Integrated reactive nitrogen budgets and future trends in China

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Reactive nitrogen (Nr) plays a central role in food production, and at the same time it can be an important pollutant with substantial effects on air and water quality, biological diversity, and human health. China now creates far more Nr than any other country. We developed a budget for Nr in China in 1980 and 2010, in which we evaluated the natural and anthropogenic creation of Nr, losses of Nr, and transfers among 14 subsystems within China. Our analyses demonstrated that a tripling of anthropogenic Nr creation was associated with an even more rapid increase in Nr fluxes to the atmosphere and hydrosphere, contributing to intense and increasing threats to human health, the sustainability of croplands, and the environment of China and its environs. Under a business as usual scenario, anthropogenic Nr creation in 2050 would more than double compared with 2010 levels, whereas a scenario that combined reasonable changes in diet, N use efficiency, and N recycling could reduce N losses and anthropogenic Nr creation in 2050 to 52% and 64% of 2010 levels, respectively. Achieving reductions in Nr creation (while simultaneously increasing food production and offsetting imports of animal feed) will require much more in addition to good science, but it is useful to know that there are pathways by which both food security and health/environmental protection could be enhanced simultaneously.

reactive nitrogen | national budget | food production | industrial nitrogen | human health

Reactive nitrogen (Nr) as both a resource and a pollutant has played an important role in the tremendous changes in China's society, economy, and environment since the end of the 1970s. Haber–Bosch N fixation (HBNF) supplied more than 35 Tg (1 Tg = 10^{12} g) Nr to agricultural and industrial uses in 2012 in China, accounting for about 30% of world total HBNF (1, 2). This Nr has contributed to substantially increased food production in China, but it has come at an enormous human and environmental cost (3). In 190 major Chinese cities in 2013, about 85% of fine particulate matter with diameter smaller than 2.5 μm ($PM_{2.5}$) concentrations exceeded the safe level of 25 $\mu g \cdot m^{-3}$ suggested by WHO (4), and Nr emissions contributed significantly to this $PM_{2.5}$ (5, 6). Surface water eutrophication and nitrate accumulation in shallow groundwater threaten the safety of drinking water (7, 8). Overapplication of N fertilizer causes soil acidification, restricting the sustainable development of agriculture (9). High atmospheric deposition of Nr across China alters species composition and plant chemistry in natural and managed ecosystems (10, 11).

Using Nr to increase food production while at the same time protecting health and the environment is a crucial challenge for China, and one with substantial global implications. The United States and the European Union (EU27) have completed holistic assessments of N sources, fluxes, and effects (12, 13), and policy measures have been implemented to reduce unintended Nr emissions and their consequences (14). Such comprehensive assessment, however, has not been done in China, despite the magnitude of Nr production and the consequent human and environmental costs (3). Some aspects of N cycling in China have been studied, including the agricultural N cycle (e.g., refs. 15, 16), the industrial N cycle (17), and the creation and fates of Nr (18, 19). However, these studies did not clearly define important subsystems within China and left out some important connections among subsystems. Here we (*i*) analyze N sources, fluxes, and fates in China as a whole and among 14 different subsystems (cropland, grassland, forest, livestock, aquaculture, industry, human, pet, urban green land, wastewater treatment, garbage treatment, atmosphere, surface water, and groundwater) and (*ii*) use this subsystem-level budget to explore management scenarios and evaluate possible trajectories of N use and their consequences. Our overall objective is to evaluate how China can use Nr in support of sustainable development on a national scale.

Results and Discussion

Nitrogen Budgets in China from 1980 to 2010.

Inputs and creation of Nr. Inputs of Nr occur through biological N fixation in natural (NBNF) and cropland (CBNF) ecosystems, through HBNF, through mobilization of geological N and thermal fixation during fossil fuel combustion, and through imports of N-containing material (mostly animal feed). Lightning can also create Nr, but we ignored this flux, which is smaller than 0.1 $Tg \cdot yr^{-1}$ (19). With the exception of NBNF, all of these inputs are reasonably well constrained. Total N input to China's terrestrial ecosystems increased from 24.7 to 61.3 $Tg \cdot yr^{-1}$ from 1980 to 2010—mostly as a consequence of an increase in HBNF from 11.4 to 37.1 $Tg \cdot yr^{-1}$—and anthropogenic Nr creation amounted to 74% of total N input to China in 1980 and 88% in 2010 (Fig. 1). Nr inputs varied spatially across China, with higher values in eastern coastal areas and lower values in western regions, in accordance with the distribution of population density across China (*SI Appendix*, *Text*).

