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Excessive N fertilization in intensive agricultural areas of China has resulted in serious environmental problems because of atmospheric, soil, and water enrichment with reactive N of agricultural origin. This study examines grain yields and N loss pathways using a synthetic approach in 2 of the most intensive double-cropping systems in China: waterlogged rice/upland wheat in the Taihu region of east China versus irrigated wheat/rainfed maize on the North China Plain. When compared with knowledge-based optimum N fertilization with 30–60% N savings, we found that current agricultural N practices with 550–600 kg of N per hectare fertilizer annually do not significantly increase crop yields but do lead to about 2 times larger N losses to the environment. The higher N loss rates and lower N retention rates indicate little utilization of residual N by the succeeding crop in rice/wheat systems in comparison with wheat/maize systems. Periodic waterlogging of upland systems caused large N losses by denitrification in the Taihu region. Calcareous soils and concentrated summer rainfall resulted in ammonia volatilization (19% for wheat and 24% for maize) and nitrate leaching being the main N loss pathways in wheat/maize systems. More than 2-fold increases in atmospheric deposition and irrigation water N reflect heavy air and water pollution and these have become important N sources to agricultural ecosystems. A better N balance can be achieved without sacrificing crop yields but significantly reducing environmental risk by adopting optimum N fertilization techniques, controlling the primary N loss pathways, and improving the performance of the agricultural Extension Service.

intensive agriculture | synthetic N fertilizer | denitrification | nitrate leaching | N deposition

The last 40 years have seen an extraordinary change in the global N cycle (1). As recently as the 1960s, N availability in most parts of the world was controlled by natural processes, but the expanded production of synthetic N fertilizer and release of N from fossil fuel combustion now matches the natural rate of formation of reactive N on a worldwide basis (2). The rate of change has been spectacular with half of the synthetic N fertilizer ever used having been applied during the last 15 to 20 years (3). Furthermore, because of the difficulty in accurately predicting N fertilizer requirements, rates exceeding plant requirements are often applied, thus inducing unintended environmental consequences such as leaching of nitrate and emission of nitrous oxide and ammonia. This has become a major concern for scientists, environmental groups, and agricultural policymakers worldwide.

From 1977 to 2005, total annual grain production in China increased from 283 to 484 million tons (a 71% increase) and the average grain production per unit area increased from 2,348 to 4,642 (a 98% increase). However, synthetic N fertilizer application increased from 7.07 to 26.21 million tons (a 271% increase) over the same period. This resulted in a partial factor productivity from applied N (PEP_N) that decreased from 55 to 20 kg/kg (4, 5). Large amounts of external N inputs with decreasing N use efficiency have contributed to severe environmental degradation since the 1990s (6, 7). For example, the annual application rate of synthetic N for conventional agricultural practices in east and southeast China as well as the North China Plain now ranges from 550 to 600 kg of N per hectare for typical double-cropping systems (5, 8).

The Taihu region and the North China Plain are 2 of the most intensive agricultural regions in China and the most economically developed areas (5, 9). Large inputs of synthetic N fertilizer, rapid development of intensive livestock production systems, and rapidly increasing consumption of fossil fuels have severely disturbed regional biogeochemical N cycling and led to a series of environmental problems including eutrophication of surface waters, nitrate pollution of groundwater, acid rain and soil acidification, greenhouse gas emissions, and other forms of air pollution. There have also been effects on human health and normal functioning of ecosystems (9, 10), details of which are given in *SI Text*.

Here, we provide the actual grain yields and total N losses associated with knowledge-based optimum fertilization strategies compared with experience-based N management practices from numerous on-farm field experiments. The 'knowledge-based optimum N fertilization' (11) hereafter refers to using large numbers of field-based academic research results or soil tests on quantity and timing of synthetic N fertilizer practically, using regional mean optimal N (RMON) application rate in rice/wheat systems and in-season N management based on the soil mineral N test in wheat/maize systems (5, 8, 12, 13). We use an integrated approach to measure the fate of ^{15}N fertilizer and to document the major loss pathways *in situ* rather than by using individual measurements of loss processes reported in other studies to understand N behavior in these 2 rotations and thus attempt to control the environmental risk. We explain the reasons for the large external N inputs currently practiced in these intensive management systems and suggest strategies for balancing N management by applying knowledge-based optimum N fertilization techniques.

This study examines double-crop rotations, which are 2 of the most intensive agricultural systems worldwide and are widely practiced in Asian countries. The frequent alternation between flooding and draining the rice/wheat rotations led to significant changes in soil N transformations as compared with upland crop rotation. In addition, the extremely high external synthetic N inputs

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Table 1. Average grain yields and total N losses of the optimum N fertilization (ON) compared with farmers' N practices (FN) (Field Study 1 and 2).

		N rate		Grain yield		Total fertilizer N loss*	
Crop and site of field experiment	N fertilization	Rate, kg of N per hectare	Ratio of FN to ON	Yield, kg·ha^{-1}	Ratio of FN to ON	Total loss, kg of N per hectare	Ratio of FN to ON
Rice in Taihu (*n* = 26)	ON†	200		8,270		102	
	FN	300	1.5	8,012	0.97	174	1.7
Wheat in Taihu (*n* = 9)	ON†	153		3,700		76	
	FN	250	1.6	4,084	1.10	155	2.0
Wheat in NCP‡ (*n* = 121)	ON§	128		6,024		25	
	FN	325	2.5	5,764	0.96	71	2.8
Maize in NCP‡ (*n* = 148)	ON§	158		8,900		52	
	FN	263	1.7	8,500	0.95	108	2.1

*Total fertilizer N losses calculated with the models of Fig. 1B simulated from ^{15}N field experiments.
†Regional mean optimal N application rate calculated from the mean of economically optimum N rates of field experiments in Taihu regain (5, 12).
‡Data including Field Study 1 and also summarized from ref. (15, 16); NCP, North China Plain.
§In-season nitrogen management based on soil Nmin test on the NCP (8, 13, 15, 16).

used in these two rotations during the last decade provided a unique opportunity to study the effects of N on environmental degradation. A detailed knowledge of N cycling and environmental impacts of these two intensive agricultural regions may also provide options for more rational N management practices in other intensive management cropping systems around the world.

