Supporting Information

Global riverine N and P transport to ocean increased during the twentieth century despite increased retention along the aquatic continuum

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Contents

SI1. Model approaches for river nutrient transport

Available estimates of global river export are often based on lumped approaches that combine all processes between the soil and the river mouth in a few parameters in regression models, e.g. models for dissolved inorganic N (Meybeck, 1982;Peierls et al., 1991;Howarth et al., 1996;Kroeze and Seitzinger, 1998;Caraco and Cole, 1999) and models for dissolved inorganic and organic, and particulate N, P, C (Seitzinger et al., 2005;Seitzinger et al., 2010;Mayorga et al., 2010). The lumped regression models are limited, however, in that they consider sources and sinks to be homogeneously distributed in space, do not separate terrestrial from in-stream processes, and rarely account for nonlinear interactions between sources and biogeochemical processes. Other hybrid approaches such as SPARROW; SPAtially Referenced Regression On Watershed attributes (Smith et al., 1997;Alexander et al., 2008) expand on conventional regression methods by using a mechanistic model structure in correlating measured nutrient fluxes in streams with spatial data on nutrient sources and landscape characteristics. However, such regression approaches generally apply to a limited time period are not appropriate for analysis of long-term changes.

There is a range of continuous or event-based distributed watershed-scale models available which simulate all the components of a landscape based on the hydrology (Table SI 1). A common aspect of these models is that they all require large amounts of data that may be difficult to obtain in all countries, especially for long-term analyses, and that many models focus on N and ignore P (Borah and Bera, 2003).

SI2. IMAGE Global Nutrient Model (GNM) Preprocessing of agricultural statistics
Description of management of statistical data

SI2.1 Data
The IMAGE-Global Nutrient Model (GNM) uses two input files: (i) The upt_<year>.csv (files with crop production, N-fixation data, fertilizer use efficiency, crop, grass and fodder crop uptake); (ii) nutdata_<year>.csv (data for livestock systems, emissions, fertilizer data). The preprocessing consists of the collection of all the data in these two files from statistics for all countries of the world from FAOSTAT (FAO, 2015b, a), and subnational data for USA (USDA-National Agricultural Statistics Service, 2015), China (National Bureau of Statistics of China, 2014;China Ministry of Agriculture, 2014;China Livestock Yearbook Editing Committee, 2014), and Europe (European Commission, 2015). FAOSTAT data cover the period 1961 till the most recent year. USDA has data cover a longer period; the start year for Chinese data is variable; for all data sources the rule is that if available 1961-most recent year is used; otherwise the earliest year with available data; missing years are interpolated; The distribution of subnational data for the first available year is used together with FAO data for the whole country for preceding years. If data for similar categories is available, the trend for that item is used to compute preceding years for the item with missing data.
Data on crop production, livestock, and fertilizer use is from FAO. For countries where subnational data is available (USA, China, India) the data are scaled so that the national total matches the FAOSTAT data.

Crops are first grouped according to the 34 crops distinguished by FAO Agriculture Towards 2030 and 2050 studies (Alexandratos and Bruinsma, 2012;Bruinsma, 2003). Grass is the 35th group. Using estimated N, P_2O_5 and K_2O contents for the 34 crop groups, the amounts of nutrients in the harvested parts are computed for each country, state (USA, India) or province (China). This information is used to distribute information generated by IMAGE at the scale of world regions to the country scale. This procedure warrants consistency between IMAGE output and GNM calculations at the country and grid scale.

IMAGE distinguishes 7 crop groups, i.e. temperate cereals, rice, maize, tropical cereals, pulses, root and tuber crops, oilcrops. The 7 crop groups are taken from IMAGE output AGRPRODC.out. Crops that are not provided by IMAGE ("other crops", crop group 8) are taken from the 34 FAOSTAT crops and added to the category of upland crops.

The upt_<year>.csv (files with crop production, N-fixation data, fertilizer use efficiency, crop, grass and fodder crop uptake) files (see B) distinguishes the 7 IMAGE crops, group 8 of "other crops", group 9 (sugar can and maize biofuels) and group 10 (woody biofuels) are taken from AGRPRODB.out and group 11 (grass) is taken from IMAGE output files AGRPRODP.out.

For calculations and presentation the 11 groups are again grouped to 4 groups, i.e. legumes, wetland rice, upland crops and grass. Fodder crops can be legumes, but these are grouped with upland crops. N fixation is calculated for the legumes group and the leguminous fodder crops, but assigned to either legumes or upland crops.

The file nutdata_<year>.csv includes country, state or province scale data and information. For each geographical unit, ten animal categories are distinguished, i.e. nondairy cattle, dairy cattle, buffaloes, pigs, poultry, sheep and goats (one category), horses, mules, asses and camels. The files also contain statistics on fertilizer use by crop, and fractions of 11 types of N fertilizers.

This preprocessing consists of the retrieval of the following data from the statistics for the following items:

- N and P_2O_5 fertilizer per country per year
- Crop production per country per year aggregated to 34 crop groups
- Fodder crop production per country per year for 15 fodder crops
- Stocks (number of heads) for animals per country per year for 10 animal groups.
- Number of slaughtered animals (heads) per country per year for 10 animal groups.
- Crop harvested area per country per year for the 7 IMAGE crop groups and group 8 ("other crops").

Data are converted to MyM-format (Beusen et al., 2011). This MyM-conversion is the transformation to files with years as columns (see C1). In the current version, the converted annual data from FAOSTAT per year is used directly, instead of the running 3- or 5-year average as in IMAGE.

SI2.2. Program

preproc.py is the main program, which generates all the upt_<year>.csv (files with crop production, N-fixation, fertilizer use efficiency, crop, grass and fodder crop uptake) and nutdata_<year>.csv (data for livestock systems, emissions, fertilizer data) files which are located in the output directory "output". The years that are generated are read from the FAOSTAT data.

SI 2.3. Methodology

SI 2.3.1. Region definition

The python programme region_info.py does the coupling between isocodes and IMAGE regioncodes by combining two files:
region_file_FAO_output = r"region_info/IsoFaoNbal.dat"
region_file_image = r"region_info/isofao26.dat"
These files are also used for converting the FAOSTAT data into the MyM file format.

SI 2.3.2. Crop properties

The crop properties such as N content, P_2O_5 content and dry matter fraction are read from:
crop_properties_file = r"input/crop-composition.csv"
nocrop_properties_file = r"input/nocrop-composition.csv" (grass, maize/sugarcane, woody biofuels)
nfix_legumes_file = r"input/nfix_legumes.csv"

IMAGE crops (9 = sugar cane and maize; 10 = woody biofuels; and 11 = grass) are read from the nocrop_properties_file.
The nfixation parameters for soybeans, groundnuts, pulses and other fodder legumes are read from the file
nfix_legumes_file.

SI 2.3.3. Crop production

Crop production of the 34 crops (excluding grass) are read from FAO. Also USDA information for US per state and for China per province. In crops_us.py the USDA file is read and the production per state is returned. Idem for China. The split_countries.py programme splits the country information of France, Italy and Spain into regions based on Eurostat information. The distribution information for the production is given in the
distribution_file_production = r"input/other_countries/production.csv"
The distribution file for production must be given for 35 cropgroups (including grass).
The distribution_file_production is used as a weight. The numbers are converted into fractions per region per year.
Example:
250;250001;Bretagne,1;130
250;250002;France_without_Bretagne,1;260
Total for dimension 1 is 390. This means 1/3 goes to Bretagne and 2/3 to rest of France.
If the FAOSTAT value for dimension 1 is 9000, then this information will result into 3000 to Bretagne and 6000 to the rest of France.

