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Global Gray Water Footprint and Water Pollution Levels Related to Anthropogenic Nitrogen Loads to Fresh Water

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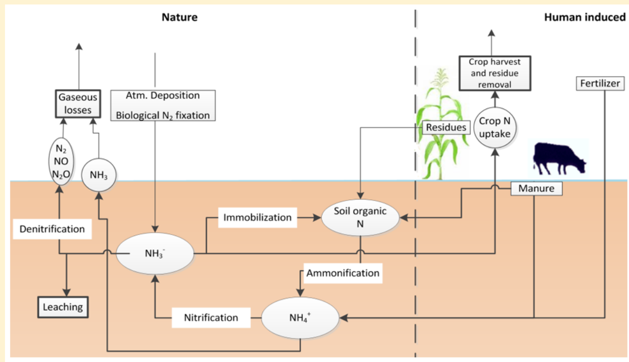
Supporting Information

ABSTRACT: This is the first global assessment of nitrogen-related water pollution in river basins with a specification of the pollution by economic sector, and by crop for the agricultural sector. At a spatial resolution of 5 by 5 arc minute, we estimate anthropogenic nitrogen (N) loads to freshwater, calculate the resultant gray water footprints (GWFs), and relate the GWFs per river basin to runoff to calculate the N-related water pollution level (WPL) per catchment. The accumulated global GWF related to anthropogenic N loads in the period 2002–2010 was 13×10^{12} m^3/y. China contributed about 45% to the global total. Three quarters of the GWF related to N loads came from diffuse sources (agriculture), 23% from domestic point sources and 2% from industrial point sources. Among the crops, production of cereals had the largest contribution to the N-related GWF (18%), followed by vegetables (15%) and oil crops (11%). The river basins with WPL > 1 (where the N load exceeds the basin's assimilation capacity), cover about 17% of the global land area, contribute about 9% of the global river discharge, and provide residence to 48% of the global population.



■ INTRODUCTION

Increased use of nitrogen (N) has both positive and negative effects.[1] The increase in crop yields over the past decades is partly due to the increased use of fertilizer in agriculture. However, a large fraction of N applied to croplands in the form of fertilizer and manure ends up entering the freshwater system causing degradation of the water quality and eutrophication of groundwater, rivers, lakes, and coastal and marine ecosystems.[2] Human-induced eutrophication of rivers, lakes, estuaries and coastal seas has already resulted in loss of biodiversity, hypoxia and fish kills in many parts of the world.[3−11]

As a measure to quantify the pressure that additional N puts on freshwater resources, we use the gray water footprint (GWF). More broadly, the water footprint is an indicator of human appropriation of freshwater resources. It measures both the direct and indirect "water use" of consumers and producers. The term "water use" refers to two different components: consumptive water use (of rainwater—the green water footprint—and of surface and groundwater—the blue water footprint) and degenerative water use (the gray water footprint). GWF is measured as the volume of water required to assimilate pollution and can be interpreted as a dilution water requirement, a concept that can be traced back to Postel et al.[12] and Chapagain et al.[13] The "gray water footprint" was first introduced by Hoekstra and Chapagain[14] and defined as the volume of fresh water that is required to assimilate the load of pollutants based on natural background concentration and existing ambient water quality standards.[15] The advantage of expressing water pollution in terms of the water volume required for assimilating the pollutants, rather than in terms of concentrations of contaminants, is that this brings water pollution into the same unit as consumptive use, as shown by Hoekstra and Mekonnen.[16] In this way, the use of water as a drain and the use of water as a resource, two competing uses, become comparable.

We will use the concept of water pollution level (WPL) to express the effect of a GWF per river basin on the water quality in this basin. WPL is defined as the GWF in a river basin divided by the river basin runoff.[15] The GWF refers to the volume of water (m^3/y) required for the assimilation ("dilution") of the load of pollutants; the maximum sustainable GWF (the "waste assimilation capacity") is given by the actual volume of water available, which is the river basin runoff (m^3/y). WPL thus shows the fraction of the waste assimilation capacity in a river basin that has been actually consumed. If WPL = 1, the waste assimilation capacity has been fully consumed. WPL > 1 implies that waste assimilation capacity of the basin is insufficient to take up the actual pollution, resulting in a violation of water quality standards.

