Biogeochemistry (2012) 108:381–394
DOI 10.1007/s10533-011-9606-y

A nitrogen budget of mainland China with spatial and temporal variation

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Received: 1 September 2010 / Accepted: 25 April 2011 / Published online: 12 May 2011

Abstract The present study evaluated nitrogen (N) input and output in mainland China using updated data of temporally and spatially-based land use maps and statistical data at national and provincial scales. The total N inputs increased from 3,081 kg km^{-2} in 1985 to 5,426 kg km^{-2} in 2007. Chemical fertilizer dominated the N input and showed an increasing trend. Biological N fixation was the second important N input till 1990 and atmospheric deposition became the second most important source after that, accounting for 24.0% in 2007. There was no net N input through food/feed import in 1985, but it accounted for 3.5% of the total N input in 2007. According to a mass balance model, we assumed total N input equal to output. The results showed that more than half of the total N was denitrified or stored in the system. Ammonia volatilization accounted for 18.9–22.9% of the total N input, and N export to water bodies accounted for 17.9–20.7%. About 5.1–7.7% of the N input was emitted to the atmosphere through biomass burning. When calculated per unit area, total N input, N export to water bodies, denitrification and storage could be very well explained by human population density. Nitrogen input and major outputs were also positively related to per capita gross domestic product and the percentage of total land area used as cropland. The N budget is compared to that of some other countries and the environmental impacts of the N cycle is discussed.

Keywords Nitrogen deposition · Chemical N fertilizer · Ammonia volatilization · Denitrification

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Introduction

Nitrogen (N) is a fundamental component of living organisms, and it has been strongly influenced by human activity. From the pre-industrial era to 1990, reactive N input to the global terrestrial system increased twofold (Galloway and Cowling 2002). The massive N input has enabled humankind to greatly increase food production. However, excessive N can induce a series of economic and environmental problems such as the greenhouse effect, destruction of the ozone layer, acid rain, nitrate pollution in groundwater, eutrophication of lakes and offshore water, and biodiversity reduction locally, nationally and globally (Vitousek et al. 1997; Drecht et al. 2003). Several N budget calculations at global or national scales have been conducted (e.g. Galloway et al. 1996, 2004; Bashkin et al. 2002; Filoso et al.

2006; Parfitt et al. 2006). The findings of these national and international research programs investigating the manifold consequences of human alteration of the N cycle have led to a much improved understanding of the scope of the anthropogenic N problem and possible strategies for managing it (Gruber and Galloway 2008).

China is the third largest country in the world and has diverse climatic conditions ranging from tropical in the south to cold temperate in the north, and from humid in the east to arid in the northwest. Rapid economic development and expansion of the human population in the past three decades has resulted in a large increase in chemical fertilizer and fossil fuel consumption, and thus greatly altered the N cycle. However, the changes in the input and fate of reactive N are not well understood. Xing and Zhu (2002) estimated N budgets in terrestrial ecosystems for a single year for all of China and its three major watersheds; however, the spatial and temporal changes were not documented. Due to the lack of measured parameters, Xing and Zhu (2002) used two values to estimate N deposition for the vast territory of the whole country, and estimated the amount of N export to water bodies by using the IPCC default values. While several other studies noted the spatial and temporal changes in the N budget of China (e.g. Fang et al. 2007; Wang et al. 2007; Qiu et al. 2008; Sun et al. 2008), all have focused on agroecosystems, without considering the vast area of other land uses.

China is a developing country with a large rural population. Biofuel (crop residue and fuel wood) used to be the major energy source in rural areas. Although the rapid economic development has increased rural access to commercial energy, and the use of biofuel is decreasing, crop residue is increasingly being burned in the field. It was estimated that in 2000, 24.3 and 19.4% of the crop residue were burned as fuel and in fields, respectively (Yan et al. 2006). The N contained in crop residues is emitted to the atmosphere as nitrogen gas (N_2) and reactive gases (Andreae and Merlet 2001), being a significant pathway of N output of the land surface. However, this part of N output was ignored in earlier budget calculations in China.

The objectives of this study were therefore to (1) compile a N budget for mainland China with spatial and temporal distribution by using more up-to-date activity data and flux parameters, and (2) analyze the changing patterns in N inputs and output and their impacts on the environment.

Data and methods

N budget model

N budgets were established based on the mass balance model of Howarth et al. (1996). N inputs included biological fixation, chemical fertilizer, atmospheric deposition, and import of food and feed. N output included ammonia (NH_3) volatilization, N export to water bodies, food and feed exports, and biomass burning emissions. The difference between N inputs and outputs was assumed to be denitrification and storage, which both are difficult to quantify directly. The N budget was calculated for the years 1985, 1990, 1995, 2000, 2005 and 2007 for each province, autonomous region or municipality of mainland China. The inputs and outputs were then spatially allocated using 1 km resolution land use maps (obtained from Data Sharing Infrastructure of Earth System Science, http://www.geodata.cn).

Basic data

Total land area of each province, municipality or autonomous region was acquired from a shape file of a mainland China map obtained from National Basic Geographical Information System (2004) and preprocessed by GIS techniques. The total area of cropland and cultivated area of major crops were obtained from the China Agricultural Yearbook (Editorial Board of China Agriculture Yearbook 1986, 1991, 1996, 2001, 2006, 2008). Forests and grassland areas in 1985, 1995 and 2000 were quantified using a 1-km resolution land use map (from Data Sharing Infrastructure of Earth System Science, http://www.geodata.cn) and the China Statistical Yearbook for 2005 and 2007. Precipitation data were obtained from http://www.cru.uea.ac.uk/cru/data/hrg/, with 30-second resolution. Data of crop yield, populations of people and livestock, and chemical fertilizer consumption were gathered from the China Statistical Yearbook (Edited by National Bureau of Statistics of China 1986, 1991, 1996, 2001, 2006, 2008) and China Agriculture Yearbook (Editorial Board of China Agriculture Yearbook 1986, 1991, 1996, 2001, 2006, 2008).