If USDA or Chinese data do not provide information per state or province for this crop, then we use a weight, i.e. the total production of all crops per state compared to the total crop production of the US. So states or provinces with large volumes of crop production will also have large share of these "missing" crops.
The difference between China/US and FAOSTAT are given in the upt_<year>.csv.

The above crop production information is used to distribute information generated by IMAGE at the scale of world regions to the country scale. This procedure warrants consistency between IMAGE output and GNM calculations at the country and grid scale.

SI 2.3.4. Nutrient contents, crop uptake

Based on the FAOSTAT crop production and the N content, P_2O_5 content and dry matter fraction, the N and P_2O_5 uptake are calculated per country and for the 3 crop groups. Nutrient contents and dry matter fractions are in input directory in file crop_composition.csv and nocrop_composition for grass and biofuels (directory input).

N fixation is calculated per country for legumes. Also the N content and P_2O_5 content is calculated for the 7 IMAGE crops plus group 8 "other crops" per IMAGE region. The oil crops (crop group 7 in uptake files) is divided into two groups. One is added to upland crops and the second (soybeans and groundnuts currently; this can be modified if necessary) is added to legumes. The N and P_2O_5 content and dry matter fraction is also calculated for these groups. This all is based on the FAOSTAT crop production.

SI 2.3.5. IMAGE crop and grass production tabular data

The IMAGE crop and grass production data is available from three files:
AGRPRODC.OUT (production of the 7 IMAGE crop groups),
AGRPRODB.OUT (biofuel crop production) and
AGRPRODP.OUT grass production).
The compilation of this data is performed by image_production.py

SI 2.3.6. IMAGE crop and grass distribution grid data

The IMAGE grid unformatted data (GFRAC_<year>.19.UNF0) on the distribution of crops and grass within grid cells is grouped into the four land use classes which GNM distinguishes: upland crops, legumes, wetland rice and grassland in get_image_area.py. IMAGE distinguishes two types of grass, i.e. extensive (marginal), and all other grassland. Here, grassland is classified as intensive (mixed) or extensive (pastoral) on the basis of the fraction cropland in the grid cell. Cropland occurring in a grid cell with extensive or intensive grassland is then also classified as such.

The total land area of each cell (excluding water and urban area) is also converted from UNF format to ascii grid.

The file GFRAC_<year>.19.UNF0 contains 19 fractions for each gridcell. The order of the GFRAC array is as follows:
IMAGE output rainfed crops (0-11):
0=grass , 1=temp.cer, 2=rice, 3=maize, 4=trop.cer.
5=pulses, 6=roots&tubers, 7=oilcrops
8=sugar cane; 9 = maize; 10 = woody; 11=nonwoody biofuel crops
IMAGE IRRIGATED crops (12-18)
12=temp.cer,13=rice,14=maize,15=trop.cer.
16=pulses,17=roots&tubers,18=oilcrops

The file GLCT_<year>.UNF1 contains the land cover type. Here only number 1 is used (agricultural land).

The fraction croparea soybean and groundnuts (soyfraction) of the total oilcrop production area is calculated from FAOSTAT (in fr_soybean.py) based on the croparea provided by FAOSTAT.

The upland crop, legumes and wetland rice are calculated from GFRAC with:
wrice = gfrac(Ducharne et al.) + gfrac[13]
legumes = gfrac[5] + gfrac[16] + soyfraction * (gfrac[7]+ gfrac[18])
upland_crops = sum(gfrac[1:18]) - wrice - legumes
Total grass = gfrac[0:]

The grassfactor (value given for each IMAGE region, fr_grass) is used for each cell. This fraction is used to decide whether the grid cell is classified as intensive or extensive. Generally grid cells are intensive when grass fraction is smaller than fr_grass (this implies that arable land exceeds this fraction, i.e. there is a system with exchange of feed and manure), else it is extensive (arable land is less than this fraction, i.e. grassland is dominant, and no exchange of feed and manure). Grassland and cropland within a grid cell have the same classification (intensive or extensive). For the regions Western Europe and Japan, all crops and grass are intensive. For US and Canada, grid cells are intensive when the grass fraction exceeds fr_grass. In IMAGE class extensive grassland (marginal), i.e. GLCT = 2 all grass or crops are classified as extensive.

All gridfiles are saved to output directory/grids. For the period 1961 – last year FAO, for all years grids are provided (interpolation between the given IMAGE years).

SI 2.3.7. Fertilizer data

The distribution of fertilizer over the four crop groups is done in fertilizer.py.
First the basic file fubc_file = r"input/FUBC_2000_mother.xlsx" with sheet fubc_sheetname = "FUBC5" is read. This is the data published by FAO, IFA and IFDC (FAO/IFA/IFDC, 2003). More recent inventories of fertilizer use by crop (http://www.fertilizer.org//En/Statistics/Agriculture_Committee_Databases.aspx) cover only 23 countries and lack data on fertilizer use in grassland, while FUBC5 includes a much larger number of countries. In FUBC5 the total N and P_2O_5 fertilizer is provided per country and per crop or grass with the area where the fertilizer is applied. This information is used to calculate an average application rate (kg/ha) per country for the four crop groups. This is used as the application rate for the year 2000.

The data for missing countries are assigned a regional average (in this case IMAGE region) application rate per crop based on the given FUBC. For grass the regional average application rate is not used and set to zero.

The next step is to obtain the crop area from IMAGE USDA and Chinese data for all crops and aggregate this for each country to the 4 crops (IMAGE_area). The N or P_2O_5 crop yield is now calculated as the N or P_2O_5 uptake (based on FAOSTAT, see C4) divided by the harvested area. The key for distributing the fertilizers is now:

Nfertbase = (Nfertilizer[year_2000]/area_fubc[year_2000])/(Nuptake[year_2000]/IMAGE_area[year_2000])

FAOSTAT does not provide data on grass production. Instead, grass production and grass area is taken from IMAGE per IMAGE region as a proxy from the output file AGRPRODP.out.
For grass we use:

Nfertbase = (Nfertilizer[year_2000]/area_fubc[year_2000])/(grassprod[year_2000]/IMAGE_grassarea[year_2000])

Prior to 1960 we assume zero fertilizer use in grassland. From 1960-1970 the calculated grass fertilizer is reduced by 0, 0.1, 0.2, 0.3, ... 1.
Based on Nfertbase and Pfertbase a distribution is created for each year for upland crops, legumes, and wetland rice based on:

Nfertilizer_distribution[iyear][icrop][ireg] = IMAGE_area[iyear][icrop][ireg] * Nfert_base[ireg][icrop] * Ncropyield[iyear][icrop][ireg]

and for grass:
Nfertilizer_distribution[iyear][igrass][ireg] = Nfert_base[ireg][igrass] * (grassprod /IMAGE_area) * IMAGE_area = Nfert_base[ireg][igrass] * grassprod

Thus each country has a fertilizer_distribution for the 3 crops (upland crops, legumes, and wetland rice) and grass. This is the distribution key to disaggregate the country N and P_2O_5 FAO fertilizer over the 4 crops in all years.