Significance

China is the world's largest producer of reactive nitrogen (Nr), and Nr in the form of synthetic fertilizer has contributed substantially to increased food production there. However, Nr losses from overuse and misuse of fertilizer, combined with industrial emissions, represent a serious and growing cause of air and water pollution. This paper presents a substantially complete and coherent Nr budget for China and for 14 subsystems within China from 1980 to 2010, evaluates human health/longevity and environmental consequences of excess Nr, and explores several scenarios for Nr in China in 2050. These scenarios suggest that reasonable pathways exist whereby excess Nr could be reduced substantially, while at the same time benefitting human well-being and environmental health.

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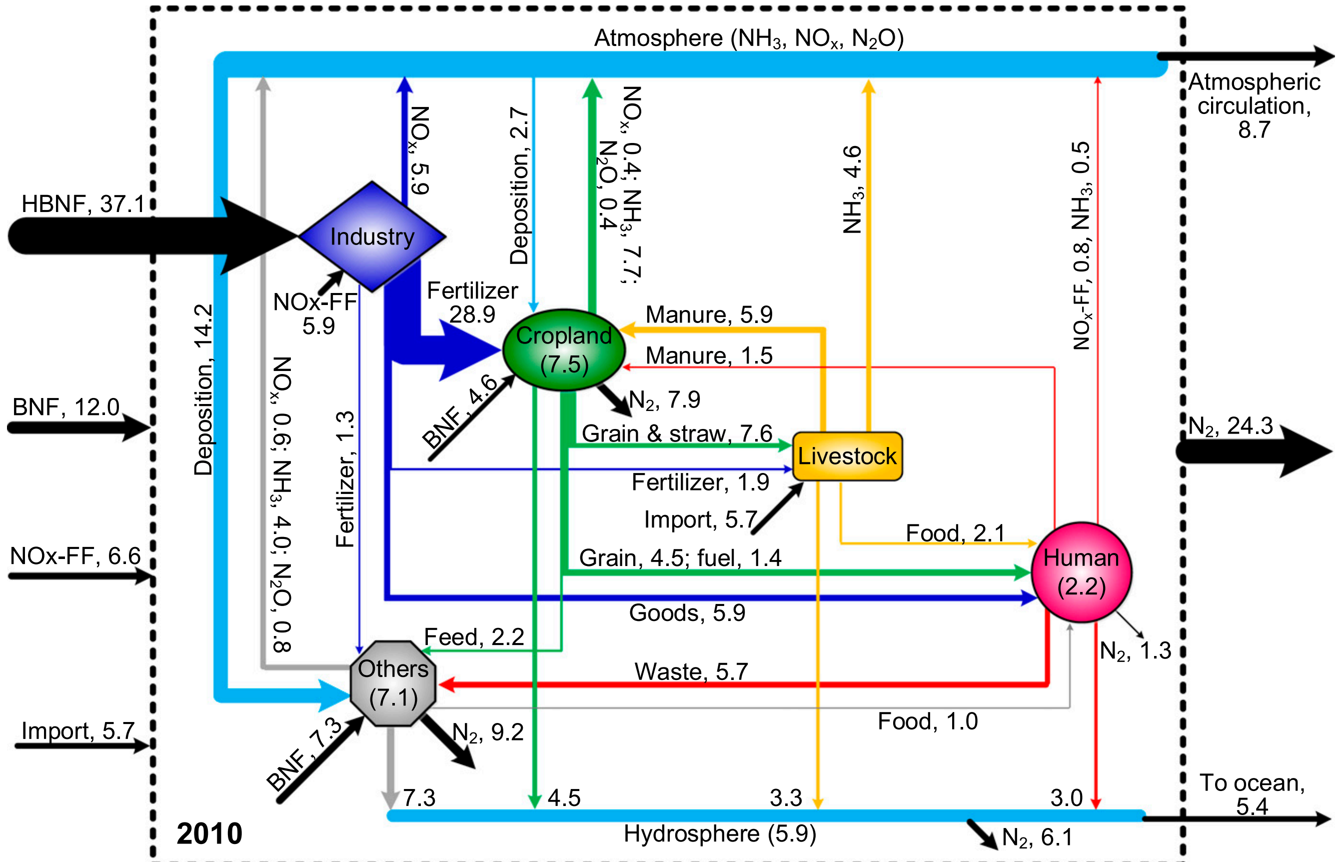

Fig. 1. Nitrogen budgets among seven important subsystems within China in 2010. The Others category includes forest, grassland, aquaculture, urban green land, pets, wastewater treatment, and garbage treatment subsystems. N fluxes smaller than 1 Tg N in 2010 are not shown, although they are included in the full 14-compartment matrix in *SI Appendix, Matrix*, and in our analyses. The comparable N fluxes in 1980 can be found in *SI Appendix*, Fig. S4. The width of arrows is proportional to the magnitude of N flux. The numbers in brackets in each box and in the hydrosphere represent Nr accumulation in each subsystem. The black arrows represent N inputs to or outputs from China. BNF, biological N fixation; HBNF, Haber–Bosch N fixation; NO_x-FF, NO_x emission via fossil fuel combustion. N_2 emission refers to Nr losses via denitrification and biomass burning. Units are in Tg $N\cdot yr^{-1}$.