Results and Discussion

We developed knowledge-based optimum N management techniques in rice/wheat systems in south China and wheat/maize system in north China during the last decade. The regional mean optimal N (RMON) application rate was calculated from the average of economically optimum N rates (i.e., the point of the N rate where the marginal grain production value is equal to the marginal N fertilization cost) based on large numbers of on-farm field experiments in the rice-based rotations and in-season N management based on the soil mineral N (Nmin: NH_4^+-N + NO_3^--N) test previously developed for upland crop rotations. In principle, RMON recommends a regional rational N rate in combination with fine tuning according to specific field conditions, taking into consideration the relative uniformity of climate, soil N fertility, and absence of a satisfactory soil N test index for rice-based rotations (5, 12). RMON gives the mean of economically optimum N rates and can be used as a reference for extension technicians. This recommendation method can be easily adapted to rural areas of China where soil testing facilities are usually not available and the cropping index is high. The short time interval between harvest of the first crop and sowing of the second makes soil testing impractical.

The improved Nmin method is based on synchronization of crop N demand and soil Nmin supply, and is considered to be a soil N supply index because of the high amounts of Nmin (mostly nitrate) frequently found in the root zone and its high availability for subsequent crop growth in the next phase of upland rotations. We found that high potential rates of mineralization and nitrification contribute to this high accumulation of nitrate in soils of the North China Plain (14). This method has been well established in wheat/maize rotations in recent years (8, 13, 15, 16) and can be easily adapted to individual fields because the nitrate test has become easy to perform in the laboratory and may even be feasible in the field (17). In the present study, we also evaluated the agronomic performance and potential environmental risk of these knowledge-based strategies in comparison with the farmer's experience-based practices.

We conducted numerous on-farm field experiments (Field Study 1) with different N levels in Taihu region and the North China Plain from 2003 to 2006. With the sole exception of wheat in Taihu region, the optimum N rate as determined using the regional mean optimal N application rate in Taihu region and the improved Nmin method on the North China Plain appeared to give slight increases in crop yield compared with farmers' current practices (Table 1). However, no significant differences ($P > 0.05$) were found, indicating that similar high crop yields can be achieved with lower N fertilizer rates because of increased soil indigenous N supply (18, 19). In contrast, total N losses (including NH_3 volatilization, denitrification and leaching from the top 1 m of the soil profile) increased significantly with increasing N inputs, indicating high environmental costs were caused by exceeding optimum N fertilizer rates.

We used ^{15}N in some of the field experiments (Field Study 2) to quantify differences in fertilizer N fate at different application rates among the four crops. We found that N recovery rates decreased with increasing N application rate for all of them (Fig. 1*A*). N rates showed polynomial, linear, polynomial, and logarithmic relationships with N recovery by rice, wheat (south), wheat (north) and maize, respectively. Each fitting produced a highly significant model ($P < 0.01$). At lower N input levels (<125 kg of N per hectare), the order was: wheat (north) > maize > wheat (south) > rice. At higher N input levels (>125 kg of N per hectare) the order was: wheat (north) > rice > maize > wheat (south), indicating that rice recovered more N at high N application rates. N loss rates increased with increasing N application rate in all four crops (Fig. 1*B*). A logarithmic model satisfactorily simulated the correlations between N rate and N loss rates for the four crops ($P < 0.01$). Rice and wheat (south) in the Taihu region showed much higher N loss rates than did wheat (north) and maize on the North China Plain. The trends of the N retention rates with increasing N rate differed among the four crops. The rice and wheat systems in the Taihu region showed lower N retention rates than wheat and maize systems on the North China Plain. Moreover, N retention rates in wheat (south) and wheat (north) decreased first and then increased with further increase in N rate, showing typical polynomial relationships ($P < 0.01$). However, N retention rates in rice and maize decreased by power function relationships when N rate increased ($P < 0.01$). These findings indicate that the upland wheat and maize rotation system has significantly higher N recovery rates, higher N retention rates, and lower N loss rates than the waterlogged rice and upland wheat rotation in the Taihu region under current soil N fertility and farmers' management practices. Moreover, on the North China Plain wheat has significantly higher N recovery rates, higher N retention rates, and lower N loss rates than maize, and in the Taihu region, rice has significantly higher N recovery rates and lower N loss rates than wheat. Rice/wheat rotations show high 'leakage' of external N inputs with resulting pollution threats to water and air. However, wheat/maize rotations show strong retention of external N inputs that can remain available for use by subsequent crops.

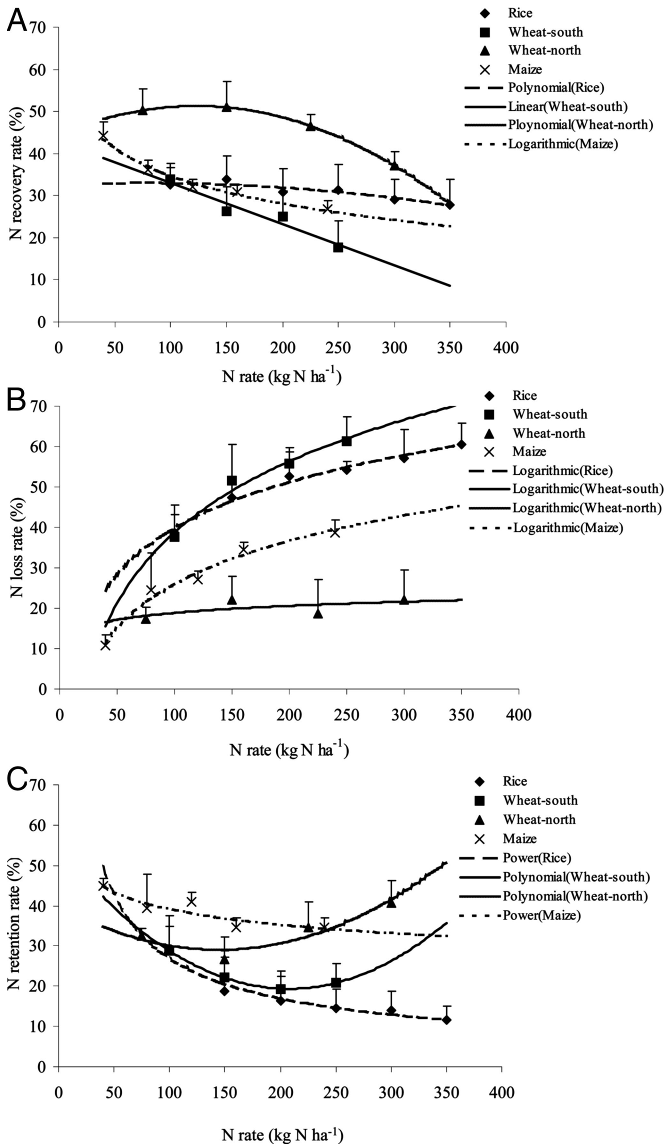

Fig. 1. Relationships of N recovery rate (*A*), loss rate (*B*), and retention rate (*C*) with N application rate in 4 crops (Field Study 2). Vertical bars denote standard deviation of the mean (average of 6 ^{15}N field experiments). Each model fitting produced a highly significant model ($P < 0.01$).

