

Supporting Information

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SI Text

Uncertainty Analysis. To account for uncertainties associated with the data, we treated all the model inputs as random variables (i.e., variables with a probability distribution) and propagated the resulting uncertainties through the model using a Latin Hypercube sampling scheme (1). The average values of input/output fluxes of nitrogen described in the main text were all assumed to have a variation $\pm 10\%$. A uniform distribution function for all variables was used, and their respective uncertainties were propagated. The uncertainties are quantified as a coefficient of variation for each pixel.

Calculation of IN and OUT. IN_{fer} was calculated by using two sources of fertilizer data and the spatially explicit data on crop distribution from the SPAM (see details in section on SPAM and crop distribution maps). The first source of fertilizer data is the International Fertilizer Industry Association (IFA), which provides crop-specific nitrogen fertilizer consumption in 88 countries for one specific year ranging from 1995 to 2001 (2). The second source, the Food and Agriculture Organization (FAO) of the United States, provides total nitrogen fertilizer consumption in individual countries from 1961 to 2002 (3). We assume that the annual nitrogen fertilizer consumption by a certain crop is proportional to the total annual consumption from the FAO in individual years. Accordingly, when crop-specific nitrogen consumption in 2000 is not available from the IFA, it can be estimated based on the crop-specific nitrogen consumption in other years. It was assumed that mineral fertilizer was only applied in irrigated and high-input rain-fed systems. For each crop, the crop production share of each grid cell in the total national crop production under irrigated and high-input rain-fed systems was calculated. Because food production has a strong linear relation to nitrogen fertilizer consumption (3, 4), the crop-specific nitrogen fertilizer consumption in a grid cell was calculated by multiplying total national nitrogen fertilizer consumption by the production share of the grid cell. The crop-specific nitrogen fertilizer application rate in a grid cell was calculated by dividing the crop-specific nitrogen fertilizer consumption by the harvest area of the crop under all systems. For the countries that are not included in the IFA dataset, national nitrogen fertilizer consumption from the FAO was evenly allocated to the total harvested area of croplands. This rough estimation will not lead to large errors because the countries not covered by the IFA only accounted for around 6% of the total world fertilizer application (3).

IN_{man} was calculated by multiplying livestock density with animal-specific excretion rates and excretion collection rates. The livestock density data for the year 2000 were obtained from the Gridded Livestock of the World (GLW) from the FAO. The GLW describes the spatial distribution of cattle, buffaloes, sheep, goats, pigs, and poultry at a resolution of 3 arc-minutes. The data were first converted into 5 arc-minutes. The density maps of other livestock, such as camels, mules, horses, and rabbits, are not available, but these livestock types only account for about 3% of the live animal stock and 0.5% of the global meat production (3). The omission of these types will only slightly influence the manure supply. Similarly, wild animal excretion on cropland was considered to be very small (5), and was therefore not considered in this analysis. To calculate the nitrogen excretion rates of various livestock types in individual countries, we followed Sheldrick et al. (6) and assumed that excretion rates within a given livestock category are proportional to the slaughter weights. Sheldrick et al. (6) provide baseline data on livestock weight and livestock nitrogen excretion rates for cattle, pigs, sheep, goats, and poultry with certain slaughter weights (50 kg of N head⁻¹.yr⁻¹ and

250 kg·head⁻¹ for cattle, 12 kg of N head⁻¹.yr⁻¹ and 80 kg·head⁻¹ for pigs, 10 kg of N head⁻¹.yr⁻¹ and 15 kg·head⁻¹ for sheep, 10 kg of N head⁻¹.yr⁻¹ and 12 kg·head⁻¹ for goats, and 0.6 kg of N head⁻¹.yr⁻¹ and 2 kg·head⁻¹ for poultry). The slaughter weights of the various livestock types in individual countries were calculated as the sums of the body weights of carcass, edible offal, slaughter fats, hides, and skins, which were obtained from the FAOSTAT (3). Excreta can be produced in either stables or meadows. Excreta produced in a stable is more efficiently collected and stored for manure application, whereas only a small part of livestock excreta from a meadow is recovered and used elsewhere (7). Here, we only consider the excreta produced in a stable for manure supply. According to Bouwman et al. (8), the shares of the nitrogen excretion produced in stables compared with total nitrogen excretion are 54% and 46% for cattle in developed and developing countries, respectively (average of dairy cattle and nondairy cattle), 33% for buffaloes, 10% for sheep and goats, and 100% for pigs and poultry. There are also losses (mainly volatilization of NH₃) during excretion, collection, and storage of excreta. The losses were calculated by multiplying the nitrogen excretion by specific volatilization rates of different livestock types as reported by Bouwman et al. (8): 36% for cattle, pigs, and poultry and 28% for buffaloes, sheep, and goats.

