Supporting Information for

Global grey water footprint and water pollution levels related to anthropogenic nitrogen loads to fresh water

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1. Method and data

1.1. Diffuse sources of nitrogen

We compute annual soil nutrient balances based on nitrogen (N) inputs and outputs at a spatial resolution of 5×5 arc-minute. We considered four inputs: application of artificial fertilizer (IN_{fer}) and animal manure (IN_{man}), wet and dry atmospheric deposition (IN_{dep}), and biological N fixation (IN_{fix}). The outputs in the N balance include N withdrawal from the field through crop harvesting (OUT_{harv}), N output through taking away crop residues (OUT_{res}) and gaseous losses (OUT_{gas}).

Inputs from mineral fertilizers (IN_{fer})

The fertilizer application rate per crop per country was calculated using three sources of fertilizer data and the spatially explicit data on crop distribution from Monfreda et al. [1]. IFA et al. [2] provide fertilizer application rates per crop for 88 countries. FAO [3] and Heffer [4] were used to complement data for crops and countries missing from the IFA et al. [2] data. Since the application rates provided in these data sources is for different years, these were adjusted to fit FAO [5] country average nutrient fertilizer consumption per year for the period 2002-2009.

Inputs from animal manure (IN_{man})

The manure input was calculated at grid cell level by multiplying livestock density by the animal-specific excretion rates. The volume of manure actually applied on cropland was estimated by accounting for the collection rate and the allocation of collected manure over croplands versus pasture. We considered manure inputs on croplands (including managed grasslands), but did not further study manure inputs on grazing lands.

Total manure N production within the grazing, mixed and industrial animal production systems for the major livestock categories (cattle, buffaloes, sheep, goats, pigs and poultry) was calculated by multiplying the spatially explicit global livestock density with animal-specific excretion rates and then adjusted for the fraction of manure available for cropland and grassland application [6-8].

The manure N production per production system per animal category per grid cell (M_{exc}, kg N/ha) was calculated as follows:

$$M_{exc}[a,s] = D[a,s] \times E[a,c,s] \quad (1)$$

where *D[a, s]* is the density of animal category *a* for production system *s* (head/ha) and *E[a,c, s]* the nutrient excretion rate of animal category *a* in country *c* and production system *s* (kg N/head).

To calculate the manure (N) excretion rate per animal category per country we followed the approach of Liu et al. [7]. Sheldrick et al. [9] provide data on animal manure excretion rates for cattle, pigs, sheep, goats, and poultry relative to the animals slaughter weight. The manure excretion rate per animal category, production system and country was calculated by combining Sheldrick et al. [9] global average manure excretion rates with slaughter weight of animals per production systems and per country:

$$E[a,c,s] = \frac{SW[a,c,s]}{SW_{shel}[a]} \times E_{shel}[a] \qquad (2)$$

where *SW[a,c,s]* is the slaughter weight of animal category *a* (kg/head) in country *c* and production system *s*, $SW_{shel}[a]$ the global average slaughter weight of animal category *a* (kg/head) and *E[a]* the global average manure excretion by animal category *a* (kg/y), both obtained from Sheldrick et al. [9]. The slaughter weights (*SW[a,c,s]*) of the different animal category per production systems per country was obtained from Mekonnen and Hoekstra [10].

We can distinguish three types of manure within each production system and country [6, 8]: (a) manure produced from animals housed in stables, (b) manure produced from livestock grazing on pasture or rangeland, and (c) manure excreted for example in urban areas, forests and along roadsides and manure used as fuel or for other purposes that are considered to fall outside the agricultural system:

$$M_{exc}[a,s] = M_{stor}[a,s] + M_{graz}[a,s] + M_{out}[a,s] \qquad (3)$$

where $M_{stor}[a, s]$ is the volume of manure of animal category *a* for production system *s* collected in storage (kg N/ha), $M_{graz}[a, s]$ the volume of manure of animal category *a* for production system *s* that is produced during grazing (kg N/ha) and $M_{out}[a, s]$ the volume of manure of animal category *a* for production system *s* that falls outside the agricultural systems (kg N/ha). The fractions of manure that are produced during grazing and the fractions of manure that are not available for spreading on crop- and grasslands for the different animal categories and production systems were obtained from Bouwman et al. [8].