There are a few previous global studies that quantify the global anthropogenic N load to fresh water from all sectors (agricultural, domestic and industrial) at a high spatial resolution.[16−28] Some other global high-resolution studies

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focused on one sector only, for example, agriculture[29] or domestic waste.[30,31] Only two earlier studies quantified the global N-related GWF,[16,25] and only one previous study estimated N-related WPLs across the globe.[25] Mekonnen and Hoekstra[32] made the first estimate of the global N-related GWF for a large number of crops at a spatial resolution of 5 by 5 arc-minute, using a simple model with a fixed N leaching-runoff fraction, thus leaving out local factors such as crop type grown, soil type and agricultural practices that can influence processes of leaching and runoff. Hoekstra and Mekonnen[16] made the first estimate of the overall N-related GWF, by supplementing the agriculture-focused data from Mekonnen and Hoekstra[32] with estimates of the GWF's from the domestic and industrial sector. The GWF was estimated roughly by assuming it to be equal to untreated return flows from the two sectors. Liu et al.[25] made another, independent estimate of the global N-related GWF, including all three sectors, and was the first to estimate resultant WPLs per river basin. The study focused on showing the geographical spread of WPLs and did not relate the water pollution to specific sectors (agriculture, domestic, industrial), or, with respect to the water pollution from agriculture, to specific crops.

The present study estimates gray water footprints and water pollution levels associated with anthropogenic N loads at global scale at a 5 by 5 arc minute resolution and relates the pollution to specific sectors and crops. The analysis is carried out for the period 2002−2010. The novelty of the study is that, for the first time, N-related water pollution levels are estimated at a high spatial resolution level and related to specific sectors, and to specific crops in the case of the agricultural sector. We achieve this by combining the strengths of three earlier studies: (1) Hoekstra and Mekonnen[16] who estimate N-related loads and gray water footprints per sector and crop, but assuming simple leaching-runoff ratios for diffuse pollution and not comparing gray water footprints to sustainable levels; (2) Bouwman et al.,[17] who use a more advanced soil balance approach for estimating diffuse N loads; and (3) Liu et al.[25] who compare gray water footprints to maximum sustainable levels at river basin level in order to calculate water pollution levels, but do not specify gray water footprints and water pollution levels by sector or crop.

■ MATERIALS AND METHODS

The Anthropogenic Nitrogen Load to Fresh Water. Diffuse N loads to fresh water from agriculture were estimated for 126 crops separately. We took spatial crop distributions from Monfreda et al.[33] The application rate of artificial fertilizer per crop per country was calculated using three sources of artificial fertilizer data. Primarily, we used the data set of IFA et al.,[34] which provides artificial fertilizer application rates per crop for 88 countries. We used FAO[35] and Heffer[36] to complement this data set. Since the application rates provided in these data sources refer to different years, these were adjusted to fit FAO[37] country average artificial fertilizer consumption per year for the period 2002−2010. The manure input was calculated at grid cell level by multiplying livestock density (taken from FAO[38]) by the animal-specific excretion rates. The animal-specific excretion rate per animal category, production system and country was calculated by combining global average manure excretion rates from Sheldrick et al.[39] with the slaughter weight of animals per production system and per country, as taken from FAO.[37] 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. In this study, we considered manure inputs on croplands (including managed grasslands), but did not further study manure inputs on grazing lands. We grouped the crops into leguminous, irrigated and nonleguminous in order to estimate the N input through biofixation. Atmospheric N deposition rates for the year 2000 were taken from Dentener et al.[40] The nutrient input through irrigation water was calculated for irrigated croplands by multiplying the N content of irrigation water (in kg of N per cubic meter) 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.:[41] 3.3 mg/L. The irrigation application rates at 5 × 5 arc minute spatial resolution for all crops under irrigation were obtained from Mekonnen and Hoekstra.[42] The N removal with harvested crops was estimated by multiplying the crop yield by the crop-specific N content. The N removal with crop residues was calculated by multiplying the yield of crop residue by the nutrient content of the crop residue and a residual removal factor. We adopted the approach of Liu et al.[29] to calculate the nutrient loss through erosion. We employed the empirical model of Bouwman et al.[43] to calculate ammonia volatilization and Bouwman et al.[44] to estimate N loss through N_2O and NO from the application of animal manure and artificial fertilizers. Denitrification (emission of N_2) in the soil was calculated as a fraction of the N surplus after accounting for ammonia volatilization and N removal with the harvest of crop and crop residue.[27] Leaching and runoff of N—the movement of N from the soil to ground or surface waters—was estimated by assuming balance of N in the soil in the long term. Finally, we estimated, still at grid cell level, the anthropogenic N load to fresh water (i.e., the load due to artificial fertilizer and manure application) by multiplying the total leached volume by the fraction of N input from artificial fertilizer and manure to the total N input (which also includes the amounts of N added through biofixation, deposition and irrigation water).