N inputs

Biological fixation

Biological N_2 fixation was estimated on an area basis. Fixation rates were assumed to be 8,000 kg N km^{-2} year^{-1} for soybean and peanut (Smil 1999), 15,000 kg km^{-2} year^{-1} for green manure (Yan et al. 2003a), and 500 kg km^{-2} year^{-1} for grassland (Bouwman et al. 2005). Flooded rice fields provide a unique set of conditions for biological N_2 fixation by autotrophic N_2-fixing cyanobacteria and heterotrophic N_2-fixing bacteria (Roger and Ladha 1992), and we used a fixation rate of 3,000 kg N km^{-2} year^{-1} (Zhu and Wen 1992) as the default. Upland crops were assumed to fix N at 1,500 kg km^{-2} year^{-1} (Burns and Hardy 1975). Fixation rates of forests vary with forest type and region due to natural and climate differences. For symbiotic N fixation in forestland, we adopted the region-specific values from 1,510 to 2,500 kg N km^{-2} year^{-1} (Table 1) of Xi et al. (2007). For non-symbiotic N fixation in forestland, we assumed a constant rate of 40 kg N km^{-2} year^{-1} (Boyer et al. 2002).

Chemical fertilizer

Amounts of chemical N fertilizer consumption at national and provincial scales were obtained from the China Statistical Yearbook (National Bureau of Statistics of China 1986, 1991, 1996, 2001, 2006, 2008). An average N content of 30% of compound fertilizer is commonly assumed in China and hence we used this value to calculate the N input as compound fertilizer.

Atmospheric wet and dry deposition

China is one of the global heavy N-deposition areas, especially in the southeast (Galloway et al. 2008), resulting from the extensive use of fossil fuels in industry and transportation, use of chemical fertilizer in agriculture and expansion in intensive animal husbandry in the last three decades. However, there is no systematic nationwide monitoring network to derive geographical and temporal distribution of deposition rate. Nevertheless, there have been many miscellaneous measurements of wet N deposition in different locations of China since the early 1980s. Previous studies suggested a relationship between N deposition and emission (Asman 1998; Goulding et al. 1998; Su et al. 2005). In the current study, we used this relationship to estimate wet N deposition from gaseous N emission data. We gathered about 600 data points of wet deposition of ammonium (NH_4^+)–N and nitrate (NO_3^-)–N over China spanning 1981–2006. For measurement data before 2003, we first averaged the NH_4^+–N and NO_3^-–N deposition rates on a yearly basis for each 0.5 $\times$ 0.5 degree-grid, if data are available, and identified the emission rates of NH_3 and NO_x for the corresponding grid of the corresponding year from the Regional Emission Inventory in Asia (Ohara et al. 2007), then we calculated the deposition/emission ratios for reduced N (NH_4^+/NH_3) and oxidized N (NO_3^-/NO_x). The deposition/emission ratios were then averaged over the main regions of China and over years (Table 1). To validate the relationship, we compared the deposition rates measured after 2003 to those predicted by the relationship. The results showed that the

Table 1 Region-specific N fluxes in forest ecosystem and deposition/emission ratios of reduced and oxidized N in mainland China

Region	N fluxes in forest ecosystem (kg N km^{-2} year^{-1})[a]			Deposition/emission ratio of reduced N (NH_4–N//NH_3–N)	Deposition/emission ratio of reduced N (NO_3–N/NO_x–N)
	Symbiotic N fixation	N runoff and leaching	Ammonia volatilization		
Northeast China	2,200	279	100	0.69	0.23
Northwest China	1,510	279	100	1.58	0.26
North China	1,730	5	100	0.42	0.15
Central China	2,500	149	150	0.46	0.17
Southeast Coastal	2,500	149	150	0.65	0.17
Southwest China	2,500	149	150	0.70	0.45
Area-weighted average	2,231	151	132	0.63	0.29

[a] Data source: Xi et al. (2007)

predicted deposition rates agreed reasonably well with observations (Fig. 1). Therefore parameters in Table 1 were used to estimate wet deposition N from the emission inventory of Ohara et al. (2007).

Due to the difficulty in measurement, only a few data are available for dry deposition. Xie (2006) reported a dry deposition rate of 716 kg N km^{-2} year^{-1}, comparing to a wet deposition rate of 2,570 kg N km^{-2} year^{-1} at a suburban site in eastern China. Similarly, Yang et al. (2010) measured a dry deposition rate of 760 kg N km^{-2} year^{-1}, comparing to a wet deposition rate of 3,200 kg N km^{-2} year^{-1}at a field site in eastern China. Lü and Tian (2007) estimated the total inorganic N deposition for China, with dry deposition being 2.9 Tg N year^{-1} and wet deposition being 9.45 Tg N year^{-1}. All these data indicate a dry:wet deposition ratio of about 2:8. However, in these studies, not all N species in dry deposition were accounted and thus they likely underestimated dry deposition. Considering that dry deposition can be nearly equal to wet deposition in forest area (Lovett and Lindberg 1993), we assumed an uniform dry:wet deposition ratio of 3:7 and then estimated dry deposition rate from that of wet deposition.

