Centennial-scale analysis of the creation and fate of reactive nitrogen in China (1910–2010)

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Human mobilization and use of reactive nitrogen (Nr) has been one of the major aspects of global change over the past century. Nowhere has that change been more dramatic than in China, where annual net Nr creation increased from 9.2 to 56 Tg from 1910 to 2010. Since 1956, anthropogenic Nr creation exceeded natural Nr creation, contributing over 80% of total Nr until 2010. There is great interest and uncertainty in the fate and effects of this Nr in China. Here, a comprehensive inventory of Nr in China shows that Nr (including recycled Nr) has continuously and increasingly accumulated on land (from 17 to 45 Tg), accompanied by increasing transfers to the atmosphere (before deposition; from 7.6 to 20 Tg), inland waters (from 2.7 to 9.6 Tg), and coastal waters (from 4.5 to 7.7 Tg) over the past 30 y. If current trends continue, Nr creation from human activities will increase to 63 Tg by 2050, raising concerns about deleterious environmental consequences for land, air, and water at regional and global scales. Tremendous amounts of Nr have accumulated in plants, soils, and waters in China over the past 30 y, but the retention capacity of the terrestrial landscape seems to be declining. There is a possibility that the negative environmental effects of excessive Nr may accelerate in coming decades, increasing the urgency to alter the trajectory of increasing Nr imbalance. Here, a conceptual framework of the relationships between human drivers and Nr cycling in China is oriented and well-targeted to Chinese abatement strategies for Nr environmental impact.

national nitrogen budget | biogeochemical cycling | chemical fertilizer | nitrogen use efficiency

Nitrogen (N) is an essential element for all organisms, and it can limit the net primary productivity of aquatic and terrestrial ecosystems (1, 2). The vast majority of global N, however, exists as stable atmospheric N_2 and thus, is unavailable to organisms, unless it is converted into reactive N (Nr) species, which can sustain food production and global population. (The term Nr includes inorganic reduced forms of N, inorganic oxidized forms, and organic compounds in contrast to unreactive N_2 gas.) Globally, the creation of anthropogenic Nr has increased dramatically over the past century from ∼15 Tg N in 1860 to ∼156 Tg N in 1995 because of production of food and energy (3). Human activities will continue to promote N mobilization and transform the global N cycle through combustion of fossil fuels, growing demand for N in agriculture and industry, and pervasive inefficiencies in N use (4).

After formed, Nr is highly mobile and widely distributed. On a global basis, Nr is cycled and distributed by international trade or hydrologic or atmospheric transport; on a national basis, the circulation of anthropogenic Nr varies by environmental systems (atmosphere, hydrosphere, and biosphere), resulting in Nr accumulation in diverse reservoirs at different rates (5). For example, Nr can be applied to and retained in the biosphere, or it can be emitted to the atmosphere and then deposited to downwind ecosystems. Nr accumulation in different reservoirs has a wide variety of consequences, magnified with time as Nr moves along its biogeochemical pathways (6). These consequences result in a cascade of environmental changes, including smog, acid rain, forest dieback, coastal dead zones, biodiversity loss, stratospheric ozone depletion, and an enhanced greenhouse effect as well as health problems (5–8). Currently, there is great interest in improving the efficiency of Nr use and balancing food production, energy consumption, and industrial needs with the aim of minimizing damages to environmental systems (9). Essential to this effort is an understanding of the correlation between human social–economic activities and Nr cycling at local, regional, national, and global scales.

A substantial literature has examined the problems of accelerated Nr creation and emission in China (10–12). However, most of these investigations have focused on a specific district or only on agro-ecosystems rather than the whole country (13–15). There are still large gaps in knowledge of the specific dynamic and trajectory of Nr cycling across the entire environmental system of China. One limitation to assess and solve Nr-driven problems is the scarcity of a detailed Nr budget for China that quantifies multiple pathways of Nr input and loss over time. Here, we present an analysis of the pathways of Nr, including its creation, flux, and accumulation in different environmental systems (land, air, and water) in China over a long-term timescale to determine how the key drivers modulate the pathways of Nr in China. The specific objectives of this study were to (*i*) analyze the temporal (1910–2010) variations of Nr creation and use in mainland China; (*ii*) develop a national-scale model for N cycling that depicts N flows between different environmental subsystems and N inputs and outputs in each subsystem; (*iii*) provide preliminary quantitative estimates of annual Nr distribution in different subsystems of China to articulate the environmental fate and effects of Nr from 1978 to 2010; and (*iv*) construct a conceptual framework to describe how the universal and specific human drivers impact the national Nr cycling and the corresponding abatement strategies in China.