Nitrogen balance of subsystems. The pathways by which Nr inputs cascaded among the full set of 14 subsystems in 1980 and 2010 are summarized in *SI Appendix, Matrix*, and discussed in *SI Appendix, Text*. Here we illustrate Nr dynamics in China through a simplified model that includes seven compartments: industry, cropland, livestock, human, atmosphere, hydrosphere, and a summed "Others" subsystem that includes forests, grasslands, aquaculture, pets, urban lands, etc. This reduced set of compartments is small enough to display, yet allows us to discuss elements of the budget relating to Nr, food security, and the environment.

Nr flows through these seven subsystems in 1980 and 2010 are summarized in Fig. 1 and *SI Appendix*, Fig. S4, and uncertainties in these calculations are described below. Most new Nr entered into circulation in China through the industrial subsystem; this new Nr increased from 12.5 $Tg\cdot yr^{-1}$ in 1980 to 43.0 $Tg\cdot yr^{-1}$ in 2010. Most of the absolute increase occurred as a consequence of increased HBNF, although because the rate of increase in fossil fuel-derived N was even greater (from 1.1 to 5.9 $Tg\cdot yr^{-1}$), the relative importance of fossil fuel N increased over time. Small quantities (totaling 1.1 $Tg\cdot yr^{-1}$ in 2010) of already-fixed Nr entered the industrial subsystem as raw material for the production of nonfood goods (e.g., cotton and leather). The most important output from the industrial subsystem was fertilizer N (32.1 $Tg\cdot yr^{-1}$, or 87% of HBNF in 2010), most of which went to cropping systems (Fig. 1). Manufactured goods accounted for 5.9 $Tg\cdot yr^{-1}$ of N transferred to human systems in 2010, and another 5.9 $Tg\cdot yr^{-1}$ of N was emitted to the atmosphere as NO_x during fossil fuel combustion.

Synthetic fertilizers, manures, and CBNF were the three main forms of N inputs to croplands; all three increased from 1980 to 2010, and the proportional contribution of industrial fertilizer to total inputs increased from 49% to 65% of total input. Thus, although recycled Nr (human and animal manure, even atmospheric deposition and irrigation water) made a meaningful contribution to total N inputs to cropping systems, N inputs to croplands were primarily—and increasingly—from industrial fertilizer. N losses and removals from cropping systems in 2010 included reactive N trace gases (8.1 $Tg\cdot yr^{-1}$ for NH_3 and NO_x), denitrification to N_2 (7.9 $Tg\cdot yr^{-1}$) and N_2O emission (0.4 $Tg\cdot yr^{-1}$), grain and straw to livestock systems (7.6 $Tg\cdot yr^{-1}$), food (4.5 $Tg\cdot yr^{-1}$) and fuel (straw, 1.4 $Tg\cdot yr^{-1}$) to human systems, feed to the Others category (2.2 $Tg\cdot yr^{-1}$, mostly to aquaculture), and losses to surface and groundwater (4.5 $Tg\cdot yr^{-1}$); all of these except the food and fuel transferred directly to human systems increased more than twofold from 1980 to 2010.

Nitrogen input to the livestock subsystem more than doubled from 5.9 $Tg\cdot yr^{-1}$ in 1980 to 16.3 $Tg\cdot yr^{-1}$ in 2010, including both domestic and imported sources of feed. Imported feed increased nearly fourfold in this interval, reaching 5.7 Tg $N\cdot yr^{-1}$ in 2010. Our estimate of feed import is about 10% higher than the national statistical data, which include only major grains (20). Major outputs of N from the livestock system in 2010 included manure returned to croplands (5.9 $Tg\cdot yr^{-1}$), food for the human subsystem (2.1 $Tg\cdot yr^{-1}$), volatilization of NH_3 (4.6 $Tg\cdot yr^{-1}$), and losses to stream and groundwater (3.3 $Tg\cdot yr^{-1}$). All of these fluxes were elevated in 2010 compared with 1980; however, the fraction of N that flowed into the livestock system (and was not consumed by humans) and then returned as manure to cropland remained 40–50% through this period.