To estimate N loads from diffuse sources, we followed the approach of Bouwman et al.,[17] which uses a full soil balance approach, accounting for precipitation and soil properties for estimating gaseous losses from the soil. The soil parameters were obtained from Batjes.[45] The precipitation data for the period 2002−2010 were obtained from the Climate Research Unit of the University of East Anglia.[46] The rooting depths for individual crops were obtained from Allen et al.[47]

N loads from point sources were estimated based on dietary per capita protein consumption per country over the period from 2002 to 2010, using data from FAOSTAT,[37] following the approach of Van Drecht et al.[30] The N intake through food is estimated by assuming an average of 16% N content in the protein consumed.[48,49] About 97% of the N intake is assumed to be excreted in the form of urine and faeces and the remainder 3% is lost via sweat, skin, hair, blood, and miscellaneous.[31,50−52] Data on connection to public sewerage system and the distribution of the different treatment types was collected from different sources.[30,53−55] Since there is lack of data on industrial emissions, we have estimated the N load from the industrial sector as a function of the urban domestic load by assuming a ratio of industrial to urban households N load of 0.10.[56−60]

An extended description of the method and the data used in estimating N loads is presented in the Supporting Information.

Gray Water Footprint. Following Hoekstra et al.,[15] the gray water footprint (GWF, m^3/y) is calculated by dividing the

N load (*Load*, kg/y) by the difference between the ambient water quality standard for N (the maximum acceptable concentration c_{max}, mg/L) and the natural concentration of N in the receiving water body (c_{nat}, in mg/L):

$$\mathrm{GWF} = \frac{Load}{(C_{\max} - C_{\mathrm{nat}})} \quad (1)$$

The natural concentration is the concentration in a water body if it were in pristine condition, before human disturbances in the catchment. In the literature we can find different values for maximum allowable and natural concentrations. Liu et al.[25] use 3.1 mg N/L for the maximum concentration and 1.5 mg N/L for the natural concentration. For this study we have taken the maximum acceptable value provided by the GWF guidelines,[61] 2.9 mg N/L, which again is based on the guideline for the protection of aquatic life as proposed by the Canadian Council of Ministers of the Environment.[62] The GWF guidelines from the Water Footprint Network suggest a natural concentration value for total N of 0.36 mg N/L,[61] which is close to the average natural concentration of N in rivers of 0.375 mg N/L reported by Meybeck.[63] In this study, we assumed a rounded off value of 0.4 mg N/L.

Water Pollution Level. The water pollution level (WPL), which measures the degree of pollution within a catchment, is estimated as the ratio of the total of GWF in a catchment to the actual runoff from that catchment (R_{act}, m^3/y):

$$\mathrm{WPL} = \frac{\mathrm{GWF}}{R_{\mathrm{act}}} \quad (2)$$

The annual actual runoff data at a 30 by 30 arc minute resolution were obtained from the Composite Runoff V1.0 database.[64]

Since many model inputs and parameters are uncertain, we assessed the sensitivity of the GWF estimation to uncertainties in input data. We used Latin hypercube sampling (LHS)[65] and assumed uncertainty ranges of ±20% for the major model inputs and parameters (artificial fertilizer and manure application rate, N biofixation rate, atmospheric deposition, N input with irrigation water, N removal with harvested crops and crop residues, protein intake, urban population, sewer connection, N removal in sewerage treatment, and the maximum allowable and natural concentrations). We did 1000 runs for the two nutrient loads (diffuse and point sources) and estimated resultant GWF ranges by accounting for the uncertainty ranges (±20%) for all the major parameters mentioned above.

RESULTS

Gray Water Footprint Related to Nitrogen. The total leaching and runoff of N from the world's croplands is estimated at 35 million tonne N/y, of which 70% (24.4 million tonne N/y) originated from anthropogenic sources (fertilizers, manure). The global N load to freshwater bodies from point sources was about 8.2 million tonne of N per year (91% domestic and 9% industry). Thus, the global anthropogenic N load to fresh water systems from both diffuse and point sources in the period 2002−2010 was 32.6 million tonnes per year. The Supporting Information provides further details on the estimated global anthropogenic N load from agriculture to fresh water. The global GWF related to the total anthropogenic N load—from both diffuse and point sources—was 13×10^{12} m^3/y (Table 1). China contributed about 45% to this global total, the U.S. about 7%, Russia 6% and India 5%.