Results and Discussion

Temporal Variation of Nr Creation in Mainland China. Nr creation and use nationwide includes industrial N fixation (INF; presented with chemical fertilizer application and industrial material production), biological N fixation (BNF; both symbiotic and nonsymbiotic), inadvertent fixation during fossil fuel combustion, and natural N fixation by lightning. Over the past 100 y, total Nr creation in mainland China increased from 9.2 Tg in 1910 to 56 Tg in 2010, an increase of more than five times with an annual growth rate of 1.8% (Fig. 1). Growth in Nr creation was relatively modest and stable before 1970 and then increased sharply, with an annual growth rate of 3.7% from 1970 to 2010. The rapid growth since 1970 was largely driven by the emergence of a domestic N fertilizer industry and the intensive agriculture during the 1970s, and as a result, chemical fertilizer of 29 Tg N

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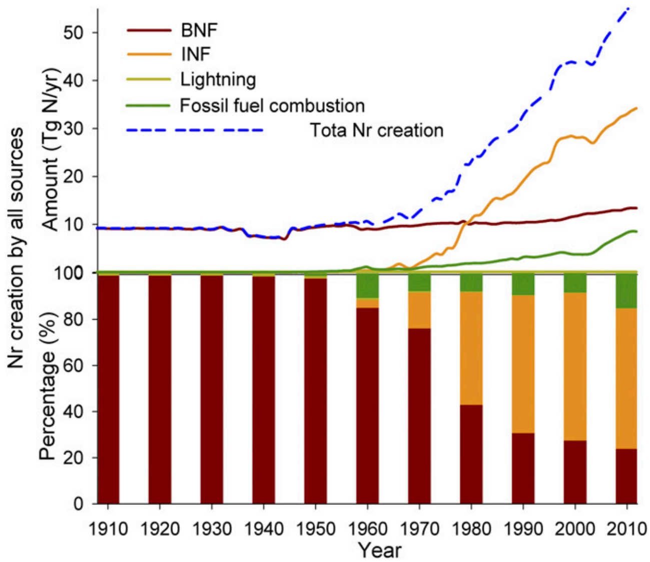

Fig. 1. Creation of Nr in mainland China for the period 1910–2010. *Upper* represents the Nr creation amount by natural sources (lightning, forest, and grassland BNF) and anthropogenic sources (agriculture BNF, INF, and fossil fuel combustion); *Lower* represents the percentage of each source.

accounted for over one-half of Nr use in 2010. To a large extent, the impact of humans on the N cycle stems from their attempt to alleviate N deficiencies in agriculture by the application of fertilizer (7). Recently, the average N fertilizer application rate on Chinese cropland has reached 240 kg/ha per year. In contrast, the rest of synthetic ammonia contributed 5.1 Tg N to produce other chemical materials and products in 2010, about 10% of the synthetic ammonia consumption. The importance of BNF, which was the single largest source of Nr before the 1980s, has declined from 99% of total input in 1910 to 24% in 2010. Nonetheless, the absolute amount showed a gradual upward trend from 9.1 to 13 Tg between 1910 and 2010 resulting from the land use changes (i.e., the recovered forest area after 1960s and the increased sown area of crops). Nr input through fossil fuel combustion has also increased continuously as the replacement of biofuel accompanied the industrial revolution, from 2.7×10^{-3} Tg in 1910 to 8.5 Tg in 2010, with particularly marked increases since 2002 when the annual growth rate reached 11%. Natural N fixation by lightning has been stable and low (9.4×10^{-2} Tg), and its share of total Nr creation has decreased from 1.0% to 0.2% in the last century.

Creation of Nr was first dominated by natural processes. In 1956, anthropogenic Nr input exceeded the natural Nr input, and it has played a primary role in N cycling since that time. In 2010, anthropogenic and natural Nr input reached 47 and 9.0 Tg, accounting for 84% and 16% of total Nr input, respectively. Given projections of population, gross domestic product per capita, energy intensity, and industrialization level, China is predicted to create 63 Tg N from human activities by 2050, estimated using the Stochastic Impacts by Regression on Population, Affluence, and Technology model (*SI Text* and Tables S1 and S2). If unabated, China will play a dominant and increasing role in the global N cycle. In addition, there has been considerable variation in the spatial heterogeneity of Nr use across China. Detailed information about disproportional distribution of Nr use and population size and affluence is given in *SI Text* and Fig. S1.