Inputs of Nr to the human system increased from 5.4 to 15.4 $Tg{\cdot}yr^{-1}$ from 1980 to 2010. Food (agricultural products, livestock, and aquaculture, the last subsumed in the Others category in Fig. 1) increased from 3.1 to 7.4 Tg $N{\cdot}yr^{-1}$, and the proportion represented by animal-derived food increased from 14% to 40% of the total. The utilization of Nr in nonfood goods, including chemical (e.g., explosives and nylon) and agricultural (e.g., cotton and leather) products, increased over 10-fold from 0.5 to 5.9 $Tg{\cdot}yr^{-1}$, reflecting the expansion of industrial N use with socioeconomic development in China. Although the quantity of agricultural residue entering the human subsystem (primarily as biofuel) decreased slightly over the interval, household use of fossil fuel increased and added 0.7 Tg $N{\cdot}yr^{-1}$ of NO_x to the atmosphere in 2010. Losses of N from the human subsystem to waste (in the Others category) increased dramatically from 1980 to 2010, whereas the amount of N returned from the human subsystem to croplands decreased in absolute terms from 2.2 to 1.5 $Tg{\cdot}yr^{-1}$, reflecting a substantial and increasing disconnect between the human and cropland subsystems.

The Others subsystem in Fig. 1 represents a combination of subsystems that have smaller and/or more limited connections to the cropland, industry, livestock, and human subsystems than those subsystems have to each other. Connections among all 14 subsystems are summarized in *SI Appendix, Matrix*. Among these subsystems, aquaculture had relatively small fluxes (2.8 Tg $N{\cdot}yr^{-1}$ total inputs in 2010) but with extremely rapid growth since 1980 (when inputs were only 0.2 Tg $N{\cdot}yr^{-1}$). Forests and grasslands represent subsystems with relatively large N fluxes overall (combined inputs via deposition and NBNF of 21.0 $Tg{\cdot}yr^{-1}$ between them) that do not connect strongly with human, industrial, cropping, or livestock subsystems.

Both the atmosphere and hydrosphere subsystems receive substantial and increasing quantities of Nr; NH_3 plus NO_x to the atmosphere increased from 8.9 to 24.6 Tg $N{\cdot}yr^{-1}$, whereas inputs of Nr to surface and groundwater increased from 4.9 to 18.3 $Tg{\cdot}yr^{-1}$ from 1980 to 2010, reflecting substantial increases in air and water pollution in China. NH_3 remained the most important flux to the atmosphere through the period, although the fraction of atmospheric emissions represented by NH_3 decreased from 80% to 69% from 1980 to 2010. More than 70% of the NH_3 plus NO_x emitted from China was redeposited there in 2010, but 8.2 Tg $N{\cdot}yr^{-1}$ left China and was deposited downwind, where it had a substantial effect on the Nr budget of the western Pacific Ocean (21). In contrast, most Nr lost to the hydrosphere did not return to circulation within China, the major exception being 0.6 $Tg{\cdot}yr^{-1}$ in irrigation water returned to cropland in 2010 (up from 0.1 $Tg{\cdot}yr^{-1}$ in 1980). About 6.1 Tg $N{\cdot}yr^{-1}$ was denitrified to N_2 (and 0.1 $Tg{\cdot}yr^{-1}$ to N_2O) within the hydrosphere in 2010.

There are numerous uncertainties in Fig. 1 and *SI Appendix, Matrix*, as well as some components that are well constrained. The latter include HBNF and its use in fertilizer versus industrial products. The most uncertain fluxes are inputs of N as NBNF, denitrification to N_2, and Nr accumulation. Neither natural biological N fixation nor denitrification is well constrained on a regional scale anywhere in the world (22), and Nr accumulation is calculated as the difference between inputs and outputs and so reflects all of their uncertainties. Irrigation, crop and straw production, livestock production, and human food consumption are derived from national statistics, and we believe their uncertainties are small (within a range of ~5%) (20). Partitioning N surplus into different loss pathways introduces larger uncertainties because particular pathways are affected by many factors, e.g., temperature, humidity, management, pH, and oxygen availability (e.g., ref. 23); however, atmospheric deposition and the chemistry of Chinese rivers provide useful constraints on estimates of flux from terrestrial subsystems within China (4, 13), and as discussed below and in *SI Appendix*, they align well with our analyses.

Losses and storage of Nr. Taking the information on 14 subsystems in China presented in *SI Appendix, Matrix*, and summarized in Fig. 1 as the most complete and coherent Nr budget currently available, we can return to the overall N balance of China as a whole. We calculate that losses of Nr increased from 15.6 to 38.9 $Tg{\cdot}yr^{-1}$ from 1980 to 2010; most losses occurred as N_2, which is environmentally benign, but losses of Nr to downwind and downstream systems increased from 5.4 to 14.2 $Tg{\cdot}yr^{-1}$ over the interval, and losses of the greenhouse gas N_2O increased from 0.6 to 1.4 Tg $N{\cdot}yr^{-1}$. There was an absolute (from 9.2 to 22.7 $Tg{\cdot}yr^{-1}$) increase in Nr stored within China, most of it in overfertilized cropland, where both organic N and nitrate accumulation have been observed (24, 25); in forests, which retain much of the Nr they receive from atmospheric deposition (26); in groundwater, through leaching from soil N accumulation and landfills [and consistent with observed shallow groundwater pollution (8)]; and in human systems, which accumulate abundant N-containing industrial materials.