The split_countries.py splits the country information for France, Italy and Spain into regions. The distribution information for the fertilizer is given in the distribution_file_Nfertilizer = r"input/other_countries/Nfertilizer.csv" for the four crops and distribution_file_Pfertilizer = r"input/other_countries/Pfertilizer.csv" for the four crops.

SI 2.3.8. Livestock

Animals are from the FAOSTAT data. For China and US the local information is read. For China not all years are given. Here the first or last year per animal given is used. Also for the US no all animals are known. Only the first five classes are included. We use the sum of cattle and sheep&goats as a proxy to fill in the other animal classes (like horses, camels, mules, asses etc.).

The split_countries.py splitting the country information of France, Italy and Spain into regions. The distribution information for the animals is given in the distribution_file_animals = r"input/other_countries/animals.csv" for the 10 animal groups.

SI 2.3.9. Fodder crop data

Fodder crop production is retrieved from FAOSTAT. Note that this is not in the regular data. The fodder crop production for the states of the US and provinces of China is not available. The distribution of fodder crop production for China, USA over the states/provinces is based on the distribution of cattle, dairy and pigs (class 1 , 2 and 4). Splitting the country data for France, Italy and Spain into regions is done in the same way.

```
# [Class] 1 ! Maize for forage and silage
# 5510 636
# [Class] 2 ! Sorghum for forage and silage
# 5510 637
# [Class] 3 ! Rye grass for forage and silage
# 5510 638
# [Class] 4 ! Grasses Nes for forage and silage
# 5510 639
# [Class] 5 ! Clover for forage and silage
# 5510 640
# [Class] 6 ! Alfalfa for forage and silage
# 5510 641
# [Class] 7 ! Green Oilseeds for silage
# 5510 642
# [Class] 8 ! Leguminous for silage
# 5510 643
# [Class] 9 ! Cabbage for fodder
# 5510 644
# [Class] 10 ! Turnips for fodder
# 5510 646
# [Class] 11 ! Beets for fodder
# 5510 647
# [Class] 12 ! Carrots for fodder
# 5510 648
# [Class] 13 ! Swedes for fodder
```

5510 649
[Class] 14 ! Forage products
5510 651
[Class] 15 ! Vegetables Roots Fodder
5510 655
[Class] 16 ! Pumpkins for fodder. Pumpkins are not used. See personal comments Luis.
5510 645

Fodder products are split in two classes, i.e. (i) fodder for nondairy and dairy cattle (cattle) (ii) fodder for nondairy, dairy cattle and pigs (all).
product_class =
["636","637","638","639","640","641","642","643","644","646","647","648","649","651","655"]
product_class_cattle = ["636","637","638","639","640","641","642"]
product_class_all = ["643","644","646","647","648","649","651","655"]
# Clover and alfalfa have the same N fixation as the N content of the production.
nfixation_crops = ["640", "641", "643"]
The cattle fodder production is distributed over cattle and dairy. For the US this group is only distributed over the dairy animals.
The all fodder class is distributed over cattle, dairy and pigs. Three fodder productions have N fixation, which is assumed to be equal to the N fodder production (production times the N content.

SI 2.3.10. Nutrients in fodder crops
Nutrients in fodder crops are added to the upland crops. To do so, a multiplier for additional N and P_2O_5 uptake of upland crops is calculated.
This multiplier is used to compute the additional N or P_2O_5 upland crop uptake by fodder crops as (upland+fodder)/upland.

SI 2.3.11. Fertilizer use efficiency
With all the above data we can compute the Partial Factor Productivity or Fertilizer Use Efficiency (FUE), calculated as the kg fertilizer per kg dry production on the 4 crop groups and per IMAGE region .

SI 2.3.12. Data for years prior to 1961
Country crop production data for the period 1961-1969 is taken directly from FAOSTAT, because IMAGE data is available from 1970 onwards. Prior to 1961 we use the data from Bouwman et al. (2013) for 1900 and 1950. For 1900-1930 we assume constant soil budgets, 1935-1945 budgets are equal to those in 1950. Prior to 1970 we use 5-year time steps, and from 1965 onwards annual data are produced.

Some changes were implemented in the data: (i) Two IMAGE regions were added (South Africa, India) to be consistent with IMAGE 3.0; (ii) Grass production is calculated with grass areas as basis; (iii) Grass areas were made consistent with Klein Goldewijk et al. [, 2010 #11015]; (iv) Crop uptake, livestock and N fixation distributions within IMAGE regions and crop N and P uptake for Upland crops, legumes, rice and grass and all other distributions within IMAGE regions are based on distributions for 1961-1965; (v) fodder crops are assumed to be produced from 1950 onwards with a linear increase to 1961; prior to that we assumed that food wastes have been used as animal feed. (vi) the year 1960 is equal to 1961.

SI 2.3.13. Output data

Write all information to output files upt_<year>.csv and nutdata_<year>.csv. For the nutdata only the number of animals and fertilizer use is calculated and put in the nutdata_<year>.csv. As basis of this file, and interpolation is done between mother files nutdata_<year>.csv generated in earlier studies. All data in this file is interpolated. All mother nutdata files which are used are located on input/nutsdata.

The upt_<year>.csv are created every year. The fixed_input.txt (directory input) which is included in upt_<year>.csv every year. At this moment only [NUMBER OF COUNTRIES WITH NO MAXIMUM FERTILIZER] and [ISOCODES OF COUNTRIES WITH NO MAXIMUM FERTILIZER] are listed in this file.

SI3. Data and methods
General descriptions are provided for the hydrological model (Section SI 3.1), nutrient delivery model (SI 3.2), and in-stream nutrient retention model (SI 3.3). The approach for the sensitivity analysis is described in SI 3.4). All calculations presented have a yearly time scale, and refer to total N and total P (total of all chemical forms).

SI 3.1. Hydrology
The grid-based global hydrological model PCR-GLOBWB (Van Beek et al., 2011) representing the terrestrial part of the hydrological cycle is used to quantify water stores and fluxes (Main text Figure 1a). For this study it is implemented with a daily time step and a spatial resolution of 0.5 by 0.5 degrees. All results presented in this paper refer to annual aggregated results. Twentieth century monthly precipitation and temperature are from *New et al.* (2000) and downscaled to daily values using the ERA-40 reanalysis (Uppala et al., 2005). *Beusen et al.* (2015) provide details on the water balance model, drainage network and handling of soil and vegetation.

Land cover is either natural vegetation, cropland or grassland, obtained from IMAGE (Stehfest et al., 2014). Depending on the land cover, water is delivered as specific runoff to the drainage network, consisting of direct runoff, interflow (shallow groundwater) and base flow (deep groundwater) (Main text Figure 1b) according to the approach presented elsewhere (Bouwman et al., 2013a). Each grid cell represents varying fractions of land and fresh water surfaces, the drainage network of laterally connected channels, lakes and reservoirs along which the locally generated specific runoff is accumulated and routed to obtain the river discharge.

The channel dimensions are parameterized on the basis of the Leopold and Woolhiser relationships for bank full discharge and the overbank storage capacity based on the Hydro1k dataset (more details in Winsemius et al., 2013; Van Beek and Bierkens, 2009). Where water storage exceeds the channel capacity, flooding occurs, water depth and extent depending on the elevation distribution within a 0.5 by 0.5 degree grid cell.