Retention of N is favorable for long-term improvement of soil N fertility and further emphasizes the need to reduce N application rates. It should be noted that N rates applied to wheat in the Taihu region are similarly high to those on the North China Plain, but wheat grain yields are much lower than in the north and show much lower N recovery rates (Table 1). One explanation is that farmers in the Taihu region try to compensate for the unfavorable growing conditions for wheat (e.g., extremely wet conditions in the root zone due to high rainfall and groundwater) by increasing N fertilizer application rates (20).

To further understand the different pathways of N loss from current N practices and to control them in the future, we conducted a lysimeter study (Lysimeter Study) in the Taihu region and a 4-year field experiment (Field Study 3) on the North China Plain, measuring in situ NH_3 volatilization, denitrification, N leaching, and N_2O emissions. We found that these N loss pathways differed sharply among the four crops because of differences in climate, soils and management practices (Table 2). Ammonia volatilization seems to be the main loss pathway for fertilizer N, with losses accounting for 19.4 and 24.7% of applied N in wheat (north) and maize seasons, respectively. Volatilization was much lower for rice and wheat (south), with losses accounting for 11.6 and 2.1% of applied N, respectively. Denitrification losses were quite small in the wheat and maize on the North China Plain (0.1 and 3.3% of applied N). In contrast, denitrification was the primary pathway for fertilizer N loss in rice and wheat within the Taihu region, accounting for 36.4 and 43.5% of applied N, respectively. Leaching accounted for only 0.3% of applied N from rice and 3.4% from wheat (south) season, but accounted for 2.7 and 12.1% of applied N for wheat and maize growing seasons on the North China Plain. We may be overestimating the denitrification loss in rice/wheat systems due to error accumulation with the different methods (5), but the order of magnitude agrees with previous studies (5).

Alternating upland wheat and waterlogged rice led to high denitrification of applied N and accumulated nitrate (after wheat harvest) in the rice season (20–22). The traditional practice of surface application of urea-N stimulated NH_3 volatilization from rice (23, 24). After plowing of paddy soil, a hard layer that formed below the plow layer due to clay deposition appeared to reduce leakage from the rice crop, but resulted in higher leaching losses during the wheat season when drainage prevails (25). The high denitrification loss for wheat (south) is attributed to wet soil conditions, higher temperatures and relatively high soil carbon (C) content in the Taihu region (26, 27). During the wheat season in the Taihu region, N fertilizer (urea) is readily nitrified and accumulates in the soil, consequently becoming available for loss through denitrification during the rice season (22). Thus the residual NO_3^--N in the soil profile is not fully available to the subsequent crop as is the case in upland crop rotations. We speculate that the very large N losses (corresponding with lower N retention rates) and large C inputs in straw under the anaerobic conditions in rice/wheat rotations may be another explanation for the structural changes in soil organic matter and increased C-rich humic acids found by Schmidt-Rohr *et al.* (28). On the North China Plain, with its calcareous soils, typical pH 7.5 to 8.5 and predominant use of

Table 2. Different N loss pathways expressed as a percentage (mean ± SD) of N application rate in farmers' N practices (Field Study 3, Lysimeter Study)

		Taihu region		North China Plain	
Component		Rice	Wheat-south	Wheat-north	Maize
N rate (kg of N per hectare)		300	250	325	263
Recovery rate (%)*		29.6 ± 4.9	18.4 ± 6.3	31.0 ± 3.6	25.5 ± 5.2
Retention rate (%)*		21.7 ± 5.1	28.5 ± 4.6	45.7 ± 5.4	33.9 ± 2.3
Loss pathway	NH_3 volatilization (%)	11.6 ± 4.7	2.1 ± 1.4	19.4 ± 5.2	24.7 ± 5.6
	Leaching out of 1 m soil depth (%)	0.3 ± 0.5	3.4 ± 2.1	2.7 ± 2.6	12.1 ± 8.5
	Denitrification (%)	36.4†	43.5†	0.1 ± 0.04	3.3 ± 1.6

*Measured from corresponding ^{15}N field experiments.
†Calculated by difference method.

urea or ammonium bicarbonate as fertilizer (accounting for 80%), NH_3 volatilization is generally considered to be the major pathway of N loss, especially in the hot maize growing season (29–31). Due to lower soil water content and soil organic matter (1.0–1.5%), most experimental results on the North China Plain have shown the magnitude of N loss by denitrification is no more than 10 kg of N per hectare and that denitrification accounts for less than 5% of the fertilizer N, most of which occurs during the hot and humid summer season (30, 32). We found that nitrate accumulation in the soil profile and gradual leaching below the root zone were the important N loss pathways in wheat and maize rotations under conventional agricultural N management practices due to concentrated rainfall in summer and flood irrigation (8, 33), contradicted conventional thinking that leaching losses are not an important pathway for N loss in semi-humid upland agricultural systems of the North China Plain.

Field Study 1 and the Lysimeter Study also showed that NH_3 volatilization increased rapidly and represented the major pathway for loss in response to increasing N rate, but N_2O emission and leaching did not increase dramatically with increasing N rate during the rice growing season (Fig. S1, rice). Leaching did increase rapidly and accounted for most of the reactive N loss with increasing N fertilizer rate, while N_2O emissions and NH_3 volatilization increased very little in response to N rate during the wheat season in the Taihu region (Fig. S1, wheat-south). Ammonia volatilization and NO_3-N leaching increased significantly with increasing N rate and N_2O emissions also showed an upward trend in the wheat growing season on the North China Plain (Fig. S1, wheat-north). During the maize growing season, NH_3 volatilization, NO_3-N leaching, and N_2O emissions all increased significantly, with N leaching showing a particularly strong increase (Fig. S1, maize). These results confirm that emissions of reactive N to the environment will increase after N fertilization exceeds the optimum rate. However, we did not find large N_2O emissions among the four crops. The total N_2O emissions in rice, wheat (south), wheat (north), and maize with different N application rates were 0.13 to 0.50, 0.27 to 0.63, 0.13 to 0.24, and 0.16 to 0.65 kg of N per hectare, respectively. Expressed as average emission factors, this accounted for 0.20, 0.30, 0.12, and 0.23%, respectively, close the IPCC 2006 guideline of 0.3% for paddy rice soil and much lower than the guideline of 1% for upland soil (27, 34, 35). We explain these results by the low ratio of N_2O to N_2O+N_2 from denitrification products (36, 37) under waterlogged rice conditions, low available carbon sources and low water-filled pore space (WFPS) for denitrification in most semi-humid upland soils on the North China Plain (14).