Net food and feed import

In China, food/feed export was greater than import before 1985, since then there has been no net food/feed export on a national scale. In terms of N mass in food/feed, the import in 2007 was ten times higher than export. In the present study, we calculated the balance between food/feed import and export at national level using statistical data from the China Statistical Yearbook (National Bureau of Statistics of China 1986, 1991, 1996, 2001, 2006, 2008). At the provincial scale, there were large imports and/or exports of food/feed due to the disproportion between food production and human and animal populations. We first calculated the value of the national average N-consumption by per human based on total national food production and national net food and feed export (or import), then multiplied that value by the total human population of a province to estimate the total N demand of a province. The difference between the total N demand and N contained in food products (crops and livestock) in a province was considered to be the net N import (export)—negative indicated a net food/feed import, otherwise an export.

N in crop and animal products was estimated from their N contents and total mass of products. Usually, protein rather than N contents, are reported for animal products, and we assumed N content to be 16% of protein content (Lu 1999). Data of crop yield and animal number were obtained from the China Statistical Yearbook (National Bureau of Statistics of China 1986, 1991, 1996, 2001, 2006, 2008), and parameters along with their sources used to calculate N mass in crop and animal products are shown in Tables 2 and 3.

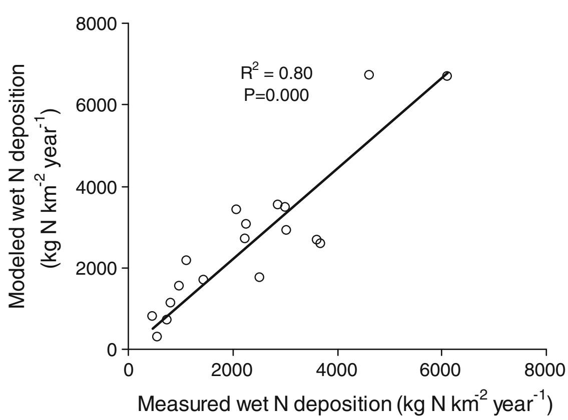


Fig. 1 Modeled versus measured wet deposition of N in mainland China

N outputs

NH_3 volatilization

NH_3 is volatilized from croplands, livestock husbandry and forestlands. We used the method of Yan et al. (2003b) to estimate NH_3 emission from croplands, which included NH_3–N emission from chemical fertilizer, animal excreta used as fertilizer, biological N_2 fixation, N from crop residues returned to cropland, and background emissions. To estimate emissions from husbandry and industry, we used the method of Wang et al. (2009). In their study, NH_3–N emission factors under different conditions of livestock housing and excreta storage were 10.3, 1.64, 0.28, 2.19, 5 and 7 kg N year^{-1} capita^{-1} for cattle, pigs, chickens, sheep, horses, and donkeys/mules, respectively. The NH_3 emissions during grazing were 2.66, 1.53 and 3.5 kg N year^{-1} capita^{-1} for cattle, sheep and horses, respectively. Background emission from grassland was 100–1,100 kg N km^{-2} year^{-1}

Table 2 N content in different grains used to calculated net food/feed export (kg N kg^{-1})

Rice	Wheat	Maize	Rape	Soybean	Cotton	Peanut	Potatoes	Sesame	Tobacco	Sugarcane	Beet
0.019[a]	0.023[a]	0.014[a]	0.039[b]	0.051[b]	0.039[b]	0.042[b]	0.0026[a]	0.303[b]	0.263[b]	0.022[b]	0.024[c]

N content in the grain for rice, maize, oil rape and wheat were measured, those for soybean and cotton were obtained from [a] Xiang et al. (2006), [b] China Fertilizer Information Web http://www.natesc.gov.cn/sfb/ and [c] Liu et al. (2006)

Table 3 N produced by main livestock and poultry used to estimate N budgets

Animal and poultry	Urine[a] (kg N $capita^{-1}$ $year^{-1}$)	Manure[a] (kg N $capita^{-1}$ $year^{-1}$)	Body weight[b] (kg)	Protein content of animal body(%)[b]
Pig	7.58	7.58	100	13
Cattle	19.45	29.34	477	17
Sheep	0.94	4.81	45.4	15
Chicken		0.37	2.04	17

[a] From Wu (2005); [b] from Liu et al. (2006)

(Zhou et al. 2008), and we chose 330 kg N km^{-2} $year^{-1}$ in this study. NH_3 volatilization from forestlands is shown in Table 1.

Reactive N export to water bodies

Leaching and run-off loss of N from soils depends on such factors as land use type, form of applied N, soil properties and rainfall. We compiled a dataset of measurements of N leaching and run-off loss from croplands. However, there was no statistically significant relationship between N loss and these influencing factors. For run-off loss of chemical N applied to croplands we therefore used an average loss rate of 5% derived by Zhu and Chen (2002). For leaching loss from chemical fertilizer applied to rice and upland crops, we averaged loss rates of 3.4 and 11.1%, respectively, from our compiled dataset. N loss through leaching and run-off from grasslands has range 63–30,000 with an average of 330 kg N km^{-2} $year^{-1}$ (Li and Chen 1997; Zhou et al. 2008). N export to water bodies from forest ecosystems varies with region and forest type, and we used the parameters of Xi et al. (2007) as shown in Table 1.

Human and livestock waste is not well processed in China. It was estimated that 25% of all livestock and human waste is discharged to water bodies (Liu 1991; Liu et al. 2005). We adopted these estimates in the current study.

Biomass burning emissions

In this study, we estimated N emissions from biomass burning including forest fires, grassland fires, field burning of crop residues, and biofuels (crop residue and wood fuel). We used the same method and data sources as Yan et al. (2006) to estimate the amount of burned biomass. Emission factors for N_2, NO_x, NH_3, N_2O and HCN from various biomass burning sources were obtained from Andreae and Merlet (2001).

Denitrification and storage

Denitrification represents a large proportion of N output. However, denitrification is difficult to quantify because the process has high spatial and temporal variation and the dominant end product (i.e. N_2) has a high background concentration in the environment (Groffman et al. 2006). In this study, we took the difference between total N input and other outputs as the total of denitrification and N storage, without distinguishing between them.

