to atmosphere. N gas emissions in land indicates sum of the emissions from both natural and agricultural ecosystems.

This is partially because the N use efficiency of fertilizers has improved over recent decades (Shang *et al* 2019). In other words, the amount of nitrogen input per area of land was reduced. On the other hand, South-East Asian rivers (i.e., Red, Chu, Mekong, Chao Phraya) showed the increase trends still in the 2000's (figure 2), could indirectly contribute to the nitrogen load in the East China Sea via the South China Sea (Chen 2008).

In this study, we focused on TIN loads around the East-China sea (including Yellow sea) as a trans-boundary sea among East-Asian countries (figure S1). During 1961–2010, total annual N inputs (the sum of N deposition, N fertilizer, manure and biological fixation) in the watershed around the East China sea have increased from 12.9 Tg-N Year^{-1} to 36.9 Tg-N Year^{-1} (figure 4(a)). While, between 1961 and 2010, the river TIN loadings from the watershed surrounding the East China Sea increased from 2.7 Tg-N Year^{-1} to 5.5 Tg-N Year^{-1} (figure 4(b)), more than doubling the loadings. Although there are few examples of studies on the assessment of nitrogen loading in the same area, some comparable estimates exist. In one study by Li *et al* (2014). the river-derived DIN loading to the East China Sea was 1.6 Tg-N Year^{-1} to 6 Tg-N Year^{-1} from 1980 to 2007. A very recent example is the total nitrogen assessment study by Wang *et al* (2020a) in which the total nitrogen load from 1970 to 2010 ranged from 1.8 Tg-N Year^{-1} to 7.9 Tg-N Year^{-1}. Although the rate of increase is different from these two studies, the number of orders of magnitude is comparable with our estimate. The Chinese Rivers' contribution of N loading to the East China Sea was the highest among the rivers and averaged 78.0% ($\pm$ 3.2% in standard deviation (S.D.)) over the simulation period (figure 4). Therefore, the time-series of total N loading to the East China Sea was almost identical to that of the major rivers in China. That is, the total N loading had a consistent upward trend from 1960 to 1990, and the increase slowed down and stagnated in the 2000s. However, rivers not mentioned here by name (i.e. others) contributed nearly 20% of the total N loadings, and their total loadings continued to increase by almost 1.0 Tg Year^{-1} at 2011.

3.2. N cascade in the watershed

To visualize the nitrogen cascade in the watershed of the East-China sea (figure S1), we summarized N flows from land to sea by Sankey diagram as in figure 5. As also shown in the time-series of N loading in major rivers (figure 2), the TIN loading to the sea nearly doubled from 3.1 Tg-N Year^{-1} to 5.5 Tg-N Year^{-1} during 1970 to 2010. It is attributed to the increase of anthropogenic N inputs, that is, N deposition, synthetic N Fertilizer, manure, and human sewage (figure 5). The nitrogen load from human sewage increased from 2.6 Tg-N Year^{-1} to 4.2 Tg-N Year^{-1} during this period, while the inflow as TIN, excluding nitrogen removal by wastewater treatment facilities and the non-organic nitrogen fraction, ranged from 0.9 Tg-N to 1.5 Tg-N. In this period, the highest increase of anthropogenic N source was in the synthetic N fertilizer and manure inputs which increased from 5.4 Tg-N Year^{-1} to 22.3 Tg-N Year^{-1}, that is, about a four-fold increase (figure 4). However, the N load from land to sea has not increased as much as the rate of increase in synthetic N fertilizer and manure. The fertilizer dataset used in this study (Wang *et al* 2020b) are based on China's sub-national scale statistics, which reflect the decline of fertilizer N application rates since 2003 partially due to the improvement of nitrogen use