Not all animal excreta is available as manure to be applied on crops and grassland. The fraction of manure that is produced and available for application on crop- and grasslands depends on a number of factors such as the degree of animal confinement or pasture grazing, cost of transport and agricultural practices [11]. Some manure is also lost during excretion, collection and storage through ammonia (NH_3) volatilization. Therefore, the quantity of manure actually applied on crops and managed grassland (IN_{man}, kg N/ha) is:

$$IN_{man}[a,s] = M_{stor}[a,s] - N_{vol,stor}[a,s] \qquad (4)$$

where $N_{vol,stor}[a, s]$ represents ammonia volatilization from animal housing and storage for animal category *a* and production system *s* (kg/ha), calculated as follows:

$$N_{vol,stor}[a,s] = N_{stor}[a,s] \times \beta[a,s] \qquad (5)$$

where $N_{stor}[a,s]$ is the quantity of N manure of animal category *a* (kg/ha) in production system *s* in animal housing and storage and $\beta[a,s]$ the ammonia volatilization rate of animal category *a* and production system *s* (%). According to Bouwman et al. [12], the volatilization rate for cattle, pigs and poultry is 36% and for buffaloes, sheep and goat 28%.

The available manure that is applied to crops and managed grassland varies from country to country. We used the data on the share of manure applied on cropland and grassland for 23 European countries from Menzi [13] and for the individual states of the US from Kellogg et al. [14]. We used the average of the 23 European countries value to other EU countries. We used the US average share of manure applied on crops and grassland for other high-income countries, including Canada, Australia and Japan. For developing countries we adopted the value provided by Bouwman et al. [6, 8]: 95% of the available manure is applied on cropland and 5% on grassland. For EU countries we used maximum application rates of 170 kg N/ha/y based on the existing EU nitrates directive.

Inputs from deposition (IN_{dep})

Atmospheric N deposition rates (including dry and wet deposition of NH_x and NO_y) for the year 2000 were taken from Dentener et al. [15]. The 30 arc-minute original data were converted to a resolution of 5×5 arc-minute.

Inputs from fixation (IN_{fix})

Following Bouwman et al. [6], total N fixation by leguminous crops was estimated by multiplying the N in the harvested product by a factor of two to account for all above and belowground plant parts. N fixation by cyanobacteria in irrigated rice ranges from 20 to 30 kg N/ha during the growing season [16]. In this study we used an average value of 25 kg N/ha. For non-leguminous crops, the non-symbiotic biological N_2 fixation rate is assumed to be 5 kg N/ha [6].

Inputs from irrigation water (IN_{irr})

The nutrient input in irrigation water is calculated for irrigated cropland by multiplying the N content of irrigation water (in kg N/m^3) by the irrigation application rate (in m^3/ha per year). We adopted the average N content of irrigation water as provided by Lesschen et al. [17]: 3.3 mg/l. The irrigation application rate at 5 arc-minute spatial resolution for all crops under irrigation condition was obtained from Mekonnen and Hoekstra [18].

Outputs from harvested crop and grass (OUT_{harv})

The N withdrawal in the harvested crops is calculated by multiplying the crop production by the N content of the crops. N loss through harvested crop (OUT_{harv}, kg N/ha) is calculated by aggregating the nutrient withdrawal from each crop harvested and adding the nutrient withdrawal due to grass consumption and harvest as follows:

$$OUT_{harv} = \sum_{p=1}^{m} \left(Y[p] \times n[p] \right) \qquad (6)$$

where *Y [p]* is the yield of crop *p* (tonne/ha) and *n [p]* the N content of crop *p* (kg N/tonne of crop).

The crop yields at 5 arc-minute spatial resolution were obtained from Mekonnen and Hoekstra [18, 19]. The N and P contents of major crop were taken from IPNI [20]. For other crops and crop groupings values from FAO [21] and Roy et al. [22] were used. For nuts and spices, we have adopted the values of fruits and vegetables respectively from FAO [21].

