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Key Points:

- Global phosphorus loads to freshwater are interpreted in terms of resulting grey water footprints
- Cereal production accounts for 31% of the phosphorus load from agriculture to water systems
- The P-related grey water footprint exceeds assimilation capacity in 38% of the global land area

Supporting Information:

- Supporting Information S1
- Table S1
- Table S2
- Table S3

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Global Anthropogenic Phosphorus Loads to Freshwater and Associated Grey Water Footprints and Water Pollution Levels: A High-Resolution Global Study

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Abstract We estimate the global anthropogenic phosphorus (P) loads to freshwater and the associated grey water footprints (GWFs) for the period 2002–2010, at a spatial resolution of 5 × 5 arc min, and compare the GWF per river basin to runoff to assess the P-related water pollution level (WPL). The global anthropogenic P load to freshwater systems from both diffuse and point sources is estimated at 1.5 Tg/yr. More than half of this total load was in Asia, followed by Europe (19%) and Latin America and the Caribbean (13%). The domestic sector contributed 54% to the total, agriculture 38%, and industry 8%. In agriculture, cereals production had the largest contribution to the P load (31%), followed by fruits, vegetables, and oil crops, each contributing 15%. The global total GWF related to anthropogenic P loads is estimated to be 147 $\times 10^{12}$ m^3/yr, with China contributing 30%, India 8%, USA 7%, and Spain and Brazil 6% each. The basins with WPL > 1 (where GWF exceeds the basin's assimilation capacity) together cover about 38% of the global land area, 37% of the global river discharge, and provide residence to about 90% of the global population.

1. Introduction

Globally, the atmospheric phosphorus (P) reservoir is very limited (Smil, 2000) and part of the P available in soluble and particulate form is removed from land to the ocean through erosion and runoff (Mackenzie et al., 2002; Smil, 2000). The amount of P in soils that is available to plants is limited because most of the P contained in rocks is either not fully soluble or becomes insoluble very rapidly (Bouwman et al., 2013; Smil, 2000). Mineral fertilizer and manure are added to supplement the P shortage, but the plants take up only part of the applied P in the same year and a large portion of the P that is not taken up by the plants accumulates in the soil as "residual P" (Syers et al., 2008). The residual P that accumulates over the years can be taken up by crops and the excess P will enter the surface water through erosion. P is transferred to the ocean mainly by erosion and runoff, while leaching of P is generally small because the phosphates in soils are soluble to a small extent only (Smil, 2000). A large portion of P (75–90%) that is transported with runoff water from cultivated land is in particulate form (Sharpley et al., 1993, 2003).

Human actions have significantly changed the global biogeochemical cycles of P. While preindustrial transfer of P from soils to waters was about 12 Mt/yr, P losses from soil to water were about 27 Mt/yr around the year 2000, mostly due to increased erosion as a result of intensified land use (Smil, 2000). A large part of this additional load of P from soils to water refers to P that is naturally contained in soils, but eroding now. On top of the P that is naturally contained in soils and flowing to water through erosion, the P contained in mineral and manure fertilizers applied in agriculture partly ends up in surface water, either directly through erosion, runoff, and leaching of P from the soils, or indirectly, e.g., through excreta of humans and animals (Bennett et al., 2001; Mackenzie et al., 1998; Seitzinger et al., 2005, 2010; Smil, 2000). The increased crop yield observed over the last decades can partly be attributed to the use of fertilizers in crop production (Smil, 2000; Tilman, 1999; Tilman et al., 2001; Vitousek et al., 2009). Unfortunately, part of the mobilized P enters surface waters and eventually ends up in coastal seas (Bouwman et al., 2009; Kanakidou et al., 2012; Seitzinger et al., 2005, 2010) with profound effects upon the quality of receiving waters through the eutrophication of lakes, rivers, and coastal waters (Bennett et al., 2001; Carpenter et al., 1998; Correll, 1998; Seitzinger et al., 2010; Smil, 2000; Smith, 2003; Tilman, 1999).

We apply the grey water footprint (GWF) concept to quantify the pressure put by anthropogenic P loads on freshwater resources. GWF is the amount of freshwater needed to assimilate ("dilute") the pollutants load based on the natural background and maximum allowable concentrations (Hoekstra et al., 2011). By putting water pollution in terms of the volume of water required to assimilate the pollutants, one is able to express water pollution and consumptive water use in the same unit (Hoekstra & Mekonnen, 2012). This allow us to compare the two competing uses of water: water as a resource and water as a sink for pollutants.

With the metric of water pollution level (WPL) one can measure the fraction of the waste assimilation capacity of a river basin that has already been used up. The WPL per river basin is calculated by dividing the GWF in the basin by the actual river runoff in the basin (Hoekstra et al., 2011). The WPL is classified as low if the GWF is smaller than the actual river runoff in the basin (WPL < 1). The basin's waste assimilation capacity is said to be fully consumed if WPL $= 1$. A WPL exceeding 1 indicates the violation of the water quality standards and that the basin's waste assimilation capacity is not big enough to take up the pollutant load. Several earlier global studies have quantified the global anthropogenic P load to freshwater from point and diffuse sources (Bennett et al., 2001; Bouwman et al., 2009; Harrison et al., 2005a, 2005b; Kroeze et al., 2012; Liu et al., 2008; Mayorga et al., 2010; Peñuelas et al., 2013; Seitzinger et al., 2005, 2010; Smil, 2000; Turner et al., 2003). Some of them focused on specific sectors, e.g., agriculture (Bouwman et al., 2013) or domestic and industrial waste (Morée et al., 2013; Van Drecht et al., 2009). Van Dijk et al. (2016) quantified the P flows for EU-27 (the 27 countries of the European Union) by considering food chains, food waste and nonfood related flows. Liu et al. (2012) is the only previous study that estimated global P-related GWF and WPLs. The study showed the geographical spread of WPLs but fails to show the contribution of particular sectors (agricultural, industrial, and domestic) or crops to the water pollution.