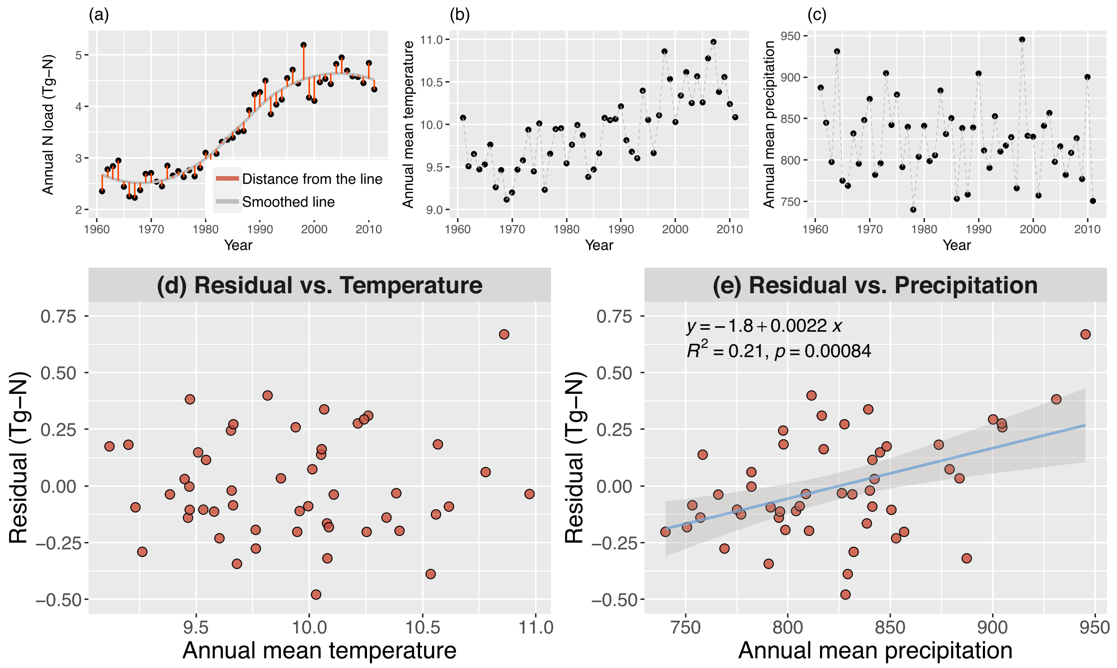

Figure 6. Relationship between TIN load and meteorological factors in the watersheds to East China Sea. Changes in TIN load over time (same as figure 4(b)) and evaluation of residue components without trend (a), and historical changes in basin-wide average annual mean temperature (b) and annual precipitation (c). Relationship between the annual mean temperature and residue components (d) and relationship between precipitation and residue components (e). In (a), trend extraction using locally estimated scatterplot smoothing (loess) (gray line) and the residuals (red line) are shown, and the vertical axis in (d) and (e) shows this detrended residue component. The blue line in (e) shows the regression line, and the gray envelope shows its 95% confidence interval.

efficiency by farmers' efforts, especially in the North China plain and near the lower reaches of the Yangtze River (Shang *et al* 2019). This reduction in fertilizer application is due to improvements in varieties and cultivation techniques, but is a regional characteristic that was not reflected in the nationally aggregated statistics (Shang *et al* 2019). The background to this trend is also related to China's recent agro-environmental policies. Chinese national policies such as National Soil Testing and Nutrient Recommendation program, which started from 2005, would contribute the improvement of nitrogen use efficiency of the crop in China (Liu *et al* 2015). In addition to these reasons, large scale farming have been gradually increased due to the rapid urbanization in China, which would contribute the reduction of N fertilizer application rates through the relatively effective nutrient management in the large-scale farms (Ju *et al* 2016). In 2015, the Chinese government introduced an action to reduce the use of chemical fertilizers to zero by 2020 (Jin and Zhou 2018). Therefore, it is expected that the contribution of nitrogen fertilizer will be reduced more recently.

Crop processes in the VISIT model are represented as grasslands of C3 plants, which are harvested annually, but harvesting of edible parts such as grains is not considered. So, the model does not mimic the actual improvement in the nitrogen management in the watershed of the East-China sea except fertilizer rate optimization (e.g., straw incorporation, improved crop varieties and mechanization; Zhang *et al* 2017), however; the decrease in nitrogen loss occurs due to the decrease in the amount of fertilizer applied per unit area. This is because the increased crop harvesting contributed to limiting the increase of N leaching. On the other hand, in natural ecosystems, there is an increase in the net accumulation of biomass N and soil organic matter nitrogen, which mitigates the increase in TIN due to increased nitrogen fallout. In addition, the increase of N_2 and N_2O emissions partially attributed to reducing the increase of N load to sea (figure 5). Nitrogen removal by denitrification on rivers has increased from 1.6 Tg-N Year^{-1} to 2.1 Tg-N Year^{-1} from 1970 to 2010, and it is calculated that about 30% of the TIN is denitrified for the TIN entering the river. This study was evaluated using the same parameters obtained in the nationwide ^{15}N tracer experiments in the US conducted by Mulholland *et al* (2009), which have not been validated in the East-Asia region. Currently, river denitrification is evaluated only in a very limited number of models Seitzinger *et al* (2006), which does not allow for sufficient comparison. Future verification is essential. Although it is a very rough comparison, Seitzinger's review (Seitzinger *et al* 2006) showed that globally, denitrification in rivers is 35 Tg-N Year^{-1} compared to 125 Tg-N Year^{-1} in terrestrial areas, a ratio of about 28%, which is generally similar to the ratio of terrestrial to river denitrification in this study. In addition, in the Jiulong River located in southeast China, DIN removal rate by denitrification has been estimated based on field measurements, and the nitrogen removal rate by denitrification in the river was estimated to be 24% (Chen *et al* 2014), which is almost the same order of magnitude. In this study, the parameter V_{max}, which defines the maximum areal N_2 emission rate, is set to 93.6 mg-N m^{-2} day^{-1}, based on the study by Mulholland *et al* (2009). On the other hand, the observed value of areal denitrification rates, which is the basis for the estimates in Chen *et al* (2014), ranged from 2 mg-N m^{-2} day^{-1} to 589 mg-N m^{-2} day^{-1}. This range is comparable to the present study. However, previous estimates made in the Yangtze River, although based on observations made over a six-month period from autumn to winter, showed a very low nitrogen removal rate of about 2% in the river (Yan *et al* 2004), indicating a large degree of uncertainty.