Allocations of manure between cropland and pasture are more relevant for developed countries, particularly for Europe and North America. According to the results of a questionnaire completed by 140 experts in 21 European countries and Canada (9), on average, 66% of the solid manure is used for cropland, whereas the remainder is used for pasture application. The shares range among countries [e.g., 50% in Switzerland and The Netherlands, 70–90% in United Kingdom (70% for manure from cattle and 90% for manure from pigs and poultry), 80% in Finland and Sweden]. When precise data were available, they were used for calculations. Otherwise, the average share of 66% is applied. For the United States, Kellogg et al. (10) calculated the nitrogen flows from manure that potentially end up in 24 crops and pasture for each state. This calculation shows that, at the national level, the capacity of cropland to assimilate nutrients is 6.7 times higher than that of pastureland. Using these results, around 87% of manure goes to cropland and the remainder goes to pasture. The shares to cropland range greatly, with high values (e.g., >90%) in the Lake States and Corn Belt and low values (e.g., <60%) in the Southern Plains. The shares in each state are used in this study. In developing countries, manure produced in stables is mainly used in cropland and is rarely used in pasture. Almost all the manure is applied to cropland. Following Smil (11), 90% of the manure is assumed to be recycled eventually to the crops studied here, whereas the remainder is used for other fodder crops.

IN_{dep} was based on modeled estimates of total wet and dry mineral (NO_y + NH_x) deposition from Dentener (12). Dentener (12) estimates the nitrogen deposition for the years 1960, 1993, and 2050. In this paper, we used the nitrogen deposition in 1993. The original data with a resolution of 0.5° were converted to a resolution of 5 arc-minutes.

IN_{fix} mainly occurs in two ways: symbiotic IN_{fix} by leguminous crops and nonsymbiotic IN_{fix} by Cyanobacteria. The mean fixation rates for the predominant nitrogen fixing crops were taken from Smil (11). His estimates are 80 kg of N per hectare for soybeans and groundnuts, 40 kg of N per hectare for beans, 60 kg of N per hectare for other pulses, and 100 kg of N per hectare for sugar cane. Cyanobacteria in irrigated rice fields can fix 20–30 kg of N per hectare during the growing season (11). Here, we used an average of 25 kg of N per hectare. Minor contributors of

biological fixation are cereals (except for irrigated rice), tubers, and oil crops (11). Typical rates are mostly less than 5 kg of N per hectare in cereal fields in dry environments, but some studies report values over 20 kg of N per hectare in humid environments (11). Here, the rates were assumed to be 12 kg of N per hectare.

IN_{sed} consists of two parts: nitrogen input in irrigation and nitrogen input in sediment as a result of erosion. The nitrogen input in sediment is calculated combined with soil output to soil erosion. The nitrogen input in irrigation water is calculated for irrigated cropland by multiplying the nitrogen content of irrigation water (in kilograms of N per cubic meter) by the irrigation application rate (in $m^3 \cdot ha^{-1} \cdot yr^{-1}$). We used an average nitrogen content of irrigation water of $3.3 \times 10^{-3} \text{ kg} \cdot m^{-3}$, after Lesschen et al. (13). The irrigation application rates for all crops under irrigated conditions were simulated with a GEPIC (GIS-based Environmental Policy Integrated Climate) model (14).