Table 1. Global Gray Water Footprint Related to Nitrogen Loads to Fresh Water Per Sector and Specification for the Ten Countries with the Largest Contribution (Billion m^3/y)

region	agriculture	domestic	industry	total
China	4916	891	68	5875
U.S.	636	224	21	881
Russia	616	107	10	733
India	458	192	23	674
Pakistan	262	23	3	288
Brazil	195	102	16	312
Egypt	149	28	4	181
Japan	41	114	12	167
Germany	107	26	2	134
Ukraine	112	32	3	147
others	2275	1235	127	3637
world total	9767	2974	288	13 029

The contributions of different product categories and different regions to the global GWF related to anthropogenic N loads to fresh water are presented in Figure 1. The largest share (75%) comes from diffuse sources, that is, N leaching and runoff from croplands. Cereal production contributes 18% to the global N-related GWF (wheat 7% and maize 6%), production of vegetables 15% (tomatoes 1.1%) and oil crops 11% (soybean and rapeseed 3.1% each and cotton 2.4%). N loads from the domestic sector account for 23% of the total and the industrial sector 2%. Looking at the regional contribution, we find that Asia (mainly China) contributed almost two-thirds to the total GWF, followed by Europe (15%), Northern America (8%) and Latin America and the Caribbean (6%).

Figure 2 shows the global GWF at high spatial resolution. The spatial variation of GWF correlates to the spatial variation of the nutrient loads, which are highest in areas of intensive agriculture and densely populated areas. Large GWFs are observed in Southeastern China, Northern India, Western Europe, Midwestern U.S., the Nile delta in Egypt, South East Brazil and the Central Valley in Chile.

Water Pollution Level. The water pollution level (WPL) related to anthropogenic N loads is shown in Figure 3. The basins with WPL > 1, together cover about 17% of the global land area (excluding Antarctica), 9% of the global river discharge, and provide residence to about 48% of the global population. River basins in most parts of Asia, Western Europe, Southwestern U.S., Northern and Southern Africa, Argentina, and Australia have WPL > 1. In most of these basins, the large human induced N loads are responsible for the high WPL, but the high WPL levels observed in the Saharan desert, Arabian Peninsula, and large parts of Australia are due to the very low runoff levels in these basins to assimilate N.

Table 2 presents the GWF and WPL related to anthropogenic N loads for the major river basins of the world. Out of the 20 river basins listed, seven basins have a WPL > 1. In order to identify to what extent each economic sector and different agricultural products contribute to the nutrient loads, we will present a detailed analysis of the nutrient loads and the WPL in these seven basins in the next section.

Water Pollution Level in Selected River Basins. The Yangtze River, or Chang Jiang ("Long River"), is world's third longest river, ranking behind the Nile and the Amazon Rivers.

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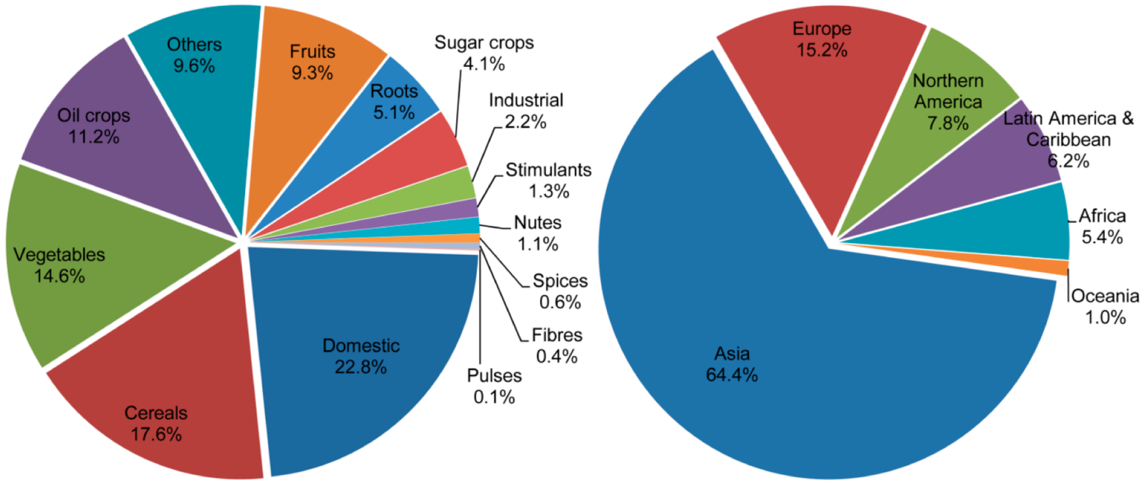

Figure 1. Relative contribution of different product categories (left) and different regions (right) to total gray water footprint related to anthropogenic nitrogen loads. Period: 2002−2010.