Thus, due to the N flows of various stages of the nitrogen cascade, the nitrogen load from the river to the East China Sea shows a moderate rate of increase relative to the nitrogen input of the basin. So, We should pay attention to the change in land use as well as the amount of reactive nitrogen input to understand the past and future trends of N load from land to sea.

3.3. Meteorological factors and TIN load

As mentioned in the previous section, the annual nitrogen load changes significantly over the calculation period, showing year-to-year variations apart from the trend. This is due to meteorological factors. In order to focus only on this inter-annual variability, the trend due to changes in nitrogen input was detrended by smoothing, and the residue was analyzed. Comparing the residue of nitrogen loading with the trend of annual precipitation and annual mean temperature averaged over the entire East China Sea basin (figures 6 and S1), a significant positive linear relationship with annual precipitation was found ($p < 0.001$). This is because as rainfall increases, the nitrogen retention time in the watershed is shortened due to the increased water flux in the soil, and N load increases (Sinha *et al* 2017). Precipitation in this region is expected to increase throughout the 21st century, and Guo *et al* (2017) estimated that annual rainfall will increase by about 80 mm in 2050 compared to the present, and there will be more years of increased rainfall if extreme events are included. Therefore, it may be necessary to pay attention to the increase in nitrogen runoff due to climate change.

There is no significant correlation between annual mean temperature and residue. This does not mean that temperature increase does not affect TIN loading. The mean annual temperature has a clear upward trend over the simulation period (figure 6(b)), and the residues do not include the increase trend due to the detrending. Therefore, the effect of temperature rising cannot be estimated by this analysis, but its inter-annual variation in temperature was not consistent with that of nitrogen loading, so it is at least not a regulation factor in inter-annual variability of the N loading in this region (figure 6). While temperature increases can have the effect of accelerating TIN load by increasing biological activities such as nitrogen mineralization and nitrification, there are also offsetting effects of increased TIN load, such as increased nitrogen assimilation in ecosystems (Greaver *et al* 2016). For example, Ballard *et al* (2019) showed in a statistical analysis of US rivers that springtime temperature has a negative effect on TIN loading. How the nitrogen budget will be balanced in the East China Sea basin under future climate change is still unclear and further research is needed.

3.4. Limitations of this study

Finally, we discuss about the shortage of our approach. Our model don't consider the N load to sea via submarine groundwater discharge (Beusen *et al* 2013). For example, in China, total N load to groundwater also increased recently (Gu *et al* 2013), which is attributed to the increase of N load from to sea. Although the direct N inputs via groundwater to sea in the coastal area is not major loading from land to sea as in the previous modelling study (Wang *et al* 2020a), groundwater processes –N loss by denitrification in underground (Bouwman *et al* 2013) and time delay of N load to rivers (Wang *et al* 2013)–could surely reduce the total TIN load from land to sea in the N cascade. While the model improvement of VISIToRN is essential to evaluate more accurate N load to the seas, our evaluation of N load are often in trust in understanding trends because the general historical trend of N load in the rivers were governed by the anthropogenic Nr inputs. In addition, as discussed in 3.2, the uncertainty of various parameters in the model has become a problem in quantitative evaluation. We believe that more integration of the findings from field observations is necessary for improvement.

4. Conclusion

Nitrogen loadings to the seas via rivers increased rapidly especially in the 1970s and 1980s ((figure 4)), but in recent years, the increase in loading has slowed in many rivers in East-Asia. In the light of the East China Sea basin, we found that the increase in various types of reactive nitrogen flows in the N cascade makes a significant contribution to mitigates the increase in TIN (figure 5), which also indicates significant changes in N stock. The implications of these considerable buildup of N have to be resolved in future studies that would also account for the N removal with harvest. Although there has been a slowing trend in recent years, however, one BAU scenario estimates that reactive nitrogen inputs will double in the region by 2050 (Gu et al 2015). So, we need a special attention to Nr cycling and improve the Nr managements in anthropogenic sources in this region.

Although slowing down in recent years, non-point sources such as fertilizer and manure applications are still the main Nr loads to the watershed. Therefore, their reduction is very important for the reduction of riverine Nr load. N fertilizer to agricultural land is expected to be further reduced by recent large-scale policies in China (Jin and Zhou 2018), but may be weakened by increased rainfall due to climate change (Bowles et al 2018). Nitrogen management adapted under climate change may become more important in this region.

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Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

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