The aim of this study is to estimate anthropogenic P loads to freshwater and the associated GWFs at global scale at 5×5 arc min resolution, assess WPLs at river basin level and attribute the pollution to particular sectors and crops. The assessment is done for the period 2002–2010. Estimating the anthropogenic P loads to freshwater and the associated GWFs will help to assess whether we are within or have passed the planetary boundaries or upper tolerable limits defined by Carpenter and Bennett (2011) and Steffen et al. (2015).

2. Materials and Method

The current study is the second part of an effort to assess the global nutrients (N and P) loads to freshwater and the associated GWF and WPL, which was supported by the CREEA (Compiling and Refining Environmental and Economic Accounts) project of the European Community. The first part of the study focused on anthropogenic N loads and the associated GWF and WPL (Mekonnen & Hoekstra, 2015). The two studies largely follow the same approach and assumptions, insofar N and P flows have the same drivers.

In the following section, we describe briefly the method used to estimate the P loads from diffuse and point sources, the GWF, and the WPL. The supporting information contains a detailed description of the methods applied and data used to estimate the P loads from point and diffuse sources [Calloway et al., 1971; Drechsel & Kunze, 2001; Eisentraut, 2010; Held et al., 2006; Kellogg et al., 2000; Liu et al., 2010; MacDonald et al., 2011; Marshall, 2005; Roy et al., 2006; CIESIN & CIAT, 2005; FAO et al., 1985; IFA et al., 2002; IPNI, 2012; USDA, 2014a, 2014b].

2.1. P Input and Output in Agricultural Production

P balances in crop fields were estimated for 126 crops separately by accounting for the P inputs to the soil through mineral fertilizer, manure and irrigation, and P removal with crop harvest and crop residues. We considered the spatial distribution of crops as given by Monfreda et al. (2008). The rate of mineral fertilizer applied for major crops per country for 88 countries was derived primarily from the data set from IFA et al. (2002). We also used data from FAO (2012b) and Heffer (2009) to complement this data set. We scaled these applied fertilizer per unit area to fit the national mineral fertilizer consumption statistics of FAO (2012a). To estimate the amount of manure applied on cropland per country, we first estimated the volume of manure produced and then estimated the fraction that is collected and applied over croplands. The manure volume was calculated by combining livestock densities data from FAO (2012c) with the excretion rates per animal. We combined global average manure excretion rates from Sheldrick et al. (2003) and the animals slaughter weight per country from FAO (2012a) to estimate the animal-specific excretion rate per animal category and country. The study considered manure applied on crop fields and managed grasslands, but left out the part that falls on grazing lands. The irrigation application rate (m^3/ha per year) was obtained from Mekonnen and Hoekstra (2010a) was multiplied by the average P content of 0.4 g/m^3 in irrigation water as obtained from Lesschen et al. (2007) to derive the P input through irrigation water. The geographic spread of global irrigated areas has been derived from Portmann et al. (2010). The volume of P removed with the crops harvested was calculated by multiplying crop yields by their crop-specific P content. In the same way, the volume of P removed with crop residues was estimated as crop residue yields times the crop-specific residue P content and a residue removal factor. Crop yields were calculated per crop at a high spatial resolution following Mekonnen and Hoekstra (2010b, 2011), scaled at national level to fit FAO's national production statistics (2012a). The crop-specific P contents of crops and crop residues and fractions of crop residue removed were obtained from different sources (FAO, 2004; Graham et al., 2007; Krausmann et al., 2008; Nelson, 2002; Perlack et al., 2005; Ravindranath et al., 2005).

2.2. P Loads From Diffuse Sources

To estimate the P loads to freshwater from diffuse sources, we used the "surplus approach" of the Grey Water Footprint Accounting Guidelines (Franke et al., 2013), in which the P load to freshwater is estimated by multiplying the surplus P by an erosion-runoff-leaching fraction. The P surplus is estimated as P input from mineral fertilizer and manure minus the removal of P with crop and crop residue harvest. The erosion-runoff-leaching fraction was estimated per grid cell as a function of five influencing factors: soil texture, soil erosion vulnerability, soil P content, rainfall intensity, and management practices. The erosion-runoff-leaching fraction (β) will lie somewhere in between the minimum (β_{min}) and maximum (β_{max}) erosion-runoff-leaching fractions, depending on the values of the influencing factors:

$$\beta = \beta_{\min} \left[\frac{\sum_i s_i \times w_i}{\sum_i w_i} \right] (\beta_{\max} - \beta_{\min}) \tag{1}$$

Per influencing factor, the score for the erosion-runoff-leaching potential (s_i) is multiplied by the weight of the factor (w_i). We adopted the minimum and maximum erosion-runoff-leaching fractions, the scores per factor, and the weight of the factors from the Grey Water Footprint Accounting Guidelines, which were developed by an international expert panel (Franke et al., 2013). Soil parameters were obtained from ISRIC-WISE (Batjes, 2012). The soil erosion vulnerability map was obtained from USDA (2014a, 2014b). The soil P content was obtained from Yang et al. (2014). The rainfall data for the period 2002–2010 was obtained from CRU (Mitchell & Jones, 2005).