Comparison with earlier N budgets of China. The current budget is the first to our knowledge to integrate and distribute agricultural and industrial fluxes of N and to consider fluxes of Nr cascading among subsystems within China. Earlier studies differed in scope and methods (e.g., refs. 15–19), with some focused primarily upon agricultural systems. Here we compare the current budget with Ti et al. (18) and Cui et al. (19), who included most components of the overall N budgets in China (Table 1). Ti et al. (18) did not consider industrial N use (other than HBNF) and NO_x-FF in their overall budget. Cui et al. (19) conducted a more complete N budget for China as a whole but only considered three subsystems in China, land, air, and water, and so we cannot compare their results with ours on the subsystem level, especially for N accumulation. Overall, information on N inputs (e.g., HBNF) and N in products (e.g., food N) in previous budgets was drawn from the same sources as ours and not surprisingly matches our numbers closely (e.g., refs. 15–16).

Our calculation of N_2 emission is close to Ti et al. (18) but double that estimated by Cui et al. (19), who estimated the N_2 emission by using a fixed ratio with N_2O emission. In fact, much N_2O emission in China is derived from the nitrification processes (27). Furthermore, rice cultivation and polluted surface water contributed to about half of the total N_2 emission with little accompanying N_2O emission owing to low redox potentials (*SI Appendix*, Table S1). Also, Cui et al. (19) estimated a very low N_2O emission, even lower than in the United States and EU27 (Table 1).

Our estimate of NO_x-FF was lower, whereas NH_3 emission was much higher than that in Cui et al. (19). Consistent with our analysis, large-scale monitoring of N deposition in China suggested an at least 2:1 ratio for NH_3-N/NO_x-N in China (11), and our estimate of the emissions ratio of NH_3 to NO_x is close to 2, compared with the ratio of 1 estimated by Cui et al. (19). High levels of NH_3 emission also are consistent with the dominance of ammonium-based fertilizers in China (28), with the common application technique of surface broadcasting, and with observations of high NH_3 emissions in Chinese croplands (23, 28). Without closed systems to produce liquid manure (12, 29), the air dry process used to produce manure for application in China also results in high emission of NH_3 from livestock. Additionally, Cui et al. (19) estimated that 1.6 $Tg{\cdot}yr^{-1}$ of Nr was transported through air from other countries or the ocean to China, a flux necessitated by their low rates of Nr emission relative to deposition. We believe that the large, opposite flux (8.7 Tg $N{\cdot}yr^{-1}$ from China to its surroundings) that we calculated is more reasonable. Prevailing winds blow pollutants from China to Korea, Japan, the Pacific Ocean, and even North America (21, 30), and the Tibet Plateau blocks most pollutant transfer from other countries in the west such as India.

Our estimate of the total N accumulation (22.5 $Tg{\cdot}yr^{-1}$ in 2010) in China was higher than that in Ti et al. (18) but lower than that in Cui et al. (19). Ti et al. (18) did not include N accumulating in groundwater, landfill, and human settlement, and their soil N accumulation was estimated by carbon sinks and so did not include well-documented inorganic N accumulation as nitrate (24,

Table 1. Comparison of the main N fluxes in China and in the United States and the EU27

N fluxes and socioeconomy	China: Ti et al. (18) (2007)	China: Cui et al. (19) (2010)	China: This study	United States	EU27
Nr creations, Tg N·yr^{-1}					
NBNF	10.3*	8.4	7.1	6.4	0.5
CBNF	—	4.6	4.6	7.7	1.0
HBNF-SF	29.1	29	32.0	10.9	11.4
HBNF-IN	—	5.1	5.1	4.2	6.0
NO_x-FF	—	8.5	6.6	5.7	3.4
Nr losses, Tg N·yr^{-1}					
N_2	20†	10.6	24.3	—	11
NH_3	10.3	10	17.2	3.1	3.2
NO_x	—	10	7.4	6.2	3.5
N_2O	—	0.4	1.4	0.8	0.7
Runoff	9.7	12	12.2	4.8	10.5
Leaching	—	2.1	5.9	4.2	—
Export by air	—	−1.6	8.7	—	2.3
Export by water	—	7.7	5.5	1.9	4.5
Accumulation, Tg N·yr^{-1}	7.9‡	32.4§	22.7		
Cropland	—	—	7.5	0.8	−3.4
Forest	—	—	6.0	0.4	0.7
Grassland	—	—	0.6	0.3	−0.03
Human	—	—	2.2	—	—
Groundwater	—	—	5.9	—	—
Aquaculture	—	—	0.3	—	—
Urban green land	—	—	0.2	—	—
Socioeconomic factors					
Land area, million km^2		9.6		9.8	4.3
Population, million		1,341.3		310.4	500.4
GDP, trillion, PPP, $2005		9.1		13.1	13.8