The river channels can be interrupted by lakes and reservoirs (Main text Figure 1c) for which the outflow is controlled by a storage-outflow relationship for lakes and by the requested downstream demand in the case of reservoirs. Each water body is specified by a volume, surface area, and depth. Lake characteristics are from the Global Lakes and Wetlands Database (GLWD1) (Lehner and Döll, 2004). Reservoirs are introduced dynamically on the basis of the reported construction year from the Global Reservoirs and Dams (GRaND) database (Lehner et al., 2011).

SI 3.2. Nutrient delivery
The calculation of surface (section SI3.2.2) and subsurface (SI3.2.3) runoff starts from soil N and P budgets (SI3.2.1) on the basis of the hydrological flows from PCR-GLOBWB. Other nutrient sources that are directly delivered to surface water included in IMAGE-GNM are discussed in section SI3.2.4.

SI 3.2.1. Soil N and P budgets
Soil N budgets have the input terms fertilizer, manure, biological N_2 fixation and atmospheric deposition; P inputs are fertilizer and manure; N and P outputs include crop and grass harvest and grazing. Annual soil N and P budgets for agricultural and natural ecosystems are calculated with the IMAGE-GNM model for the years 1900-2010 for 0.5 by 0.5 degree grid cells. Country data

are taken from FAO statistics for most countries; for USA, China, France, Spain and Italy subnational data were used. Details on the data collection and handling are in the SI2. Soil nutrient budgets for grid cells covered by natural vegetation or agricultural land represent the difference between inputs to the soil and withdrawal by plants. For agriculture, IMAGE-GNC distinguishes upland crops, legumes, rice, and grass. These land cover types are either mixed systems (with a large arable component generally close to urban areas) or pastoral systems (dominated by grassland in remote regions).

Uptake by natural ecosystems is neglected, assuming all vegetation is in a mature stage. Biological N_2 fixation rates in natural ecosystems are based on the low estimate for area coverage of leguminous plants and free-living N fixing bacteria from Cleveland et al. (1999). Global N_2 fixation calculated thus (74 Tg N yr^{-1} in 1900 and 54 Tg N yr^{-1} in 2000) brackets a recent estimate of 58 Tg yr^{-1} for pre-industrial times (Vitousek et al., 2013).

In case of a surplus of inputs over withdrawal, the soil nutrient budget is the potential loss to (i) the aquatic environment via surface runoff, leaching and groundwater transport, or (ii) gaseous loss to the atmosphere through denitrification and ammonia volatilization, or (iii) accumulation in the soil. Input terms for natural ecosystems include atmospheric deposition and biological N_2 fixation.

SI 3.2.2. Soil N and P delivery by surface runoff

Two nutrient surface runoff pathways are distinguished, i.e. erosion losses from recent nutrient applications in the form of fertilizer and manure, and soil loss including a “memory” effect related to long-term historical changes in soil nutrient inventories. Loss from recent nutrient applications of fertilizer, manure or organic matter are calculated from the N and P input terms on the basis of slope (based on Bogena et al. (2005)), land use and soil texture (based on Velthof et al. (2009; 2007)). Estimates of soil erosion losses based on slope, soil texture and land cover type are used to estimate country aggregated soil-loss rates for arable land, grassland and natural vegetation. The model keeps track of all inputs and outputs in the soil P budget, to calculate the actual P content. All inputs and outputs of the soil budget are assumed to occur in the top 30 cm; the model replaces P enriched or depleted soil material lost at the surface by erosion with fresh soil material with the initial soil P content at the bottom. Soil organic C content is used as a basis to calculate N in eroded soil material using land-use specific C:N ratios. The assumption that C content of soils for different land cover types are constant has a minor effect on N (10% change in C means only ~1% change in N); however, N in soil erosion loss will vary with changing land use.

SI 3.2.3. Subsurface N transport and loss

N leaching to groundwater and riparian zones and flow to surface water have recently been described (Bouwman et al., 2013a). Annual soil N leaching is calculated as a fraction of the surplus of the soil N budget corrected for surface runoff, based on temperature, the residence time of water and NO_3^- in the soil, soil texture, soil drainage and soil organic C.

We assume that croplands in arid climates are irrigated, allowing for leaching and denitrification. In arid regions under natural vegetation (e.g. deserts) there are various fates of N, including accumulation of nitrate in the vadose zone below the root zone (Walvoord et al., 2003), surface runoff, ammonia-N volatilization, nitrification, denitrification (Peterjohn and Schlesinger, 1990;

Schlesinger and Peterjohn, 1991). It is not possible to quantify the relative contribution of each process, but it is clear that only a negligible part of N surpluses in arid climates is lost by denitrification. It is therefore assumed that N surpluses are not prone to denitrification in soils under natural vegetation and grassland with annual precipitation < 3 mm. The global amount of this N surplus in the 3100 Mha of arid lands was 18 Tg in the year 2000.

The model ignores P leaching and transport through aquifers; given the low solubility of phosphates, it is not surprising that annual losses of this element owing to leaching are very small, apart from P-saturated soils in some industrialized countries (references in Smil, 2000b).

Denitrification in shallow groundwater (5 m thick) is a function of travel time and half-life of nitrate. The travel time distribution is calculated over the thickness of 5 m, with a maximum travel time of 100 years within a grid cell. The mean travel time is a function of the aquifer thickness, porosity and the inflow. Porosity and the half-life of nitrate depend on the lithology.

We assume that denitrification is negligible in the deep groundwater system (50 m thick), and the modeled NO_3^- outflow from deep groundwater is thus a maximum estimate. The calculation of denitrification in riparian zones is similar to that in soils with two differences. Firstly, a biologically active layer with a thickness of 0.3 m is assumed (compared to 1 m in soils), as riparian zones show strong vertical gradients. Denitrification rates are high in this topsoil due to high organic matter contents. Secondly, the effect of pH on denitrification rates is included.

SI 3.2.4. Direct delivery of nutrient sources

Recent estimates show that allochthonous organic matter input to rivers is an important flux in the global C cycle (Cole et al., 2007), but so far the global nutrient contribution of this process for river nutrient dynamics have not been investigated. Here, estimates of NPP are taken from IMAGE for wetlands and floodplains. Part of annual NPP is assumed to be deposited in the water during this flooding. The ratio of litter to belowground inputs of organic matter ranges from 30:70 to 70:30 (Trumbore et al., 1995; Vogt et al., 1986); here it is assumed that 50% of total NPP ends in the surface water. N and P inputs to the water are estimated based on a C:N ratio of 100 and a C:P ratio of 1200 (Vitousek, 1984; Vitousek et al., 1988).

Release of P from weathering of parent rock is calculated according to Hartmann et al. (2014) using runoff, background concentration based on lithological class (Dürr et al., 2005), temperature and soil type. Discharge of N and P in wastewater is taken from Morée et al. (2013), and N and P release from aquaculture is from two recent inventories (Bouwman et al., 2013b; Bouwman et al., 2011) with a spatial allocation using three weighing factors, i.e. population density, presence of surface water and mean annual air temperature.

Atmospheric N deposition to water bodies is from Dentener et al. (2006) for the year 2000, and deposition in the years before that were scaled with grid-based emissions of ammonia (Bouwman et al., 2013d). Deposition in floodplains, wetlands and river channels is ignored, because it is already part of the soil N budget.