Uncertainty analyses of N budgets

To evaluate the uncertainties in the N inputs and outputs, Monte Carlo simulation was performed to quantify the overall uncertainty in the response variables. The Monte Carlo method assumes that the uncertainty of the 'model inputs' can be characterized by their statistical distribution functions. For N fixation rates, a coefficient of variation (CV) of 35% was assumed. For wet and dry deposition rates, CVs of 30 and 60% were assumed, respectively. N content of compound fertilizer is not a fixed value and thus a CV of 5% was assumed. For NH_3 volatilization rate, CVs of 3–35% were used for different sources. For N export to water bodies, CVs of 5–55% were used for human and livestock waste, croplands and other land uses. For the amount of biomass burning, CVs of 5–20% were assumed. A total of 10,000 Monte Carlo simulations were performed using Matlab software (MatlabR2009a, The MatWorks^{TM}).

Results

Sources of N input

Estimated N inputs and their proportion to total N input at a national scale are shown in Table 4 and Fig. 2. Total N input increased from 3,081 kg N km^{-2} in 1985 to 5,426 kg N km^{-2} in 2007, a 76% increase. Chemical fertilizer N consumption dominated N input and accounted for 53.6% of the total N input in 2007. Atmospheric N deposition increased continuously, from 767 to 1,300 kg N km^{-2} during 1985–2007. While the total amount of N_2 fixation changed little from 1985 to 2007, its contribution to total N input decreased from 32.5 to 18.9%. Net N input through food/feed import increased steadily. Although there was net export of grains such as rice and maize in the past two decades, import of soybean with high N concentration increased greatly during 1995–2007. And China became the largest soybean importing country from 2000 (Chu et al. 2006), leading to net N import.

For each province, we distributed different N inputs to various land uses with a 1-km spatial resolution. N deposition was assumed equal for all land uses. Chemical fertilizer N was evenly distributed to cropland, and N input through food/feed import was evenly distributed to developed land. N_2 fixation rate of different land uses varied, as explained in the 'Data and methods' section. We summarized the N inputs for each grid (results for 2007 are in Fig. 3). At a provincial scale, there was large spatial variability in total N inputs, ranging from 588 to 50,582 kg N km^{-2} year^{-1} for the Tibet Autonomous Region and the Shanghai Municipality, respectively. Total N inputs of different provinces in different years were significantly correlated with cropland areas ($R^2 = 0.51$, $P < 0.01$), since chemical N fertilizer was the dominant source of N input. As a result, there was a large total N input in provinces in eastern and central China (e.g. Jiangsu, Sandong, Henan and Anhui) where land use is predominantly agriculture (Fig. 3). Relatively large N inputs were also found in southern and southeastern provinces (e.g. Zhejing, Fujian, and Guangdong) where with high per capita Gross Domestic Product and 50% of the land area was forest, which has a higher N_2 fixation rate. It is no wonder that the vast western area had very low N input as the major land use was desert. N input was relatively low in the most northeastern part of China due to the low chemical N fertilizer application rate and low crop index.

At a provincial scale, total N input steadily increased in all provinces during 1985–2007 (Fig. 4). The highest increase was in Ningxia and Tianjing, where total N input increased about tripled from 1985 to 2007. Tibet had the lowest increase in total N input, with a rate <10%, due to N_2 fixation being the dominant source of N input in Tibet, which changed little during the period. For most regions, the increase rate of total N input was 0–200 kg N km^{-2} year^{-1} (Fig. 4), with greater increase rates in eastern and central China.

N output and storages

NH_3 volatilization accounted for about 21% of the total N input in different years, but increased from 705 kg N km^{-2} in 1985 to 1,144 kg N km^{-2} in 2005. Due to the decrease in livestock population (especially pigs) in 2007, NH_3 volatilization was lower in 2007 than in 2005 (Table 4). Percentages of NH_3 volatilization from croplands, livestock husbandry, forestland and grassland were 60.8, 18.9, 4.3 and 11.3% in 1985, respectively; and 59.4, 19.4, 3.3 and 14.4% in 2005.

The amount of N exported to water bodies increased by 53% in the 22 years, and showed similar trends to that of NH_3 volatilization, peaking in 2005 and slightly decreasing in 2007. Our results showed that about 36% of the N exported to water bodies come from croplands and 48% from human and livestock excreta in 2007.

The amount of N output through biomass burning was relatively stable but its share in total N output decreased from 7.7% in 1985 to 5.3% in 2007 (Fig. 5).

Table 4 Nitrogen budget in mainland China on national scale from 1985 to 2007 (mean ± standard deviation)

Year	N input (kg N km^{-2} year^{-1})				Total (kg N km^{-2} year^{-1})	N output (kg N km^{-2} year^{-1})				
	N deposition	N fixation	Chemical fertilizer	Net food and feed import		Denitrification and storage	Ammonia volatilization	Export to water bodies	Net food and feed export	Biomass burning
1985	767 ± 262	1000 ± 177	1,314 ± 3	0	3,081 ± 391	1,499 ± 361	705 ± 156	637 ± 129	4 ± 0.2	236 ± 46
1990	950 ± 347	986 ± 175	1,820 ± 6	22 ± 1	3,778 ± 465	1,971 ± 465	813 ± 185	755 ± 159	0	238 ± 47
1995	1,089 ± 412	976 ± 171	2,329 ± 12	24 ± 1	4,418 ± 532	2,285 ± 532	1,009 ± 229	860 ± 185	0	263 ± 46
2000	1,169 ± 455	999 ± 173	2,394 ± 15	48 ± 1	4,610 ± 562	2,397 ± 562	1,053 ± 238	924 ± 200	0	236 ± 39
2005	1,232 ± 491	1,063 ± 181	2,767 ± 22	176 ± 5	5,238 ± 590	2,828 ± 590	1,144 ± 254	991 ± 221	0	279 ± 50
2007	1,300 ± 519	1,027 ± 178	2,907 ± 26	193 ± 6	5,426 ± 616	3,140 ± 698	1,027 ± 237	972 ± 207	0	287 ± 50