Outputs from crop residues (OUT_{res})

Part of the crop residues is removed from croplands and used, for example, as biofuel or for animal feeding. The nutrients withdrawal with crop residue (OUT_{res}, kg N/ha) was calculated by multiplying the yield of crop residue by the nutrient content of the crop residue and adjusting this by a removal factor:

$$OUT_{res} = CR[r] \times n[r] \times \gamma[r] \tag{7}$$

where *CR[r]* is the volume of crop residue *r* (tonne/ha), *n[r]* the N content of residue *r* (kg N/tonne of crop residue), and *γ[r]* the removal factor of the crop residue *r*. The nutrient contents of the crop residues were taken mainly from IPNI [20] and FAO [21] and for a few crops from Roy et al. [22]. Missing values for nuts and spices were filled by adopting the values of fruits and vegetables respectively from FAO [21]. The volume of crop residue was calculated by multiplying dry crop yield with a residue-to-product ration (RPR). The RPR values for a large number of crops and crop groupings were obtained from Eisentraut [23]. For spices, we took the RPR values of vegetables. The crop residue removal factors in Ghana, Kenya and Mali for various crops were obtained from FAO [21]. Removal factor for 18 crops or crop groups in India were derived from Ravindranath et al. [24]. For other crops which are not covered by Ravindranath et al. [24], we used the average removal factor of the 18 crops. For the USA, crop residue removal factors for maize and wheat for a large number of states were obtained from Graham et al. [25] and Nelson et al. [26], respectively. Removal factors of maize and wheat in other states were taken as the average removal factor in the states with data. For other crops, the residue removal factors in the USA were adopted from Perlack et al. [27]. For other countries with no data, removal factors were adopted from Krausmann et al. [28], who provide residue removal factors for major crop groupings and geographic regions.

Outputs through gaseous losses (OUT_{gas})

A large quantity of N is lost from animal manures and fertilizers by volatilization of NH_3 [16]. We adopted the empirical model of Bouwman et al. [29] to calculate ammonia volatilization from the application of animal manure and N fertilizers. The empirical model takes into account the influence of crop type, fertilizer type, manure or fertilizer application mode, soil cation exchange capacity (CEC), soil pH and climate.

N loss through gaseous emission (OUT_{gas}, kg/ha) is the sum of NH_3 volatilization and N_2O-N or NO-N emission:

$$OUT_{gas} = N_{vol,spr} + N_{emission} \tag{8}$$

where $N_{vol,spr}$ is NH_3 volatilization (kg/ha) during spreading of manure on the field and $N_{emission}$ the emission of N_2O-N or NO-N (kg/ha).

Following Bouwman et al. [29], NH_3 volatilization ($N_{vol,spr}$, kg/ha) during spreading of fertilizer and manure is calculated as follows:

$$N_{vol,spr} = IN_{fer,man} \times \varphi \tag{9}$$

where $IN_{fer,man}$ is the fertilizer/manure application rate (kg/ha) and φ the NH_3 volatilization rate, calculated as:

$$\varphi = \exp(\textit{factor value for crop type} + \textit{fertilizer type} + \textit{application mode} + \textit{soil pH} + \textit{soil CEC} + \textit{climate}) \tag{10}$$

where the factor values (for crop type, fertilizer type, application mode, soil pH, soil CEC and climate) were taken from Bouwman et al. [29]. To estimate the NH_3 volatilization, we grouped the crops into rice and other crops following Bouwman et al. [29].

Nitrification (oxidation of NH_4^+) and denitrification (reduction of NO_3^- or NO_2^-) are the main sources of NO_x and N_2O emitted from the soil [16]. Generally, soil denitrification occurs in or just below the root zone under high soil water content and limited oxygen availability, forming N_2, N_2O and NO [30]. In this study we followed Bouwman et al. [8] and estimated the three gases (N_2, N_2O and NO) separately.

Denitrification (emission of N_2) in the soil is calculated as a fraction of the available surplus N after accounting for N withdrawal with harvested crop, crop residue and ammonia volatilization [30]:

$$N_{denitrification} = f_{den} \times N_{surplus} \tag{11}$$

where $N_{surplus}$ is the N surplus which is calculated as the difference between N surface balance and ammonia volatilization. The N surface balance is calculated as the difference between the total N input (IN) and the N uptake by crops ($=OUT_{harv} + OUT_{res}$). The denitrification fraction (f_{den}) is calculated with a model that combines the effect of temperature, crop type, and soil and hydrological conditions [30]:

$$f_{den} = \min[(f_{climate} + f_{text} + f_{drain} + f_{soc}), 1] \tag{12}$$

where $f_{climate}$ represents the effect of climate on denitrification rates and where f_{text}, f_{drain} and f_{soc} are factors representing the effects of soil texture, soil drainage and soil organic content on denitrification rates, respectively. For rice the f_{den} is set at 0.75. The climate factor ($f_{climate}$) was estimated following Van Drecht et al. [30] and the factors f_{text}, f_{drain} and f_{soc} were adopted from Van Drecht et al. [30] for the respective soil parameters. The soil texture, drainage class and SOC were obtained from the derived soil properties on a 5×5 arc-minute global grid (version 1.2) from ISRICWISE [31].