To quantify atmospheric and irrigation water contributions (hereafter 'environmental N inputs') to agricultural systems in the Taihu region and the North China Plain, we measured deposition and N inputs from irrigation water (Monitoring Study). Total environmental N inputs reached about 89 kg of N per hectare in the Taihu region and about 104 kg of N per hectare on the North China Plain. Comparisons with observations made from both regions in the 1980s shows more than 2-fold greater environmental N inputs with time. N deposition increased from 15 to 33 kg of N per hectare and irrigation water N inputs from 15 to 56 kg of N per hectare in Taihu region (Fig. S2) (38). During the same period, N deposition increased significantly from 21 to 89 kg of N per hectare and irrigation water N inputs from 8 to 15 kg of N per hectare (Fig. S2) (38, 39) on the North China Plain. Inorganic N and soluble organic N are the primary environmental N sources that can be used directly by crops or after transformation in the soil (40). The absence of crop yield response to N application in most field experiments since the 1990s (5, 8, 15, 16, 20) is not only attributable to increased indigenous soil N supply, but also to high environmental N inputs (41, 42). Moreover, high N deposition may also contribute to surface water eutrophication, acid rain, and soil acidification (43) in these regions. High total N concentrations (7 to 8 mg·N·L^{-1}) in the irrigation surface water in the Taihu region and high NO_3^--N concentrations in the irrigation shallow groundwater on the North China Plain also reflect the heavy N pollution of water resources in both areas.

We calculated annual N balance for both rotations using two scenarios: conventional N practice versus optimum N fertilization (Tables S1 and S2). We computed an annual N surplus of 87 kg of N per hectare for current practices with large losses by denitrification for the rice/wheat system. Synthetic N fertilizer was the primary input followed by N in irrigation water, biological N_2 fixation, and N deposition. A better N balance can be achieved by adopting optimum N fertilization strategies designed to maintain relatively high yields but reduce environmental risk. However, dinitrification could be further reduced by improving carbon management and controlling the water regime (25–27). We also computed a 212 kg of N per hectare surplus for current practice with large losses by NH_3 volatilization within the wheat/maize system. A large proportion of surplus N accumulated as nitrate in the soil profile after harvest (33), and partly existed as N in organic form due to manure application. However, the unusually large quantity of annual surplus N might also be caused by underestimating the leaching loss due to drought conditions in our observation years with 24–46% reduction in rainfall (8). Previous studies showed that strong N leaching losses only occurred in some years with heavy summer rainfall (33), leading to high nitrate accumulation in the deep subsoil (8, 33) and the groundwater (43). In the future, a slightly negative N balance could be achieved using an optimum N rate of about 286 kg of N per hectare to maintain relatively high yields (15, 16). The slightly negative balance would be conducive to making sure the plants fully use accumulated nitrate and further reduce nitrate leaching (8, 33). A high N retention rate is conducive to maintenance of soil N fertility. Changing the N application techniques and using deep placement of urea or ammonium bicarbonate could substantially reduce NH_3 volatilization losses from calcareous soils (29).

A question that needs to be addressed is why farmers in China continue to use such excessively high rates of N fertilizer. Before the 1990s, scientists, government and extension staff encouraged farmers to increase synthetic fertilizer inputs to increase yields and feed an increasing population. As a result, synthetic fertilizer inputs increased continually throughout the country, especially in the Taihu region and North China Plain. By 2000, the rates were far greater than crop demand and serious environmental degradation had begun. Although this trend has been recognized by the scientific community since the late 1990s, on-farm practices are difficult to reverse after 10 to 15 years of effort (43). Persuading farmers to limit fertilizer inputs is difficult because many of them still hold to now traditional opinions that higher crop yield will be obtained with more fertilizer. Application rates of N therefore often include an extra 'insurance' component to prevent yield loss rather than matching inputs to crop demand. The high off-farm incomes and relatively low retail prices for N fertilizers (with government subsidies for production and transportation) compared with U.S. and European prices are also important factors (43, 44). A third major reason is the poor infrastructure of the Extension Services and poor transfer of knowledge to farmers (43). A critical objective of the optimum N techniques developed using our studies are to substantially lower fertilizer N application rates by accounting for indigenous soil N and environmental N inputs for maintenance of the yields needed to feed an increasing population. This strategy also limits losses of total N to the water and atmosphere with the aim of establishing sustainable agricultural systems (8, 11, 12).

Crop production in intensive agricultural practice currently relies too heavily on synthetic N fertilizer inputs and cereal monocropping, especially in Asian countries under pressure to feed large and growing populations. It is now time to change this situation by balancing yield and environmental consequences. Integrated management packages need to be developed for major cropping systems that include efficient recycling of manures and crop residues, the use of legume crops in rotations to increase internal N cycling and further reduction in the reliance on synthetic N fertilizers (45).

Conclusions

It took nearly 50 years to achieve food sufficiency in China. An unanticipated cost has been that massive fertilizer inputs have led to significant environmental degradation (6). Over-fertilization is a serious problem in intensive agricultural production areas in China, resulting in enrichment of reactive N in the air, soil and water with consequent impairment of ecosystem services. Our studies show that more efficient use of N fertilizer can allow current N application rates to be reduced by 30 to 60%. This would still maintain crop yields and N balance in rotations, while substantially reducing N losses to the environment. The over-application of N also represents an unnecessary economic expenditure for farmers. The new recommendations should fully take into account the N supplying capacity of the soil and N deposited from air and irrigation water. The characteristics of N behavior among the 4 crops were sharply different depending on climatic, soil and management practices. This must be taken into consideration to further reduce N losses. Only by reducing fertilizer N inputs can degraded environments be gradually restored, enhanced and protected. Although several environmental standards have been set in the past, there are still no legislative controls in China equivalent to those in the European Union (33). China would benefit from adopting and enforcing relevant agricultural regulations (7). All these goals could be achieved by removing government subsidies, introducing an N fertilizer tax, improving local Extension Services, educating farmers for environmental awareness, and employing practices that avoid serious environmental degradation (43).

Materials and Methods

Study Areas. Two different representative intensive agricultural regions were selected as study areas: (1) the Taihu region in east China (30–32 °N, 119–122 °E) in the alluvial delta of the Yangtze River and (2) the North China Plain in northeast China (32–41 °N, 113–120 °E) in the alluvial plain of the Yellow River (Fig. S3). Details of the climate, soils, and crops are given in *SI Text*.