IN_{res} was calculated by multiplying OUT_{res} with a removal factor (γ) to account for the ratio of the residues removed from the field to the total crop residues ($IN_{res} = (1 - \gamma) \times OUT_{res}$). Part of the crop residues is removed from cropland and used, for example, as biofuel or for animal feeding. No country keeps comprehensive statistics of crop residue uses; hence, removal factors are commonly not available. The removal factors for various crops in Ghana, Kenya, and Mali were collected from the FAO (15). The removal factors in other African countries were adapted from data provided by Herrero et al. (16), which indicated removal factors of 11 crops (wheat, rice, maize, sorghum, millet, barley, soybeans, potato, sweet potato, cowpea, and groundnut) in East Africa, Southern Africa, Central and West Africa, and North Africa. Removal factors of 18 crops or crop groups in India were calculated based on the reported quantity of agricultural residue uses from Ravindranath et al. (17). The average removal factor (83%) of the 18 crops was used as the removal factors of crops in India not covered by Ravindranath et al. (17). Removal factors of 11 crops in South Asia, West Asia, Southeast Asia, East and Central Asia, Latin America, and Europe were adapted from data provided by Herrero et al. (16). For 48 states of the United States, Graham et al. (18) quantified the amount of maize stover and collectable stover. The removal factor for maize was calculated as the ratio of collectable stover to the total corn stover. Corn production in other states was marginal. Removal factors of wheat in 37 states of the United States were calculated based on data provided by Nelson (19), who quantified the annual quantity of crop residues and removable residues for 1997. Removal factors of wheat in other states were taken as the average removal factor in the 37 states. Low-residue crops, such as soybean, rarely produce enough residues to maintain adequate soil cover (20). In addition, the conservation tillage rate is high for soybean, and soybean residues have almost no alternative use in the United States. Hence, it was assumed that no residues are removed from soybean fields. For other crops in the United States, it was assumed that 30% of the residues were removed from the field according to Smil (11). Because maize, wheat, and soybean accounted for 75% of the total crop residues produced (21), this simplified assumption will not lead to large errors. For all other countries, data on removal factors are not available. It was assumed that 35% and 50% of the crop residues were removed in developed and developing countries, respectively (11).

OUT_{crop} was calculated by multiplying the dry crop yield by the nutrient content of the crops. The crop fresh yield for various crops in different systems was obtained from the spatial allocation model. Moisture content of various crops was obtained from Milbrant (22). The nitrogen contents of different crops were obtained from the FAO (15). OUT_{res} was calculated by multiplying the dry yield of the crop residue by the nutrient content of the crop residue. Dry crop residue yield was calculated by multiplying dry crop yield with a residue-to-product ratio (RPR). The RPR values of barley, maize, cotton, beans, groundnuts, rice, sorghum, soybeans, sugar cane, and

wheat were obtained from Milbrant (22). The RPR values of millet, cassava, and coffee were obtained from Koopmans and Koppejan (23). The RPR values of other pulses, other fibers, and other oil crops were assumed to be the same as those of beans, cotton, and soybeans, respectively. For all other crops, the RPR was calculated from a harvest index (HI) as $(1 - HI)/HI$ (24). The HI values of the crops were obtained from Smil (24).

OUT_{lea} was calculated with a regression model (Eq. S1) developed by De Willigen (25), which has been used by the FAO (26, 27) and many other researchers [e.g., Smaling et al. (28), Haileslassie et al. (29), Lesschen et al. (13), Marenya and Barrett (30) Haileslassie et al. (31)]

$$OUT_{lea} = (0.0463 + 0.0037 \times (P/(C \times L))) \times (F + \gamma \times D - U), \quad [S1]$$

where P is the annual precipitation in $mm \cdot yr^{-1}$, C is clay content in %, L is layer thickness or rooting depth in meters, F is mineral and manure fertilizer nitrogen in kilograms of $N \text{ ha}^{-1} \cdot yr^{-1}$ and is equal to the sum of IN_1 and IN_2 , γ is the decomposition rate of manure matter in % per year, D is soil nitrogen density in kilograms of N per hectare, and U is uptake by crop in kilograms of $N \text{ ha}^{-1} \cdot yr^{-1}$. D is calculated by multiplying bulk density (BD, in $kg \cdot m^{-3}$), total nitrogen in soil (TOTN, in $kg \cdot kg^{-1}$), and layer thickness L . The data on C , BD, and TOTN were obtained from the derived soil properties on a 5×5 arc-minute global grid (version 1.0) from the World Inventory of Soil Emission Potentials database (32) (<http://www.isric.org>). The $0.5^\circ P$ data in 2000 were obtained from the Climate Research Unit of the University of East Anglia (version CRU TS 2.0) (<http://www.cru.uea.ac.uk>) and were converted into 5 arc-minute resolution data. Values of L for individual crops were obtained from the FAO (15). γ was assumed as 1.6% per year according to the FAO (15).

