



# Variations and its driven factors of anthropogenic nitrogen loads in the Yangtze River Economic Belt during 2000–2019

Meng Cui<sup>1,2</sup> · Qingjun Guo<sup>2,3</sup> · Yujun Wei<sup>4,5</sup> · Xuening Yu<sup>6</sup> · Jian Hu<sup>7</sup> · Liyan Tian<sup>8</sup> · Jing Kong<sup>9</sup>

Received: 13 December 2021 / Accepted: 6 July 2022 / Published online: 5 August 2022  
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## Abstract

Since the millennium, China has economically taken off with rapid urbanization, and anthropogenic nitrogen emission intensity has undergone remarkable changes. To better understand the impact of urbanization on anthropogenic nitrogen, this study calculated the spatio-temporal heterogeneity of anthropogenic nitrogen in the Yangtze River Economic Belt (YREB) since 2000, based on the estimation, using obstacle analysis to quantify the driving of industry and agriculture on N growth and using the gray model to analyze the impact of urbanization on N changes. Additionally, using the environmental pressure model to predict the future N load. The results indicated N load in the YREB increased rapidly from 21.4 Tg in 2001 to a peak of 24.5 Tg in 2012 and then decreased to 22.2 Tg in 2019. Although N flux gradually increased from the west to the east of the YREB, the growth rate had an opposite trend with a negative growth in the eastern region. Hotspots are mainly concentrated in urban agglomerations, which contributed to ~60% N load of the YREB, and the YREB contributed to ~90% N load of the Yangtze River Basin. Obstacle degree scores indicated wastewater was a major industrial driver of N growth before 2010, and then became waste gas; increased mechanization and fertilizer control effectively reduced nitrogen emissions during agricultural development. The gray analysis of urbanization indicated urban population, industry, and services had the strongest correlation with N load changes. Scenario simulations suggest N loads of the YREB remain at a high level by 2030; however, there are still opportunities to effectively control N growth through high technological innovation and reducing the proportion of industry under an enormous population. This research contributes to a better understanding of the impact of urbanization on anthropogenic nitrogen and helps developing countries to precisely control nitrogen hotspots and sources.

**Keywords** Nitrogen loads · Urbanization · Human activities · Yangtze River Economic Belt

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Responsible Editor: Baojing Gu

✉ Qingjun Guo  
guojq@igsnr.ac.cn

<sup>1</sup> National Marine Data and Information Service, Ministry of Natural Resources, Tianjin 300171, China

<sup>2</sup> Center for Environmental Remediation, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

<sup>3</sup> College of Resources and Environment, University of Chinese Academy of Sciences, Beijing 100190, China

<sup>4</sup> Research Center for Intelligent Society and Governance, Zhejiang Lab, Hangzhou 310000, China

<sup>5</sup> Key Laboratory of Regional Sustainable Development Modeling, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing 100101, China

<sup>6</sup> School of Geography and Ocean Science, Nanjing University, Nanjing 210023, China

<sup>7</sup> Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

<sup>8</sup> Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China

<sup>9</sup> School of Environmental and Chemical Engineering, Jiangsu Ocean University, Lianyungang 222005, China

## Introduction

Human activities are increasingly disturbing the global nitrogen cycle (Fowler et al. 2013; Zhang et al. 2020b). Large-scale nitrogen production and use have drastically improved industrial and agricultural production efficiency, but also caused many negative effects on the environment, such as water eutrophication (Baulch 2013; Liu et al. 2018), soil acidification (Lu et al. 2015), and air pollution (Chen et al. 2012; Huang et al. 2021). Presently, more than half of severe global environmental problems are related to nitrogen cycle changes caused by human activities, and the nitrogen cycle has become one of the three major security threats beyond the planetary boundary (Rockstrom et al. 2009; Vitousek et al. 2013). Environmental pollution caused by nitrogen cycle imbalance in many countries and regions is becoming more serious (Kim et al. 2014; Sun et al. 2021), and the world faces huge challenges in reducing reactive nitrogen and coping with negative environmental problems caused by excessive nitrogen (Erisman et al. 2013; Houlton et al. 2019). In developing countries, in particular, rapid urbanization and unbalanced regional development are constantly changing nitrogen emission intensities, which in turn leads to more complex, comprehensive environmental management.

A series of urban agglomerations have formed through urbanization and regional development, which are key to promoting national economic development and sensitivity to the ecological environment (Cui et al. 2020; Wu et al. 2015). Cities in agglomerations cross administrative boundaries and interact and influence each other's economy, transportation, and population movement, leading to expansion and complexity of environmental pollution (Seto et al. 2010). In recent decades, China has economically taken off (Bekkers et al. 2021). However, the Yangtze River Economic Belt (YREB) and its urban agglomerations are in the initial stage of development and accompanied by a series of environmental problems, especially eutrophication caused by nitrogen and phosphorus (Chen et al. 2017). Although a series of articles have analyzed the hotspots and spatio-temporal heterogeneity of anthropogenic nitrogen flux in the Yangtze River Basin (YRB) and identified agricultural sources as major nitrogen contributors (Cui et al. 2021; Chen et al. 2016; Wang et al. 2014), there remains a lack of in-depth discussion on how urbanization processes affect nitrogen. The basin contains multiple administrative units, making it difficult to ascertain clear liability. Meanwhile, because of the unbalanced development of various regions within the economic belt (Li and Liu 2017) and significant differences in environmental management levels, it is difficult to

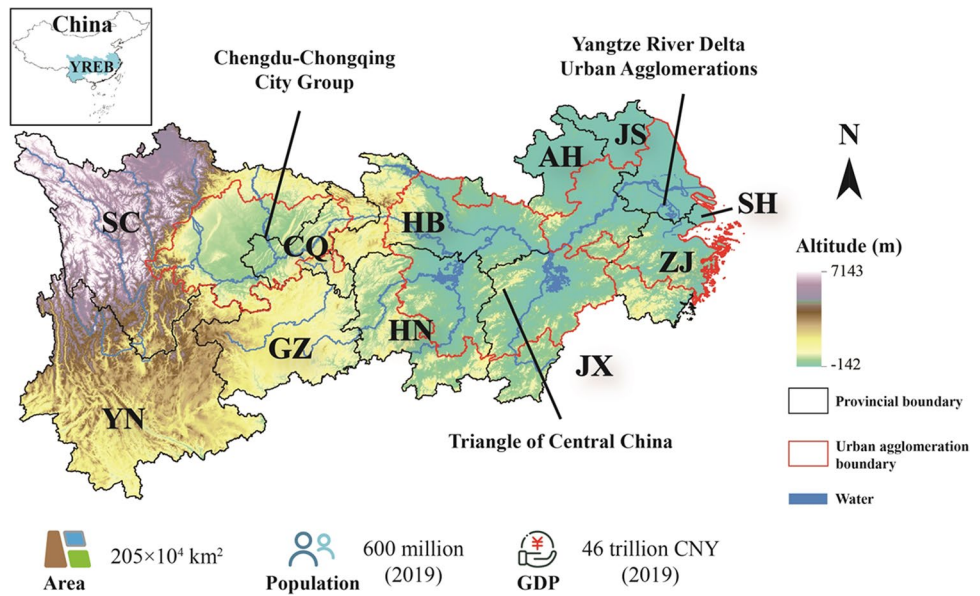
effectively control water pollution in the YRB. Therefore, investigating the impact of human activities on nitrogen loads during socioeconomic development will not only promote sustainable development of the economic belt but also maintain the ecological environment of the basin and estuary area.

Evaluating spatial–temporal variations and identifying emission hotspots are prerequisites for understanding current nitrogen problems in the YREB. However, there are some difficulties in estimating nitrogen loads according to established practice and experience:

- (i) Prefecture-level statistical data are conducive to the establishment of a detailed spatial distribution. However, prefecture data frequently contain fewer indicators and more missing data. Although provincial data cannot directly cover smaller administrative regions, there are more types and larger amounts.
- (ii) Normally, prefecture-level statistical data issued on paper will not be corrected, updated, or republished. However, provincial statistics are often corrected and updated based on nationwide censuses and other work.
- (iii) The YREB includes more than 100 prefecture-level cities. Annual data compilations for each city consume considerable manpower and time, and data quality may also be poor. For greater feasibility, some studies select statistics for specific years rather than continuous time series.
- (iv) Administrative region changes cause statistical data changes. In recent years, China's frequent urban expansion, and administrative unit divisions and mergers have caused substantial difficulties for researchers.

Although our previous work analyzed the nitrogen sources and spatiotemporal differentiation characteristics of the YRB, there are relatively few studies on how human activities trigger changes in nitrogen during urbanization. In the YREB, a more accurate estimation of anthropogenic nitrogen, the impact of socioeconomic development on nitrogen loads, and identification of spatial–temporal characteristics are key to effective control of nitrogen loads and reduction of environmental risks. Based on the above, the contents and objectives of this study mainly include (i) calculating interannual nitrogen loads since 2000 using the mass balance model and socioeconomic data; (ii) expanding nitrogen load data from provincial resolution to kilometer grid using the population density weighting method, and obtaining nitrogen load spatial–temporal characteristics; (iii) determining industrial and agricultural development and urbanization drivers of changing nitrogen loads by the gray relational

**Fig. 1** The overview of the Yangtze River Economic Belt (YREB). The YREB has 11 provincial administrative regions including Shanghai (SH), Jiangsu (JS), Zhejiang (ZJ), Anhui (AH), Jiangxi (JX), Hubei (HB), Hunan (HN), Chongqing (CQ), Guizhou (GZ), Sichuan (SC), and Yunnan (YN)



model; (iv) estimating the impact of nitrogen loads on YRB with spatial superposition analysis; and (v) simulating future nitrogen load changes under different scenarios and proposing countermeasures. By achieving the above goals, we will help developing countries control nitrogen hotspots and nitrogen sources more precisely with more explicit management objects, and support to better understand the driving effect of the urbanization process on anthropogenic nitrogen.