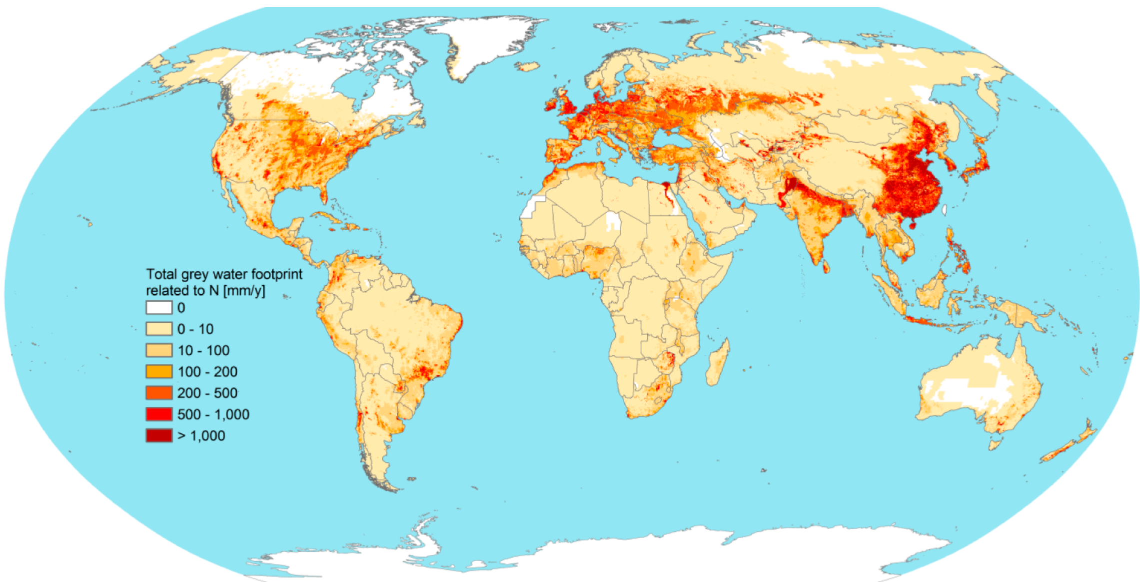

Figure 2. Gray water footprint related to anthropogenic nitrogen loads from diffuse and point sources. Period: 2002−2010. The data are shown in mm/y on a 5 by 5 arc minute grid. Data per grid cell have been calculated as the gray WF within a grid cell (in m^3/y) divided by the area of the grid cell (in 10^3 m^2).

Over the last 50 years, the Yangtze has experienced a 73% increase in pollution levels. About 42% of the country's sewage and 45% of the industrial waste are discharged into the river annually.[66] Discharge of such large amounts of sewage and industrial wastes as well as diffuse loads of nutrients, pesticides and herbicides from the agricultural sector have made the Yangtze one of the most polluted rivers in the world.[66] According to Yan et al.,[67] both nitrate concentrations and fluxes in the Yangtze have increased more than 10-fold from 1968 to 1997, mainly due to artificial fertilizer use. Our results show that the WPL of the Yangtze River Basin is about 2.0, indicating that the river's waste assimilation capacity for N has been overused by a factor two. About 96% of the N load to the freshwater system within the basin comes from the agricultural sector and another 3% from the domestic sector. Among the agricultural crops, production of vegetables account for about 26% of the total N load, following by oil crops (23%) and cereal crops (mainly rice and wheat, 19%).

With 343 people per km^2 in 2000, the Ganges River Basin is one of most densely populated river basins in the world. The river is one of the world's most polluted ones. Disposal of large quantities of untreated and partially treated sewage combined with massive water abstraction have resulted in severe water quality degradation of the river.[68] With an average WPL of 1.2, the N assimilation capacity of the basin has been exceeded by 20%. About 56% of the N load to freshwater bodies within the basin comes from the domestic sector and another 39% from the agricultural sector. Among the agricultural crops, cultivation of cereals (mainly rice and wheat) accounted for about 30% to the total N loads.