The first part of the regression equation determines the fraction of mobile nitrogen leached, and the second part determines the quantity of mobile nitrogen available (28). Although this model is based on an extensive literature review and is valid for a wide range of soil and climate conditions (25), it may give unreasonably high values when extremely high precipitation occurs. Here, we set an upper limit as $(0.2 IN_{fer} + 0.5 IN_{man})$ for OUT_{lea} . According to a survey of 40 agroecosystems in three continents, nitrogen losses in terms of leaching rarely exceed 20% of the total mineral fertilizer application (11). Losses exceeding 50% of nitrogen from manure application are also rarely reported in the literature (11).

Both nitrification and denitrification remove soil nitrogen as N_2 , NO , and N_2O (11). Volatilization of NH_3 is responsible for large nitrogen losses from both animal manure and all ammonia fertilizers (11). Stehfest and Bouwman (33) summarized information from 1,008 N_2O and 189 NO emission measurements from agricultural fields around the world and reported the factors that can significantly influence the emissions of N_2O and NO . For N_2O , the factors included nitrogen application rate, crop type, fertilizer type, soil organic carbon content, soil pH, and texture, whereas for NO , they included nitrogen application rate, soil nitrogen content, and climate. These investigators also developed statistical models explicitly considering these factors to simulate global annual emissions of N_2O and NO in cropland with high spatial resolution. In our paper, crops were classified into three groups (i.e., rice, legumes, other crops) and emissions of N_2O and NO were simulated with the same statistical models from Stehfest and Bouwman (33). Bouwman et al. (34) summarized information from 1,667 NH_3 volatilization measurements from around the world documented in 148 research papers to assess the influence of different influencing factors (e.g., crop type, fertilizer type, fertilizer application rate, soil organic carbon, texture, pH) on NH_3 volatilization. Statistical models were developed for NH_3 volatilization of nitrogen fertilizer and manure in lowland rice and upland crops. In this paper, crops were classified into two groups (i.e., rice, other

crops) to estimate NH_3 volatilization with the same statistical models from Bouwman et al. (34).

In Eq. S2, OUT_{ero} was calculated as

$$\text{OUT}_{\text{ero}} = E \times \text{TOTN} \times (1 - \alpha), \quad [\text{S2}]$$

where E is soil erosion in $\text{kg} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$, TOTN is total nitrogen in soil in kilogram of N per kilogram, and α is a redeposition coefficient. At least a quarter of the eroded soil is redeposited on adjacent cropland or on more distant alluvia (11). The term α is assumed to be 0.25 here. The 5 arc-minute erosion data were converted from Ito (35), who estimated soil erosion with a spatial resolution of 30 arc-minutes with a widely used revised universal soil loss equation (RUSLE) (36).

SPAM and Crop Distribution Maps. Nutrient balance in cropland is closely related to crop management practice; hence, crop distribution maps indicating different management patterns are the basis for spatially explicit assessment in nitrogen flows. The latest global rain-fed and irrigated crop distribution data (version 2000/3.0) are used here, which are generated using the SPAM (37, 38).

The SPAM applies a cross-entropy approach to make plausible allocations of crop production in geopolitical units (country or state) into individual pixels through judicious interpretation of all accessible evidence, such as production statistics, farming systems, satellite images, crop biophysical suitability, crop prices, local market access, and prior knowledge (37). A more detailed description and applications of the model can be found in the literature (37–39).