## Materials and methods

### Study area

The YREB covers 2.05 million square kilometers, accounting for approximately 21% of China's land area (Fig. 1). More than 40% of China's total population and GDP comes from this area, as does one-third of its freshwater resources and food production. The YREB has more than 100 prefecture-level cities, including 9 provinces such as Sichuan, Yunnan, Guizhou, Hubei, Hunan, Anhui, Jiangxi, Jiangsu, Zhejiang, and 2 municipalities directly under the Central Government, such as Chongqing and Shanghai. To promote central and western region development and modern urbanization construction, the Chinese government approved development plans for three city clusters in 2015, 2016, and 2018, respectively, including Chengdu-Chongqing City Group (CCCG), Triangle of Central China (TCC), and Yangtze River Delta Urban Agglomeration (YRDUA). The detailed information on city clusters is shown in the Supplementary.

## Nitrogen loads and visualization

### Estimation of nitrogen loads

Our team successfully built and implemented a model for calculating nitrogen loads in the YRB (Cui et al. 2021), which does not consider details of nitrogen flow and establishes a black box for all human activity for the final input source (including fertilizer, recycling nitrogen, biological nitrogen fixation, aquaculture, atmospheric deposition, wastewater discharge). In this study, we use the model to study the nitrogen loads in the YREB. The calculation framework is as follows:

$$N_{INPUT} = \sum_{i=1}^{11} (N_{FER_i} + N_{BIO_i} + N_{REC_i} + N_{AQU_i} + N_{PS_i} + N_{DEP_i}) \quad (1)$$

where  $N_{INPUT}$  represents total nitrogen loads in the YREB (Tg/year);  $i$  represents the province in the economic belt.  $N_{FER}$  represents the amount of fertilizer nitrogen that is not absorbed by crops and is lost to the environment (fertilizers include nitrogen fertilizer and compound fertilizer).  $N_{BIO}$  is the amount of biological nitrogen fixation. Biological nitrogen fixation is a process whereby symbiotic nitrogen fixation of plants or the non-symbiotic nitrogen fixation of soil microorganisms convert  $N_2$  in the atmosphere into free nitrogen. Crop nitrogen fixation is mainly calculated according to planting area and crop nitrogen fixation rate. Nitrogen-fixing crops include rice, peanuts, beans, and other crops in this study.  $N_{DEP}$  is atmospheric nitrogen deposition ( $NO_x$  and  $NH_3$ ).  $N_{REC}$  is recycling nitrogen (sources mainly include manure produced by rural residents and livestock, and residual crops returned to the field).  $N_{AQU}$  and  $N_{PS}$  are aquaculture (fish, shrimp, crabs, and shellfish) and wastewater

nitrogen (industrial and domestic discharge), respectively. Interannual changes of various nitrogen end members are shown in the Supplementary section, and details of each end member are shown in published work by Cui et al. (2021).

To improve result accuracy, we revised our previous work including atmospheric deposition and wastewater discharge. This study used two global human activity nitrogen emission databases, CAMS (Copernicus Atmosphere Monitoring Service) (Granier et al. 2019) and ACCMIP (Atmospheric Chemistry and Climate Model Intercomparison Project) (Lamarque et al. 2010), to estimate nitrogen deposition in the YREB. By superimposing the  $\text{NO}_x$  and  $\text{NH}_3$  grids of the two databases, total nitrogen deposition was obtained by averaging. Wastewater nitrogen includes industrial wastewater and domestic sewage discharge. In the current statistics, ammonia nitrogen data are relatively complete records, while total nitrogen data only covers the period 2011–2017. Therefore, we used the 7-year total nitrogen and ammonia nitrogen data of 11 provinces in the YREB to establish a regression relationship to estimate total nitrogen for the missing years ( $R^2$  reached 0.88,  $p < 0.05$ ) (Figure S1 in Supplementary). The formula for estimating wastewater total nitrogen in this study is:

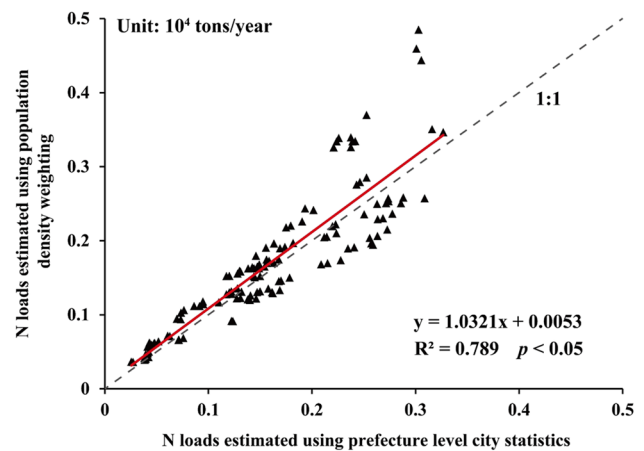
$$N_{PS} = 1.4998 \times N_{NH} - 0.412 \quad (2)$$

where  $N_{PS}$  is the total N of wastewater discharge ( $10^4$  tons/year), and  $N_{NH}$  is the sum of ammonia discharge from industrial and domestic wastewater ( $10^4$  tons/year).

### Mapping of nitrogen loads

We expanded the nitrogen distribution from the provincial scale to  $1 \text{ km} \times 1 \text{ km}$  through population density weighting. Population density data were derived from the Resource and Environment Data Cloud Platform, Chinese Academy of Sciences. Since these data also included a series of human activity characteristics, such as land use types, we did not weigh other factors to avoid double counting. Additionally, nitrogen deposition raster data comprehensively considered numerous influencing factors, and so, weighting was unnecessary.

To verify the feasibility of this method, we randomly selected a series of prefecture-level cities. According to data availability and completeness for each city and the matching degree of indicators, 48 cities that could be used to calculate nitrogen loads were selected that had at least 2 years of complete data. ArcGIS software was utilized to extract and calculate nitrogen loads of these cities. By calculating nitrogen load using population density weighting, and directly using prefecture-level city statistics (Fig. 2), we found that the population density-weighted estimation of regional nitrogen loads was feasible. Determination coefficient by the two methods was 0.79,  $p < 0.05$ , relative error range was  $-0.34$ – $0.34$



**Fig. 2** Comparison of two methods for estimating nitrogen loads at the prefecture-level city scale. The  $x$ -axis represents the nitrogen loads estimated directly from the statistics of prefecture-level cities; the  $y$ -axis represents the nitrogen loads estimated by using population density weighting to convert the provincial scale results into the prefecture city scale

(Table S1 in Supplementary), and the number of samples with an error of more than 0.3 accounted for 12.5% of all samples. Additionally, due to incomplete statistical data in prefecture-level cities, most of the indicators used for nitrogen loads were fewer than for provincial data. Therefore, theoretical calculation results should be lower than the actual values, which is also reflected in the correlation of the results obtained by the two methods. The regression coefficient was 1.03, which means that the result of directly using prefecture-level city statistics was slightly lower.

### Uncertainty analysis

Original data used to estimate other nitrogen, except for atmospheric deposition, were all obtained from the National Bureau of Statistics, China Statistical Yearbook, China Environmental Statistical Yearbook, China City Statistical Yearbook, China Rural Statistical Yearbook, province and municipality statistical yearbooks, and prefectural-level city statistical yearbooks. Publication year were from 2001 to 2020, and so available was from 2000 to 2019. The original data is displayed in the form of figures in Supplementary. Additionally, we implemented the Monte Carlo simulation to determine result uncertainty (Yan et al. 2011; Huang et al. 2014), including average, minimum, maximum, and 95% confidence.

Although differences in population and agricultural distribution may be small in cities with a relatively high proportion of agricultural production, there may be relatively large errors in cities dominated by services and industries. Provincial capital cities with concentrated populations have relatively large N load relative errors, such as Chengdu (0.34), Hefei ( $-0.32$ ), and Changsha (0.28). In cities with a relatively high proportion of services and industries, the errors are also

relatively large (Table S1). For example, the ratios of three industrial activities in Chuzhou in 2019 were 8.6 (agriculture), 49.1 (industry), and 42.3 (service), respectively, and relative errors reached  $-0.32$ . Huainan ( $-0.34$ ), Tongling (0.27), and Maanshan (0.28) had similar characteristics. Additionally, prefecture-level statistical data completeness is usually lower than for provincial and national data, which impacts estimation. However, the proportion of these cities with high errors was small, therefore, did not significantly impact our study.

## N changes under the influence of socioeconomic development

### The impact of industrial and agricultural development

Obstacle analysis is often used to quantify the negative impact of various elements on the system and has been widely used to analyze the impact of economic, resource, environmental, and other factors on the healthy development of society (Zhang et al. 2019; Gao et al. 2021). We used the obstacle degree model to determine the impact of human activities on nitrogen load growth in the YREB. The higher the factor's obstacle degree, the smaller the promotion of nitrogen growth, which is more conducive to reducing nitrogen loads, and vice versa. We divided anthropogenic nitrogen emissions into two sub-systems, agriculture and industry. To avoid repeating calculation, we did not consider life source, because nitrogen contribution of residents' excrement is lower, and all kinds of daily necessities such as food and other consumables come from industrial and agricultural production. The agricultural subsystem includes 7, and the industrial subsystem includes 3 indicators, as shown in Table 1. The total power of agricultural machinery and industrial pollution control investment are negative indicators. The higher the level of agricultural mechanization, the lower the nitrogen loss caused by agricultural production; the higher the investment in pollution control, the fewer pollutants will be emitted. The calculation details are shown in Supplementary.