According to Bouwman et al. [32], the major factors influencing the emission for N_2O include N application rate, crop type, climate, soil organic carbon (SOC) content, soil texture, drainage and soil pH, and for NO they include N application rate, SOC and soil drainage. To estimate the NO and N_2O emission, we grouped the crops into four groups (i.e, rice, legumes, grass and other crops) and applied the statistical model developed by Bouwman et al. [32]. Following Bouwman et al. [32], the emission of N_2O-N or NO-N ($N_{emission}$, kg/ha) is calculated as follows:

$$N_{emission} = IN_{fer,man} \times \exp\left(constant + \sum_{i=1}^{n} Factor\,class(i) \right) \tag{13}$$

where the *constant* and the *Factor classes* (for N_2O: N application rate per fertilizer type, crop type, climate, SOC, soil texture, drainage and soil pH; for NO: N application rate per fertilizer type, SOC and soil drainage) were adopted from Bouwman et al. [32]. The SOC, soil texture, drainage class and pH were obtained from the derived soil properties on a 5×5 arc-minute global grid (version 1.2) from ISRICWISE [31].

1.2. Point sources of N

Domestic N loads

The N load from the domestic sector is estimated following Van Drecht et al. [33]:

$$N_{dsw} = N_{hum} D(1 - R_N) + N_{hum}(1 - D) f_{sw} \tag{14}$$

where N_{dsw} is the N load from households to surface water (kg/person/y), N_{hum} the human N emission (kg/person/y), D the fraction of the total population that is connected to public sewerage systems (dimensionless), f_{sw} the fraction of non-sewered human N which is discharged to surface water, and R_N the overall removal of N through wastewater treatment (dimensionless). The first part of the equation refers to the population connected to sewerage systems and the second part to the population not connected to sewerage systems.

Human N emissions

The human N emission is estimated based on dietary per capita protein consumption per country over the period from 2002 to 2010, from FAOSTAT [5]. The N intake in the food is estimated by assuming an average 16% N content in dietary per capita protein consumption [34, 35]. About 97% of the N intake is assumed to be excreted in the form of urine and faeces and the remainder 3% of N intake is lost via sweat, skin, hair, blood and miscellaneous [36-39].

Data on connection to public sewerage system (D) was collected mainly from UN Statistics Division [40], for European countries from Eurostat [41], and for OECD countries from OECD [42]. For other countries with no data on D, we used the regional average values from Van Drecht et al. [33].

To estimate the N removal through wastewater treatment (R_N), we distinguished three wastewater treatment types with differing N removal efficiencies based on the work of Van Drecht et al. [33]: primary treatment (10% N removal), secondary treatment (35% N removal), and tertiary treatment (80% N removal). Data on the distribution of the different treatment types for European countries was obtained from Eurostat [41] and for OECD countries from OECD [42]. For other countries, we adopted regional average values from Van Drecht et al. [33] .

Part of the human N emission from populations that are not connected to sewerage systems is recycled in agriculture [43], about 20% of the N is lost through ammonia volatilization [38, 39] and some part is lost by leakage and seepage to soils and groundwater [44, 45]. We have assumed that the fraction of non-sewered human N that is discharged to surface water (f_{sw}) is 10%.

Industrial N load

Industries responsible for nutrient loads include the food, textile, paper & cardboard, grease & oil, tannery, and soap industries [46]. Since there is a lack of data on industrial loads per sector, we have estimated the N load from the industrial sector as a whole as a function of the emission from the gross human urban nutrient load. The ratio of N load from industrial to urban domestic ranges from 0.05 to 0.17 [46-49]. We have assumed an average value of 0.10. We further assumed that about 30% of the total N load is lost in wastewater stabilization ponds or through ammonia volatilization following the works of Morée et al. [38] and Billen et al. [46]. Therefore, the N load from the industrial sector as function of the load of urban human households is:

$$N_{isw} = 0.1 \times 0.7 \times [N_{hum} U (1 - R_N)] \quad (15)$$

where N_{isw} is the N load from industries to surface water (kg/person/y) and U fraction of the total population living in urban areas.