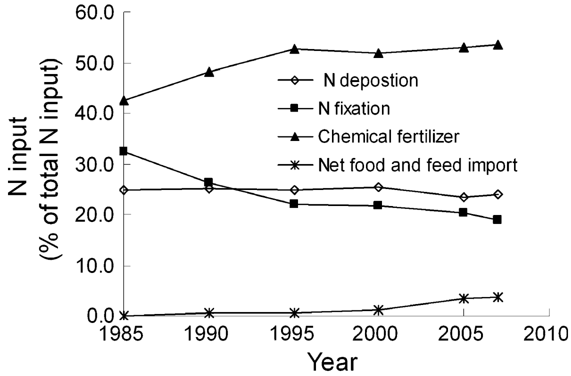


Fig. 2 Changes in the shares of various N input sources

There was a minor net N output of 4.0 kg N km^{-2} though food/feed exports in 1985 and no net export after that.

Based on a mass balance model, the N that is stored and denitrified was estimated to amount to 1,499 kg N km^{-2} in 1985 to 3,140 kg N km^{-2} in 2007 (Table 4), accounting for 48.7 and 57.9% of total N input, respectively (Fig. 5). Both denitrification and storage are difficult to quantify. However, the N storage in terrestrial ecosystems may be roughly estimated from the carbon storage. A recent study showed that carbon storage was 92–105 Tg C year^{-1} for vegetation and 75 Tg C year^{-1} for soils during 1980s and 1990s (Piao et al. 2009) in Chinese terrestrial ecosystems. No significant changes in the C/N ratio of cropland soils of China between 1980s and 2007 were detected (Yan et al. 2010). Therefore by assuming a C/N ratio of 250 for vegetation and a C/N ratio of 10 for soils, the N storage in terrestrial ecosystems in China could be estimated at about 7.9 Tg N year^{-1}, and thus the total denitrification in Chinese terrestrial ecosystem would be over 20 Tg N year^{-1} during the period, indicating that about 16% of the total N input was stored and about 42% was denitrified in Chinese terrestrial ecosystems in recent years.

Discussion

Factors influencing N budget

Human population in mainland China increased by 24.7% from 1985 to 2007 (China Statistical Yearbook 1986, 2008), but total N input increased by 76%

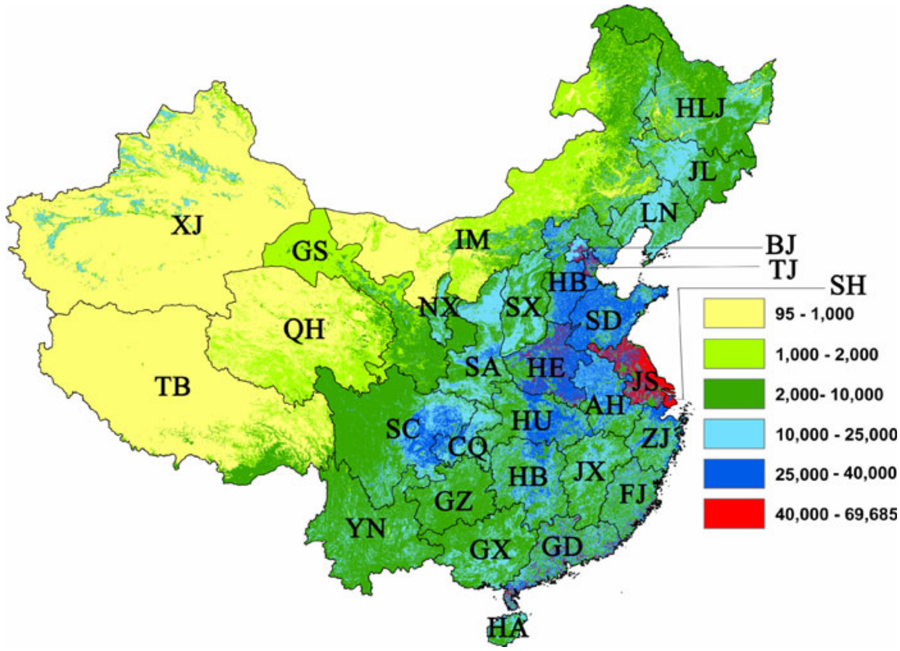
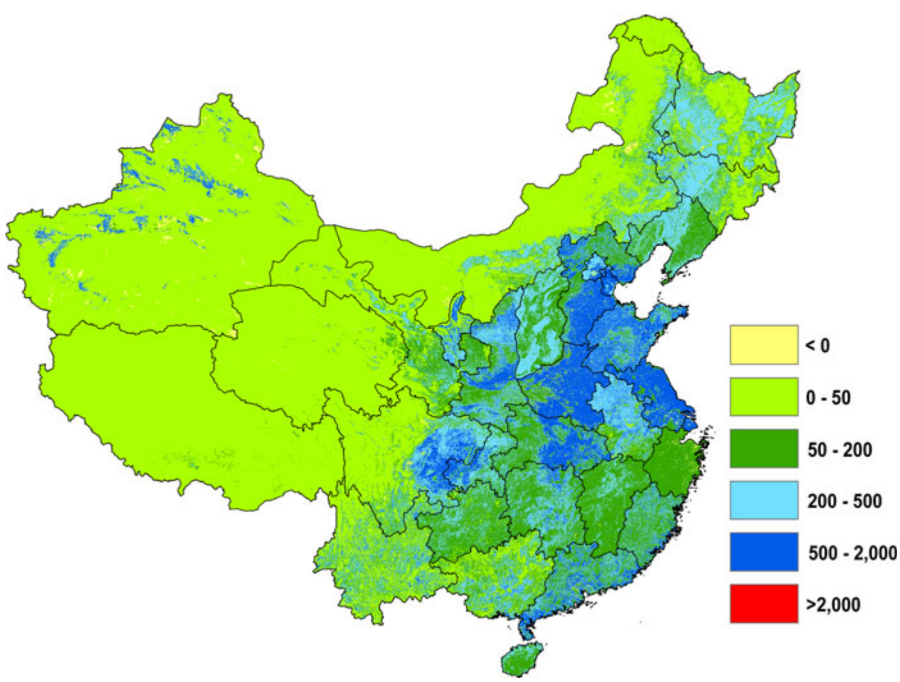