### The impacts of urbanization

Presently, there is no uniform definition of urbanization (Kuznets 1955; Huang and Fang 2003; Chen et al. 2009), and research foci include many perspectives such as demography, geography, economics, and sociology. Therefore, we used the gray model, which has been widely used in data analysis to study urbanization impacts on changes in YREB's nitrogen load from population, land, and economic perspectives. Population urbanization indicators include total population, urban population ratio, and agricultural population ratio. Land urbanization indicators include construction land area, and economic urbanization indicators include the proportion of the output value of three industries (agriculture, industry, service) and regional GDP.

### Scenario simulation

We used the STIRPAT (Stochastic Impacts by Regression on Population, Affluence, and Technology) model to establish a nitrogen emission environmental pressure model of the YREB to quantify the impact of socioeconomic development on N loads. STIRPAT is a quantitative analysis model that assesses human activity impacts on the environment (Dietz and Rosa 1997; York et al. 2003) and, due to its flexibility and scalability of research variables, has been widely used in carbon footprint, water footprint, and some environmental pressure studies (Fan et al. 2006; Liddle and Lung 2010; Poumanyong and Kaneko 2010; Arshed et al. 2021). STIRPAT includes population, affluence, and technology. This study extends the STIRPAT model by including the level of industry and service as an indicator of technology, population index (divided into urban and rural population), and using GDP as an economic indicator (see Supplementary for modeling details). We preset 6 development scenarios and imported them into the N environmental pressure model to predict possible N loads of the YREB in 2030. In scenarios I to III, only the average annual increments in different periods

**Table 1** Industrial and agricultural factors of obstacle analysis

System	Index	Unit	Attribute
Agriculture	Proportion of rural population	%	+
	Total power of agricultural machinery	$10^4$ kilowatts	-
	Total sown area of crops	$10^3$ hectares	+
	Consumption of chemical fertilizers	$10^4$ tons	+
	Grain yield	$10^4$ tons	+
	Number of large livestock	$10^4$ heads	+
	Total output of aquatic products	$10^4$ tons	+
Industry	Industrial wastewater discharge	$10^4$ tons	+
	Industrial waste gas emissions	$10^9$ m <sup>3</sup>	+
	Industrial pollution control investment	$10^4$ CNY	-

“+” means positive effect, “-” means negative effect

are selected as the basis for estimating future variables, scenarios IV to VI are the three development states of high, medium and low speed respectively. The parameters of the various scenarios are shown in Table 2.

## Results and discussion

### Spatial–temporal variations of N loads

#### Interannual variation of N loads

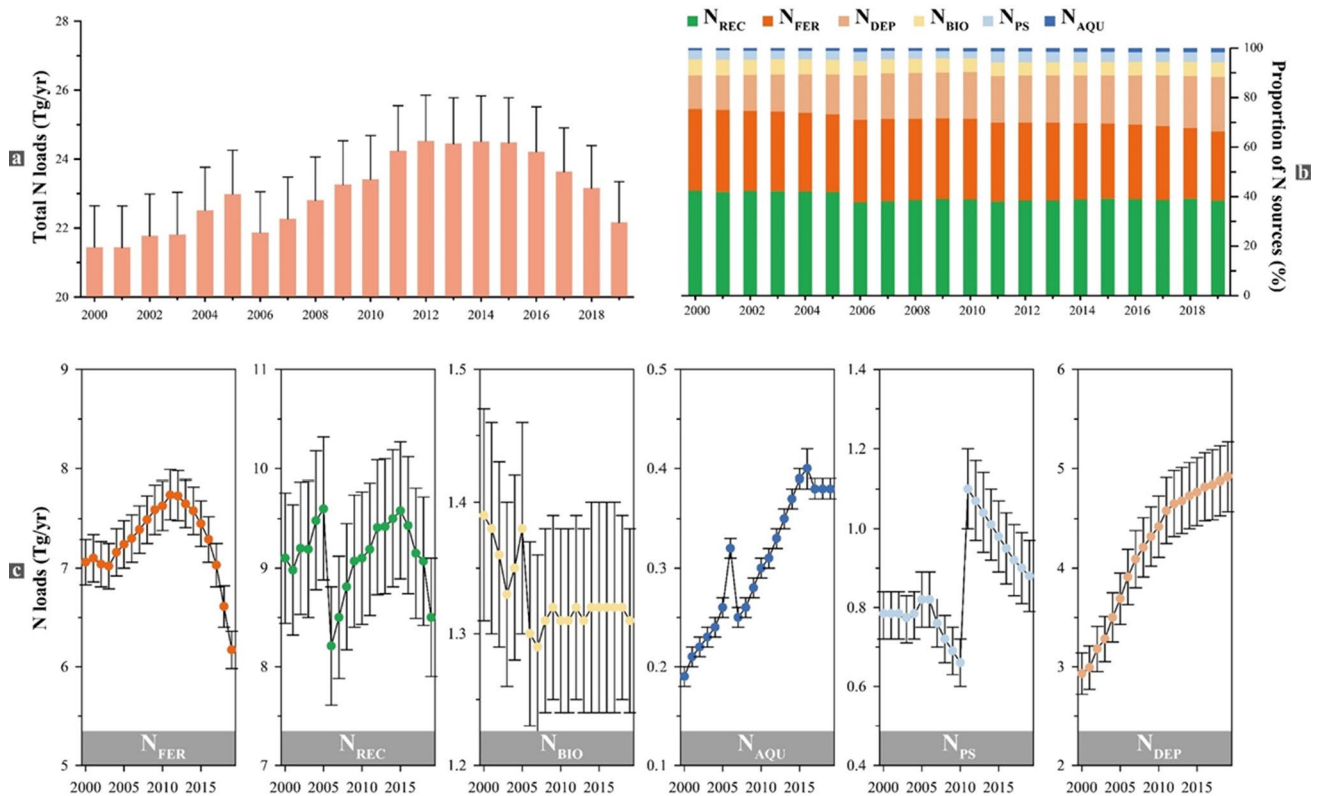
Since 2000 in the YREB, N loads experienced three periods of rapid growth (2000–2011), stability (2012–2015), and rapid decline (2016–2019) (Fig. 3a). The lowest N load was 21.4 Tg in 2001, and the peak appeared in 2012, reaching 24.5 Tg. The coefficient of variation in the rapid growth and

decline phases reached 0.037 and 0.032, respectively, while in the stable phase (0.001) was much lower. Fertilizer N and recycling N were the main contributors to nitrogen loads in the YREB (Fig. 3b), accounting for 31.5% and 39.6%, respectively (2000–2019 average, the same below). In 2015, the Ministry of Agriculture of the People’s Republic of China promoted the Action Plan for Zero Growth of Fertilizer Application, and by 2020, the contribution of fertilizer N to the YREB continued to decline (Fig. 3c). However, this action has not yet had a significant effect in reducing the application of compound fertilizers (Figure S2). In compound fertilizers, nitrogen content is relatively limited and has a small contribution to fertilizer N, while nitrogen fertilizer contributes more than 90% to fertilizer N. The contribution of nitrogen fertilizer to fertilizer N dropped from 95.6% in 2000 to 89.7% in 2019, and therefore, fertilizer N changes are mainly affected by the change in nitrogen fertilizer. The

**Table 2** Simulation indicators under different scenarios in 2030

	Population (100 million)		GDP per capita (10 <sup>4</sup> CNY)	Percentage of GDP (%)		Scenario setting method
	Urban	Rural		Sec-ondary industry	Tertiary industry	
2019	3.6	2.4	8	40	54	
Scenario I	4.9	1.4	29	35	60	(I) Using the average annual growth rate of each indicator from 2000 to 2019 as the predicted average annual growth rate in the future
Scenario II	5.1	1.3	22	30	65	(II) The method is the same as above, the average annual growth rate is the value from 2010 to 2019
Scenario III	5.0	1.5	20	25	70	(III) The method is the same as above, the average annual growth rate is the value from 2015 to 2019
Scenario IV	4.5	1.8	20	25	70	(IV) Socioeconomic development continues the trend of rapid growth. The society is at a high level of willingness to have children. By 2030, China’s population will reach 1.47 billion, and the average urbanization rate in various regions will reach 71% (Luo et al. 2016). The GDP growth rate and the growth of the proportion of the secondary and tertiary industries are the same as in 2015–2019
Scenario V	4.4	1.9	19	34	60	(V) Socioeconomic development gradually slowed down. In 2030, China’s population will reach 1.45 billion. At present, the urbanization rate of the YREB is increasing by about 1% every year, and the average annual growth rate of the urban population proportion is about 2%. In this scenario, the urban population proportion is set to 70% in 2030. In recent years, GDP growth rate has remained between 7 and 11%. In this scenario, it is assumed that the average annual growth rate of GDP is 9%. From 2010 to 2019, the secondary and tertiary industries in the YREB will fall by 9% and increase by 11% respectively. It is assumed that due to aging and the increase in labor costs, the changes in the proportions of secondary and tertiary industries will change by 6% in 2030
Scenario VI	4.2	2.0	16	37	57	(VI) Socioeconomic development slowed down significantly. Considering China’s current declining birth rate and serious aging, it is assumed the average population growth rate is 2‰ in 2030. Affected by demographic changes and the continuity of COVID-19, economic growth begins to slow down. It is assumed that the average annual growth rate of GDP is 7%, and the proportions of the secondary and tertiary industries change by 3%. The urbanization rate will be 68% in 2030 (Li et al. 2017)

To simplify calculations, GDP used in this study is nominal GDP, which has not been converted by the base year price



**Fig. 3** Interannual variations of anthropogenic nitrogen loads in the Yangtze River Economic Belt. **a** Interannual variations of total nitrogen loads. **b** Proportion of different nitrogen sources. **c** Interannual variations of different nitrogen sources

correlation coefficient between fertilizer N load and nitrogen fertilizer application confirmed this effect, as it reached 0.952 ( $p < 0.01$ ) (Table S2).