Comparative Analysis on the Global and Chinese Nr Creation. There are several important common points and discrepancies in Nr creation in China and the rest of the world (Table 1). (*i*) Over the past 100 y, natural Nr creation by terrestrial BNF decreased from 120 to 98 Tg globally because of conversion of natural lands and forests to croplands (16), but it increased from 7.2 to 8.9 Tg in China, primarily because of the forest area recovered quickly after the 1960s. (*ii*) Rates of anthropogenic Nr creation and use increased sharply both globally and in China because of food and energy production (17, 18), but the proportion of global creation used by China has nearly doubled, from 13% in 1910 to 22% in 2005. The annual growth rate of human-induced Nr use in China (3.3%) has been higher than the global growth rate (2.7%). (*iii*) Nr created by the Haber–Bosch process has dominated anthropogenic Nr both globally and in China. However, N fertilizer production in China started later and then developed extremely rapidly only after the 1970s (18). So far, China has become the largest producer and consumer of N fertilizer, accounting for one-third of world fertilizer production in 2010 (19). (*iv*) Nr creation associated with fossil fuel combustion has grown globally and in China, increasing from 0.3 Tg in 1910 to 25 Tg in 2005, with only 0.9% of the global emissions coming from China in 1910, rising to 25% by 2005 because of the coal-based energy supply and use in China. Energy consumption in China has increased drastically since the 1970s and now accounts for nearly one-fifth of the global energy consumption (20). (*v*) By 2050, global anthropogenic Nr creation will reach 267 Tg, with 63 Tg or a 24% contribution from China.

Nr Balance and Accumulation in Subsystems of China. Both new and recycled Nr migrates and cascades through the environment from land and air to fresh water and ultimately, the open ocean (6, 21). There is a strong need to determine just where Nr is transported and accumulating so that efforts to reduce its environmental impact can be successfully targeted and implemented. N balances (i.e., the difference between inputs and outputs) can be used to document where surplus N is being retained. A detailed analysis of input and output patterns (Fig. 2 and Fig. S2) as well as the assumptions and uncertainties in the methodology are given in *SI Text*. The uncertainties of Nr accumulation in subsystems are estimated through Monte Carlo simulation and summarized in Table 2.

Nr balance of the land subsystem. Total inputs of Nr to the land subsystem in China were 32 Tg in 1978 and 76 Tg in 2010, and new and recycled Nr comprised about 60% and 40% of the total Nr inputs during this period, respectively. The most important Nr input was chemical fertilizer, which contributed from 24% in 1978 to 38% of Nr input in 2010, followed by N deposition, BNF, and manure. Nr input from manure started to decline after 2000. Total outputs of Nr to other environmental subsystems were 16 and 31 Tg in 1978 and 2010, respectively, mainly through NH_3 volatilization and denitrification to N_2 and N_2O. Nr accumulation increased from 17 to 45 Tg from 1978 to 2010, with an accumulation rate of over 50%. These results are broadly similar to a previous agricultural mass-balance study in China (i.e., 45–65% N fertilizer applied was recovered in plants and stored in soil, and the other 35–55% was lost through gaseous loss, runoff, and leaching) (22) but still highly uncertain.

Additional analysis was conducted to examine Nr accumulation patterns in the land subsystem (Table S3). Our results showed that 6.6 and 17 Tg Nr accumulation in land was taken up and recovered by crop plants in 1978 and 2010, respectively (then partly transformed to biomass, food, or product), accounting for more than 40% of total N accumulation in the land. Overall, N use efficiency (i.e., the recovery rate of crop plants as well as the ratio of crop harvest N to N input into land surface) was relatively low (only 21–25%), and thus, the imbalance of N between agronomic inputs and harvest removals was considerable (23). Furthermore, most of the N removed by crops was lost to the environment as excreta and sewage through human food consumption (24); 3.7×10^{-2} and 4.8 Tg Nr were accumulated in chemical materials and products. The remaining N was either stored as wastes on land or retained in soils and other pools (e.g., natural vegetation and plant residues) at harvest. Most previous studies have discounted the fate of solid wastes. However, in the progress of urbanization and industrialization in China, except for