2.3. P Loads From Point Sources

We followed the Van Drecht et al. (2009) approach to estimate P loads from point sources over the period from 2002 to 2010. The P intake was estimated from the nitrogen (N) intake based on an N:P ration of 10:1 (on mass basis) (Hiza & Bente, 2011; Sette et al., 2011). The food-related N intake was estimated by assuming 16% N in consumed protein (Block & Bolling, 1946; FAO, 2003). The protein consumption per capita per country was obtained from FAOSTAT (FAO, 2012a). We assumed that around 97% of the P consumed is excreted through urine and feces and the other 3% to be lost via sweat or else (FAO et al., 1985; Morée et al., 2013). Based on Morée et al. (2013) and Billen et al. (1999), we assumed that 30% of the industrial P load is assimilated in wastewater stabilization ponds. We used different data sources for the public sewerage system connection percentage per country and for the distribution of the different treatment types (European Commission, 2014; OECD, 2014; UNSD, 2014; Van Drecht et al., 2009). The P removal through wastewater treatment was estimated by distinguishing three wastewater treatment types with different average P removal efficiencies according to Van Drecht et al. (2009): 10% P removal for primary treatment, 45% P removal for secondary treatment, and 90% P removal for tertiary treatment. We further assume that a part of the P from people not connected to drainage systems is recycled on cropland, that another part of the human excreta enters soils and groundwater through leakage and seepage, and that some part is disposed into septic tanks and lagoon systems. The remainder of the nonsewered human waste is assumed to enter the surface water through dumping of human wastes in open water or through surface runoff of wastes. We have assumed that 10% of the nonsewered P enters surface water. Due to lack of data, industrial P loads were assumed to be equivalent to 15% of urban household P loads (Billen et al., 1999; Brion et al., 2008; Liu, 2005; Luu et al., 2012; Quynh et al., 2005).

2.4. Grey Water Footprint

The grey water footprint (GWF, m^3/yr) was estimated based on Hoekstra et al. (2011):

$$GWF = \frac{L}{(c_{\max} - c_{nat})} \tag{2}$$

whereby L is the P load (in g/yr), and c_{max} and c_{nat} are the maximum allowable concentration of P and the natural concentration of P in the receiving water body, respectively (in g/m^3).

The natural concentration of P is the concentration that would occur under pristine conditions, i.e., undisturbed by human activities. Different values of maximum allowable and natural concentrations are provided in the literature. Liu et al. (2012) use 0.95 and 0.52 mg P/L for the maximum and the natural concentrations, respectively. In this study, we took the maximum allowable value 0.02 mg P/L that is provided by the GWF Accounting Guidelines from the Water Footprint Network (Franke et al., 2013). This value is based on the guideline for the protection of aquatic life as proposed by the Canadian Council of Ministers of the Environment (CCME, 2004) and very close to the planetary boundary of 0.024 mg P/L for lakes and reservoirs as suggested by Carpenter and Bennett (2011). For the natural concentration, we assumed a value of 0.01 mg P/L as suggested by the GWF Accounting Guidelines (Franke et al., 2013). Different ecosystems have different natural concentrations and react differently to P loads, requiring unique natural and maximum allowable concentration values. Yet, for a global study like this one, finding basin-specific values is an elaborate task and almost impossible. Therefore, we have taken single values for both variables for the whole world.

2.5. Water Pollution Level

We estimate water pollution level (WPL) as follows:

$$WPL = \frac{GWF}{R_{act}} \tag{3}$$

where R_{act} is the actual runoff from a catchment (m^3/yr). We took annual actual runoff data from the Composite Runoff V1.0 database, at a 30 $\times$ 30 arc min (Fekete et al., 2002).

2.6. Sensitivity Assessment

To estimate the sensitivity of both the total P load and the GWF as a result of uncertainties in the input variables, we carried out a Monte Carlo analysis with 10,000 simulation runs, varying the major input variables and parameters within a standard deviation of 20% of their central estimate, using a normal distribution. For the diffuse loads, for each of the 126 crops we varied the following parameters: fertilizer application rate, manure application rate, P removal with harvested crop and crop residue, and the erosion-runoff-leaching fraction. For the point loads, we varied: per capita protein consumption, P to N ratio in protein intake, N content in protein intake, P excreted as urine or faeces, P load from nonsewered wastewater, P lost in the stabilization pond, the coverage of sewage connections, P removal with sewage treatment, and P load from detergent. Finally, for the GWF we varied the difference between the maximum allowable and natural concentrations. The Monte Carlo simulation was done at 30 arc min spatial resolution (close to 30,000 grid points globally).

3. Results

3.1. Phosphorus Loads From Diffuse Sources

Table 1 presents a global overview of P inputs to agricultural soils (through mineral fertilizer, manure, and irrigation water) and P outputs (through P removal with crop harvests and crop residues, and through erosion, runoff, and leaching) per major crop category. In the period 2002–2010, the total global P input was 24 Tg of P per year. Mineral fertilizer contributed most to the input of P in croplands, accounting for 71% of the global P input. Manure and irrigation water contributed another 24% and 5% of the total input, respectively. The P input from mineral fertilizer is dominated by cereals, which account for 55% of the total P input from mineral fertilizer. Next come oil crops with a contribution of 17% and vegetables with 6%. The P input through manure application is largest for fruits and vegetables, which contribute 26% and 20% to the total P input through manure, respectively. Cereal crops account for the largest P removal through crop harvest (62%) and crop residues removal (60%). Next are oil crops, which contribute 18% to the total P removed

Table 1
Global P Inputs From Mineral Fertilizer, Manure, and Irrigation Water, P Removal With Crop Harvest and Crop Residues and P Erosion, Runoff, and Leaching to Freshwater Systems per Crop Category (Gg/yr)

Component in P balance	Cereals	Fruits	Vegetables	Oil crops	Pulses	Roots and tubers	Sugar crops	Nuts	Other crops[a]	Total
P in mineral fertilizer	9,391	757	1,024	2,859	245	553	546	124	1,500	17,000
P in manure	872	1,530	1,181	652	583	210	126	227	513	5,895
P in irrigation water	647	68.5	43.5	163	21.5	20.3	50.1	13.7	198	1,227
Total P inputs	**10,911**	**2,355**	**2,248**	**3,675**	**850**	**784**	**723**	**365**	**2,210**	**24,121**
P removed with crops harvested	6,228	179	83.8	1,842	227	136	135	1.77	1,163	9,996
P removed with crop residues	1,702	41.7	104	718	72.8	32.0	29.4	1.51	138	2,839
Total P removed with crop and crop residues	**7,930**	**220**	**188**	**2,561**	**300**	**168**	**164**	**3.28**	**1,301**	**12,835**
P surplus (available for erosion, runoff, and leaching)	**2,981**	**2,135**	**2,061**	**1,114**	**550**	**616**	**559**	**361**	**910**	**11,286**
P erosion, runoff, and leaching	185	85.8	84.4	85.5	28.0	26.9	22.2	15.2	52.2	585
Total P outputs	**8,115**	**306**	**272**	**2646**	**328**	**195**	**186**	**18.4**	**1,353**	**13,420**
P erosion, runoff, and leaching from anthropogenic sources[b]	**174**	**83.3**	**82.7**	**81.7**	**27.3**	**26.2**	**20.7**	**14.6**	**47.5**	**558**