Recycling N has undergone a wave-like change process, with a peak of 9.6 Tg in 2005 (Fig. 3c). Livestock breeding manure contributed the most to recycling N, followed by residual crops that were returned to the field (Figure S3). In recent years, frequent poultry and livestock flu outbreaks and other epidemics have caused feeding instability, and a series of policies to promote food production adopted to ensure domestic food security (Yu et al. 2011), which may explain the changes in the two sources of recycling N. Additionally, rural resident nitrogen contributions have also gradually declined, as a result of rapid urbanization in China.

Atmospheric nitrogen deposition is the third largest contributor (~18.0%), showing an upward trend from 2.9 Tg in 2000 to 4.9 Tg in 2019. Ammonia nitrogen increased from 1.8 to 2.4 Tg, nitrate nitrogen increased from 1.1 to 2.5 Tg, and nitrate nitrogen exceeded ammonia nitrogen in 2015 (Figure S4). The nitrogen deposition of the YREB exhibited an S-shaped growth pattern, where the growth rate slows annually. This phenomenon may benefit from the implementation of the Air Pollution Prevention and Control Action

Plan in China (Ministry of Agriculture and Rural Affairs, PRC 2015). Nitrogen fertilizer application and animal husbandry are the main  $\text{NH}_3$  emission sources, and exhaust gases emitted from fossil energy consumption are the main sources of  $\text{NO}_x$  (Dentener et al. 2006; Ianniello et al. 2011). A decrease in the amount of nitrogen fertilizer in recent years has had an important influence on nitrogen deposition, which is confirmed by the gradual decrease in the proportion of ammonia nitrogen (Figure S4). Additionally, a national atmospheric nitrogen deposition study indicated that China's measures to control the application of nitrogen fertilizers have significantly reduced  $\text{NH}_y$  (Yu et al. 2019).

In the YREB, the amount of biological nitrogen fixation changed little, as the amount of nitrogen fixed decreased from 1.4 Tg in 2000 to 1.3 Tg in 2019. Pearson correlation analysis indicated the most crucial components affecting the change in biological nitrogen fixation were peanuts and beans (Table S3), with correlation coefficients of 0.844 and 0.626, respectively ( $p < 0.01$ ). Although the sown area of rice and other crops was much higher than peanuts and beans (Figure S5), its nitrogen fixation rate was relatively low, approximately  $3,000 \text{ kg km}^{-2} \text{ year}^{-1}$ , while the latter two types both exceeded  $8,000 \text{ kg km}^{-2} \text{ year}^{-1}$  (Chen et al. 2016; Li and Jin 2011; Zhang et al. 2017).

China's five largest freshwater lakes are all located in the YREB, which has natural aquaculture advantages and a developed river fish catering industry chain. Meanwhile, with improving consumption levels, people's demand for fish is very high, and aquaculture development has led to the continuous growth of nitrogen emissions (Fig. 3c). Since 2000, in the YREB, nitrogen emissions from aquaculture approximately doubled from 0.19 to 0.38 Tg. Although there was no significant change in shellfish farming, which has always maintained an annual output of ~300,000 tons, fish farming increased by about 72%, and shrimp and crabs, by more than 400% (Figure S6). Additionally, the government implemented a series of policy measures from 2016, especially in 2019, to ban fishing in the Yangtze River for 10 years (Zhang et al. 2020a), and the YREB's freshwater fishing production fell from 1,449,342 tons in 2015 to 1,165,582 tons in 2019. However, this policy may have stimulated aquaculture industry development, as freshwater farming output increased from 18,455,566 tons in 2015 to 18,861,965 tons in 2019, which significantly promoted aquatic nitrogen emissions.

Additionally, Yangtze River conservation efforts led to a downward trend in ammonia nitrogen discharge from industrial wastewater, with a decrease of 81%, but ammonia nitrogen from domestic sources is still high (Figure S7). Affected by the amount of living ammonia emissions, wastewater nitrogen after 2011 was much higher than the value before then. By reviewing a large number of statistics, we found that this pattern appears to be a national phenomenon, not an abnormality for an individual region. Unfortunately, relevant departments have not provided an explanation for it. We suspect that it may reflect improvements in detection and analysis methods for aquatic ammonia nitrogen or changes in statistical approach.

The nitrogen source composition in the entire YREB was similar to those in most provinces (Figure S8). Recycling and fertilizer nitrogen were the highest contributors, accounting for 40% and 32%, respectively. When the proportion of rural residents' manure nitrogen in  $N_{REC}$  is excluded, the contribution of the YREB's agricultural source can be obtained, i.e., the sum of fertilizer nitrogen, biological nitrogen fixation, aquaculture, livestock manure, and residual crop, which is about 78%. Therefore, agricultural production is the major nitrogen contributor to the YREB, while this proportion is relatively small in the developed industrial and commercial areas of Shanghai and Zhejiang province.

### Spatial distribution and N load hotspots

In the YREB, total nitrogen loads in each region did not show obvious regional characteristics affected by area (Fig. 4a, b). Among them, Sichuan Province has the

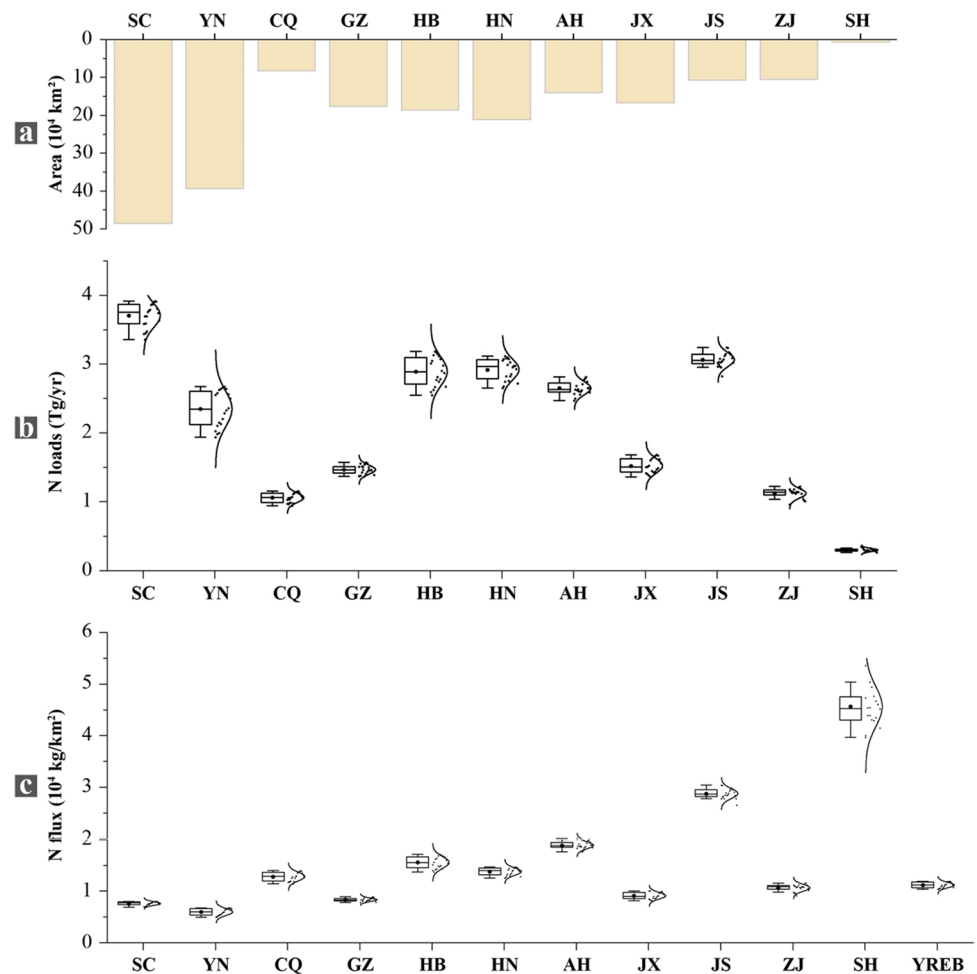
highest total nitrogen loads with the largest area due to developed agriculture. Jiangsu and Zhejiang areas in the eastern coastal region were similar, but total nitrogen load differences were nearly 2 Tg/year, and mainly affected by agricultural emission sources. Jiangsu has developed agriculture, while Zhejiang is mostly mountainous and hilly, and not suitable for large-scale agricultural production. Fertilizer N and recycling N in Jiangsu were approximately 1.3 Tg/year and 0.9 Tg/year, while Zhejiang's were 0.4 Tg/year and 0.3 Tg/year, respectively. Notably, flux per unit area shows that nitrogen flux north of the Yangtze River was higher than the south, and higher in the east than in the west. (Fig. 4c). An analysis of terrain and land use data found that western and southern regions of the YREB are mostly mountains and hills with a large area of forest land (Fig. 1, Figure S9), which may explain why nitrogen from agricultural sources is lower than in northern and eastern regions. This pattern is also consistent with the spatial characteristics of potential crop yield in China (Liu et al. 2015). Although Shanghai's total nitrogen load was the lowest in the entire economic belt, due to the smallest area and intense human activities including developed transportation and industry, flux was the highest, reaching  $4.6 \times 10^4$  kg/year, much higher than the average of  $1.1 \times 10^4$  kg/year in YREB.