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Table 1. Comparative analysis of global and Chinese N budgets (Tg N/y)

Input pattern*	1910 Global	1910 China	1990 Global	1990 China	2005 Global	2005 China	2050 Global	2050 China
Natural input								
Terrestrial ecosystem[†]	120 (16)	7.2	107 (16)	6.0	98 (16)	8.2	98 (16)	8.9
Anthropogenic input								
Agriculture BNF	15 (3)	2.0	34 (17)	4.5	40 (4)	4.6	50 (3)	—
INF[‡]	0 (3)	0	85 (17)	20	121 (4)	31	165 (3)	—
Fossil fuel burning	0.3 (3)	2.7×10^{-3}	21 (17)	3.2	25 (4)	6.3	52.2 (3)	—
Amount of anthropogenic Nr	15.3	2.0	140	28	186	42	267	63
Amount of Nr	135	9.2	216	34	284	50	365	72

*Excluding natural N fixation by lightning.

[†]BNF by terrestrial organisms, including forest and grassland BNF.

[‡]The global values for INF are the industrial Nr amounts created by the Haber–Bosch process (mainly for fertilizers), and the values for China are the N fertilizer application and materials production.

agricultural wastes (e.g., crop residues), industrial and domestic wastes in cities are increasing considerably. Apart from a few solid wastes used as compost, which reentered the N cycle, the rest in stocks (including landfill in the ground and pile in land surface) contributed to Nr accumulation in the land subsystem, about 2.1 Tg in 1978 increasing to 5.8 Tg in 2010. This considerable fraction is likely to be reduced and recycled through the improvement of the disposal and utilization technologies for solid wastes in China. The Nr stored in soils and other sinks reached 7.9 and 17 Tg in 1978 and 2010, respectively. The striking increase in Nr accumulation in soil mainly resulted from the high mineral fertilizer application in China, although there was a negative soil N balance (i.e., N depletion) in cropland globally (25). However, likely because of a delayed and missing sink of N from the land subsystem (e.g., denitrification and leaching), Nr accumulation in soils did not equal net N retention. In the long run, Nr accumulation in soils after harvest, ultimately, can either be taken up by crops, if it remains in the root zone, or be subjected to loss to the environment (26).

Nr balance of the atmosphere subsystem. Total Nr inputs to the atmosphere subsystem were 7.6 Tg in 1978 and 20 Tg in 2010, most of which were delivered to terrestrial and aquatic systems through N deposition (6.3 Tg in 1978 and 22 Tg in 2010). The imbalance between N emission and N deposition could arise from the inherent uncertainties of our Nr budget and the omission of air transport between neighboring countries. The change of Nr inputs from fossil fuel burning was most significant, estimated at 1.8 Tg in 1978 and 8.5 Tg in 2010. Especially after 2002, the release of Nr by fossil fuel combustion increased suddenly and sharply, consistent with an enhancement of energy intensity during this period (27). Similar to fossil fuel burning, NH_3 volatilization from land contributed a substantial and increasing amount of Nr to the air from 5.1 to 10 Tg over the past 30 y, mainly from fertilizer and excreta (28), which was much higher than the global average emission rate because of the patterns of animal densities and type and intensity of chemical fertilizer use in China (29). Compared with these two sources, other N inputs to the atmosphere were stable and marginal. Of