Some known nutrient sources were ignored. Atmospheric P deposition is not modeled, because deposition rates are generally <1 kg P/ha (Graham and Duce, 1979; Meybeck, 1982; Gibson, 1997; Mackenzie et al., 1998), which is negligible compared to agricultural inputs in animal manure or fertilizers. Geological sources of N were not included and may lead to underestimation of N delivery in areas with parent rock with elevated N content (metamorphic, igneous, and, particularly, sedimentary and metasedimentary rocks) (Holloway and Dahlgren, 2002). However, the lithology data (Dürr et al., 2005) do not provide enough detail to produce a first-order estimate of N release from weathering. Finally, aquatic N_2 fixation is not considered, which may lead to underestimation of N inputs to surface water columns.

SI 3.2.5. Nutrient release from aquaculture

A model was developed by Bouwman et al. (2011) to estimate nitrogen (N) and phosphorus (P) country budgets for aquaculture production of individual species within crustaceans, bivalves, gastropods and seaweed, using country production data for the 1970-2010 period from the Food and Agriculture Organization, and scenarios based on the Millennium Assessment for 2010-2050.

A similar global model was developed by Bouwman et al. (2013a) to calculate feed and nutrient budgets for freshwater and marine omnivorous and carnivorous aquaculture finfish production. The model uses national production data for the period 1970-2010 and the Millennium Ecosystem Assessment scenarios for production and management for 2010-2050.

The data from the latter study was used in a paper (Bouwman et al., 2013b) that compares the magnitude and changes of nutrient release in different countries (China, Chile, Mexico). Nutrients from freshwater, brackishwater marine aquaculture (mariculture) are allocated in specific parts of Chile and Mexico.

Here we describe the spatial allocation of nutrient release at 0.5 by 0.5 degree resolution by aquaculture for freshwater aquaculture and mariculture for all countries of the world.

SI 3.2.5.1. Allocation of nutrient release from freshwater aquaculture

General

The production of aquaculture is now given by the Food and Agriculture Organization of the United Nations (FAO) FishStat database (FAO, 2013) for each country and for each species, type of environment (marine, brackish, freshwater), and sea. Although the Food and Agriculture Organization of the United Nations (FAO) has a wealth of information available on aquaculture production by farm or production system within a range of countries for different species and environments (see http://www.fao.org/fishery/collection/naso-maps/en and http://www.fao.org/fishery/naso-maps/fact-sheets/en/), it is still difficult to obtain global spatially explicit data on where aquaculture production is located.

For distributing the aquaculture production (P) of a country spatially (here 0.5 by 0.5 degree grid cells), we developed an allocation procedure based on a weighting factor map. To construct the weighting map, we made use of expert judgment (see next paragraph). The weighting factors W range from 0 (no chance to find any P), to an arbitrary maximum value (very likely to find P), so W and P must be positively correlated. For China we used provincial data on marine and freshwater aquaculture production to better allocate within the country (Bureau of Fisheries Ministry of Agriculture, 2014).

The weighing factor map construction

The weighing factor map is built as a combination of three factors that influence the location of the aquaculture production:

- Population density
- Presence of water bodies
- Temperature.

Population density

Population density was used as a proxy for aquaculture production, assuming that most fish production takes place close to populated areas. Two major assumptions are: (i) when there are no people living in a grid cell, a low probability of aquaculture production is assumed ($q = 0.01$); (ii) for high population densities (above x_{end} =10000 inhabitants/km^2), the probability of aquaculture production is also low ($q = 0.01$). The optimum population density (x_{opt}) is around 1000 inhabitants per square kilometer. We use the following approach based on a two parabolic functions with the equation:

$$W_{\mathrm{population}} = ax^2 + bx + c$$

Where x = population density, and $W_{\mathrm{population}}$ =probability of aquaculture production based on population. The values for a, b and c depend on the population density:

For x < 0 and $x > 10000$: $y = q$

For the left part of the function ($x < x_{\mathrm{top}}$):
$a = (q-1)/x_{\mathrm{top}}^2$
$b = -2a*x_{\mathrm{top}}$
$c = q$

For $x_{\mathrm{top}} < x < 10000$:
$a = (q-1)/(x_{\mathrm{top}}-10000)^2$
$b = -2a*x_{\mathrm{top}}$
$c = 1 + ax_{\mathrm{top}}^2$

Presence of water bodies

The Global Lakes and Wetlands Database (GLWD) (Lehner and Döll, 2004) has twelve different types of water bodies (Table SI 2). For each type of water body, we estimated the probability that freshwater aquaculture can occur. The probabilities $W_{\mathrm{waterbody}}$ for each type of water body are in Table SI 2.

Temperature

Aquaculture is not possible in regions with low temperatures. We therefore use the criterion that there is no aquaculture production in cold regions. Since global water temperature data are not available, we use mean annual air temperature as a proxy, and the limit is taken as 0°C, hence $W_{\mathrm{temperature}} = 0$ for annual temperature <0°C. This is slightly lower than the limit for air temperature, because water temperatures normally lag behind those of air.

Overall weighing factor

The overall probability of finding aquaculture in a grid cell is calculated as follows:

$$W_{\mathrm{overall}} = W_{\mathrm{population}} W_{\mathrm{waterbody}} W_{\mathrm{temperature}}$$

We use the population density and water temperature for the year 2000 and use these weighing factors for all years. So weighing factors are independent in time.

Allocation

Allocation for finfish and shellfish is calculated separately. The allocation takes place in two steps: (i) all grid cells with $W_{overall}$ < 10% of the maximum value in that country. This excludes the grid cells with lowest probability. (ii) aquaculture production is allocated to the remaining grid cells based on the ratio of the probability of the grid cell and the sum of probabilities of all grid cells within that country. (ii) Subsequently, all grid cells with a production <1000 kg fresh fish are excluded. The weighing factor for these cells are set to 0, and the allocation procedure is repeated. Where the country production is smaller than this minimum, one grid cell is allocated.

Allocation by weighing is done by the following equations:

$$P(i) = [\frac{W(i)}{SW(j)}]$$

$$SP(j) \leq Pmax(i)$$

Where *P*(i) = allocation variable in cell i (unit), *W*(i) = weighing variable in cell i (any unit), *SW*(j)= sum of weights *W*(i) of region j (any unit), *SP*(j)= sum of all values of allocated variable *P*(i) of region j (unit), and *P*max(i) = maximum value of *P*(i) in cell i (unit).

In the process of allocating the production *P* to grid cells i of country i, temporally *P*(i) may exceed *P*max(i). In that case, we have a residual Pres in grid cell i. The sum of all residual *P*'s of region j, *SP*res(j) must be allocated in the next round in cells, where we still have allocation space left, i.e. *P*max-*P* > 0. The allocation process is completed when all the *SP* is allocated. Grid cells that remain after these two steps, are assigned N and P emissions to surface water from freshwater aquaculture on basis of $W_{overall}$.

SI 3.2.5.2. Allocation of nutrient release from mariculture

Mariculture production consists of brackish water aquaculture and marine aquaculture. These two types have different allocation procedures.

Nutrients released by brackish water aquaculture production are allocated to coastal land grid cells (bordering the sea) with human population density and temperature as a weighing factor, following the procedures discussed above for freshwater aquaculture.