Nitrogen hotspots in the YREB were mainly concentrated in the Sichuan Basin and the plains of the middle and lower reaches of the Yangtze River (Fig. 5a), where the YRDUA, the TCC, and the CCCG are located. The core cities of the three urban agglomerations are areas with extremely high nitrogen loads. For instance, nitrogen flux in Chengdu, Wuhan, and Shanghai, respectively, exceeded  $3 \times 10^4$  kg/km<sup>2</sup>. In contrast, there are plateaus and mountains in the western region, with sparse populations and underdeveloped industry and agriculture, where nitrogen flux in most areas is  $< 5 \times 10^3$  kg/km<sup>2</sup>, or only about half the YREB average. However, the spatial distribution of nitrogen growth is opposite to the nitrogen flux intensity, that is, an increase in the western region and a decrease in the central and eastern regions (Fig. 5b). The increase in the western region generally exceeds 30%, and in Kunming and surrounding areas even exceeds 80%.

The three urban agglomerations are not only nitrogen hotspots in the YREB but also major contributors, accounting for nearly two-thirds of the nitrogen loads of the YREB. The YRDUA, the TCC, and the CCCG contributed 19.5%, 23.6%, and 15.9%, respectively, to nitrogen loads of the economic belt (2000–2019 average). Each region's nitrogen contribution to the urban agglomeration is different (Fig. 5c). Three-quarters of the nitrogen loads in CCCG come from Sichuan Province, while in TCC, Hubei's nitrogen contribution is approximately 44%. Contributions from different YRDUA regions varied significantly, with Jiangsu accounting for nearly half (48%), and Shanghai accounting for only 7%.



**Fig. 4** Anthropogenic nitrogen loads of various regions in the Yangtze River Economic Belt. **a** Area of different regions. **b** N loads and **c** N flux in different regions. YREB: Yangtze River Economic Belt, SH: Shanghai, JS: Jiangsu, ZJ: Zhejiang, AH: Anhui, JX: Jiangxi, HB: Hubei, HN: Hunan, CQ: Chongqing, GZ: Guizhou, SC: Sichuan, YN: Yunnan



## Industry and agriculture impacts on N growth

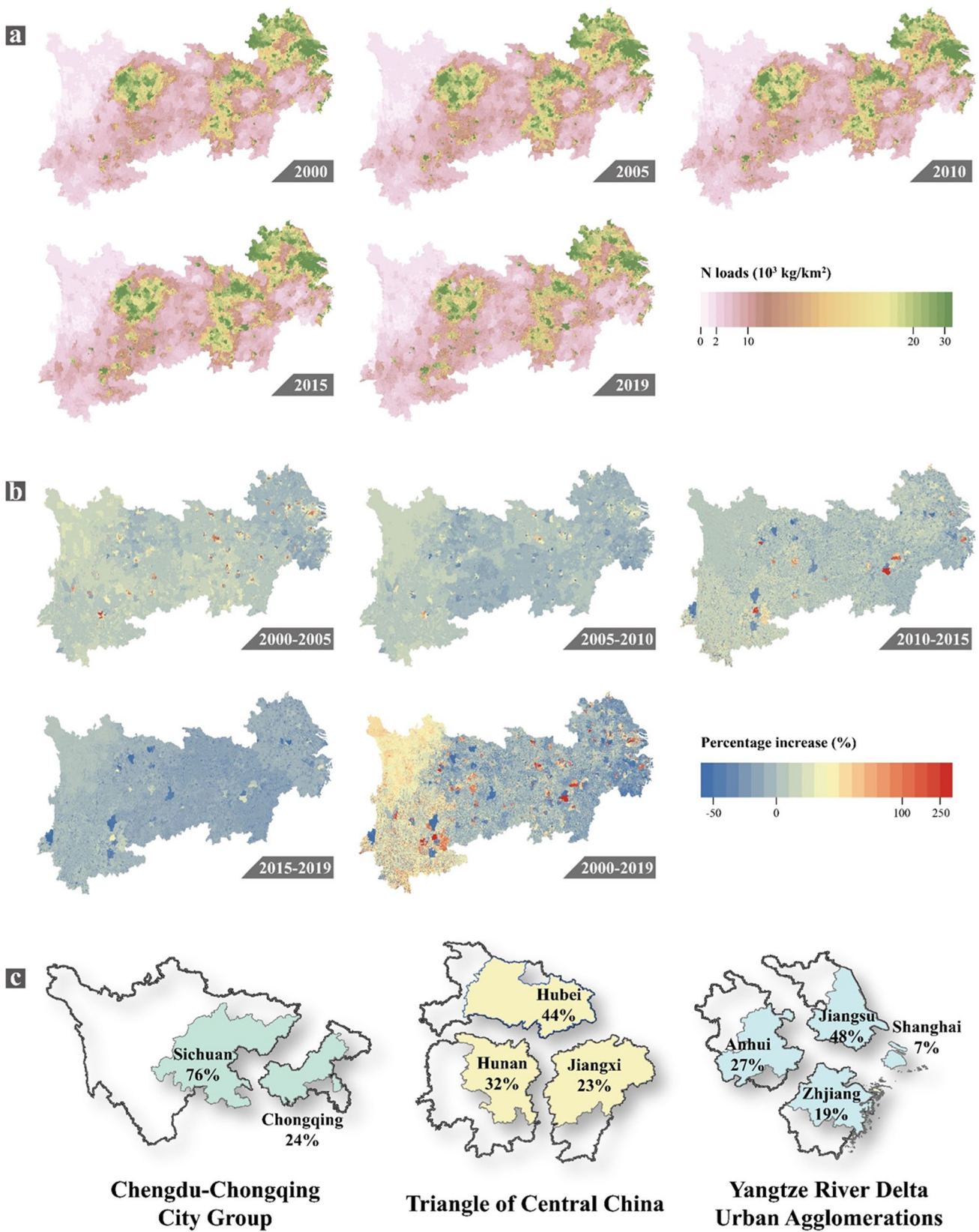
Industry and agriculture are the direct drivers of nitrogen emissions (Galloway et al. 2008). Since 2000, China's industrial and agricultural production structure has been continuously optimized and adjusted, which has inevitably impacted nitrogen loads. We divided sources of YREB's anthropogenic nitrogen emissions into agricultural and industrial subsystems, and used the obstacle degree model to analyze the influence of each element on the growth of nitrogen loads in their development. We took 5 years as the time period and calculated obstacle degree scores for factors that affect nitrogen loads in four time periods. The factors were sorted according to their obstacle degree score, the lower the score, the stronger the promotion of nitrogen growth (Fig. 6).

### Industrial subsystem

For the industrial subsystem, exhaust gas emission had the highest score in the YREB before 2010, followed by wastewater discharge, and pollution control investment was the lowest. This shows that there were insufficient

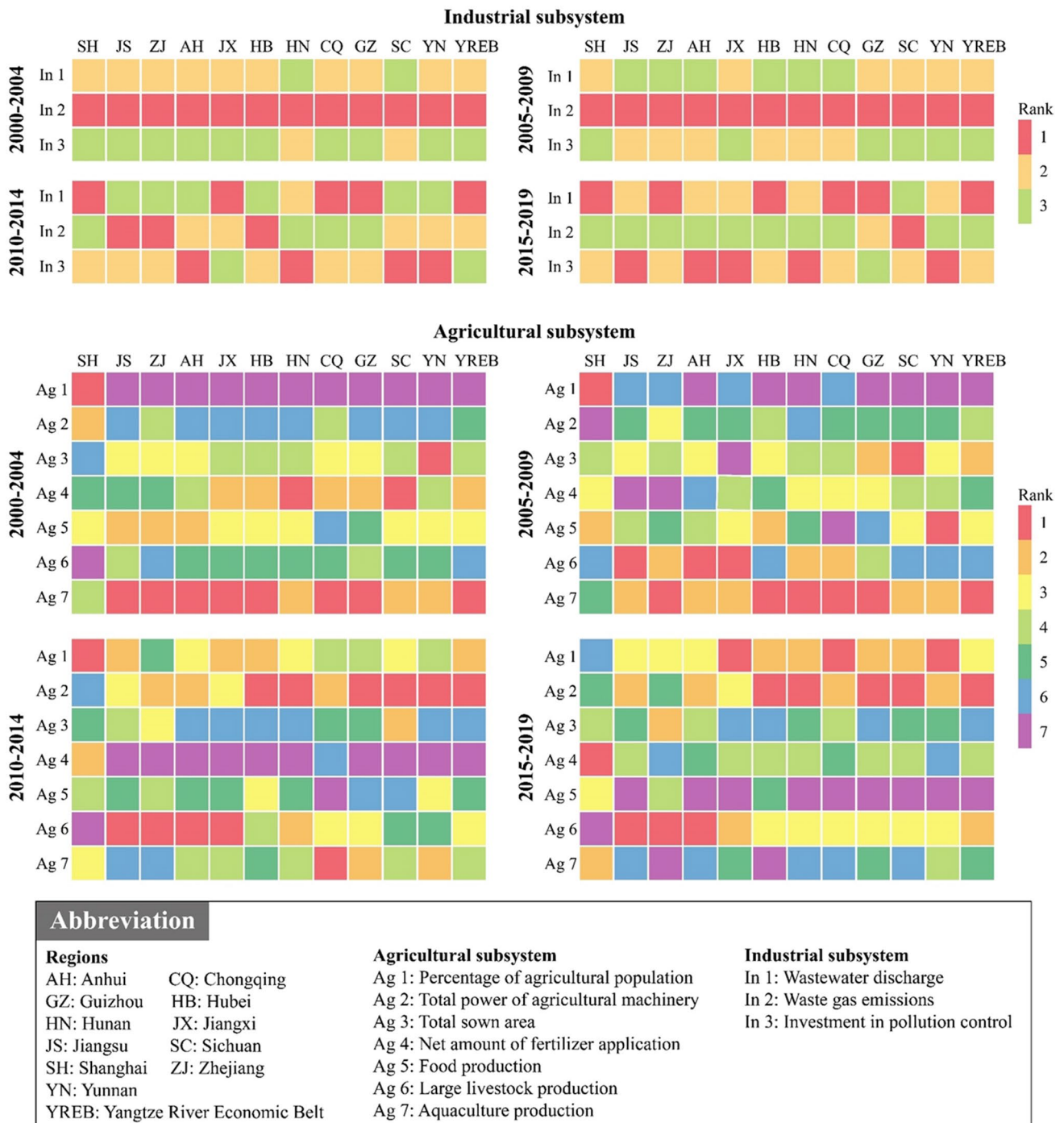
environmental measures and limited effects, and that wastewater discharge had a stronger effect on nitrogen growth than exhaust gas emissions. After 2010, wastewater discharge had the highest obstacle degree score, and exhaust gas emissions had gradually decreased. The obstacle degree of investment in industrial environmental pollution control increased after 2015, indicating that the initial pollution control effect was achieved. Sewage treatment achieved some nitrogen reduction, but industrial waste gas treatment was still not optimistic. An increase in the industrial environmental governance investment obstacle degree is closely related to the increasing intensity of investment in various regions (Figure S10).