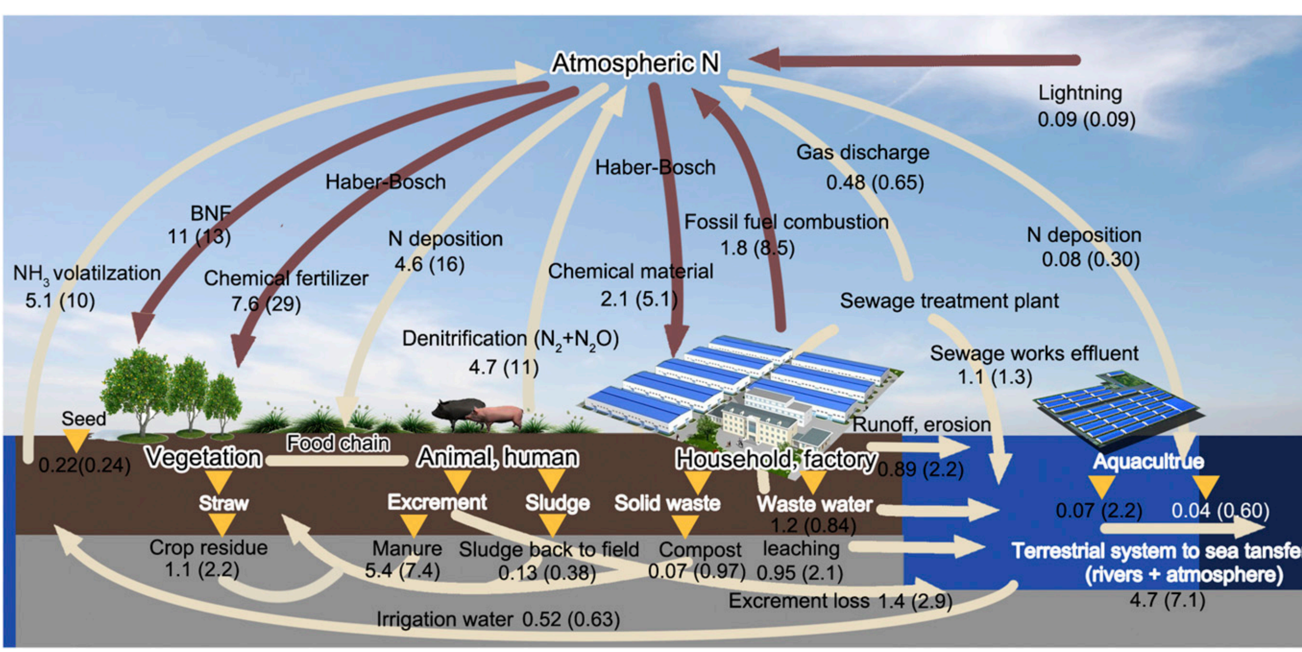

Fig. 2. Schematic model of the national Nr cycling (Tg N/y). The land subsystem is brown, and the inland water and coastal water subsystems are light and dark blue color, respectively. The red lines represent new Nr creation; the yellow lines represent recycled Nr flows. The numbers in front of the parentheses represent the Nr flux in 1978; the number in the parentheses represent the Nr flux in 2010.

Table 2. Nr accumulation (or balance) in different subsystems of China in 1978 and 2010 (Tg N/y)

Subsystem	1978 Median	1978 Minimum (5th)	1978 Maximum (95th)	2010 Median	2010 Minimum (5th)	2010 Maximum (95th)
	Range*					
Land subsystem	17	12	22	45	33	57
Plants and products	6.7	5.3	8.0	22	19	26
Solid waste storage	2.1	1.5	2.8	5.8	4.1	7.6
Storage in soils and others†	7.9	4.8	11	17	11	24
Atmosphere subsystem‡	7.6	6.4	8.9	20	16	24
Atmosphere subsystem (deduct deposition)§	1.3	−0.1	2.7	−1.6	−6.4	3.1
Inland water subsystem	2.7	2.0	3.4	9.6	8.4	11
Coastal regions	4.5	4.0	5.0	7.7	7.1	8.3
Total	25	18	33	61	43	79

*Reports the overall uncertainty as the 90th percentile of the resulting probability distribution (the 5th and 95th percentiles and the median).
†Including Nr stored in soils, natural vegetation that is not harvested, and plant residues.
‡Total Nr input to the atmosphere.
§The net input to the atmosphere (i.e., input to the atmosphere minus deposition back to the land and water surface).

these inputs, a small percentage of the Nr lost through denitrification was released as N_2O globally (6), which is a powerful greenhouse gas (30) and mainly from agricultural soils (31). Widespread use of N fertilizer and manure increased N_2O emissions from 0.18 Tg in 1978 to 0.41 Tg in 2010. Increased emissions of Nr species to the atmosphere since the 1980s have aroused widespread concern about air pollution in China (32). As a result, N deposition to the terrestrial and aquatic systems has increased by a factor of three from 1978 to 2010, which was in marked contrast to the United States and Europe, where rates of N deposition have leveled off or stabilized since the late 1980s with the implementation of legislation to limit atmospheric pollution (33, 34).