Nutrient releases from mariculture are allocated to coastal grid cells (sea cells bordering coastal land cells) on the basis of length of the coastline (obtained from ARCGIS), as a proxy for the presence of bays or other coastal waters partly sheltered from the influence of the open sea, i.e. places where aquaculture production could occur.

In addition to length of the coastline, aquaculture production is allocated preferentially in specific coastal types, taken from the work of Dürr et al. (2011). Tidal systems (estuaries, rias and embayments), Fjords and Fjaerds are assigned a weighing factor of 10, small deltas are assigned a weighing factor of 5, and all other coastal types a weighing factor of 1 (endorheic or glaciated, lagoons, large rivers bypassing the near-shore coastal zone, large rivers with tidal deltas, karst and arrheic coasts).

Finally, temperature was used as a weighing factor following the procedure described above for freshwater aquaculture allocation. During the allocation, grid cells that are assigned fresh-weight production of <1000 kg yr^{-1} are excluded; the weighing factor for these cells are set to 0, and the allocation procedure is repeated.

SI 3.3. In-stream nutrient retention

The spiraling approach combines hydrological (defined by the hydraulic load) and biological and chemical factors (defined by net uptake velocity) controlling retention, assuming first order kinetics is applicable (i.e., areal uptake changes linearly with concentration). Net uptake velocity is different for each element (N or P). The basic value for all water body types is modified based on temperature (following Wollheim et al. (2008a) for N and Marcé and Armengol (2009) for P). For N the net uptake velocity is further modified by N concentration, describing electron donor limitation in the case of high N loads based on Mulholland et al. (2008). The drainage network of PCR-GLOBWB represents rivers of Strahler order (Strahler, 1957) six and higher. Since small streams play an important role in nutrient retention (Wollheim et al., 2008a;Ensign and Doyle, 2006), our approach includes the parameterization of lower order streams according to Wollheim et al. (2008b).

SI 3.4. Model sensitivity

The sensitivity of the modeled delivery, retention and river export for the year 2000 to variation of 48 model parameters for N and 34 for P, respectively, and for three years (1900, 1950, 2000) is based on parameter-specific distributions between a minimum and maximum value around the standard parameter values (Table SI3). In order to limit computational load in the sensitivity analysis, the Latin Hypercube Sampling (LHS) technique (Saltelli et al., 2000) is used. LHS offers a stratified sampling method for the separate input parameters, based on subdividing the range of each of the *k* parameters into disjunct equiprobable intervals based on a uniform distribution. By sampling one value in each of the *Num* intervals according to the associated distribution in this interval, *Num* sampled values are obtained for each parameter. The number of runs (*Num)* for each year was 500 for P and 750 for N.

The sampled values for the first model parameter are randomly paired to the samples of the second parameter, and these pairs are subsequently randomly combined with the samples of the third source, etc. This results in an LHS consisting of *Num* combinations of *k* parameters. The parameter space is thus representatively sampled with a limited number of samples.

LHS can be used in combination with linear regression to quantify the uncertainty contributions of the input parameters to the model outputs (Saltelli et al., 2000;Saltelli et al., 2004). The output Y considered (see columns in Tables SI4 and SI5) is approximated by a linear function of the parameters X_i expressed by

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 \cdots + \beta_n X_n + e$$

where β_i is the so-called ordinary regression coefficient and e is the error of the approximation. The quality of the regression model is expressed by the coefficient of determination (R^2), representing the amount of variation Y explained by $Y - e$. Since β_i depends on the scale and dimension of X_i, the standardized regression coefficient (SRC_i) is used. SRC_i is a relative sensitivity measure obtained by rescaling the regression equation on the basis of the standard deviations σ_Y and σ_{Xi}:

$$SRC_i = \beta_i \frac{\sigma_{X_i}}{\sigma_Y}$$

SRC_i can take values in the interval [-1,1]. *SRC* is the relative change $\Delta Y/\sigma_y$ of Y due to the relative change $\Delta X_i/\sigma_{xi}$ of the parameter X_i considered (both with respect to their standard deviation σ). Hence, SRC_i is independent of the units, scale and size of the parameters, and thus sensitivity analysis comes close to an uncertainty analysis. A positive SRC_i value indicates that increasing a parameter value will cause an increase in the calculated model output, while a negative value indicates a decrease in the output considered caused by a parameter increase.

The sum of squares of SRC_i values of all parameters equals the coefficient of determination (R^2), which for a perfect fit equals 1. Hence, SRC_i^2/R^2 yields the contribution of parameter X_i to Y. For example, a parameter X_i with $SRC_i = 0.1$ adds 0.01 or 1% to Y in case R^2 equals 1.

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SI 4. SI Tables

Table SI1. Examples of models for watershed-scale distributed simulation models of nutrient transport in river basins.

Model	Temporal scale	Description	Reference
AnnAGNPS	Day or less	Annualized Agricultural nonpoint-source pollution model, annualized version of AGNPS for continuous simulation of hydrology, erosion, transport of nutrients, sediment and pesticides	Young et al. (1995); Bingner and Theurer (2013)
ANSWERS-continuous	Day or less	Areal Nonpoint Source Watershed Environment Response Simulation, expanded with elements from other models (GLEAMS, EPIC) for nutrient transport and inputs	Bouraoui et al. (2002)
Hydrological Simulation Program - Fortran	Hour	Continuous watershed simulation of water quantity and quality at any point in a watershed developed for US-Environmental Protection Agency (EPA).	USEPA (2011); Skahill (2004)
SWAT	Day	Soil Water Assessment Tool to predict the impact of management on water, sediment and agricultural chemical losses in large ungauged river basins	Arnold and Fohrer (2005)
MIKE-SHE	Variable, depending on numerical stability	Comprehensive, distributed, physically based model to simulate water, sediment and water quality parameters in 2-dimensional overland grids, one-dimensional channels, and 1-dimensional unsaturated and 3-dimensional saturated flow layers, with both continuous and single event simulation capabilities	Refsgaard and Storm (1995)
Riverstrahler	Reach, decade	Riverstrahler allows for analyzing, apart from other disturbances, the impact of changing nutrient load and changing nutrient ratios, and potential saturation of retention processes such as denitrification and P retention by sediment. While in-stream processes are modelled with a mechanistic model, the delivery processes are described with coefficients, lumping soils, aquifers and riparian zones	Garnier et al. (1995); Billen et al. (2000)
INCA	Day	Integrated flow and nitrogen model for multiple source assessment in catchments	Wade et al. (2002); Whitehead et al. (1998b); Whitehead et al. (1998a)
IBIS-HYDRA	Variable, 1 day to 1 year	Land surface and terrestrial ecosystem model model IBIS with hydrology model HYDRA, used for modeling dissolved inorganic nitrogen fluxes and removal	(Donner et al., 2002;Donner et al., 2004)

Table SI 1. Probability of occurrence of freshwater aquaculture for water bodies distinguished in the Global Lakes and Wetlands Database (GLWD) (Lehner and Döll, 2004)

GLWD class		$W_{waterbody}$
1	No data, or no waterbody.	0.1
2	Lake	1.0
3	Reservoir	1.0
4	River	1.0
5	Freshwater Marsh, Floodplain	0.5
6	Swamp Forest, Flooded Forest	0
7	Coastal Wetland (incl. Mangrove,	0
8	Pan, Brackish/Saline Wetland	0
9	Bog, Fen, Mire (Peatland)	0
10	Intermittent Wetland/Lake	0.5
11	50-100% Wetland	1.0
12	25-50% Wetland	1.0
13	Wetland Complex (0-25% Wetland)	0.5

Table SI 3. Model parameters included in the sensitivity analysis, their symbol and description, for which nutrient it is used, and the standard, minimum, mode and maximum value considered for the sampling procedure. Parameters are listed in alphabetical order of their symbol.