Note. Period: 2002–2010. The bold values are the sum/total of the group - eg the total input is the sum of fertilizer, manure, and irrigation.
[a]Including fodder crops, cocoa, tea, coffee, spices, and fiber crops.
[b]The amount of P erosion, runoff, and leaching that can be attributed to anthropogenic sources is estimated based on the total P erosion, runoff, and leaching times the ratio of P inputs through mineral fertilizer and manure to total P input.

through crop harvest and 25% to the total P removed with crop residues. The annual values (for the period 2002–2010) for P inputs and outputs are presented in the supporting information.

The total average anthropogenic P erosion, runoff, and leaching from agricultural fields was estimated at 0.56 Tg of P per year. The anthropogenic P loads from the agricultural fields have grown by 27% over the study period (from 525 Gg in 2002 to 666 Gg in 2010). This increase is due mainly to the increase in P from the mineral fertilizer that has increased by 31% from 2002 to 2010 (supporting information Table S4). Our results show that globally, on average about 2.4% of the total P input in the form of mineral fertilizer and manure enters the freshwater through erosion, runoff or leaching. Close to one-third (31%) of the anthropogenic P load from applied fertilizers and manure was due to cultivation of cereal crops (mainly wheat and rice, each accounting for 12%). Other crop categories with large contributions to total P erosion, runoff, and leaching were fruits, vegetables, and oil crops, each contributing 15%. Among the oil crops, soybean, and rapeseed contributed most (5% and 3%, respectively).

3.2. Phosphorus Loads From Point Sources

The global P load to freshwater from anthropogenic point sources was about 0.91 Tg of P per year (87% domestic and 13% industry). Looking at the breakdown per country, the largest contribution to this global load from point sources was by China (27%), followed by the USA (9%) and India (6%). In terms of load per capita, the highest P load to freshwater systems from point sources occurs in countries with low treatment and P removal percentages. In Japan, with an urban wastewater treatment coverage of 67% and a P removal rate of 10%, the load from point sources is 0.26 kg P per year per capita, while in Germany, with an urban wastewater treatment coverage of 100% and a P removal rate of 86%, this is 0.07 kg P per year per capita. Table S5 in the supporting information provides the annual P load from anthropogenic point sources.

3.3. Total Anthropogenic Phosphorus Load to Freshwater

In the period 2002–2010, the global total P load to freshwater systems from the sum of anthropogenic diffuse and point sources was estimated to be 1.47 Tg/yr. About 62% of this total load was from point sources (domestic, industrial) while diffuse sources (agriculture) contributed the remainder. China contributed most to the total global anthropogenic P load, about 30%, followed by India (8%), the USA (7%), and Spain and Brazil (6% each). The spatial variation in the intensity of anthropogenic loads of P is shown in Figure 1. Relatively large loads of P per hectare are observed in South-eastern China, Spain, some places in Western Europe and the Nile Delta in Egypt. Although there is difference in intensity of P loads in these areas, there