Fig. 3 Estimated total N input (kg N km^{-2}) in 2007, based on 1 km resolution land use map. *AH* Anhui Province, *BJ* Beijing Municipality, *CQ* Chongqing Municipality, *FJ* Fujian Province, *GD* Guangdong Province, *GS* Gansu Province, *GX* Guangxi Zhuang Autonomous Region, *GZ* Guizhou Province, *HA* Hannan Province, *HB* Hebei Province, *HE* Henan Province, *HLJ* Heilongjiang Province, *HN* Hunan Province, *HU* Hubei Province, *IM* Inner Mongolia Autonomous Region, *JL* Jilin Province, *JS* Jiangsu Province, *JX* Jiangxi Province, *LN* Liaoning Province, *NX* Ningxia Hui Autonomous Region, *QH* Qinghai Province, *SA* Shaanxi Province, *SC* Sichuan Province, *SD* Shandong Province, *SH* Shanghai Municipality, *SX* Shanxi Province, *TB* Tibet Autonomous Region, *TJ* Tianjin Municipality, *XJ* Xinjaing Uygur Autonomous Region, *YN* Yunnan Province, *ZJ* Zhejiang Province

Fig. 4 Average annual change rate of total N input (kg N km^{-2} year^{-1}) between 1985 and 2007, based on 1 km resolution land use map



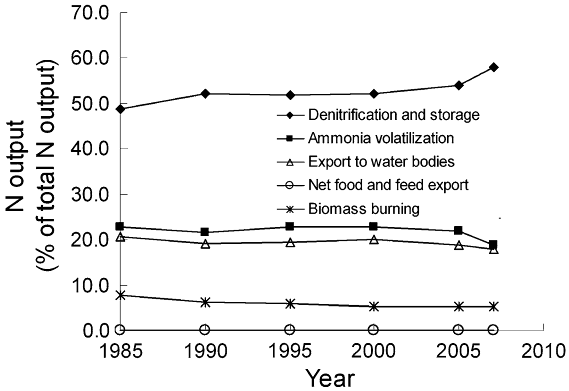


Fig. 5 Trend in the shares of N outputs from 1985 to 2007

during the same period. Obviously, rapid economic growth enhanced the demand for food, fiber and energy of the huge population and, accordingly, the consumption of chemical fertilizer and fossil fuel.

To more precisely account for the spatial and temporal variability in total N input and various N outputs, we conducted a correlation analysis between these N fluxes and land use types, human population density and per capita Gross Domestic Product (GDP). Total N input, N export to water bodies, denitrification and storage were highly correlated with population density (Fig. 6), implying that most of the N is of anthropogenic origin. Total N input, N export to water bodies, denitrification and storage also had significant positive correlations with per capita GDP (Table 5), indicating that economic development may enhance N load. Because of chemical fertilizer N which applied to cropland accounts for about half of the total N input, the percentage of cropland of total land area showed significant positive correlations with total N input and all N outputs (Table 5). In contrast, the percentage of grassland and forestland of total land area were negatively correlated with total N input and all N output. The exception is biomass burning emission, which was positively correlated with the percentage of forestland of total land area since wood fuel was a major source of biomass burning.

Comparison with N budget in other countries

When compared for similar years (e.g. around 1995), national average N input per unit area in China was higher than the global average N input but was lower than for South Korea and Europe/former Soviet Union (Fig. 7). In addition to population density and economic development, N input is also affected by climate, land use and management. For countries such as New Zealand and Brazil, due to large areas of pasture legumes and N_2-fixing crops, N_2 fixation makes up the largest N input (Filoso et al. 2006; Parfitt et al. 2006), while in South Korea and China, to pursue high crop yield, chemical fertilizer N application rate is high and dominates the N input. The high consumption of chemical fertilizer N also leads to relatively high N deposition rates in these countries.

Although the absolute amount of various N outputs changed greatly with time in China, their shares in the total N output remained relatively stable, and with denitrification and N storage the dominant output, sequentially followed by NH_3 volatilization, export to water bodies, and biomass burning emissions. Van Breemen et al. (2002) constructed N budgets for 16 large watersheds in the northeastern USA and found that, on average, soil storage and biomass increase accounted for 18% of the N input, which is close to our estimate of 16%. However, soil denitrification in China accounted for a higher percentage of the total N input (42%) than in the northeastern USA (37%). Our estimate of N export to water bodies varied from 17.9 to 20.7% of the total N output. Given that about 30% of the N loaded to water bodies could be removed or buried before reaching the river mouth (Billen et al. 1991), our estimate of N export to water bodies is significantly lower than that estimated for the northeastern USA (Van Breemen et al. 2002). There is a large discrepancy in the share of NH_3 volatilization in total N output in the two studies. In China, NH_3 volatilization accounted for 18.9–22.9% of the total N output, but only 3.0% of the total N output in northeastern USA. This is partially because most chemical N fertilizer used in China is urea, which has a high NH_3 loss rate. Compared with the N budget of New Zealand (Parfitt et al. 2006), China had a higher denitrification and storage rate and no net N output through food/feed; while in New Zealand, denitrification was only 14% of the total N output and N in exported produce accounted for 15% of the total N export.