Symbol	Description	Nutrient	Distribution[a]	Standard	Min.	Max.
A	Width factor [m]	N/P	U3	8.3	7.5	9.1
A_1	Drainage area first order stream [km^2]	N/P	U3	2.6	2.3	2.9
$A_{flooding}$	Area of flooding areas [-]	N/P	U1	1.0	0.9	1.1
B	Width exponent [-]	N/P	U3	0.52	0.47	0.57
B_{soil}	Bulk density of the soil [-]	N/P	U1	1.0	0.9	1.1
CN_{gnpp}	CN weight ratio of GNPP in flooding [-] areas	N	U3	100	90	110
$CN_{soil,crop}$	CN weight ratio of soil loss under crops [-]	N	U3	12	11	13
$CN_{soil,grass}$	CN weight ratio of soil loss under grassland [-]	N	U3	14	12.5	15.5
$CN_{soil,nat}$	CN weight ratio of soil loss under natural ecosystems [-]	N	U3	14	12.5	15.5
CP_{aomi}	CP weight ratio of gnpp in flooding areas [-]	P	U3	1200	1080	1320
$C_{sro,N}$	Correction coefficient for N in surface runoff [-]	N	U3	0.3	0.27	0.33
$C_{sro,P}$	Correction constant for P in surface runoff [-]	P	U3	0.3	0.27	0.33
D_{dgrw}	Thickness of deep groundwater system [m]	N	U3	50.0	45	55
$D_{flooding}$	Depth of flooding areas [-]	N/P	U1	1.0	0.9	1.1
D_{rip}	Thickness of riparian zone [m]	N	U3	0.3	0.27	0.33
D_{sgrw}	Thickness of shallow groundwater system [m]	N	U3	5.0	4.5	5.5
$dt50_{den,dgrw}$	Half-life of nitrate in deep groundwater [yr]	N	U3	∞	50.0	100.0
$dt50_{den,sgrw}$	Half-life of nitrate in shallow groundwater [-]	N	U1	1.0	0.9	1.1
F_{aomi}	Reduction factor for litter load to surface water [-]	N/P	U1	0.5	0.45	0.55
$F_{leach,crop}$	Reduction fraction of *N* towards the shallow groundwater system [-]	N	U3	1.0	0.9	1.0
$F_{leach,grass}$	Reduction fraction of *N* towards the shallow groundwater system [-]	N	U3	0.36	0.32	0.4
$F_{leach,nat}$	Reduction fraction of *N* towards the shallow groundwater system [-]	N	U3	0.36	0.32	0.4
f_{qgwb}	Fraction of q_{eff} that flows towards the deep system [-]	N	U1	1.0	0.9	1.1
f_{qsro}	Overall runoff fraction [-]	N/P	U1	1.0	0.9	1.1
f_{qsro}(crops)	Land-use effect on surface runoff for soils under crops [-]	N/P	T2	1.0	0.75	1.0
f_{qsro}(grass)	Land-use effect on surface runoff for soils under grassland [-]	N/P	T1	0.25	0.125	0.5

f_{qsro}(nat)	Land-use effect on surface runoff for soils in natural ecosystems [-]	N/P	T3	0.125	0.1	0.3
AOMI	Litterfall in flooding areas [-]	N/P	U1	1.0	0.9	1.1
L_1	Mean length first order stream [km]	N/P	U3	1.6	1.4	1.8
N_{aqua}	N load from aquaculture [-]	N	U1	1.0	0.9	1.1
$N_{budget,crops}$	N budgets in croplands [-]	N	U1	1.0	0.9	1.1
$N_{budget,grass}$	N budget in grasslands [-]	N	U1	1.0	0.9	1.1
$N_{budget,nat}$	N budget in natural ecosystems [-]	N	U1	1.0	0.9	1.1
$N_{conc,high}$	Retention multiplier for retention at high N concentrations [-]	N	U3	0.3	0.2	0.4
$N_{conc,low}$	Retention multiplier for retention at low N concentrations [-]	N	U3	7	6	9
N_{depo}	N deposition on surface water [-]	N	U1	1.0	0.9	1.1
N_{point}	N from point sources [-]	N	U1	1.0	0.9	1.1
$N_{uptake,crops}$	N uptake in croplands [-]	N	U1	1.0	0.9	1.1
$N_{uptake,grass}$	N uptake in grasslands [-]	N	U1	1.0	0.9	1.1
P_{aqua}	P load from aquaculture [-]	P	U1	1.0	0.9	1.1
$P_{budget,crops}$	P budgets in croplands [-]	P	U1	1.0	0.9	1.1
$P_{budget,grass}$	P budget in grasslands [-]	P	U1	1.0	0.9	1.1
$P_{budget,nat}$	P budget in natural ecosystems [-]	P	U1	1.0	0.9	1.1
Poros	Porosity of aquifer material [-]	N	U1	1.0	0.9	1.1
P_{point}	P from point sources [-]	P	U1	1.0	0.9	1.1
P_{soil}	P content of the soil [-]	P	U1	1.0	0.9	1.1
$P_{uptake,crops}$	P uptake in croplands [-]	P	U1	1.0	0.9	1.1
$P_{uptake,grass}$	P uptake in grasslands [-]	P	U1	1.0	0.9	1.1
$Pv_{f,wetland}$	Net uptake velocity for wetlands [m yr^{-1}]	P	U3	44.5	40	49
$P_{weathering}$	P content of per lithology class [-]	N	U1	1.0	0.9	1.1
$_{qtot}$	Runoff (total) [-]	N/P	U1	1.0	0.9	1.1
R_a	Drainage area ratio [-]	N/P	U3	4.7	4.2	5.2
R_b	Stream number ratio [-]	N/P	U3	4.5	4.05	4.95
R_L	Mean length ratio [-]	N/P	U3	2.3	2.0	2.6
Temp	Mean annual air temperature [C]	N/P	U2	0.0	-1.0	1.0
$v_{f,lake}$	Net uptake velocity for lakes [m yr^{-1}]	N	U3	35	32	38
$v_{f,lake}$	Net uptake velocity for lakes [m yr^{-1}]	P	U3	44.5	40	49
$v_{f,reservoir}$	Net uptake velocity for reservoirs [m yr^{-1}]	N	U3	35	32	38
$v_{f,reservoir}$	Net uptake velocity for reservoirs [m yr^{-1}]	P	U3	44.5	40	49
$v_{f,river}$	Net uptake velocity for rivers [m yr^{-1}]	N	U3	35	32	38
$v_{f,river}$	Net uptake velocity for rivers [m yr^{-1}]	P	U3	44.5	40	49
$v_{f,wetland}$	Net uptake velocity for wetlands [m yr^{-1}]	N	U3	35	32	38
V_{water}	Water volume of all water bodies [-]	N/P	U1	1.0	0.9	1.1