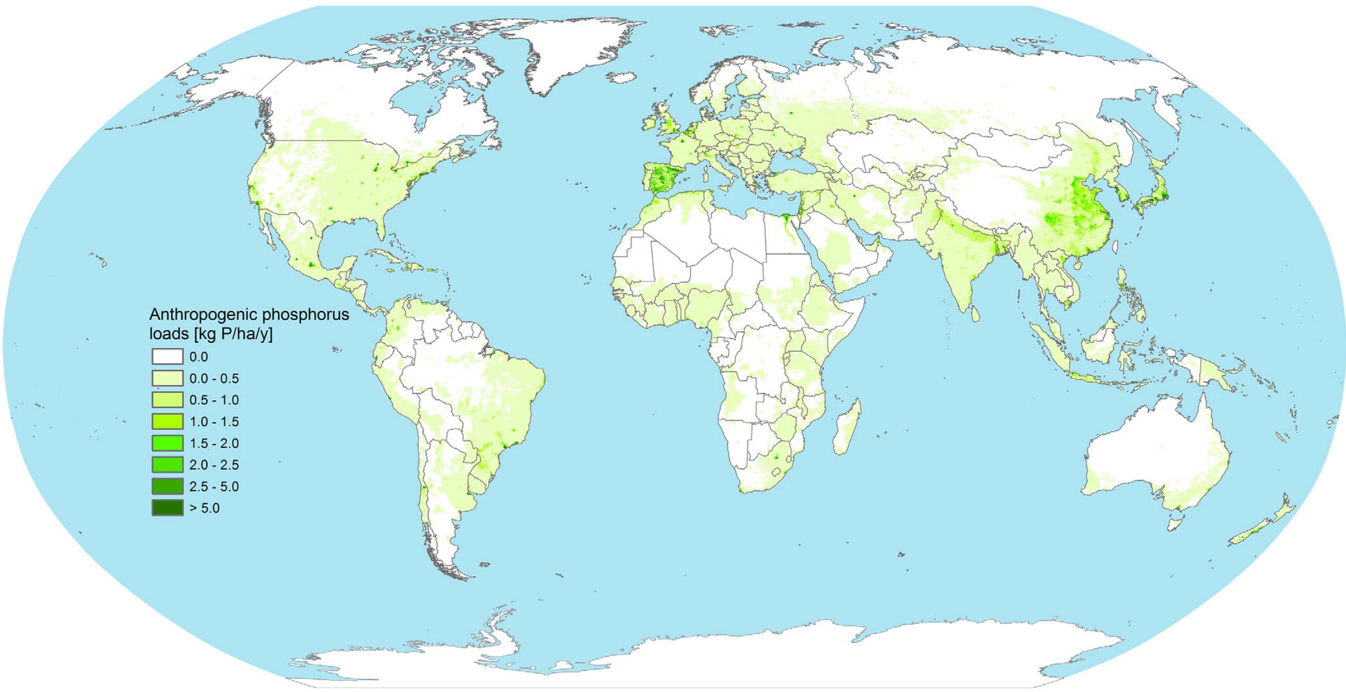

Figure 1. Global spread of anthropogenic phosphorus loads to freshwater from agriculture, industrial, and domestic sectors at a 5 × 5 arc min grid. Period: 2002–2010.

is a good agreement in identifying areas of high P loads between the current study and the result from Steffen et al. (2015). The high P loads closely follow the intensity of mineral fertilizer and manure application rates and population densities.

Figure 2 shows the contribution of major products and regions to the total global human-induced P load to freshwater. The largest contribution to the global P load (54%) comes from the domestic sector, then agriculture (38%) and finally industry (8%). About 12% of the global P load is due to cereal production (4.3% wheat and 4.4% rice), vegetable production (6.3%), and production of oil crops (5.5%, of which 1.9% related

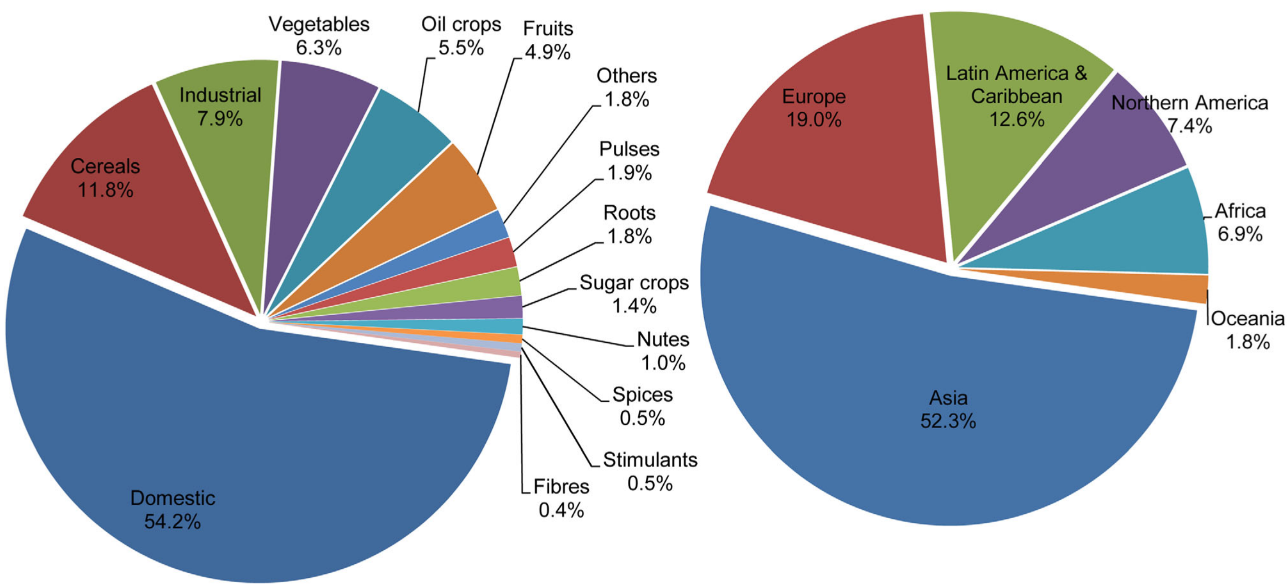

Figure 2. (left) Share of major product categories and (right) regions in the global anthropogenic phosphorus load to freshwater. Period: 2002–2010.

Table 2
Global GWF Related to Phosphorus Loads to Freshwater, Shown per Economic Sector and for the 10 Countries With the Greatest Share in the Global Total (10^9 m^3/yr)

Country	Agriculture	Domestic	Industry	Total
China	19,360	22,270	2,540	44,170
India	5,980	4,810	860	11,650
USA	1,790	7,100	967	9,850
Spain	7,340	1,010	100	8,450
Brazil	4,370	3,170	760	8,300
Russia	3,000	3,130	420	6,540
Japan	920	2,840	450	4,210
Mexico	270	2,020	430	2,710
Turkey	520	1,670	200	2,390
France	540	1,230	130	1,910
Others	11,710	30,610	4,760	47,070
World total	55,780	79,870	11,610	147,250

Note. Period: 2002–2010.

to soybean and 1.1% to rapeseed). About 52% of the global anthropogenic P load occurs in Asia (30% China). The second polluter is Europe, contributing about 19%, followed by Latin America and the Caribbean (13%) and North America (7%).

3.4. Grey Water Footprint Related to Phosphorus

In the period 2002–2010, the global GWF related to human-induced P loads to freshwater systems was 147 $\times$ 10^{12} m^3/yr (Table 2). The global GWF has increased by about 15% within the study period, with the agricultural sector showing the largest growth (by 27% from 2002 to 2010) (supporting information Table S6). Different product categories and regions contribution to this global total is the same as their contributions to the global anthropogenic P load presented in Figure 2. China dominated by contributing about 30% to the global total. India took a share of 8%, the USA 7%, and Spain and Brazil 6% each.