Nr balance of the inland water subsystem. In 1978, Nr inputs to inland waters were 5.8 Tg, and Nr outputs were 3.1 Tg, with 2.7 Tg N accumulation. By 2010, Nr inputs were 12 Tg, and Nr outputs were 2.3 Tg, with 9.6 Tg N accumulation. N inputs from various sources changed greatly during 1978–2010. These changes were driven by the low efficiency of fertilizer use and animal husbandry, which led to increases in fluvial transport of N, reflecting growing runoff, leaching, and excreta losses from land to inland waters (22). In contrast, N discharges from untreated wastewater of households and factories decreased significantly, especially after 2001, because of the high disposal rate of sewage. Our results are roughly in agreement with previous studies showing that ∼20% of anthropogenic N input to land is delivered to rivers (35, 36); this ratio ranged from 16% to 19% in our study. Interestingly, riverine N fluxes to the sea peaked at 2.54 Tg in 2004 and then slightly decreased to 1.6 Tg in 2010. This result is inconsistent with the view that riverine export of N into coastal oceans is strongly driven by N inputs (37, 38). This inconsistency is likely owing to the interception of river flow for irrigation or other purposes (e.g., south-to-north water transfers engineering beginning in 2004) and the maximization of freshwater use in China. Because inland and lentic water bodies acted as the important sinks for this Nr, nitrate pollution in groundwater, eutrophication, and algal blooms in rivers and lakes have become increasingly common (39).

Nr flux to coastal regions. Human activities have greatly increased the transport of biologically available N through land to potentially sensitive coastal ecosystems (40). The total N fluxes from terrestrial ecosystems to the sea (through rivers and atmosphere) were estimated at 4.5 Tg in 1978 and 7.7 Tg in 2010. The general consensus is that most of the riverine N is denitrified in coastal and shelf environments, never reaching oceanic regions (41). As our results showed, the riverine N fluxes (1.6–2.5 Tg) approached N losses through denitrification (1.6–3.6 Tg; i.e., 3.2–7.5 $\mu mol/m^2$ per hour) (42) during 2004–2010. Therefore, the anthropogenic increase in river-derived N had hardly any impact on the open ocean (43). Atmospheric N flux extends far downwind of major population centers in the southeastern coastal regions of China (44) and was deposited directly to the estuary and the open ocean (45). N deposition has surpassed river input and become the single largest N source for many offshore regions (e.g., the East China Sea and the Yellow Sea) (46). Combined with other direct discharges from sewage and industrial activities, these inputs have led to a series of problems in the marine environment. Over the past century, the occurrence of red tides has increased markedly and expanded from local to offshore areas. Most estuaries, especially in the Yellow and East China Seas, showed serious symptoms of eutrophication (39). In the long run, the ocean responds dynamically to Nr, because this element limits biological production in much of the world's oceans (47).

Nr Environmental Fate and Flux in China. Our assessment suggested that only around 20% of annual Nr creation and use was denitrified to N_2; the rest accumulated in the environment from terrestrial landscapes to coastal waters. Recent (1978–2010) increases in Nr accumulation in China have been extreme and unprecedented globally, ranging from 25 to 61 Tg, with an annual growth rate of 2.8%. The annual absolute amount and proportion of Nr accumulation in each sink (including soil, crop plant, solid waste, air, inland water, and coastal water) has also changed greatly with time (Table 2). The land subsystem was the main N pool in China, accounting for over two-thirds of annual N accumulation, followed by water bodies and then the atmosphere (deducting N deposition). Moreover, Nr accumulation in soils and inland waters has increased most quickly between 1978 and 2010. Clearly, the land subsystem is the critical regulator of the national N cycle and most directly affected by anthropogenic perturbations, controlling the key spatial interactions inherent in the Nr cascade. However, when the land can no longer absorb or break down the increasing fixed N, growing quantities of N compounds will end up in rivers, lakes, estuaries, and oceans (35). Hence, the potential for improved N management is highest in the land subsystem and can be used to address the multiple land, air, and water environmental problems that result from excessive N.