There are differences in the order of obstacle degree scores for industrial factors in diverse provinces. Before 2010, the factors with the highest obstacle degree in all YREB regions were exhaust gas emissions. In the early 2000s, China joined the WTO, and the economy entered a period of rapid economic growth. However, the level of industrial development and technology at this time was still relatively backward, and China was experiencing a developing model of high pollution, energy consumption, and



**Fig. 5** Spatial and temporal heterogeneity of nitrogen loads in the Yangtze River Economic Belt (YREB). **a** Spatial distribution of nitrogen loads in the YREB. **b** The percentage increase of nitrogen loads

in the YREB. **c** The contribution of various regions to the nitrogen loads of the three urban agglomerations (average value from 2000 to 2019)



**Fig. 6** Analysis of obstacles affecting nitrogen load growth. The higher the rank of the factor in industrial and agricultural subsystems, the smaller the promotion of nitrogen increase, and the rank 1 is the highest

high emissions (Nan and Gao 2011). From 2000 to 2004, wastewater obstacle degree in Hunan and Sichuan was the lowest, but in the remaining regions, the lowest was industrial pollution control investment. Except for Shanghai and Jiangxi in 2005–2009, industrial pollution control

investment obstacle degree in the rest of the central and eastern regions increased, indicating that environmental management in most YREB areas began to improve, but the western region was still lagging behind. Meanwhile, wastewater discharge obstacle degree in most of the central and

eastern regions began to decrease, indicating that sewage has increased its role in promoting nitrogen growth. After 2010, the industrial environmental investment obstacle degree in most YREB areas increased significantly, especially in the upper and middle reaches of the Yangtze River. From 2015 to 2019, except for Guizhou and Sichuan, the rest had the lowest exhaust emission obstacle degree, indicating that industrial exhaust gas become the most important driving factor at this stage, which was also confirmed in YREB's annual increase in atmospheric N deposition (Figure S4). In short, current exhaust gas emissions have become the main driver for nitrogen, and the effect of environmental governance is gradually improving.

### Agricultural subsystem

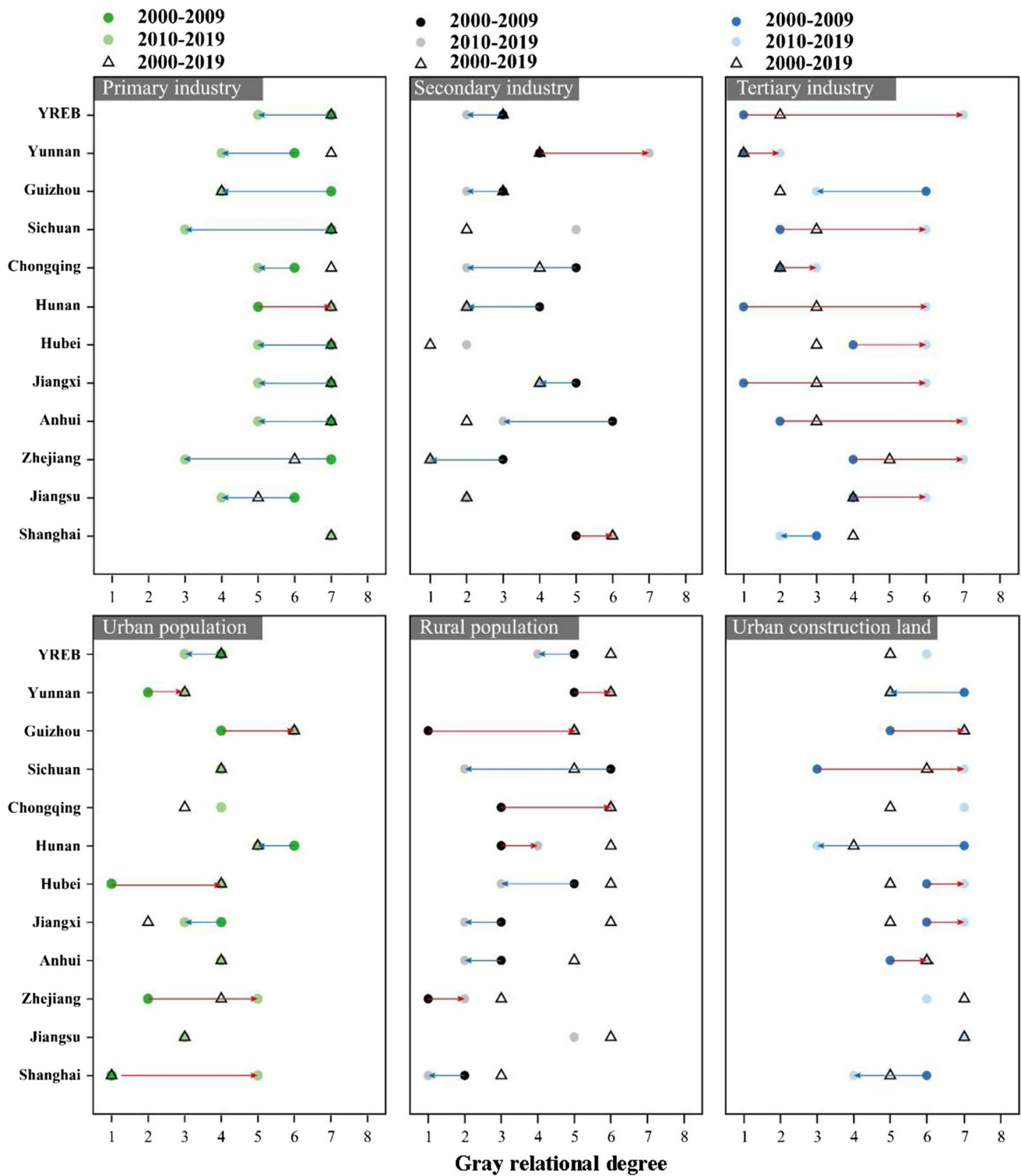
In the agricultural subsystem, the factor with the highest obstacle degree in the YREB before 2010 was aquaculture. During this period, the aquaculture industry was still underdeveloped, which had a slight impact on nitrogen emissions. Living conditions in rural areas and the level of livestock breeding were relatively backward, and pollutants such as manure had not been collected and treated effectively (Du et al. 2014) and were prone to secondary pollution. Meanwhile, the level of agricultural mechanization was relatively low, and extensive agricultural production caused a large amount of nitrogen loss during this period. After 2010, agricultural machinery became the most influential factor, followed by agricultural population and livestock breeding, indicating that the improvement of agricultural production level significantly reduced nitrogen emissions in the planting process, and rural living environment improvements and urbanization had a positive impact on reducing pollutant loss. Additionally, an increase in chemical fertilizer obstacle degree after 2015 may have been affected by the Action Plan for Zero Growth of Fertilizer Application by 2020 implemented by the Ministry of Agriculture in recent years, whereby the fertilizer application rate has been effectively controlled.

Due to differences in agricultural production structure in various regions, changes in agricultural factors have different characteristics. From 2000 to 2004, excluding Shanghai where the agricultural population was the highest obstacle degree, in the rest of the regions, it was the lowest. This reflects that high urbanization, such as in Shanghai, has a minor impact on nitrogen growth. Except for Shanghai, Zhejiang, and Chongqing, the agricultural machinery obstacle degree in other regions was relatively low, that is, the backward agricultural production level during this period played a role in promoting agricultural nitrogen loss. From 2005 to 2014, affected by the increase in urbanization and agricultural production level, the agricultural population and agricultural machinery obstacle degrees gradually increased,

which slowed down nitrogen load growth. Meanwhile, the ever-expanding food demand has gradually reduced the obstacle degrees of the sown area of crops and food production, which has promoted nitrogen load growth. From 2010 to 2014, except for the two municipalities directly under the central government (Shanghai and Chongqing), chemical fertilizer application had the lowest obstacle degree score in the other regions, indicating that it was the most important factor promoting nitrogen load increases, but its obstacle degree score increased significantly in recent years. From 2015 to 2019, the agricultural population obstacle degree factor in most regions ranked among the top three, and the urbanization rate had a significant impact on nitrogen reduction. In short, the improvements in agricultural mechanization and fertilizer control have effectively reduced nitrogen emissions in recent years.