The Xi Jiang River is shorter than the other important Chinese rivers—the Yangtze and Yellow Rivers—but its annual discharge is second after that of the Yangtze. The total anthropogenic N load to freshwater bodies was 1240 ktonne/y over the period 2002−2010, resulting in a total GWF of 500 billion m^3/y and a WPL of 2.3. Almost all (98%) of the N load

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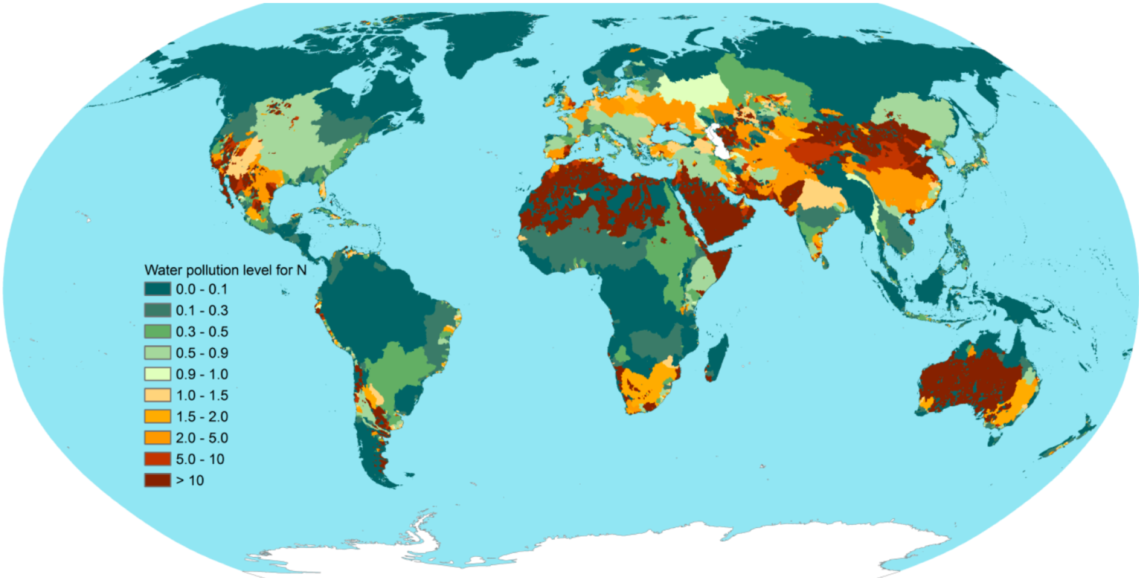

Figure 3. Water pollution level per river basin related to anthropogenic N loads from diffuse and point sources. Period: 2002−2010.

Table 2. Gray Water Footprint and Water Pollution Level Related to Anthropogenic N Loads for 20 Major River Basins, Period: 2002−-2010

basin	annual runoff (billion m^3/y)	population (million)	GWF (billion m^3/y)	WPL
Amazon	6590	26	60	0.009
Congo	1270	66	13	0.01
Yangtze	903	384	1800	2.0
Mississippi	623	73	410	0.65
Parana	542	69	170	0.30
Mekong	482	52	70	0.15
Ganges	397	417	480	1.2
Ob	396	26	160	0.39
Amur	362	66	290	0.80
Niger	330	74	50	0.15
Nile	326	145	130	0.39
Zambezi	325	30	44	0.14
Volga	269	59	240	0.90
Xi Jiang	221	63	500	2.3
Danube	208	82	130	0.64
Indus	148	150	440	3.0
Rhine	76	50	55	0.71
Aral Drainage	70	27	280	4.0
Huang He (Yellow)	49	121	410	8.3
Murray-Darling	18	2	32	1.8

within the basin was due to intensive agriculture, mainly related to the production of rice, maize, and vegetables.

The Indus River Basin is a densely populated basin facing severe water scarcity almost three-quarters of the year[69] and high nutrient pollution due to intensive agricultural activities. The total N load to fresh water in the basin was 1100 ktonne/y. A large part (59%) of the N load came from agriculture; the domestic sector contributed 38%. Cultivation of cereal crops (wheat and rice) contributes about 34% to the total anthropogenic N load in the basin; oil crops contribute 14%. The total GWF within the basin related to N was 440 billion m^3/y, resulting in a WPL of 3.0.

The Drainage Basin of the Aral Sea is shared by Uzbekistan, Turkmenistan, Kazakhstan, Afghanistan, Tajikistan, and Kyrgyzstan. Due to intensive use of the rivers flowing to the Aral Sea for irrigation, the sea has been shrinking since the 1960s.[70] Three quarters of the anthropogenic N load within the basin is due to the untreated sewage from the domestic sector. A total of 700 ktonne/y was emitted in the basin. The total GWF in the basin was 280 billion m^3/y, which is close to 4 times larger than the actual runoff of the river basin, indicating that the assimilation capacity of the basin was overused by a factor of 4.

