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

Ju et al. www.pnas.org/cgi/content/short/0813417106

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 P₂O₅ per hectare as superphosphate and 60 kg of K₂O 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 10extended-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 ¹⁵N-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

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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 ¹⁵N-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 as¹⁵N-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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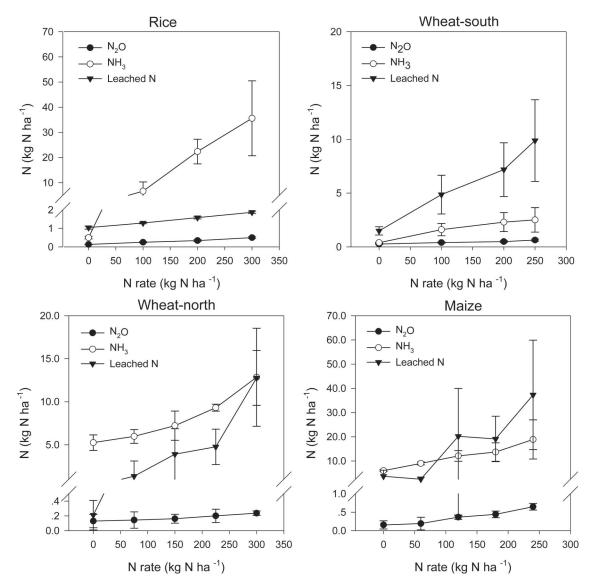


Fig. S1. Changes in NH₃ volatilization, N₂O emissions, and leached-N with different N rates in 4 crops (Field Study 1, Lysimeter Study). Vertical bars denote standard deviation of the mean.

Irrigation water Deposition

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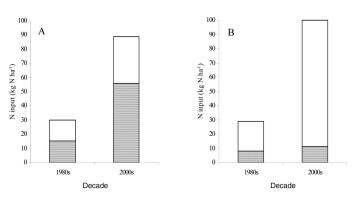
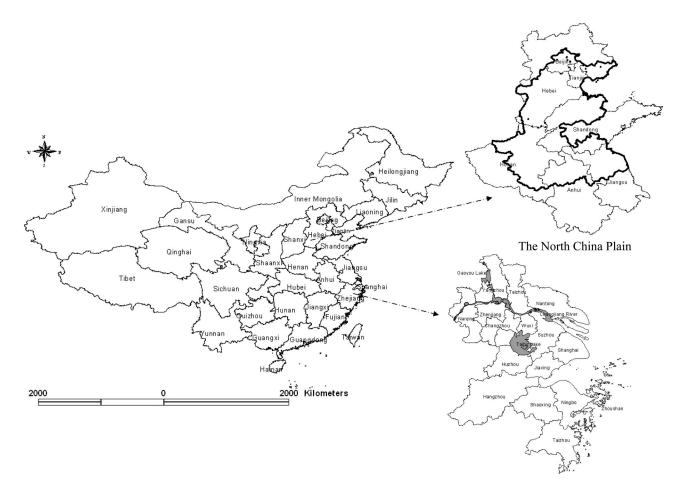


Fig. S2. N inputs from deposition and irrigation water in Taihu region (A) and the North China Plain (B) in the 1980s and 2000s. Data from the 1980s summarized from the literature (12, 14); data from the 2000s from the present study (Monitoring Study).



Taihu region in the Yangtze River delta

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.

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[†]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.

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