Field Study 1. In the Taihu region, 26 rice (8 or 9 locations each year) and 9 wheat (3 locations each year) on-farm field experiments with 7 (rice) or 5 (wheat) N rates were conducted from 2003 to 2006. The 7 rice N treatments were 0, 100, 150, 200, 250, 300, and 350 kg of N per hectare and the 5 wheat N treatments were 0, 100, 150, 200, and 250 kg of N per hectare. The plots (42 m^2 in area, 7×6 m) were arranged in a randomized complete block experimental design with 4 replicates.

On the North China Plain 6 winter wheat and 6 summer maize on-farm field experiments with 5 (winter wheat) or 6 (summer maize) N rates were conducted from October 2003 to October 2005 at Dongbeiwang near Beijing, Huimin County in Shandong province, and Baoding County in Hebei province. The 5 winter wheat N treatments were 0, 75, 150, 225, and 300 kg of N per hectare and the 6 summer maize N treatments were 0, 40, 80, 120, 160, and 240 kg of N per hectare. The plot size was 63 m^2 (9×7 m). The plots were arranged in a randomized complete block experimental design with 3 replicates. The fertilizer types and application times are given in the *SI Text*. Except for fertilizer application and grain harvest, each experimental field was managed using the individual farmer's current management practices in both rotations. Aboveground biomass was measured by hand in all plots. The central area of 16 m^2 (4×4 m) in each plot was harvested to determine grain dry matter yield.

Field Study 2. In the Taihu region, 6 rice (2 locations per year) and 6 wheat (2 locations per year) field experiments were selected to conduct ^{15}N studies in microplots to determine the fate of N fertilizer at different N levels. On the North China Plain, the ^{15}N study was carried out in microplots in the on-farm field experiments (*SI Text*). In both rotations plant and soil sampling and analysis for total N and ^{15}N abundance are described as in (44). The N recovery rate is expressed as the percentage of applied ^{15}N fertilizer taken up by the aboveground plant parts and the N retention rate as the percentage of applied ^{15}N fertilizer recovered in the top 100 cm of the soil profile. The loss rate was calculated by subtracting the recovery rate and retention rate from 100. The mean and standard variation was calculated across all of the experimental years and sites in same crop species.

Lysimeter Study. Twenty-four lysimeters containing undisturbed soil profiles were used at the research station at Changshu, Jiangsu province (31° 31.93′ N, 120° 41.88′ E) in the Taihu region. They contained a Typic Epiaquept (46) formed on alluvial loess with a silty clay loam (46) texture. A steel cylinder 1-m deep with 1.14-m inner diameter was pushed into the soil, cut at the base, and then removed. After collection, the lysimeters were prepared for free drainage by replacing approximately 0.08 m of soil at the base of each column with a nylon mesh, a gravel layer, and a porous plastic sheet (47). The lysimeters were then placed in plots permanently installed belowground at the station. Each lysimeter was used to grow a waterlogged summer rice/upland winter wheat rotation from October 2003 to October 2005 with 2 rice and 2 wheat growing seasons. Details of treatments are given in *SI Text*. Water leaching through the soil columns was collected in plastic containers placed in an underground measuring station. The leachate volume was measured at 3-day intervals and subsamples were taken from the accumulated leachate and stored at −18 °C before chemical analysis. During the winter wheat growing seasons, leachate could be collected only during rainfall periods. Leachate NO_3^-, NH_4^+, and total N concentrations were determined using a continuous flow analyzer (TRAACS 2000, Bran and Luebbe Inc.). The amount of N leached at each collection time was calculated by multiplying the volume and the N concentration of leachate. The total amount of N leached over the whole growing period was obtained by summing the individual amounts of N leached at the different collection times. The N leached from fertilizer was calculated as the difference in leached N between fertilized treatments and unfertilized controls. The mean and standard variation of each N treatment was calculated across 3 replicates over 2 years.

NH_3 volatilization and N_2O measurements were conducted in both rice and both wheat growing seasons from October 2003 to October 2005. Ammonia volatilization rate was measured with a continuous airflow enclosure method (48). The total amount of NH_3 volatilized over the whole growing period was obtained by summing NH_3 volatilization measured each day. N_2O fluxes were usually measured once a week throughout each crop growing period with static closed chambers, and measurements were made more frequently after basal fertilization and topdressing. Detail measurements and calculations are described in (49). Similar NH_3-volatilization and N_2O measurements at different N levels were conducted in the on-farm field experiment at Baoding County in Hebei province (Field Study 1). The amount of fertilizer derived NH_3 volatilization or N_2O emission was calculated as the difference between volatized NH_3 or N_2O in fertilized and unfertilized treatments. The mean and standard variation of each N treatment was calculated across 3 replicates over 2 years.

Field Study 3. A field experiment was conducted from September 2000 to September 2004 at Dongbeiwang research station near Beijing (40.08 °N, 116.28 °E) on a calcareous alluvial soil [Fluvaquents (46)] considered typical of the North China Plain. Eight successive crops (4 winter wheat and 4 summer maize) were grown on the same plots over the 4-year period. Detailed descriptions of the field experiment were reported previously (8).

^{15}N studies in microplots were also conducted in the conventional N and optimized N fertilization treatments from September 2002 to September 2004. NH_3 volatilization was measured from September 2002 to September 2004 with an automatic wind tunnel system that covered an experimental soil area of 0.7×1.5 m (1.05 m^2) (50). Field denitrification was monitored by a soil core incubation system using the C_2H_2-inhibition method (51) during the 2-year study from September 2002 to September 2004 (32). Nitrate leaching was quantified using TerrAquat passive samplers filled with ion-exchange resin. These were used to quantify the total annual amount of leached nitrate below the rooting zone with ten replicates per plot (52). The mean and standard variation of NH_3, N_2O, and nitrate leaching were calculated across 4 replicates over 2 years.

Monitoring Study. Measurements of wet and dry deposition were conducted from October 2003 to October 2005 in the 3 representative locations of the Taihu region, that is, Changshu, Nanjing, and Hangzhou. Three automatic wet-only samplers (APS series, Wuhan Tianhong Inc.), which collected precipitation samples only during precipitation events (controlled by sensors) were separately installed at the above 3 sites for the collection of wet-only deposition (53). Particulate dry N samples were collected using a 0.5-m^2 polyethylene sheet-based sampler (41). Before sunrise (about 6:00 AM in winter and around 5:00 AM in summer), 250 mL of 2 M KCl solution was used to collect deposited dew and the particles in the collector. In the present experiment, the N in dry deposition is defined as only the part of the inorganic N adsorbed to particles and exchangeable with 2 M KCl.