### Nitrogen changes under rapid urbanization

For the entire economic belt, gray relational degree between total population and nitrogen loads from 2000 to 2019 was the strongest (0.95), followed by construction land (0.80), and GDP was the weakest (0.58) (Fig. 7). Generally, in economic urbanization, the higher the degree of urbanization, the lower the proportion of agricultural output. We show that service output value has the highest correlation with nitrogen loads (0.89), followed by industrial output value (0.88), and agriculture is the lowest (0.75). However, the complex structure and economic benefits of different industries cause the economic output value to not necessarily be positively correlated with nitrogen emissions. Although agriculture is the major source of nitrogen emissions, its economic benefits are low and prices fluctuate greatly with changes in market supply and demand (Yang et al. 2008) (Figure S11, S12). For example, a substantial increase in the number of pigs raised will cause a large release of nitrogen in excrement, but farmer incomes may not necessarily increase due to the falling prices. In contrast, industry and service industries are the main contributors to GDP. To pursue higher economic benefits, industries often consume more resources and emit more pollutants. The indirect flow of nitrogen in the service industry such as transportation exhaust emissions (He and Jiang 2021), will also have a significant impact on nitrogen emissions. Additionally, urban construction land index has a strong correlation. The expansion of urban construction land will inevitably lead to rural land reduction, and cultivated land occupation will reduce N loss from agricultural activities. Previous studies have also shown that cultivated land reduction and fertilizer application reduction have similar effects on nitrogen (Wang et al. 2017). In terms of population urbanization, the degree of the urban population proportion



**Fig. 7** Gray relational analysis of urbanization factors affecting changes in nitrogen loads. The red and blue arrows indicate the rise and fall of gray relational degree, respectively. The total population and GDP rankings are not shown in the figure

correlation (0.86) is higher than that of the rural population (0.78), indicating that complex urban human activities have a stronger impact on nitrogen changes.

From 2000 to 2019, the gray correlation results of each region were similar to the overall YREB situation. However, each indicator differed in different periods. From

2000 to 2010, YREB's primary industry ranked 7th, secondary industry 3rd, and tertiary industry first. After 2010, the ranking of the first two industries rose while tertiary industry fell 6 places, indicating an increasing influence of agriculture and industry. In terms of population, both the urban and rural population rose by one place, indicating that the correlation between population and changes in nitrogen emissions had increased. The correlation degree (0.80) of urban construction land after 2010 was significantly higher than in the previous stage (0.72), showing that it was an enhanced factor. There were also differences in indicator changes in various regions. For agriculture, Shanghai has a high level of urbanization with less agricultural production, therefore, its industrial relevance ranking did not change, while in the remaining regions it increased significantly. In tertiary industry, apart from Shanghai and Guizhou where correlation rankings rose, it declined in other regions. The secondary industry did not show any obvious changing characteristics. In the urban population gray correlation ranking, nearly half of the regions declined, and agricultural population index increased or decreased in other regions equally. The urban construction land index was unchanged in 3 regions, and in 5 regions declined. The impact of urban construction land on nitrogen loads was slightly higher than the overall economic development level. However, as the contradiction between the protection of cultivated land and urban construction in China has become increasingly prominent in recent years (Wang et al. 2018), construction land may be further restricted in the future, and its impact will gradually decrease.

In short, in economic urbanization, affected by unit economic benefits, secondary and tertiary industries have a greater impact on nitrogen load; in population urbanization, the impact of urban human activities is stronger; and urbanization of land may not have a remarkable impact on the nitrogen load in the future because the area of construction land will not change significantly.

### The impact of N loads of the YREB on the basin and estuary

Previous studies have focused on nitrogen loads in the YRB, and studying the impact of the YREB on the YRB will help clarify the rights and responsibilities of regional environmental management. The YRB is about 1.8 million square kilometers which is smaller than the YREB, flowing through 19 provincial administrative units. Superposition analysis shows the overlapping area of economic belt and river basin is about 1.35 million square kilometers, accounting for 75% and 66% of the two areas, respectively (Fig. 8a). The YREB and YRB non-overlapping areas include the Qinghai and Tibet plateaus, and small parts of some provinces.

We updated our previous nitrogen load results for the YRB (Cui et al. 2021) to make the data source and calculation method consistent with this study and comparable with our YREB results.

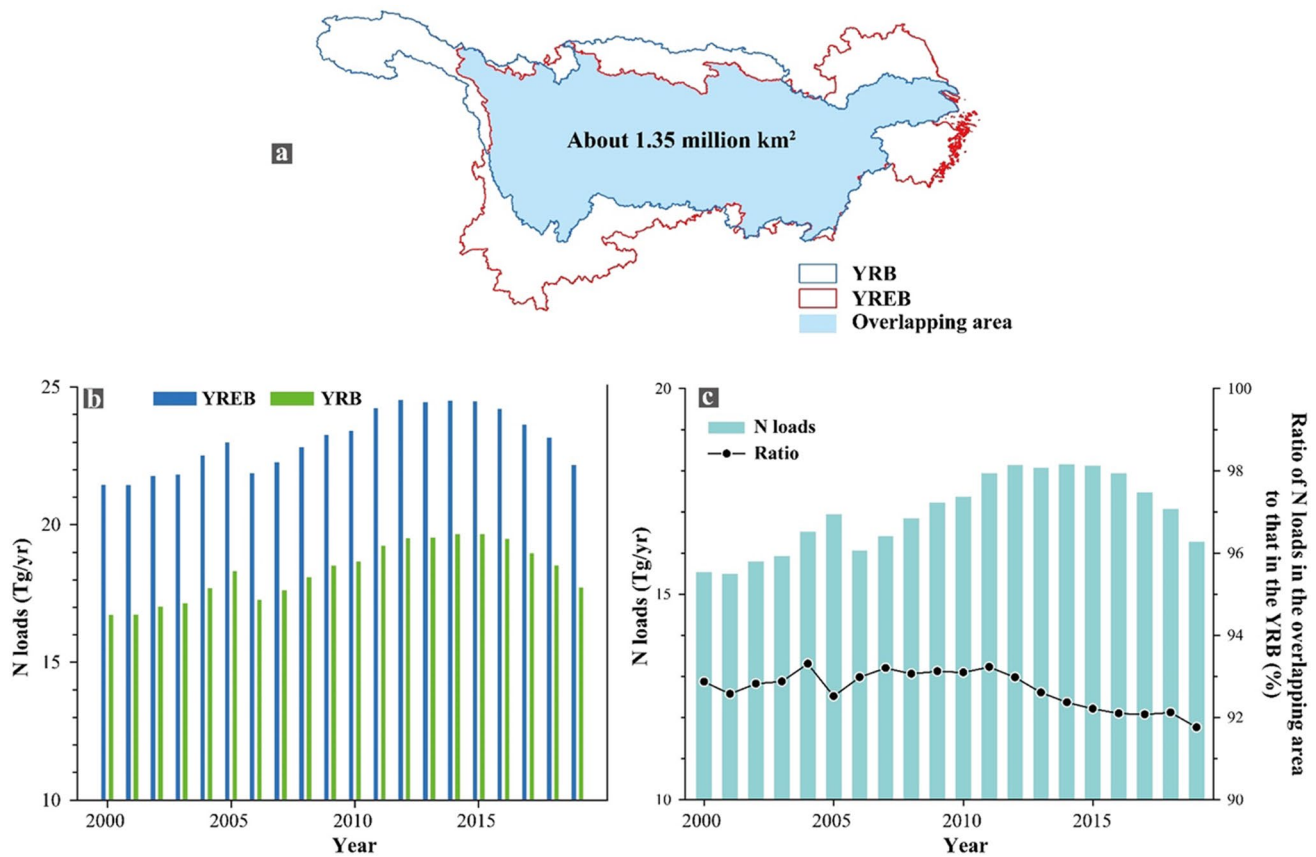
By comparing YREB and YRB nitrogen loads, input of the overlapping area accounts for 91.8–93.3% of N loads in the basin, and the CV is only 0.005, that is, the YREB area contributes more than 90% of the nitrogen loads to the YRB (Fig. 8b, c). Since the other areas not in YREB are mainly the sparsely populated Qinghai-Tibet Plateau, as well as a small part of some provinces, these areas have a low contribution to YRB nitrogen loads. Regression analysis also shows YREB's nitrogen emissions have a strong impact on the YRB ( $R^2=0.99$ ,  $p<0.05$ ) (Figure S13).

YRB inputs a large amount of nutrients into the estuary annually, among which the content of dissolved inorganic nitrogen (DIN) has shown an increasing trend, which is also an important reason for estuary red tides (Yan et al. 2010; Dai et al. 2011). Chen et al. (2016) found that annual anthropogenic nitrogen emissions accounted for ~50% of DIN output from the YRB to the estuary. Another study of the Jiaojiang River Basin in Zhejiang province from 1980 to 2010 also showed a similar result, with a contribution rate of ~57% (Huang et al. 2014). Using these estimated data and our results, it is estimated that the nitrogen contributed by the YREB each year accounts for ~50% of the DIN output from the river basin to the estuary. Although there are differences in the ratio of anthropogenic nitrogen migration and conversion to DIN in various regions, considering that the nitrogen loads from the YREB account for more than 90% of the anthropogenic nitrogen in the YRB, the difference in migration and conversion rates can be ignored, and therefore the estimated result is reasonable.

Additionally, although human activities contributed less than 10% to N loads in the basin outside the YREB, its development has spillover effects on nitrogen loads of these regions. By comparing spatial distribution of N flux in the YRB (Cui et al. 2021), we found that in the source area of Yangtze River (the part of the Qinghai-Tibet Plateau outside YREB), nitrogen growth was very significant with growth rates exceeding 50%, and even some regions exceeded 100%. It is related to the overall growth of China's economy including the YREB, and the increase in income has led to more people traveling to the plateaus, especially people in Sichuan and Yunnan. Moreover, transportation advancements in Sichuan and Yunnan contributed to the development of Tibet, resulting in a rapid increase in nitrogen load.