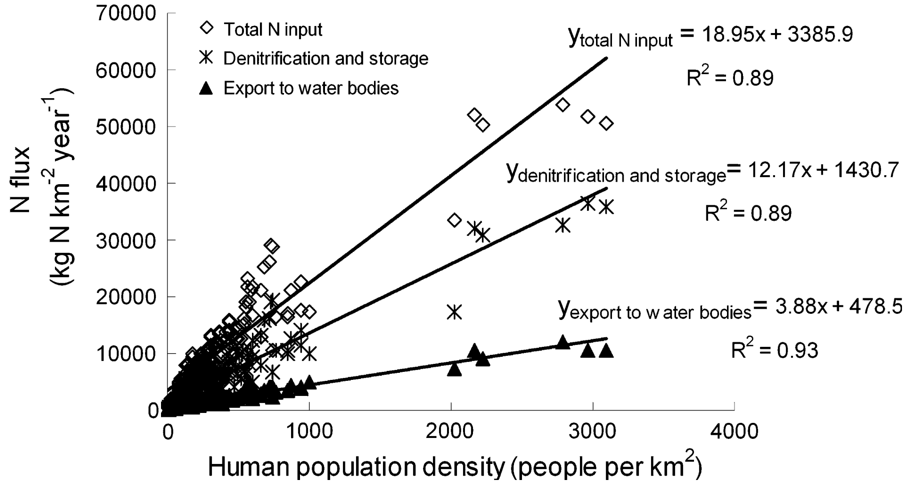


Fig. 6 Relationship between human population density and estimated total N input, N export to waterbody, denitrification and storage. Data were averaged for different provinces in different years

Riverine N export of the three major rivers

Rivers are an important link between terrestrial and oceanic ecosystems. In estuaries and coastal areas, eutrophication is a common phenomenon caused by excess loading of N and N transported to water bodies. Algal blooms have become a more and more serious problem in coastal waters in China, having occurred 12–38 times each year since 1989 (Jin et al. 1996). The Yangtze River, Yellow River and Zhujiang River are the three major out flowing rivers in China, located in the central, northern and southern parts of the country, respectively. The export of N by the three rivers has been of great concern (e.g. Huang and Huang 2002; Shen et al. 2003; Yan et al. 2003a; Chen and Yu 2004; Duan et al. 2008).

We summarized the amount of N export to water bodies for the three river basins. However, not all the N loaded to water body reaches the river mouth. Nitrogen retention in river networks, in dammed reservoirs and N loss via consumptive water use, via harvesting and grazing can occur in both basins and rivers. River-specific retention factor is not available for the three rivers. Billen et al. (1991) suggested that river retention is generally 30% of N loading and Caraco and Cole (1999) used this retention factor to estimate nitrate export in 35 large rivers in the world and found good agreement between prediction and observation. We therefore applied a river retention factor of 0.3 to estimate riverine N export of the three rivers.

Measured value of riverine N export of the Yellow River was available for years 1995 and 2000. For the Yazngtze River, export of NO_3^-–N was measured from 1985 to 2000 (Wang et al. 2006) and Yan et al.(2003a) showed that NO_3^-–N accounts for 60% of the total N in Yangtze River. For the Zhujiang River, Meybeck and Ragu (1995) reported measured value of dissolved inorganic nitrogen (DIN) for 1995, and Lin et al. (2004) showed that on average, DIN accounted for 70% of the total N in the Zhujiang River. We used these measured values and ratios to calculate riverine N export of the Yangtze River and the Zhujiang River. Table 6 showed that, the modeled riverine N exports agree reasonably well with the measured one for the Yangtze River basin and Zhujiang River basin. For the Yellow River basin, however, the estimated riverine N exports are much larger than the observed ones. This is likely due to the interception of the river water for irrigation and other purposes that may lead to low or zero flow in the lower part of the river in certain periods of the year (Meng et al. 2007).

Environmental consequences of the N budget

Although there are many uncertainties in the calculation of the N budget, our results showed that the rapid growth in human population and economic development in the last three decades have greatly enhanced N input, and consequently led to a large amount of reactive N loss into the environment. For example, river N export in Yangtze River basin was 1.93 Tg (1071 kg N km^{-2} year^{-1}) in 2005. Of the

Table 5 Relationship between N inputs/outputs and the impact factors

Impact factors	Total N input/output (kg N km^{-2} year^{-1})	Ammonia emission (kg N km^{-2} year^{-1})	Transport to water body (kg N km^{-2} year^{-1})	Denitrification and Storage (kg N km^{-2} year^{-1})	Biomass burning (kg N km^{-2} year^{-1})
Grassland (% of total area)	−0.542**	−0.559**	−0.472**	−0.505**	−0.676**
Forestland (% of total area)	−0.182*	−0.170*	−0.222**	−0.175*	0.288**
Cropland (% of total area)	0.716**	0.799**	0.675**	0.641**	0.585**
Population density(per km^2)	0.941**	0.765**	0.965**	0.941**	0.359**
Per capita GDP(Yuan)	0.555**	0.291**	0.554**	0.613**	0.110

** Correlation is significant at the 0.01 level and * correlation is significant at the 0.05 level

Yangtze River Estuary and its adjacent sea, 56% was heavily polluted in terms of N concentration (Quan et al. 2005). Algal blooms frequently occurred in recent years in Taihu Lake, located at the lower reaches of the Yangtze River (Qin et al. 2007). These environmental problems are thought to be directly related to the enhanced N export to water bodies (Xu et al. 2009).