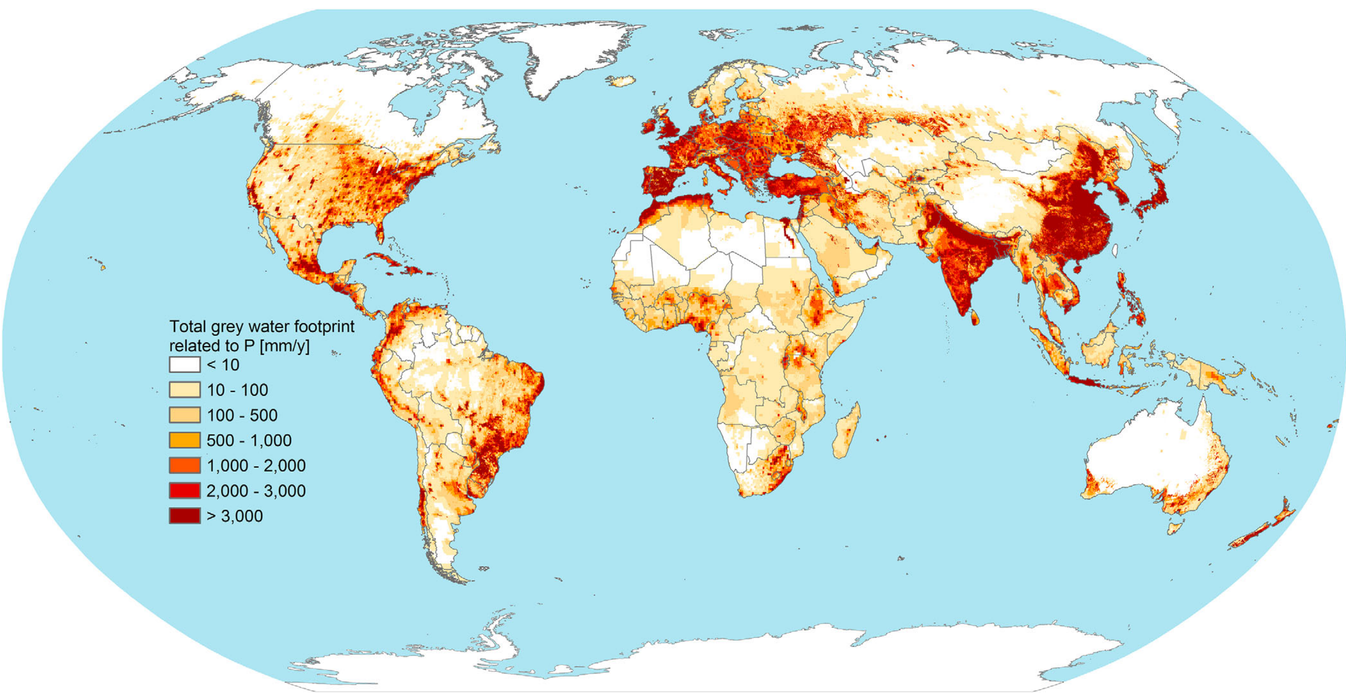

Figure 3. The GWF related to anthropogenic phosphorus loads from the agricultural, industrial, and domestic sectors in the period 2002–2010. The values are presented in mm/yr on a 5 $\times$ 5 arc min resolution. The values were calculated by dividing the GWF within a grid cell (in m^3/yr) by the grid cell's surface (in 10^3 m^2).

The geographic spread of the global GWF is shown in Figure 3. GWFs are highest in densely populated areas and regions with intensive agriculture. Large GWFs are observed in the eastern half of China, India, and Europe. Other areas with large GWF are the eastern half of the USA, the Nile delta in Egypt, Northern Africa, Turkey, and many of the coastal areas of South America.

3.5. Water Pollution Level

Figure 4 shows the WPL per river basin related to anthropogenic P loads. Together, the basins with WPL above 1 cover around 38% of the global land area (apart from Antarctica) and 37% of the global river runoff, and give habitation to around 90% of the global population. Low WPLs can be found in the basins of the Congo and Amazon and in the northern subarctic parts of Asia, Europe, and North America. Otherwise, river basins in most parts of the world have WPL > 1. In most of these basins the high WPL is due to the large human induced P loads, but in places like the Arabian Peninsula, Saharan desert, and big pieces of Australia the high WPL levels are explained by the very low runoff volumes in these areas available to assimilate P.

Grey water footprints and WPLs associated with human-induced P loads for the world's main river basins are presented in Table 3. Out of the listed 20 river basins, only three have a WPL below 1. Both the GWF (in mm/yr) and WPL indicate that the Aral drainage is the most polluted basin among the 20 basins shown, with a GWF of 84 times the assimilation capacity of the basin. About 89% of the anthropogenic P load within the Aral basin relates to domestic sector waste and 10% from industrial sector. Other severely polluted river basins include the Huang He (WPL of 45), Indus (WPL 38) Murray-Darling (WPL 23) and Ganges (11) and Yangtze (9.5) and Danube (9.3). The contributions of the different economic sectors and agricultural products to the P loads in these basins differ. For example, in the Indus and Ganges river basins, the disposal of large quantities of sewage from the domestic sector contributed 75% and 70% to the total anthropogenic P loads, respectively. The industrial sector further contributed 15% and 9% in the Indus and Ganges river basins, respectively. The production of cereal crops in the Indus and Ganges basins contributed about 10% and 17% to total the anthropogenic P load, respectively. On the other hand, in the Yangtze river basin, intensive agriculture, primarily linked to the production of vegetables, wheat, rice, maize, oil crops, and fruits, was dominant, contributing 80% to the total P load in the basin. The discharge of domestic wastewater contributed 18% to the total human-induced load in the basin.