### Future challenges

Compared with anthropogenic nitrogen fluxes around the world (Figure S14), such as major watersheds in the USA, India, and Europe, China's nitrogen flux is much higher (Han



**Fig. 8** Nitrogen loads in the overlapping area between the Yangtze River Economic Belt and the Yangtze River Basin. **a** The overlapping area between the YREB and the YRB. **b** Interannual variations

of nitrogen loads in the YREB and the YRB. **c** Changes in nitrogen loads in overlapping areas and their proportion in the YRB

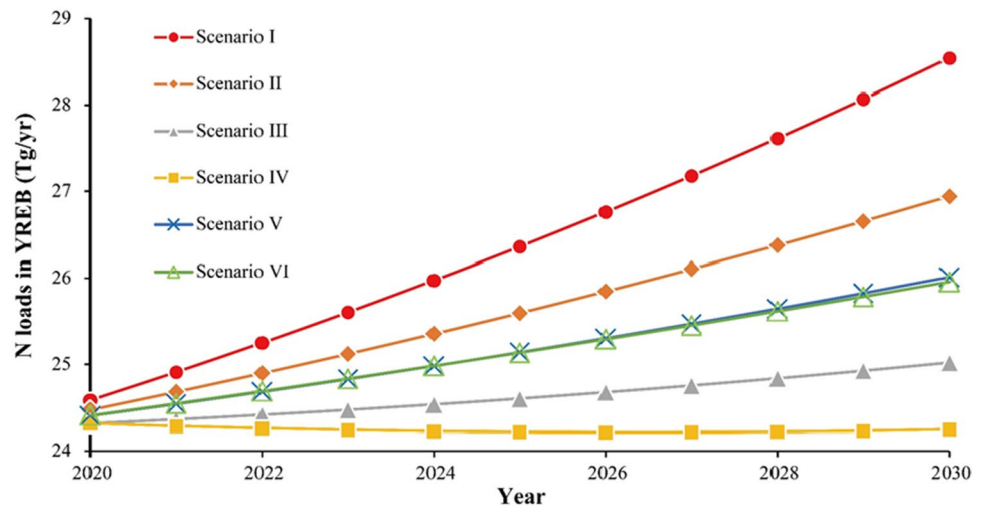
et al. 2014). China's Yangtze River Basin (Chen et al. 2016), Pearl River Basin (Cui et al. 2020), and Yellow River Basin (Wu et al. 2022) are populated and developed agricultural areas, and their nitrogen fluxes are several times higher than those in the United States and European basins (Schaefer and Alber 2007; Goyette et al. 2016; Hong et al. 2012). As a strong zone of human activities, the YREB nitrogen flux is much higher than global average (Han et al. 2020), and it is facing huge environmental pressure. In the context of a growing population, how to achieve nitrogen reduction in highly urbanized areas, especially the YREB with a dense population, and what challenges it faces in the future should be discussed in depth.

Nitrogen load changes in the YREB based on STIRPAT simulation are shown in Fig. 9. China has experienced tremendous development since 2000, especially changes in GDP and secondary industry. In scenario I, nitrogen loads reach 28.5 Tg by 2030, which is the highest among the scenarios. N loads in Scenario II and III are 27 Tg and 25 Tg, respectively. These two scenarios are based on the average growth rate of the last decade and five years, respectively. Therefore, secondary and tertiary industry changes

are higher than the 20-year average, and GDP growth rate dropped significantly compared to the initial period of joining the WTO. Additionally, China canceled the one-child policy in 2015, and population growth rates of Scenario II and III are both slightly higher than the 20-year overall level.

Scenarios IV, V, and VI indicate that N loads will reach 24.25 Tg, 26.01 Tg, and 25.96 Tg in 2030, respectively. The N load of scenario IV is the lowest among all scenarios and is the only one that shows a downward trend. In these 3 scenarios, although population growth and urbanization rate of scenario IV are relatively high, the proportion of secondary industry is the lowest, and the proportion of tertiary industry is the highest. N loads of scenarios V and VI are close to scenario III. Although there was no significant population difference between these two scenarios, the higher industrial value-added played a role in promoting nitrogen emissions. In scenarios III and IV, although GDP per capita and secondary and tertiary industrial value-added were similar, population growth and urbanization rates were significantly different. The relatively low population of Scenario IV reduces nitrogen loads by approximately 0.8 Tg. Therefore, improving science and technology levels, increasing the proportion

**Fig. 9** Trends of nitrogen change in the YREB by 2030 based on the STIRPAT model



of the tertiary industry, optimizing secondary industry structure, and promoting cleaner production, are the most effective ways to reduce future nitrogen loads.

### Countermeasures

Numerous studies show that China's future population growth rate will continue to slow down, and decline after peaking around 2030 (Luo et al. 2016; Sun et al. 2017; Department of Economic and Social Affairs, Population Division, United Nations 2019). However, “demographic dividend” is gradually disappearing, which will impose higher requirements on food demand and agricultural production quality. How to reduce N loss during agricultural production while ensuring food security is critical to reducing N loads in the YREB in the future.

Although, since the 1990s, China has implemented a series of agricultural policies, regulations, and measures to reduce nitrogen emissions, China's current fertilizer utilization efficiency and agricultural production mechanization level are relatively low. Meanwhile, affected by strong domestic demand and grain trade fluctuations in the international market, China is still facing a huge threat to food security (Anderson and Strutt 2014). If ensuring food supply and improving fertilizer utilization efficiency are not substantially achieved, N loss from fertilizers may still remain relatively high for a period of time in the future. The resource utilization rate of organic fertilizers in China is generally low, since chemical fertilizers are deemed important, while organic fertilizers are despised. Agricultural wastes rich in nutrients, such as crop stalks and poultry manure should be used more effectively. The YREB has traditional agricultural areas such as the Chengdu Plain and Middle and Lower Yangtze Plains, which are rich in organic fertilizer resources. Optimizing the fertilization structure and increasing the ratio of livestock manure and crop straw nutrients to

the field would dramatically improve the fertilization mode and reduce N loss of chemical fertilizers. Meanwhile, the Yangtze River natural water and heat resources should be fully utilized to promote green manure planting. For nitrogen hotspots, local governments should establish an active coordination mechanism and form a regional mutual assistance model, to encourage and guide farmers to establish low-polluting green planting models.

NO<sub>x</sub> released from fossil fuel combustion in the industrial production process is the main source of atmospheric nitrogen. Realizing energy structure transformation by reducing the proportion of traditional coal-fired power plants, and making full use of the advantages and potential of hydroelectric power generation in the Yangtze River would greatly lower atmospheric nitrogen deposition. Scenario predictions also show that even in the context of rapid population growth, a high level of technological innovation and a low proportion of highly polluting industries will remarkably reduce nitrogen loads. Meanwhile, during urbanization, it is necessary to rationally arrange sewage and garbage treatment plants in densely populated areas, and promote garbage classification and resource recycling to reduce nitrogen loss. Additionally, the Yangtze River Protection Law, which was formally implemented at the beginning of 2021, needs to be expeditiously applied.

### Conclusions

Anthropogenic N loads of the YREB experienced growth, stable, and decline phases. In the YREB in 2000, nitrogen flux gradually increased from western to eastern regions. But in 2019, nitrogen changes showed an opposite flux pattern, with a decline in the central and eastern regions and a significant increase in the western regions. N load hotspots



were mainly concentrated in city cluster areas, which contributed nearly 60% of N to the YREB. Approximately three-quarters of YRB land is located in the YREB, and ~90% of YRB anthropogenic N comes from this area. About half of the DIN exported from the basin to the estuary is derived from the economic belt.

During industrial development, wastewater was the main driver of nitrogen growth before 2010, after which waste gas superseded it, and the impact of industrial environmental management on nitrogen reduction gradually increased. In agricultural development, mechanization that promotes nitrogen reduction has gradually increased, and the positive effect of fertilizers on nitrogen growth has been weakened through national policies in recent years. Among the various urbanization indicators, urban population and secondary/tertiary industry development have had the most significant impact on N load changes. Moreover, with continued restriction of urban construction land, the impact of land urbanization on N loads will decrease in the future.

Predictions show that by 2030, N loads in the YREB will be significantly higher than in 2019, and the YREB will still face high N load pressure in the future. However, even in the context of rapid population growth, as long as the level of technological innovation increases and the proportion of the highly polluting secondary industry is reduced, N loads will be controlled.

**Supplementary Information** The online version contains supplementary material available at <https://doi.org/10.1007/s11356-022-21943-y>.

**Author contribution** Meng Cui: Conceptualization, Methodology, Writing—Original Draft, Visualization. Qingjun Guo: Supervision, Funding acquisition. Yujun Wei: Writing—Review and Editing, Software, Funding acquisition. Xuening Yu: Validation, Project administration. Jian Hu: Formal analysis. Liyan Tian: Investigation. Jing Kong: Data Curation.

**Funding** This research was supported by the National Natural Science Foundation of China (grant nos. 41890824, 41890822) and a project funded by the China Postdoctoral Science Foundation (grant no. 2021M693146).

**Data availability** The availability of data and materials are described in detail in “[Uncertainty analysis](#).” Data sources and uncertainty analysis. The data, calculation methods, and other additional content in this article can be found in the Supplementary materials.

## Declarations

**Ethics approval and consent to participate** This research does not involve human participants and/or animals.

**Consent for publication** All authors approved this version to be published and agreed to be accountable for all aspects of the work in ensuring that questions related to the accuracy or integrity of any part of the work are appropriately investigated and resolved.

**Competing interests** The authors declare no competing interests.

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