Precipitation samples were collected from 11 monitoring sites located on the North China Plain during the period 2003–2006. Bulk deposition (wet deposition plus sedimenting dry deposition such as dust during dry periods) was collected

AGRICULTURAL SCIENCES

(41, 53). The mean of the bulk deposition was calculated across all of the sites over the measuring years. Total deposition was determined by the ITNI system at Dongbeiwang, Beijing and Wuqiao and Quzhou in Hebei province. The ITNI system is based on ^{15}N isotope dilution and allows the inclusion of direct N uptake by the plant as well as the absorption of gaseous N compounds by the soil-plant system (40, 54, 55).

The N inputs from irrigation were calculated from the sum of the amounts of water in the periods of irrigation and the corresponding concentrations of ammonium and nitrate in the irrigation water. The measurements were conducted from October 2003 to October 2005 at 12 locations in the Taihu region and 10 locations on the North China Plain. The mean value was calculated across all of the years and sites.

Statistical Analysis. Most of the data are presented as mean ± SD (standard deviation) in the Tables and Figures. All data were analyzed using the SAS software package (SAS Institute). Means were compared using LSD (Least Significant Difference) by one-way ANOVA analysis. Regression analysis of N fertilizer rate vs. recovery rate, retention rate, and loss rate was performed using the PROC REG procedure.

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Supporting Information

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SI Text

Water pollution, especially eutrophication, has become an increasingly serious problem (1). A survey of eutrophication in the major lakes in China revealed that about 50% of the lakes investigated were eutrophic (2). As controls of point sources of pollution have been gradually introduced, non-point sources have become the major factor contributing to the eutrophication of Chinese lakes. Emissions of reactive N compounds to the atmosphere from agriculture, industry, and motor traffic exhausts all contribute to the eutrophication of water bodies after deposition (3). The annual N loading in the Yangtze River Delta has reached 3.39 million tons, with one-half of the inputs in the form of synthetic N fertilizers in 2002 (4). Currently, the annual application rate of synthetic N is about 550–600 kg of N per hectare in summer rice/winter wheat double-cropping systems in the Taihu region. Total N concentrations in major rivers in this region have already reached 7–8 mg of N per liter. On the North China Plain the annual application rate of synthetic N has also reached 550–600 kg of N per hectare in winter wheat and summer maize rotation systems, leading to a significant increase in nitrate concentrations in soil profiles and groundwater (5, 6). According to our measurements of soil nitrate after harvest in 47 winter wheat fields, 56 vegetable greenhouses, and 34 apple orchards in Huimin county, Shandong province in 2003, nitrate concentrations in the top 90 cm of the soil profile were 52–609, 270–5,038, and 32–2,406 kg of N per hectare, respectively. The corresponding ranges of values at 90–180-cm depth were 224–3,273, 68–1,047, and 228–2,430 kg of N per hectare. The amounts of nitrate in the 0–90- and 90–180-cm depth zones in vegetable greenhouse soils were significantly higher than in the other 2 cropping systems (7). Meta-analysis of 800 records in 120 publications in the literature showed that the average amount of nitrate in the top 100 cm of the soil profile of agricultural fields on the North China Plain has reached about 200 kg of N per hectare, the highest so far recorded among agricultural regions in China. A survey of groundwater nitrate-N concentrations conducted in the provinces of Beijing, Tianjin, Hebei, Shandong, and Shanxi showed that about 45% of 800 groundwater samples exceeded the WHO and European limit for nitrate in drinking water of 11.3 mg of N per liter (50 mg of NO_3^- per liter), with the highest nitrate-N concentration reaching 113 mg of N per liter (8, 9). The enrichment of N in soils, water, and air has brought about serious environmental problems in both regions.

Materials and Methods. ***Study areas.*** The first study area is in southern Jiangsu province at the juncture between the northern and central subtropical monsoon climatic zones. The average elevation is about 3–7 m above sea level (asl) and the mean annual number of sunshine hours is 1,800–2,200 h. The annual mean temperature is 15–17 °C and the annual cumulative mean temperature (days with mean temperatures over 10 °C) is 4,500–5,300 °C. The annual precipitation of 1,000–1,800 mm occurs mainly from May to September. The main soil type is Typic Epiaquept paddy soil (10). Current agricultural practice is a very intensive double-cropping system with waterlogged summer rice and upland winter wheat (or rapeseed) rotations. Rice is sown at the beginning of June and harvested at end of October, and then wheat is immediately sown and harvested at the end of May the following year. Application of organic manures (e.g., waterlogged compost) has been virtually abandoned, mainly due to high labor costs and rising incomes from off-farm activities. Production of winter green manures has been abandoned in favor of cash crops. Straw is partly returned to the fields. Mineral N fertilizer application rates are very high, leading to average annual surpluses of 217–335 kg of N per hectare (11). Due to the alternating water regime, transformation losses of N are high, resulting in low N recovery rates by crops of 28–41% (12).

The North China Plain includes the city of Tianjin, most parts of Beijing, Hebei, and Shandong provinces, and parts of Anhui, Jiangsu, and Henan provinces. It has a warm-temperate sub humid continental monsoon climate with cold winters and hot summers. Average elevation is around 20 to 40 m asl. The annual cumulative mean temperature for days with mean temperatures over 10 °C is 4,000–5,000 °C and the annual frost-free period is 175–220 days. Because of the abundance of solar radiation and relatively high temperatures, shortages of water and nutrients are the main factors limiting crop yields (13). The annual precipitation is 500–700 mm with 60–70% of the rainfall occurring during summer (July-September). The amount and distribution pattern of rainfall vary widely among years as affected by the continental monsoon climate. The calculated annual potential evaporation is close to 800 mm. The climatic water budget during the winter wheat growing period is almost always negative and during the summer maize season it is positive in some humid years. The soils are formed from alluvial loess transported to the North China Plain by the Yellow river and its tributaries. According to the soil classification of the World Reference Base for Soil Resources (WRB) of the FAO, most soils are calcaric Fluvisols or calcaric Cambisols with silt texture or Fluvaq-uents in the USDA (1994) classification (10). Soils are calcareous with a pH of 7.5–8.5 and organic matter content of around 1.0–1.5%. Current agricultural practice is a very intensive double-cropping system with irrigated winter wheat and rain-fed summer maize rotations. Winter wheat is sown at the beginning of October and harvested at beginning of June the following year and then summer maize is immediately sown and harvested at the end of September. Application of organic manures has declined in recent years, mainly due to high labor costs and rising incomes from off-farm activities. Straw is partly returned to the fields. Farmers in this region usually irrigate with large amounts of water and apply large amounts of N fertilizer to obtain high yields (6). These practices lead to substantial accumulation of nitrate in the soil profile (5, 7). The residual nitrate is readily leached down to deeper soil layers during the summer maize growing season during heavy rainfall, resulting in pollution of shallow groundwater bodies (8, 9).