Data year for China was 2010 in this study; the years listed for each study represent the data years for previous studies within China. Data year for the United States was 2002, and data year for EU27 was 2000. The data for the United States and the EU27 were adopted from Doering et al. (13) and Leip et al. (32), respectively. NBNF, natural biological N fixation; CBNF, cultivated biological N fixation; HBNF-SF, Haber–Bosch N fixation for synthetic fertilizer use; HBNF-IN, Haber–Bosch N fixation for industrial N use; NO_x-FF, NO_x emission via fossil fuel combustion. Nr losses include both natural and anthropogenic sources; these are difficult to separate because a large fraction of natural emissions derive ultimately from anthropogenic sources; for example, much of the N_2O emission from forest derives from anthropogenic Nr deposition. In this study, we listed the detailed sources of Nr losses from 14 subsystems in *SI Appendix*, *Matrix*, to illustrate these issues. The socioeconomic factors are all on a 2010 basis. GDP is at 2005 prices and adjusted for purchasing power parity (PPP). Units are in Tg N·yr^1. We cannot compare our results with Ti et al. (18) and Cui et al. (19) on the subsystem level; they did not report these data.

*Including CBNF.

†Including N_2O.

‡Only including organic N accumulating in soil and biomass.

§Including 17 Tg N·yr^{-1} in soil and biomass, 5.8 Tg N·yr^{-1} in solid waste, and 9.6 Tg N·yr^{-1} in inland water.

25). Cui et al. (19) estimated a total 33.4 Tg N·yr^{-1} accumulating in China, with accumulation in water bodies at 9.6 Tg N·yr^{-1}. We believe the high denitrification potentials in surface water (31) could not support much Nr accumulation; this difference also explains why Cui et al. (19) report lower N_2 emission.

We suggest that in comparison with previous studies, the Nr budget in Fig. 1 and *SI Appendix*, *Matrix*, provides a reasonable summary of the current status of Nr in China and how it has changed since 1980. It also provides a useful basis for comparing China with the United States and the EU27, for which similarly detailed N budgets are available (13, 32).

Comparison with N budgets and health costs in the United States and the EU27. We compare our N budget of China with recent budgets for the United States and the EU27 in Table 1. These regional budgets differ in a number of important respects, with higher rates of creation of anthropogenic Nr in China compared with the United States and the EU27 and higher emissions to the atmosphere and fluxes to the hydrosphere in China as well. Some of these differences are very well characterized; for example, the fact that China produces more Nr through HBNF and applies substantially more N fertilizer than the United States and the EU27 is well established. Other differences are less well constrained but are consistent both internally and with other sources of information. For example, NH_3 emissions in China (calculated at 17.2 Tg N·yr^{-1}) are well in excess of those in the United States or EU27 (3.1 and 3.2 Tg N·yr^{-1}, respectively); as discussed above, this difference is consistent with fertilizer types, management practices, and observations of very high rates of ammonium deposition in China (11). In contrast, the difference in NBNF (with very low inputs in the EU27 compared with China or the United States) probably reflects differences in how these inputs were calculated more than a real difference in fluxes; NBNF is difficult to determine on regional scales, and there are wide differences among the regional and global estimates of NBNF that have been proposed (33, 34).

The EU27 assessment went to considerable effort to calculate the human and economic costs of increased Nr (12). Information therein (32, 35) can be used to calculate that Nr emissions to the atmosphere cause the loss of 2.6 million years of life each year in Europe, the majority associated with the contribution of N emissions to $PM_{2.5}$ (this calculation is described in *SI Appendix*, *Materials and Methods*). The overall US assessment did not attempt a comparable evaluation, but other studies have reported substantial health effects of Nr in the United States. For example, Sobota et al. (36) calculated that the potential health and environmental damages of anthropogenic Nr in the United States totaled \$210 billion·yr^{-1}.

Emissions of both NH_3 and NO_x are higher in China than in the United States or the EU27, and intensive agriculture and industry are more closely intermingled with population centers in China than is now the case in the United States or the EU27, contributing to widespread and remarkably high levels of $PM_{2.5}$ in many Chinese cities (4). Scaling the emissions and population exposures from Europe to China (3), we calculate that using 2010 Nr fluxes, Nr emissions cause the loss of 17.4 million life years annually in China (calculation in *SI Appendix, Materials and Methods*).