The Yellow River, China's second longest river, originates in the Bayankala Mountains in Qinghai province in western China and flows through nine provinces of China before it drains into the Bohai Sea. The Yellow River is the water source for Northwest and North China. Deterioration of water quality is a very serious issue in the basin.[71] During the period 2002−2010, a total of 1000 ktonne/y N was emitted to freshwater bodies in the basin. The agricultural sector (mainly wheat, maize, and vegetables) accounted for 95% of the total N load and the domestic sector 5%. The total GWF associated with the anthropogenic N load was 410 billion m^3/y, resulting in a WPL of 8.3, indicating that the GWF exceeded the assimilation capacity by more than a factor of 8.

The Murray-Darling Basin is Australia's most important agricultural area, known as the country's breadbasket. The surface waters of the basin have exhibited toxic blue-green algal blooms due to excessive nutrient levels.[72] During the period 2002−2010, the total anthropogenic N load in the basin was 80 ktonne/y. The agricultural sector (mainly cultivation of fruits, cereals and "other crops"), accounted for 96% of the total N load. The domestic sector contributed a further 3%. The total GWF was 1.8 times larger than the actual runoff of the basin, resulting a WPL about 1.8.

■ DISCUSSION

Our estimate of the global N leaching-runoff from diffuse sources is 52% larger than the estimate by Liu et al.[29] and 15−39% smaller than the estimates by Bouwman et al.[17,20] (Table 3). The two studies by Bouwman et al.[17,20] include values for

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grassland in addition to croplands, which may explain their higher values.

Table 3. Comparison of the Estimated Overall Diffuse N Load from the World'S Croplands to Fresh Water with the Results from Previous Studies

study	N leaching and runoff to fresh water from diffuse sources (million tonne N/y)	study period
Liu et al.[29]	23	2000
Bouwman et al.[20]	41	2000
Bouwman et al.[17]	57	2000
current study	35	2002–2010

There is a large difference between the global GWF estimate in the current study and the earlier conservative estimate by Hoekstra and Mekonnen.[16] The GWF estimate related to diffuse N sources in the current study is about 13 times larger than the earlier estimate, which can be explained by a number of factors: (1) in the current study we have taken a 4× stricter assumption on the assimilation capacity of freshwater bodies (a difference between the maximum allowable and natural N concentration of 2.5 mg/L instead of 10 mg/L); (2) the computed global-average leaching-runoff fraction of applied N in the current study was 1.8× larger than the assumed constant fraction in the earlier study (leaching-runoff of 18% of the N application instead of 10%); (3) the global artificial fertilizer application in the current study is 1.3× larger (partly because a more recent period of analysis was taken); and (4) in contrary to the earlier study, the current study includes the contribution of manure, which results in a 1.4× higher total N application. The GWF estimate related to N from domestic wastewater in the current study is about ten times larger than the earlier estimate, which can be explained by the fact that the current study is based on a model accounting for protein consumption per country, wastewater treatment coverage and N removal ratios, while the earlier study made a rough, conservative estimate based on the assumption of a dilution factor of 1 for untreated wastewater discharged to the freshwater systems. The GWF estimate related to N from the industrial sector in the current study is about 80% of the earlier estimate.

The spatial distribution of the WPLs estimated in the current study is roughly in line with the earlier study by Liu et al.,[25] but in many river basins, estimates differ substantially. The two studies differ in many respects—in both model and data sources used—so that at this stage it is difficult to explain specific differences in outcomes. A more detailed comparative analysis of different approaches will be necessary.

The estimated N loads to fresh water from both diffuse and point sources are based on a number of assumptions and global data sets, leading to significant uncertainties. First, due to a lack of spatially distributed data, a number of assumptions had to be made regarding, for example, artificial fertilizer application rates per crop and per country, nutrient removal by crop harvest and removed crop residues, and manure production and application rates. Second, to estimate N leaching and runoff, the study assumed a long-term steady state condition in the soil regarding N content, which might not hold true in all places. Third, emissions from domestic sources were based on protein consumption, wastewater treatment coverage and nutrient removal in the wastewater treatment plants, while other point sources such as household solid waste, urban livestock and other domestic animal wastes were not included. Fourth, due to a lack of data, the emission from the industrial sector was estimated as a certain fraction of that from the domestic sector.