2. Detailed results on the estimated global anthropogenic N load from agriculture

The global anthropogenic N load to fresh water per crop category is presented in the last row of Table S1. The table shows all terms in the N balance of the soil: both N inputs (artificial fertilizer, manure, bio-fixation, atmospheric deposition and supply through irrigation water) and N outputs (N removal with harvested crops and crop residues, erosion, gaseous losses and leaching). The largest share of the N input in croplands comes from artificial fertilizers, which account for about 50% of the total input. N input from manure accounts for 20% of the total input, and biofixation for 18%. Cereal crops account for about 62% of the N input from artificial fertilizer and 32% of the manure. Oil crops contribute 11% of the N input from artificial fertilizer, followed by 'other crops', which contribute 9%. The 'other crops' category (mainly fodder crops) also accounts for a large contribution (25%) to the N input in the form of manure. About three-quarters of N input from bio-fixation came from oil crops. Relative to total N input, bio-fixation was most important in the case of oil crops (e.g. soybean, groundnuts) and pulses. Cereal crops account for the largest N removal with harvested crops (42%) and crop residues (60%). Oil crops are second in this respect, responsible for 24% of the N removed with harvested crops and 25% of the N removed with crop residues.

The total N leaching from croplands was 35 million tonne N/y, of which 70% (24.4 million tonne N/y) originated from anthropogenic sources (fertilizers, manure). Looking at the contribution of the different crop categories to the anthropogenic N load to fresh water, we see that the largest share (23%) came from cereal crops, followed by vegetables (19%), oil crops (15%) and fruits (12%). Our result shows that globally about 18% of the total N input in the form of artificial fertilizer and manure leaches to freshwater systems.

Table S1. Global nitrogen inputs to and outputs from croplands per crop category (million tonne per year). The leaching from anthropogenic sources is calculated as a fraction of total N leaching. Period: 2002-2010.

Balance term	Cereals	Veget-ables	Oil crops	Fruits	Roots & tubers	Sugar crops	Pulses	Nuts	Other crops[1]	Total
Artificial fertilizer	60	5.8	11	4.3	3.0	2.6	0.8	0.5	8.7	96
Manure	12	4.2	5.5	3.1	1.9	0.5	0.6	0.3	9.4	38
Bio-fixation	5.3	0.2	25	0.2	0.2	0.1	2.2	0.03	0.7	34
Atmospheric N deposition	6.7	0.6	2.3	0.5	0.5	0.2	0.6	0.1	1.3	13
N supply in irrigation water	5.3	0.4	1.3	0.6	0.2	0.4	0.2	0.1	1.6	10
Total N inputs	**90**	**11**	**45**	**8.7**	**5.7**	**3.9**	**4.4**	**1.0**	**22**	**191**
N removed with harvested crops	31	0.8	15	0.9	1.0	0.5	1.6	0.02	12	63
N removed with crop residue	12	0.2	4.7	0.3	0.1	0.1	0.8	0.01	1.0	20
Total N removed with crop and crop residues	**43**	**1.0**	**20**	**1.2**	**1.1**	**0.5**	**2.4**	**0.0**	**12.5**	**82**

N budget (available for gaseous & leaching loss)	46	10	24	7.5	4.6	3.4	2.0	1.0	9.2	108
Erosion	8.8	0.7	2.9	0.9	0.8	0.4	0.9	0.1	2.2	18
NH_3 volatilization	5.1	0.6	1.1	0.4	0.3	0.2	0.1	0.04	0.5	8.3
Denitrification (N_2)	24	3.5	9.9	2.5	1.4	1.0	0.7	0.3	1.6	45
N_2O emission	0.4	0.04	0.2	0.1	0.1	0.03	0.1	0.01	0.1	1.0
NO	0.6	0.04	0.2	0.05	0.05	0.02	0.1	0.01	0.2	1.2
N leaching	**7.1**	**5.3**	**10**	**3.6**	**2.0**	**1.7**	**0.1**	**0.5**	**4.6**	**35**
Total N outputs	**90**	**11**	**45**	**8.7**	**5.7**	**3.9**	**4.4**	**1.0**	**21.7**	**191**
Leaching from anthropogenic sources	**5.7**	**4.8**	**3.7**	**3.0**	**1.7**	**1.3**	**0.04**	**0.4**	**3.9**	**24**

[1] Including fodder crops, coffee, tea, cocoa, spices and fibre crops.

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