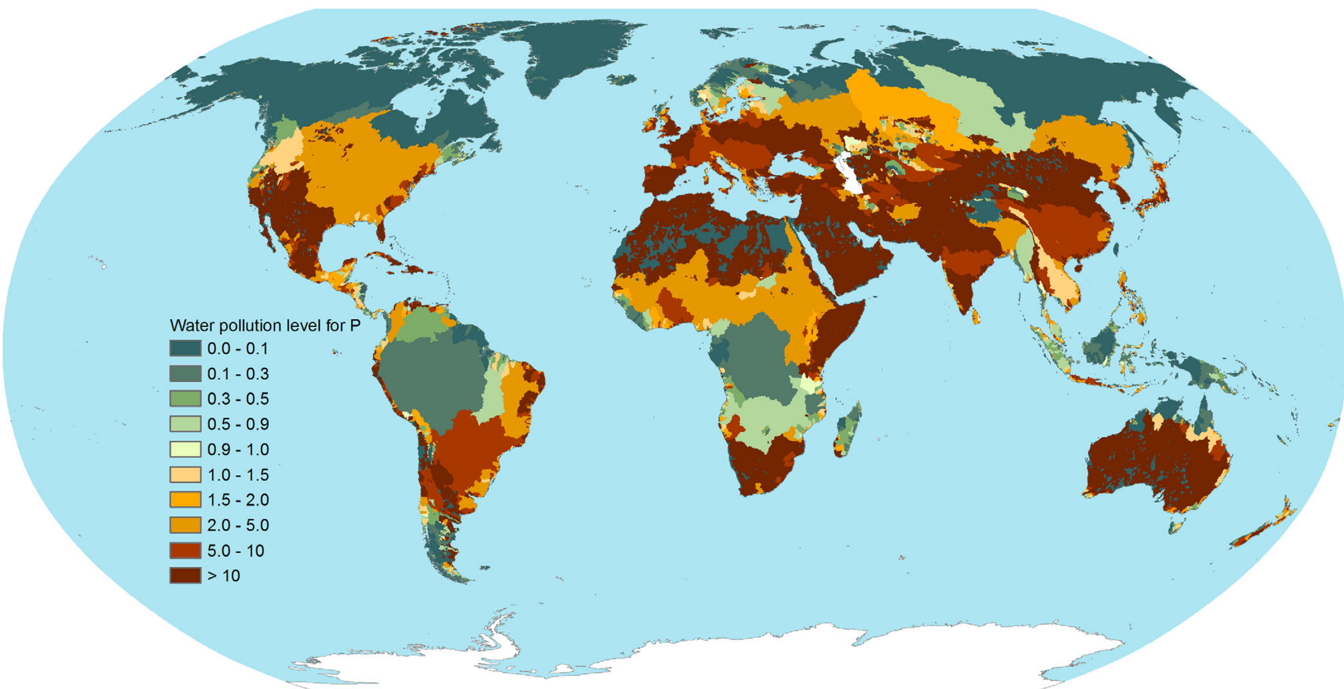

Figure 4. WPL per river basin related to human-induced P loads from the agricultural, industrial, and domestic sectors. Period: 2002–2010.

Table 3
GWF and WPL Associated With Anthropogenic P loads for 20 River Basins

River basin	Annual runoff (10^{12} m^3/yr)	Basin area (10^3 km^2)	Population (10^6)	Grey water footprint		WPL
				10^{12} m^3/yr	mm/yr[a]	
Amazon	6,590	6,070	26	1,540	254	0.23
Congo	1,270	3,760	66	280	75	0.22
Yangtze	903	2,340	384	8,600	3,689	9.5
Mississippi	623	5,630	73	2,960	527	4.8
Parana	542	3,090	69	3,400	1,100	6.3
Mekong	482	880	52	690	782	1.4
Ganges	397	1,220	417	4,200	3,459	11
Ob	396	8,780	26	790	90	2.0
Amur	362	4,830	66	1,760	365	4.9
Niger	330	2,270	74	860	378	2.6
Nile	326	3,090	145	1,340	432	4.1
Zambezi	325	1,490	30	250	170	0.78
Volga	269	4,560	59	1,330	292	4.9
Xi Jiang	221	420	63	1,630	3,894	7.4
Danube	208	1,670	82	1,940	1,158	9.3
Indus	148	1,210	150	5,570	4,592	38
Rhine	76	380	50	650	1,696	8.5
Aral Drainage	70	1,030	27	5,860	5,684	84
Huang He (Yellow)	49	1,200	121	2,200	1,832	45
Murray-Darling	18	1,100	2	400	366	23

Note. Period: 2002–2010.
[a]GWF in mm/yr has been computed as the GWF within a basin (in m^3/yr) divided by the basin's area (in 10^3 m^2).

4. Discussion

4.1. Comparison With Other Studies

Our estimate of global P erosion, runoff, and leaching from diffuse sources is 40–85% smaller than the earlier estimates by Bouwman et al. (2009, 2013), 90% lower than the estimate by Peñuelas et al. (2013), and 3.75 times larger than Seitzinger et al. (2005) (Table 4). In addition to croplands, Bouwman et al. (2009, 2013) include P inputs to grassland, which may (partly) explain their higher values. But most importantly, Bouwman et al. (2013) and Peñuelas et al. (2013) have assumed a larger fraction of the total P input in the

Table 4
Total P Loads From Diffuse Sources as Estimated in the Current Study Compared to Previous Studies

Study	P erosion-runoff-leaching to freshwater (Tg P/yr) from diffuse sources	Study period	Erosion-runoff-leaching fraction[c]
Bouwman et al. (2013)	4	2000	33% (12.5%)
Bouwman et al. (2009)	1	2000	9% (4.8%)
Peñuelas et al. (2013)	5[a]	2005–2011	(23%)
Seitzinger et al. (2005)	0.16[b]	1995	
Current study	0.6	2002–2010	5.2% (2.8%)