The GWF and WPL estimates are subject to the assumed maximum allowable and natural concentration values. Both GWF and WPL relate linearly to the assimilation capacity of a freshwater system, that is, the difference between the maximum allowable and natural concentration. Natural concentrations vary from basin to basin and different ecosystems may have a different response to N loads, requiring different maximum allowable concentration values. However, obtaining or estimating basin-specific values is an elaborate task and in this stage impossible for a global study like this one, which has been the reason why we have taken single values for both the maximum allowable and natural concentrations for the whole world. The WPL estimates from this study can be improved once better spatially distributed data on natural and maximum allowable concentrations become available. A complication though with differentiating maximum allowable concentrations per basin or country is that different governments apply different methods to establish water quality standards, so that differences in standards among countries do not only reflect differences in the sensitivity of basins, but also subjective choices regarding the method to establish the standard. This has been another reason for us to use one standard in the current study for the whole world rather than different standards per country.

We note that WPLs inversely relate to basin runoff, which can lead to high values and great sensitivity in dry basins. Further, we observe that we measure GWFs at the point where anthropogenic N loads enter the water system, not based on what is left of anthropogenic loads downstream. WPL thus reflects total load divided by critical load, measured at the point where loads enter the water system. When measured downstream, WPL could be lower than estimated in this study due to the effect of in-stream retention and transformation of N. Besides, the study does not account for spatial heterogeneities within catchments and variability within the year, which means that the data presented are annual averages at catchment level.

If we assume uncertainty ranges of ±20% for all important inputs and parameter values (artificial fertilizer and manure application rate, N biofixation rate, atmospheric deposition, N input with irrigation water, N removal with harvested crops and crop residues, protein intake, urban population, sewer connection, N removal in sewerage treatment, and the maximum allowable and natural concentrations), we find an uncertainty range of −33% to +60% in the overall global GWF estimate.

In this study we have focused on water pollution levels through anthropogenic N loads. It is important to note that a more comprehensive picture of water pollution can be obtained only if other pollutants are considered as well and if the possible interaction of pollutants is taken into account. Through its focus on N loads, the study provides information about the *pressure* on the water system from nitrogen rather than about the final *impacts* within the system, which depend on various processes within the water system.

The nitrogen-related gray water footprint has some similarity to the more recently introduced nitrogen footprint concept, which is generally defined as the total amount of reactive nitrogen (all forms of N except N_2) released to the

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environment in relation to a specific product or consumption pattern.[73,74] The difference is that the N-related gray WF focuses on release of N to freshwater systems and translates the N load into a volume of water to assimilate. In this way, the gray WF concept allows different forms of water pollution be expressed in one unit, namely the volume of water needed for assimilation. Both the gray water footprint and the nitrogen footprint concept focus on pressure on the environment, rather than at impacts, which will depend on the scale of the overall emission and processes of dispersion and removal later on.

With the growth in international trade of agricultural products, importing countries increasingly externalize water pollution to the producing countries.[16,75] In the current study we have quantified gray WFs within geographies; the results from this study can be taken forward to study the gray WF from a consumer perspective by linking consumption volumes in certain countries to the places of production and to the related gray WFs in these places of production.

Despite the uncertainties, our results provide an insight into the magnitude and spatial distribution of the N-related GWFs and WPLs. The study shows that the total GWF related to a global anthropogenic N load of 32.6 million tonnes per year was 13×10^{12} m^3/y. Close to half of this (45%) was contributed by China; the U.S., Russia, and India together contributed another 18%. The study also shows that the WPL in a large number of river basins was above 1, which means that the waste assimilation capacity in these basins has been fully consumed just by N pollution alone. The river basins with WPL > 1 cover about 17% of the global land area, 9% of the global river discharge, and provide residence to about 48% of the global population.

In some developing countries (particularly in Africa), raising crop yield may require additional N input, whereas in many regions of the world, crops receive excessive amounts of N. In these regions, excessive application of N can be reduced without affecting agricultural productivities.[2] Besides, wastewater treatment coverage and N removal rates can dramatically increase in many parts of the world by applying advanced tertiary treatment techniques. Nevertheless, in a business-as-usual scenario, N-related water pollution is expected to increase over the coming decades.[25] We propose that national governments develop GWF reduction targets that account for the assimilation capacity of river basins. Trans-boundary river basin will require international cooperation in formulating and implementing such targets.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b03191.

Additional information as noted in the text (PDF)

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Notes

The authors declare no competing financial interest.

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