Field study 1. In the rice growing seasons 50% of the N fertilizer was applied before transplanting and incorporated into the soil, 20% was broadcast before irrigation at the tillering stage, and 30% was broadcast before irrigation at the earing stage as urea. In the wheat growing season N fertilizer was applied before sowing (50% of total, incorporated into the soil) and at shouting stage (50% of total, broadcasted before rain). All plots received 60 kg of P_2O_5 per hectare as superphosphate and 60 kg of K_2O per hectare as potassium chloride before planting in each growing season based on soil P and K test results.

On the North China Plain, N fertilizer was applied before wheat planting (40% of total, incorporated into the soil) and at shooting stage (60% of total, broadcast before sprinkler irrigation) as urea in all experiments. Half of the N was band applied at the 3-extended-leaf stage (early July) and the remainder was top-dressed before rainfall or sprinkler irrigation at the 10-extended-leaf stage (early August) in summer maize. All plots received 90 kg of P_2O_5 per hectare as superphosphate and 60 kg of K_2O per hectare as potassium sulfate before wheat planting based on soil P and K test results.

Field study 2. In the Taihu region, the microplots (PVC cylinders 60-cm long with 50-cm inner diameter in the center of each main plot of the field experiments) were inserted into the soil to a depth of 40 m with a collar of 20-cm aboveground. On the North China Plain, the microplots (1×1 m, set up in the center of each main plot of the on-farm field experiments) were delineated with zinc-galvanized iron sheet to a depth of 0.35 m with an aboveground collar of 5 cm. All microplots were left unfertilized when the main plots received the N fertilizer treatments. Except for the N fertilizer using ^{15}N-labeled urea (abundance 10.32 atom%, produced by the Institute of Chemical Industry, Shanghai) and the conduct of all of the operations by hand, the microplots were managed in the same way as the main plots of the field experiments.

Lysimeter study. In the rice growing seasons, 12 of the lysimeters received 4 N levels, that is, 0, 100, 200, and 300 kg of N per hectare as ^{15}N-labeled urea, arranged in a randomized complete block design with 3 replicates. The remaining 12 were treated as controls with the local N rate to investigate the leaching of applied N during each rice season. During the wheat growing seasons the remaining 12 lysimeters received 4 N levels, that is, 0, 100, 200, and 250 kg of N per hectare as^{15}N-labeled urea, arranged in a randomized complete block with 3 replicates, and the 12 rice lysimeters received the local N rate to investigate the leaching of applied N in the wheat season.

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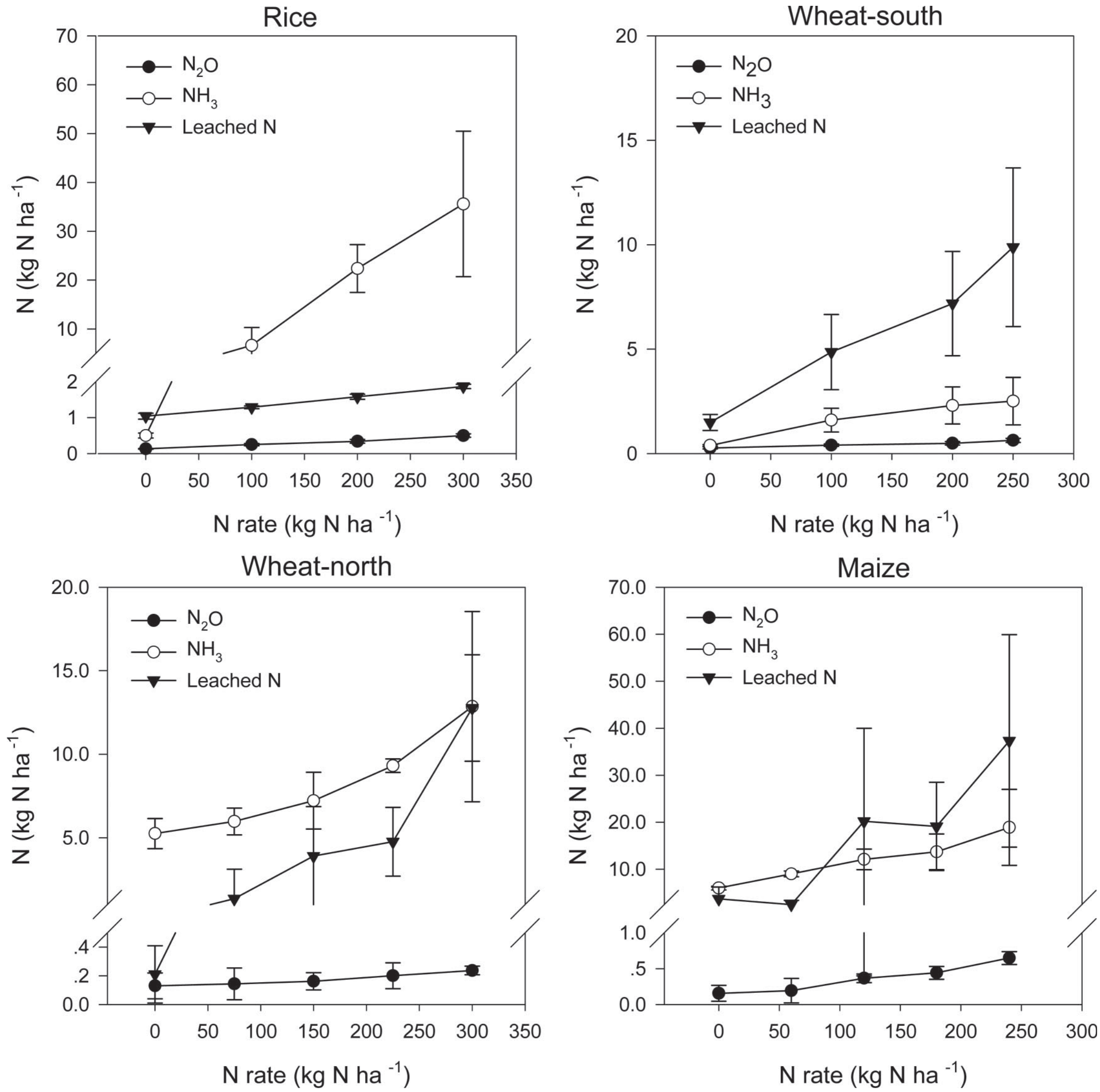

Fig. S1. Changes in NH_3 volatilization, N_2O emissions, and leached-N with different N rates in 4 crops (Field Study 1, Lysimeter Study). Vertical bars denote standard deviation of the mean.