A simplified picture of N fluxes between different environmental subsystems (Fig. S3) considers land and inland water as the terrestrial landscape. The rates of Nr flux depend on the efficiency of the transformations between reservoirs. For example, in 2010, 64 Tg N_2 were transferred from the atmosphere into the landscape, 30 Tg N were returned to atmosphere, and 2.7 Tg N were transferred to the sea, suggesting that a total 31 Tg N were stored in soil, biomass, products, and inland water or

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a retention rate of 49%. According to our assessment, over the past 30 y, the retention rate of Nr in the landscape increased gradually from 41% in 1978, peaked at 53% in 2002, and then decreased to 49% in 2010 (Fig. 3). These results suggested that the critically important ability of Chinese landscapes to retain new Nr seemed to decline. This result suggests that the potential for the landscape to retain annual new added N through increased production and organic matter storage is limited. Ultimately, this limited capacity can be reflected and manifested as N saturation in forests (48) and/or increased N losses from the land surface. Thus, improving the Nr recycling rate, as an alternative to new Nr application, is needed to increase retention in the landscape and mitigate environmental problems.

Human Drivers and Abatement Strategies for China. Clearly, the accelerating Nr creation and declining N retention capacity of the landscape are in conflict. There is great interest in determining how this conflict can be altered by harmonizing the relationships between Nr cycling and human economic drivers (including the universal and specific human drivers), which is visualized by a conceptual framework (Fig. S4). However, the efforts to control population, transform economic growth patterns, and promote energy efficiency (discussed in *SI Text*) require a long time to implement. More urgent efforts should, thereby, be given priority to mitigating and abating the environmental degradation induced by human activities in China.

Agricultural production in China has increased dramatically since ∼1975 accompanied by the emergence of a domestic N fertilizer industry. Normally, a fraction of N fertilizer use in China depended on the import until ∼1998; then, the N fertilizer production increasingly surpassed its use. Because of population growth and urban expansion, increasing food requirements and limited arable land resources inevitably led to the intensification of agriculture in China. Excessive chemical N fertilization has resulted in serious environmental problems in the atmosphere, soil, and water (49). More specifically, N fertilizer is mostly applied to vegetable-producing areas, where more serious nitrate pollution of ground and drinking water is found (50). Evidently, the imbalance of N in China far exceeds N imbalance of other countries (23). In China, manure has been used as fertilizer for centuries (51); however, recently, the use of organic Nr in agriculture has been declining. The unused portion of the N in excreta and crop residue eventually ends up in air, lakes, and rivers as a pollutant through direct discharge, runoff, and burning, etc. Hence, there are ample opportunities to reduce the environmental impact of agriculture by refining management of N fertilizer application in China.

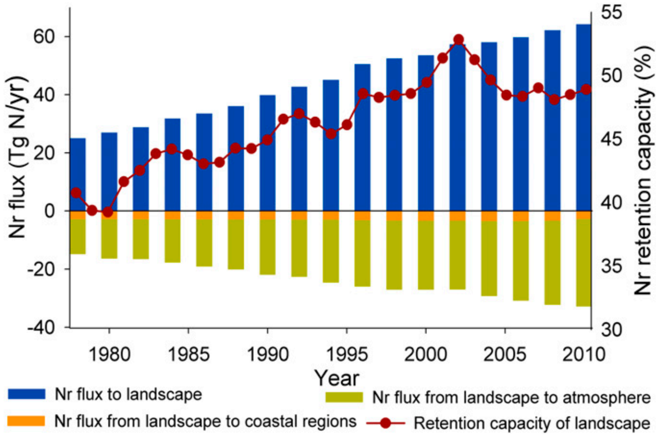

Fig. 3. Nr retention capacity of the Chinese landscape for the period 1978–2010.

China must adopt and enforce relevant agricultural regulations to eliminate N fertilizer overuse, such as removing government subsidies, introducing an N fertilizer tax, educating humans for environmental awareness (52), prescribing recommended N application rates, and issuing conventional fertilization recommendations for small-scale farming systems (53). There are many approaches to enhance N use efficiency (more efficient use of N fertilizer can allow current N application rates to be reduced by 30–60%) (49). These approaches include systematic crop rotation, optimum fertilizer timing, placement, and formulation, effective use of nitrification inhibitors, and watershed management to mitigate or redirect N losses from fields (53, 54).

Except for agriculture activities, other human social–economic activities also result in changes in Nr cycling. With the rapid urbanization progress in China since ∼1978, increasing consumption of calorie- and meat-intensive diets (55) is expected to increase N losses to the environment relative to grain-dominated diets (56). Therefore, reasonable and balanced dietary structure is encouraged. In addition, the expanding use of new energy to shift the coal-based consumption structure, improved manure and waste management, and new efficient technologies that are ideal for Chinese conditions are needed. These proactive measures need relevant sectors and agencies (e.g., water resources, environmental protection, and energy) to coordinate and cooperate.