Nr in China: Benefits and Costs, Challenges and Opportunities. The comparison in Table 1—and the associated loss of life discussed in the previous section—emphasizes the costs of Nr in China. The benefits of increased Nr have been substantial as well; HBNF-created fertilizer was a major contribution to the 148% increase in crop production from 1980 to 2010, at which time per capita consumption reached 5.5 kg N·yr^{-1} in China (close to the 6–7 kg N·yr^{-1} in the United States and Western Europe). The share of food contributed by animals increased from 14% to 40% from 1980 to 2010 with an average annual increase rate of 4%, much higher than the world average of 0.4% (1). However, N fertilizer use became less efficient (in terms of food produced per unit of N applied) from 1980 to 2010. And the increase in animal products in diets meant that despite increased crop production, China became the largest feed importer worldwide, relying on international markets for 5.7 Tg N·yr^{-1} in imported feed by 2010. As a consequence, the substantial increase in Nr creation and in crop production did not bring China closer to self-sufficiency in food production. Although food security and food self-sufficiency are not equivalent, the massive dependence of China's livestock systems on imported feed makes China both vulnerable to and in danger of driving price shocks in international markets.

The central challenge raised by the Nr budget of China and its connection to food production is that China must continue to enhance crop yields, to sustain a still-growing and ever-wealthier population and to offset the large imports of animal feed that it considers to be a threat to food security. At the same time, the environmental and health consequences of excessive Nr must be reduced far below their current levels. It is no longer feasible to trade off health and environment for yields: the human and environmental costs are already excessive and unacceptable, and returns to inputs of N fertilizer have diminished. Fortunately, there are a number of pathways available to reduce the quantity and costs of anthropogenic Nr in China. One such pathway is diet: interventions that prevent future increases in consumption of animal protein could reduce Nr. A second pathway is increasing the N use efficiency (NUE) of food production systems. At present, the NUEs of food production subsystems in China are much lower than those in the United States and the EU27 (*SI Appendix, Text*). A third pathway arises from the fact that cropland in China is poorly coupled to the livestock and human subsystems; the overall N recycling ratios of livestock excreta and human waste were only 43% and 23%, respectively, in 2010 in China.

Here we evaluate a series of scenarios based on these three pathways of intervention by which China could reduce its creation of Nr and thereby reduce associated health and environmental damages, while at the same time enhancing food security. Both industry and food production contribute to the excessive production of Nr in China, and both will have to reduce their emissions. Because of our interest in food security—and because of the dominant role of agriculture in Nr creation—we focus our analysis on the food system here. For industrial emissions we simply contrast a business-as-usual increase in combustion-related emissions and industrial (as opposed to fertilizer) use of HBNF with a transition to the best current global practices in industrial emissions.

Our scenarios assume that the total demand for Nr-containing goods and Nr-producing energy will increase in China because the per capita demand will increase with economic development (although the population will be slightly smaller in 2050 than at present) (37). Also, all of our scenarios incorporate the goal of no imports of human food or animal feed in 2050. Our reference scenario [business as usual (BAU)] assumes an increase in the fraction of food N from animal sources from 40% to 61% and NUEs at the 2010 level; together with continued increase in Nr emissions from fossil fuel combustion and in HBNF going into industrial goods, total Nr creation would increase from 48.2 to 98.4 Tg·yr^{-1} from 2010 to 2050 (Fig. 2). This increased N input would result in an increase of 63% for the total Nr losses in 2050 compared with the 2010 level. A sensitivity analysis, varying the population and per capita consumption, yielded an uncertainty of total N input ranging 89.7–106.0 Tg·yr^{-1} under BAU in 2050. Improving industrial NO_x emission to standards of emission per unit energy currently achieved in some western countries and similarly reducing Nr going into industrial goods would reduce Nr creation to 79.6 Tg·yr^{-1}.

Scenario S1 changed the total demand by reducing the share of food N derived from animals in 2050 to 40%, the current level, following the Chinese Dietary Guidelines (38). This change would reduce the total N input to 82.3 Tg·yr^{-1} in 2050, or to 63.5 Tg·yr^{-1} if coupled with improvements in industry. Although better than BAU, it still increased total N input by 32% compared with 2010, mainly through offsetting feed imports with local production. Scenario S2 is based on an improved NUE in each sector, to a level comparable to the current best level worldwide (39). Under this scenario (and with improvements to industry), Nr creation would be reduced to 46.5 Tg·yr^{-1}, about the same as in 2010. If cropland could achieve the levels of NUE and increased crop yields calculated by Chen et al. (40), Nr input could be reduced even further, to 37.0 Tg N·yr^{-1} in 2050. In scenario S3, we explored the consequences of increasing recycling of N from livestock and human systems back to cropland, by increasing recycling rates of livestock and humans from 43% and 23%, respectively, to 80% and 50%, respectively. This scenario, in combination with improvements to industry, would decrease Nr creation to 66.8 Tg N·yr^{-1} in 2050. Finally, S4 involves addressing diet, NUE, and recycling simultaneously; together with improving industry, it would reduce anthropogenic Nr creation to 31.0 Tg N·yr^{-1}, 64% of the current level, and total Nr losses in 2050 would be reduced to 52% of their current level.