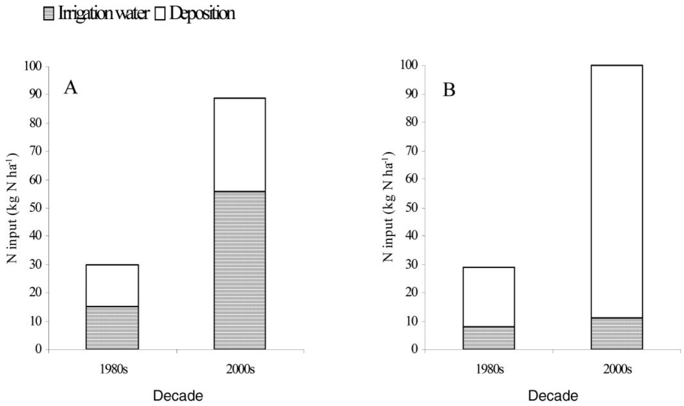

Fig. S2. N inputs from deposition and irrigation water in Taihu region (*A*) and the North China Plain (*B*) in the 1980s and 2000s. Data from the 1980s summarized from the literature (12, 14); data from the 2000s from the present study (Monitoring Study).

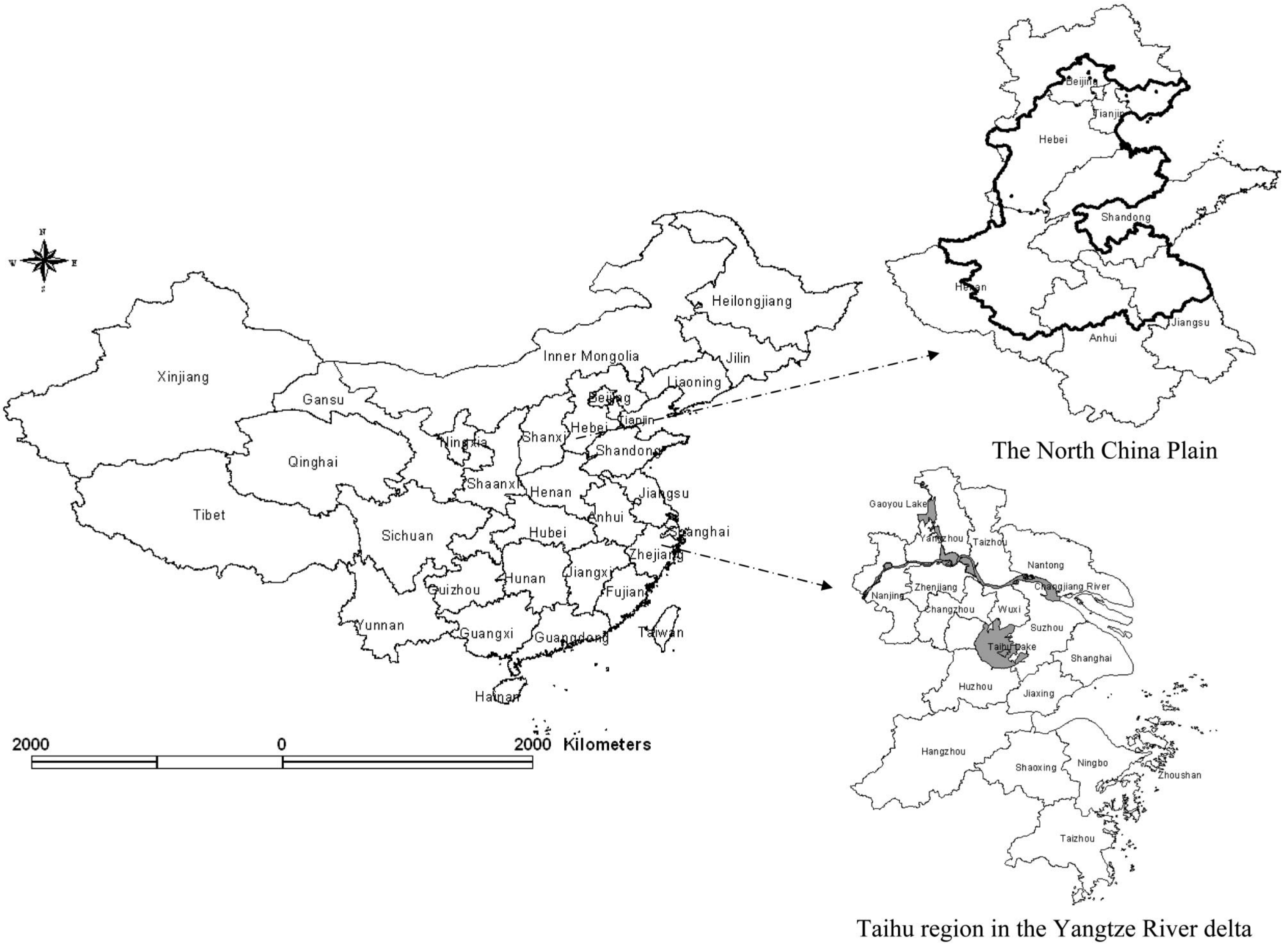

Fig. S3. Map showing the 2 study regions

Table S1. N balance of current farmers' N practices (FN) compared with the optimum N fertilization (ON) in future in rice/wheat rotation of the Taihu region (kg of N per hectare per year)

Input			Output		
Measured component	FN	ON	Measured component	FN	ON
Synthetic N fertilizer	550	353	Crop removal	341	324
Deposition	33	15*	NH_3 volatilization	38	24
Biological N fixation	45[†]	45[†]	Denitrification	206	75
Irrigation water	56	15*	Leaching loss	12	8
Seeds	4	4	Runoff loss	4[†]	4[†]
Total input	688 (A)	432 (A′)	Total output	601 (B)	435 (B′)
Balance	+87 (A-B)	−3 (A′-B′)			

*Assumes recovery to 1980's level.
[†]Data obtained by summarizing large numbers of published values according to previous studies (12, 15).

Table S2. N balance of current farmers' N practices (FN) compared with the optimum N fertilization (ON) in future in wheat/maize rotation on the North China Plain (kg of N per hectare per year)

Input			Output		
Measured component	FN	ON	Measured component	FN	ON
Synthetic N fertilizer	588	286	Crop removal	361	365
Manure	61*	61*	NH_3 volatilization	135	46
Deposition	89	21†	Denitrification	9	3
Non-symbiotic fixation	15*	15*	Leaching loss	56	23
Irrigation water	15	8†			
Seeds	5	5			
Total input	773 (A)	396 (A′)	Total output	561 (B)	437 (B′)
Balance	+212 (A-B)	−41 (A′-B′)			

*Data obtained by summarizing large numbers of published values according to previous studies (16).
†Assumes recovery to 1980's level.