The SPAM provides estimates of harvested areas, crop production, and crop yield with a spatial resolution of 5 arc-minutes for 20 crops or crop groups that, together, account for almost 90% of the world's total harvest area. These crops include six cereal crops (wheat, rice, maize, barley, millet, and sorghum), three roots and tubers (potatoes, sweet potatoes, and cassava and yams), two pulse crops (dry beans and other pulses), two sugar crops (sugar cane and sugar beet), three fiber crops (coffee, cotton, and other fibers), three oil crops (soybeans, groundnuts, and other oil crops), and one fruit (plantain and banana). The SPAM results include harvested area and production of three systems for each crop: irrigated, high-input/commercial rain-fed, and low-input/subsistence rain-fed. The complete SPAM datasets have been released to the public and can be downloaded from a dedicated Web site (www.mapSPAM.info).

High-resolution data for other cereals, except for the six cereal crops, were not simulated with the spatial distribution model mainly because of insufficient information for these crops. It was assumed that the harvested area of other cereal crops is proportional to the total harvested area of the six cereal crops in all grid cells within a country. A similar assumption was used for crop production of the other cereals. Because the other cereals only accounted for about 2% of the total arable area, it is believed that the assumptions will not lead to large errors for the global nitrogen assessment.

Comparison with Other Studies. Global consumption of mineral fertilizers is the most accurately known input from statistical data. In 2000, the actual synthesis of nitrogenous fertilizer was 86 Tg of N per year, of which almost 81 Tg of N per year was consumed and the remainder was lost during processing and transportation in chemical industries (3). Most mineral fertilizers were used on arable land, but in some regions, such as the countries of Western Europe and Oceania, a significant amount of fertilizers was applied in pastures. The calculation showed that the arable cropland accounted for about 84% of the global consumption of the nitrogen fertilizers (i.e., 67.84 Tg of N per year). This estimate was very close to that (i.e., 72 Tg of N per year in the mid-1990s) reported by Smil (11). Sheldrick et al. (40) reported a higher value largely

because the mineral fertilizers were assumed to be completely used in cropland.

Our results show good agreement with the reported values of IN_{fer} , IN_{man} , IN_{fix} , and IN_{sed} by Smil (11) (Table S1). IN_{dep} calculated here is almost 20% lower than that calculated by Smil. In our study, IN_{dep} is estimated with 0.5° resolution data on atmospheric deposition and with 5 arc-minute resolution data on crop distribution. The atmospheric deposition data were generated from a global transport–chemistry model with high spatial resolution (41), and they are more refined than those used by Smil (11). IN_{fix} in both this study and that by Smil (11) is much higher than that reported by Sheldrick et al. (40), in which the contribution of many crops, except for soybean and pulses, is not taken into consideration (e.g., irrigated rice, peanuts). The results of IN_{man} in both of the studies are much lower than that in the study by Sheldrick et al. (40), partly because Sheldrick et al. (40) assume that all the excreta generated by animals is applied in cropland.

Our OUT of 148.14 Tg of N per year was also very close to the mean estimate of Smil (11) after adjustment (Table S1). The nitrogen balance in cropland was -11.53 Tg of N per year based on our calculation. Sheldrick et al. (40) gave a higher value of negative soil balance (-18.30 Tg of N per year) partly because of the very high estimates of OUT_{crop} and OUT_{res} . Sheldrick et al. (40) calculated OUT_{crop} by multiplying statistical crop production by nitrogen content in crops. However, nitrogen content of crops is commonly expressed in terms of dry weight, whereas crop production is generally reported in terms of wet weight. Appropriate conversions must be made first with moisture content in crops (11). Similarly, the dry weight of crop residues is needed for the calculation of OUT_{res} . Ignoring moisture content can lead to considerable errors. For example, according to Smil (11), the total fresh weight of harvested crops (excluding forages) was 5,450 Tg in the mid-1990s, whereas the total dry weight was 2,750 Tg, only half of the fresh weight. In contrast, Smil (11) reported a positive balance of 7.0 Tg of N per year (Table S1). Our estimate resulted in higher nitrogen outputs to leaching, gaseous losses, and erosion than the reported maximum values from Smil (11).

The global NRR was estimated to be 59%. This estimate is close to the upper limit of the estimate (i.e., 58%) by Smil (11) but higher than the mean estimate of 50%. The NRR from both this study and the study by Smil (11) is smaller than the NRR of 64% reported by Sheldrick et al. (40), although higher than that of 43% reported by Bouwman et al. (42). The overestimation of OUT_{crop} and OUT_{res} by Sheldrick et al. (40) is one important reason for their much higher NRR. Bouwman et al. (42) estimated a low NRR because they did not include leguminous crops, which generally have a higher NRR than other crops.