There are numerous uncertainties in this scenario-based analysis of the future of Nr in China, but it nevertheless leads to three clear and strong conclusions. First, BAU is not an acceptable path. Given diminishing returns of yields to N inputs (41), it may not be possible to produce sufficient food in China under BAU, but even if that can be done, the health and environmental consequences

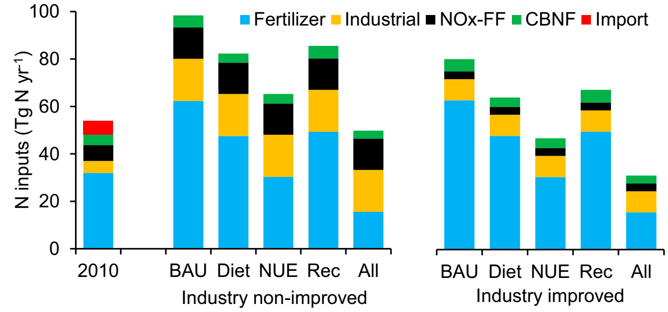

Fig. 2. Total N input to China in 2050 under different scenarios. Industry nonimproved represents a case in which industrial N use and NO_x from fossil fuels (NO_x-FF) remain the same as the BAU across all of the scenarios, and industry improved represents improvement of industrial N use and NO_x-FF to levels that have been reached in developed economies elsewhere, for all scenarios. Diet (S1) retains the animal food share of the human diet at 40%; NUE (S2) increases the NUEs of cropland, livestock, and grassland from 40%, 15%, and 6% at present to 60%, 20%, and 10%, respectively; Rec (S3) increases the recycling rates of N from livestock and humans to cropland from 43% and 23% to 80% and 50%, respectively; and All (S4) combines Rec, Diet, and NUE. Units are in Tg N·yr^{-1}.

would be overwhelming. Second, meaningful reductions in Nr creation and circulation require that China follow multiple pathways simultaneously. Only by reducing industrial emissions, not exceeding the Chinese Dietary Guidelines, improving NUE, and increasing recycling can substantial reductions in Nr (relative to present levels) be achieved, and those reductions must be achieved to ensure the sustainability of agriculture as well as human health and the environment. Third, this analysis shows that a potential pathway exists that would reduce Nr, increase food production, and improve human health and the environment.

The combined scenario S4 suggests the potential for a bright future for China's N use. Of course, it is one thing to say that the biophysical potential for a (relatively) safe and (relatively) sustainable future exists and quite another to find the political, social, and economic means to achieve that potential.

Materials and Methods

System Boundary. The system framework used for this study is shown in *SI Appendix*, Fig. S1. The study area covers the entire terrestrial land of China; Taiwan, Hong Kong, and Macao were excluded owing to data limitations. In the vertical direction, N deposition within was considered as internal flux because it was usually derived from domestic emissions (42). We considered the lower boundary of the system to be the bedrock surface; mineral resources (e.g., fixed N in coal) were not included (3). N cycling starts from the entry of Nr into the system from N_2 or (less abundantly) from imports of Nr or mobilization of fossil fuel N; N cycling within China terminates when Nr is reduced to N_2 or lost to the systems outside China. Within the boundary, the system was divided into 14 subsystems: cropland, grassland, forest, livestock, aquaculture, industry, human, pet, urban green land, wastewater treatment, garbage treatment, atmosphere, surface water, and groundwater.

Model Description. We used the urban rural complex N cycling (URCNC) model (42) to quantify N fluxes within China. The URCNC model incorporates and integrates all Nr fluxes and their interactions that can be identified, together with the linkages among subsystems (42). The basic principle of the URCNC model is mass balance for the whole system and each subsystem:

$$\sum_{h=1}^{m} \mathrm{IN}_h = \sum_{g=1}^{n} \mathrm{OUT}_g + \sum_{k=1}^{p} \mathrm{ACC}_k,$$

where IN_h and OUT_g represent the N inputs and outputs, respectively, and ACC_k represents the N accumulations. Most Nr input cascades among different subsystems; for example, NO_x emission from fossil fuel combustion deposits onto three major landscapes, natural land (i.e., forest and grassland), water bodies, and cropland, and it can undergo further transformations and fluxes in and from these landscapes. N output includes riverine N transport to coastal waters, atmospheric circulation that advects Nr away from China, denitrification, and N-containing product exports.

We identified and calculated over 6,000 N flows from 1980 to 2010 in China, using the N Cycling Network Analyzer (NCNA) model to compile the dataset and calculate all N fluxes as total Nr (43). The combined NCNA and URCNC models standardize parameters for the N flux calculations and calculate N fluxes and their interactions automatically based on a mass balance approach (*SI Appendix*, Fig. S2). More detailed methods and the data sources we used are summarized in *SI Appendix*, *Materials and Methods*.

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