[a]Taking only the erosion, runoff, and leaching from fertilizer and manure P input. The total P erosion, runoff, and leaching was 13.5–25 Tg, the largest share coming from land use, which includes changes in land use, crop management, and deforestation.
[b]Taking only anthropogenic contribution to the dissolved form of P (DIP and DOP). Seitzinger et al. (2005) estimate the DIP = 1.09 Tg P/yr with 4.4% coming from diffuse anthropogenic and DOP = 0.67 Tg P/yr with 17% from diffuse anthropogenic. The particulate P estimate by Seitzinger et al. (2005) was 9.03 Tg/yr but they do not show the contribution of human-induced versus natural sources.
[c]Values outside the brackets refer to the erosion-runoff-leaching fraction over the surplus (Input-Output) and values within the brackets refer to the erosion-runoff-leaching fraction over the total input.

form of mineral fertilizer and manure to enter freshwater through erosion, runoff, and leaching. In the current study, we have used the surplus approach as suggested by the GWF Accounting Guidelines (Franke et al., 2013), which takes into account local conditions such as precipitation and soil characteristics. The P surplus that is available for erosion, runoff, and leaching as calculated in the current study is in good agreement with the estimate by Bouwman et al. (2013), but because of the large difference in the erosion-runoff-leaching fraction between the two studies, the estimated final P loads are quite different.

The erosion-runoff-leaching fraction in the current study (2.8% of the total P input or 5.2% of net P surplus) is quite low compared to values used in the other studies (Table 4). There is a need to further re-evaluate the erosion-runoff-leaching fractions suggested in the GWF Accounting Guidelines (Franke et al., 2013). An extensive literature review and further field studies will be needed to come up with better estimates.

4.2. Uncertainties and Limitation of the Study

There are a number of uncertainties involved in the estimated P loads to freshwater from both point and diffuse sources. These uncertainties are mainly due to the various assumptions that had to be made to fill the data gaps. Beside the uncertainties mentioned in our earlier study for N (Mekonnen & Hoekstra, 2015), which are valid for the current study as well, there are other uncertainties and limitations specific to the current study. Particularly, there could be underestimation in the loads because diffuse sources from grassland were not accounted for. According to Bouwman et al. (2013), the bulk of the P budget (92%) is from croplands, while the contribution of grassland to the total P budget is only 8%. Nevertheless, the underestimation in P loads by not including the P leaching and runoff from grazing lands from manure that falls on grassland could be significant.

The sensitivity assessment shows that with assumed ±20% ranges in the input variables, the total anthropogenic P load will have a standard deviation from the average estimate of ±12% and the total GWF will have a standard deviation of ±40% (Table 5). When we assume that the ±20% ranges in the input variables sufficiently capture the uncertainties in the inputs, the standard deviations for P load and GWF reflect the uncertainties in these estimates. The relatively large standard deviation in GWF results from the fact that GWF is very sensitive to the assumed maximum allowable and natural concentrations. The sensitivity of estimated P loads and GWF values to the various input variables highlights the need for better quality input data in general and basin-specific data on the maximum allowable and natural concentrations in particular. Regarding uncertainties around the other input variables, we observe that the manure P excretion and application are less reliable than the mineral P fertilizer use that are derived from IFA et al. (2002), FAO (2012b), and Heffer (2009). Removal of P with crop and crop residue is also less reliable. P uptake by crops was calculated with a fixed global average value per crop, which does not take into account differences in crop varieties. Besides, the residue removal also does not take into account differences within a country. Future research areas include the development of a database on the maximum allowable and natural concentrations per basin at the global level. The application of more sophisticated models to estimate P loads and the derivation of erosion-runoff-leaching fractions is another area for future research.

The results of this study may stimulate national governments to formulate GWF reduction targets, which account for the assimilation capacity of the water system. These targets can further be transformed into maximum acceptable loads per river basin and downscaled to the different sources. Many national water

Table 5
Results From the Monte Carlo Analysis, in Which the Major Input Variables and Parameters are Varied Within a Standard Deviation of 20% of Their Central Estimate, Using a Normal Distribution

	(Gg/yr)		(SD/Mean)
Model output	Mean	SD	CV
P Input (mineral fertilizer + manure)	22,895	1,130	4.9%
P removal with crop and residue	12,835	742	5.8%
Diffuse P loads	558	52	9.4%
Point P loads	910	224	25%
Total P load	1,468	175	12%
The Global Total Grey WF Related to P and the Standard Deviation (km^3/yr)			
Grey WF	147,250	59,546	40%

policies lack a proper translation of what agreed water quality standards imply for the maximum loads from different sources. The current global assessment shows in which catchments P loads exceed critical loads and relates those loads to their source, down to the level whereby we even know whether it is the wheat fields in a specific catchment that contribute most or the maize fields, or one of the other 126 crops distinguished. We would recommend more detailed studies in specific catchments for the purpose of formulating specific policy. We also think that it will be useful to relate to the work on planetary boundaries or the down-scaled upper tolerable P application rates defined by Carpenter and Bennett (2011) and Steffen et al. (2015).

In order to reduce the loads from point sources, wastewater treatment coverage and P removal rates need to increase radically across the world. Reducing the application of P in agriculture as necessary environmentally, is possible in many cases without affecting agricultural productivities, and economically beneficial as well because of the savings in fertilizer use (Vitousek et al., 2009). One way of reducing the application of P without affecting crop yield levels is by utilizing residual soil P. The accumulated residual soil P or legacy P from past excessive mineral fertilizers and animal manure applications is a valuable resource but can at the same time cause water pollution (Rowe et al., 2016). Therefore, utilization of legacy P will reduce the need for additional fertilizer P application thereby reducing P that potentially will erode and runoff to freshwater (Rowe et al., 2016; Sattari et al., 2012). Sattari et al. (2012) estimate that by accounting for the legacy P, average global P mineral fertilizer requirement in 2050 will be as much as 50% less than other estimates that did not account for the legacy P. The global meat production is expected to double in the period 2000–2050 (Steinfeld et al., 2006) with further intensification of the livestock sector (Pelletier & Tyedmers, 2010). Intensive animal production often produces large quantity of manure that cannot be fully recycled on the nearby land and can seriously affect freshwater systems. Therefore, managing manure is very important for improving the soil nutrient without further causing water pollution.

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