For nitrogen outputs, OUT_{crop} from our estimate is almost identical to the estimate by Smil (51.65 vs. 50 Tg of N per year) (11), although the estimate of OUT_{res} is 17% higher than that of Smil (11). Both our estimates and those of Smil (11) for OUT_{crop} and OUT_{res} are much lower than the estimates of Sheldrick et al. (40), largely because the latter ignored moisture content of crops and residues, as stated before. Our estimate of OUT_{lea} (22.99 Tg of N per year) is a quarter higher than the maximum estimate by Smil (11). Lin et al. (43) estimated global nitrate leaching of 26.22 Tg of N per year in terrestrial ecosystems. This is much larger than our estimate because it included nitrate leaching not only in cropland but in pastureland. For OUT_{gas} , our estimate is close to the lower limit of the calculation by Smil (11). There are a few other studies available for the estimation of nitrogen gaseous losses. Bouwman et al. (34) estimated global emissions of NH_3 of 10.98 Tg of N per year in cropland for the year around 1995. Stehfest and Bouwman (33) calculated the global emissions of N_2O and NO of 4.65 Tg of N per year in cropland. Based on the above two sources, the IFA/FAO (44) reported a global estimate of gaseous emissions of 15.63 Tg of N per year in cropland for the year around 1995. This estimate was 23% lower

than our estimate and 48% lower than the average estimate of Smil (11). The difference stems from several reasons. For example, fertilizer consumption in 1995 was 2% lower than that in 2000, but the harvested area was 2% higher for cereals and 8% higher for pulses (45). Hence, the fertilizer use in each unit of harvested area was lower in 1995. This may lead to higher NRRs in cropland, and hence lower nitrogen losses.

Limitations of This Study. This paper provides an encouraging and reasonable approximation for global nitrogen flows in cropland with a high spatial resolution. Nonetheless, several limitations in our methodology and results still remain. First, our methods and results are data-constrained. The results may be largely influenced by the lack of several spatially explicit data, including crop-specific fertilizer, volatilization rates, removal factor, and crop-specific IN_{fix} . The uncertainty analysis shows that the results are very sensitive, particularly to IN_{fer} (Fig. S3). This also indicates the importance of collecting crop-specific fertilizer data. Second, apart from the unavailability of data, we regard the quality of the data used in this paper as a secondary influence. For instance, the data on atmospheric deposition and soil erosion are not from direct measurement but from simulated results by other models, and the accuracy of the data remains unclear at the grid cell level. Third, a regression equation is used to calculate nitrate leaching in which clay content in the soil is one of the major controlling factors. Although this equation is based on an extensive literature search and is valid for a wide range of soils and climates

(26), an extrapolation of it to a global scale study may lead to simulation errors. Particularly, many middle- to high-clay soils in the developed world are often artificially drained. In such tile-drained systems, leaching is very high (e.g., in the upper Midwest Corn Belt of the United States) and the regression model may underestimate nitrate leaching substantially. The leaching of nitrate is caused by many factors, including geological, hydrological, and plant–soil processes as well as management. It is still too complicated a process to be accurately modeled by any mathematical model (46). Fourth, emission of N_2 can be an important loss of nitrogen from agroecosystems. However, the few available studies have limited comprehensive measurements on the relation between N_2 emission and its driving forces. It is still difficult to assess the amount and its spatial distribution (47). Without considering correctly the dinitrogen component, our estimate is conservative for the estimation of nitrogen gaseous losses as well as total nitrogen output (OUT). According to Schlesinger (47), the mean ratio $N_2O/(N_2O + N_2)$ is about 0.375 in agricultural soils. Taking this ratio into account, our OUT is underestimated by about 4%. Last but not least, an unequivocal validation of our results is difficult because this study provides a previously undescribed comprehensive assessment of global nitrogen flows on a global scale. Further improvements are only possible once improved spatial statistics become available. Given the current focus on international data collection at the level of individual countries, progress is likely to take decades rather than years.

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