Conclusions

The initial results from our paper are considered to be reasonable in most circumstances. Nonetheless, there are uncertainties in estimating Nr fluxes and balance, especially in the land subsystem that involves diverse activities, and improvements are needed in the national N cycling model, including better parameterization of interactions with the neighboring countries, the N cycling in ocean subsystem including the natural feedback, and more direct measurements.

Our analysis suggests that, in China, the intensification of Nr use, which is a hallmark of the development process, has occurred more dramatically than in any other part of the world. China will soon (by 2050) be responsible for nearly 24% of global anthropogenic Nr creation. Meanwhile, the capacity of environmental systems in China to absorb Nr is close to being exceeded. A surfeit of fixed Nr has ended up in water bodies or atmosphere, causing critical environmental impacts. Although substantial Nr accumulated in plants, soils, and inland waters in China as it has in other regions, the retention of Nr in the landscape leveled off, suggesting that the ability of Chinese landscapes to retain new Nr has declined and that the environmental effects of Nr may be poised to greatly accelerate.

The anthropogenic dynamics of Nr in China represent three results. (*i*) Excessive N fertilization (especially in vegetable-producing areas) and low N use efficiency in intensive agricultural system are the direct drivers to Nr imbalance. (*ii*) Support of the rapid population and economic growth is the inherent driver of human-induced Nr creation. (*iii*) The progress of urbanization and industrialization aggravates Nr imbalance by influencing the different aspects of social production and consumption. These results suggest that there is a critical need to improve the use of N in agriculture, reduce Nr production associated with energy generation, and develop improved technologies for handling Nr-rich waste streams in China. These improvements will require the development of new institutions and practices. Because the nature and extent of Nr problems have developed uniquely and intensely in China, the solutions will also have to be novel and effective.

Materials and Methods

System Definition. The content of this study was broadly divided into two parts. For the first analysis, we considered temporal variation of Nr creation from 1910 to 2010 for all of mainland China. Within the administrative boundary, more detailed analyses were done at the regional and provincial levels to include 31 areas (divided to seven geographical regions), excluding

Hong Kong, Macao, and coastal islands (except for Hainan Island) because of unavailability of data (Fig. S1). For the second analysis, we considered the mainland and coastal regions affected by the terrestrial N cycle in China for analysis of Nr accumulation from 1978 to 2010.

Model Description. In this study, the national N cycle model (Fig. 2) divides the environment into four subsystems: atmosphere, land (the surface of the earth excluding the ocean, rivers, and lakes), inland waters, and coastal waters. Each subsystem can be viewed as a dynamic system with interaction of biological communities and the physical environment and as an N reservoir. The N balance framework of each subsystem is the extended input–output system, which is described in *SI Text*. For coastal regions, only the input fluxes from the terrestrial system are taken into account because of large uncertainties for other inputs and outputs (e.g., from/to the ocean). Nr accumulation is calculated as shown in Eq. **1**, and its range is estimated through propagating the uncertainties of each input and output flux using Monte Carlo simulation. Change in an N input or output element with time is governed by a differential equation that is numerically integrated on an annual basis (Tables S3 and S4). The N input and output elements include both new and recycled Nr (e.g., manure N). Results are aggregated to the national level to analyze Nr fate and flux across subsystems throughout the whole country:

$$N_{accumulation} = \sum N_{input} - \sum N_{output}. \qquad [1]$$

Dataset. Data were collected from the following sources: (*i*) a number of national and provincial statistical databases, including "China Statistical Yearbook (1948)," "Modern Chinese industrial records (1938)," "New China sixty years compilation of statistics 1949–2008," "China Statistical Yearbook (CSY)," "China Rural Statistical Yearbook (CRSY)," "China Industrial Economic Statistical Yearbook (CIESY)," "China Energy Statistical Yearbook (CESY)," "China Agricultural Statistical Yearbook (CASY)," "Chinese Animal Husbandry Yearbook (CAHY)," and "China Environmental Status Bulletin (CESB)"; (*ii*) national resource survey statistics such as the national forest resources inventory and the national survey of grassland resources information; and (*iii*) the existing research results and methodologies. A detailed description of parameter values is given in Tables S5 and S6.

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