We have shown that about 42% of the total N input was denitrified in terrestrial ecosystems. In addition, about 30% of the N export to water bodies was assumed to be denitrified before reaching estuary (Billen et al. 1991). This would have resulted in significant direct and indirect emission of N_2O (Williams et al. 1992; Seitzinger and Kroeze 1998), a gas that contributes greatly to global warming and the depletion of stratospheric ozone. In addition, the high N input in China has significantly contributed to soil acidification. Guo et al. (2010) reported that soil pH declined significantly from the 1980s to the 2000s in the major Chinese crop-production areas, with N fertilizer being the major contributor.

Uncertainties in N budget

Due to the variability in measured N fluxes and scattered data from various references, there are large uncertainties in the estimated N budget (Table 4). In the year 2007 for example, the total N input averaged 5426 kg N km^{-2}, but it may range from 4219 to 6,633 kg N km^{-2} year^{-1} (95% uncertainty range). The biggest contributor to the uncertainties in N input is the uncertainty of N deposition, especially that of dry deposition. This is due to the scarcity of measurements of dry deposition and the large spatial and temporal variability in measured wet deposition. Biological N fixation is another important source of uncertainty in total N inputs because a wide rang of values is quoted in the literature. For example, N fixation by soybean may range from 1,500 to 45,000 kg N km^{-2} year^{-1} (Smil 1999), and N fixation in forestland may range from 100 to 16,000 kg N km^{-2} year^{-1} (Xi et al. 2007). Various N outputs usually have a coefficient of variation of 17–23% except food and feed export, with denitrification and storage showing the largest uncertainty range. In the year 2007, denitrification and storage may have ranged from 1,772 to 4,508 kg N km^{-2} year^{-1} (95% uncertainty range).

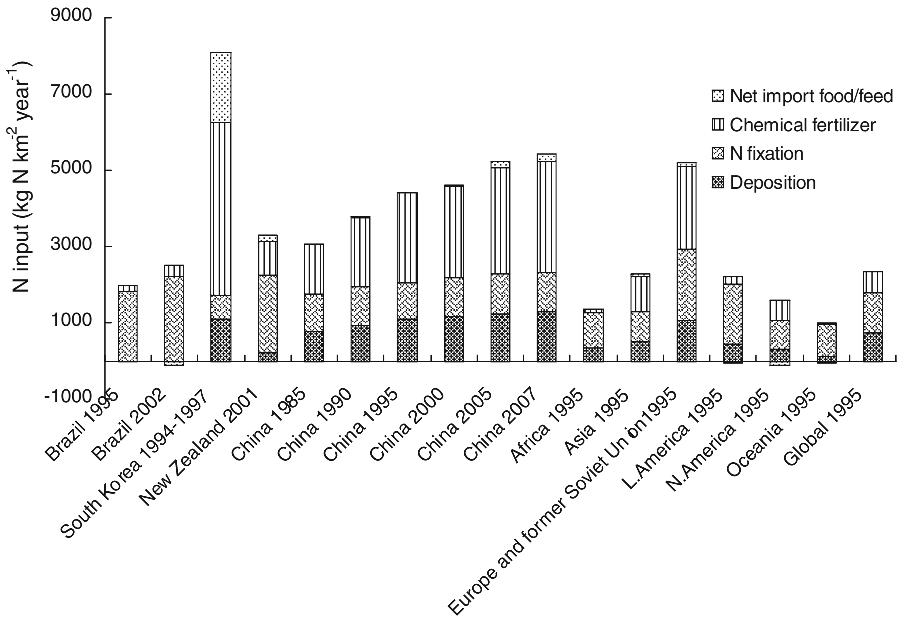


Fig. 7 Comparison of N inputs of China with other countries or regions. Data for Brazil, South Korea and New Zealand were from Filoso et al. (2006), Bashkin et al. (2002) and Parfitt et al.(2006) respectively. Data for the globe and its regions were from Galloway et al. (2004)

Table 6 Riverine N export of the three major rivers in China (kg N km^{-2} year^{-1})

River basin	Year	Modeled data (kg N km^{-2} year^{-1})	Measured data (kg N km^{-2} year^{-1})
Yangtze River (180 × 10^4 km^2)	1985	741	724[a]
	1990	812	988[a]
	1995	946	1005[a]
	2000	1004	1206[a]
	2005	1,071	
	2007	1,014	
Yellow River (75 × 10^4 km^2)	1985	364	
	1990	441	
	1995	507	129[b]
	2000	546	187[b]
	2005	598	
	2007	588	
Zhujiang River (44.21 × 10^4 km^2)	1985	637	
	1990	833	
	1995	869	772[c]
	2000	962	
	2005	997	
	2007	919	

[a] Estimated from Wang et al. (2006), [b] from Chen and Yu (2004); [c] estimated from the DIN data in Meybeck and Ragu (1995)

Conclusions

Total N input in China increased by 76% during 1985–2007. Chemical fertilizer N (use of which doubled during 1985–2007), dominated input. Biological N fixation was the second important N input till 1990 and deposition became the second important source after that. More than half of the N input was denitrified or stored in soil. NH_3 volatilization accounted for nearly 21% of the total N input, and N export to water bodies accounted for 17.9–20.7%. About 5.1–7.7% of the N input was emitted to the atmosphere through biomass burning. At a provincial scale, total N input and various N outputs per unit area correlated well with human population density. N input and major outputs were also positively related to per capita GDP and the percentage of total land area as cropland.

Acknowledgments This work was financially supported by the National Natural Science Foundation of China (No. 40621001, 41061140515). We are grateful to Profs. Guangxi Xing and Zhaoliang Zhu for their useful discussions and suggestions.

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