[a] Samples values are applied to all grid cells. For sampling, either uniform of triangular distributions are used. A triangular distribution is a continuous probability distribution with lower limit a, upper limit b and mode c, where a ≤ c ≤ b. The probability to sample a point depends on the skewness of the triangle. In the case of $dt50_{den,dgrw}$, ac=bc, and probability to sample a point on the left and right hand side of c is the same. In other cases, for example f_{Qsro}(crops) is a fraction [0,1], with standard value of 1.0. To achieve a high probability to sample close to 1.0, the triangle is designed with b=1 and c is close to 1. For some of the above distributions the expected value is not equal to the standard. Since the calculated R^2 for all output parameters exceeds 0.99, this approach for analyzing the sensitivity is still valid. The distributions used are:

U1. Uniform; values are multipliers for standard values on a grid cell basis.
U2. Uniform; values are added to the standard values on a grid cell basis.
U3. Uniform; values are used as such.
T1. Triangular; values between 0.125 and 0.5 with an expected value of 0.25.
T2. Triangular; values between 0.75 and 1.0 with an expected value of 0.995.
T3. Triangular; values between 0.1 and 0.3 with an expected value of 0.125.

Table SI 4. Standardized regression coefficient (SRC)[a] representing the relative sensitivity of N delivery, N retention and river N export representing global model results (columns) to variation in 48 parameters.

Year	1900	1950	2000	1900	1950	2000	1900	1950	2000
Parameter	N delivery			N retention			River N export		
qtot	0.15	0.15	0.24	-0.11	-0.12	-0.23	0.14	0.15	0.28
D_{rip}	-0.01	-0.01	-0.02	0.01	0.01	0.01	-0.01	-0.01	-0.02
$N_{budget,crops}$	0.05	0.08	0.26	-0.02	-0.03	-0.06	0.03	0.05	0.16
$N_{budget,grass}$	0.03	0.04	0.05	-0.01			0.02	0.02	0.02
$N_{budget,nat}$	0.38	0.30	0.20	-0.09	-0.05	-0.02	0.21	0.16	0.10
$N_{uptake,crops}$	0.01	0.01	0.06						0.03
$N_{uptake,grass}$	0.01	0.02	0.03					0.01	0.01
B_{soil}									
$CN_{soil,crop}$	-0.06	-0.09	-0.13	0.01	0.01		-0.03	-0.04	-0.06
$CN_{soil,grass}$	-0.01	-0.02	-0.03					-0.01	-0.01
$CN_{soil,nat}$	-0.05	-0.05	-0.04	0.01	0.01		-0.03	-0.03	-0.02
C_{sro}	0.04	0.07	0.18	-0.01	-0.01	-0.01	0.02	0.03	0.09
f_{qgwb}	-0.01	-0.06	-0.09		0.02	0.02		-0.04	-0.06
f_{qsro}	0.03	0.05	0.15	-0.01	-0.01	-0.01	0.02	0.03	0.07
f_{qsro}(crops)	0.01	0.03	0.11		-0.01	-0.01	0.01	0.02	0.06
f_{qsro}(grass)	0.06	0.10	0.16				0.03	0.04	0.07
f_{qsro}(nat)	0.06	0.07	0.07	-0.01			0.03	0.03	0.03
$F_{leach,crop}$	0.03	0.04	0.10		-0.01	-0.02	0.01	0.02	0.06
$F_{leach,grass}$	0.03	0.04	0.04	-0.01	-0.01	-0.01	0.02	0.03	0.03
$F_{leach,nat}$	0.40	0.31	0.19	-0.09	-0.05	-0.02	0.22	0.16	0.10
D_{dgrw}	0.00	-0.01	-0.02	0.01	0.01	0.01	-0.01	-0.01	-0.02
D_{sgrw}	-0.07	-0.07	-0.13	0.01	0.01	0.01	-0.04	-0.03	-0.07
$dt50_{den,dgrw}$	0.02	0.02	0.02						
$dt50_{den,sgrw}$	0.07	0.07	0.14	-0.02	-0.01	-0.01	0.04	0.04	0.07
Poros	-0.07	-0.07	-0.15	0.02	0.01	0.01	-0.04	-0.04	-0.08
$A_{flooding}$	0.37	0.39	0.34	-0.17	-0.15	-0.11	0.27	0.28	0.23
AOMI	0.38	0.40	0.35	-0.16	-0.15	-0.10	0.27	0.28	0.24
CN_{aomi}	-0.38	-0.41	-0.35	0.16	0.15	0.10	-0.28	-0.28	-0.24
F_{aomi}	0.38	0.40	0.35	-0.16	-0.15	-0.10	0.27	0.28	0.24
A				0.15	0.15	0.16	-0.12	-0.12	-0.12
A_1				-0.03	-0.03	-0.04	0.02	0.02	0.03
B				0.08	0.08	0.09	-0.06	-0.07	-0.07
$D_{flooding}$				-0.01	-0.01	-0.01	0.01	0.01	0.01
L_1				0.19	0.20	0.21	-0.15	-0.15	-0.16

$N_{conc,high}$				0.11	0.12	0.16	-0.09	-0.09	-0.12
$N_{conc,low}$			-0.01	0.49	0.50	0.40	-0.39	-0.39	-0.31
R_a				-0.06	-0.05	-0.08	0.04	0.04	0.06
R_b				0.07	0.07	0.08	-0.06	-0.06	-0.06
R_L				0.50	0.51	0.53	-0.38	-0.40	-0.41
Temp	-0.09	-0.08	-0.09	0.34	0.34	0.41	-0.30	-0.30	-0.36
$v_{f,lake,N}$				0.03	0.04	0.06	-0.02	-0.03	-0.04
$v_{f,reservoir,N}$						0.07			-0.05
$v_{f,river,N}$				0.38	0.38	0.38	-0.30	-0.30	-0.30
$v_{f,wetland,N}$									
V_{water}				0.01	0.01	0.01			
N_{aqua}			0.03			-0.01			0.02
N_{depo}	0.02	0.02	0.03			0.01			
N_{point}	0.05	0.11	0.22	-0.03	-0.04	-0.06	0.04	0.08	0.14

[a] Cells with no values represent insignificant SRC values; all cells with values have significant SRC, cells with no color indicate values -0.2<SRC < 0.2; green and salmon colors indicate values exceeding +0.2 and -0.2, respectively. An SRC value of 0.2 indicates that the parameter concerned has an influence of $0.2^2 = 0.04$ (4%) on the model variable considered.

Table SI 5. Standardized regression coefficient (SRC)[a] representing the relative sensitivity of P delivery, P retention and river P export representing global model results (columns) to variation in 34 parameters.

Year	1900	1950	2000	1900	1950	2000	1900	1950	2000
Parameter	P delivery			P retention			River P export		
qtot	0.27	0.23	0.17	-0.39	-0.40	-0.47	0.48	0.47	0.48
$P_{budget,crops}$		0.01	0.07					0.01	0.05
$P_{budget,grass}$									
$P_{budget,nat}$									
$P_{uptake,crops}$	0.02	0.02	0.06				0.01	0.01	0.04
$P_{uptake,grass}$	0.01	0.01	0.02					0.01	0.01
B_{soil}	-0.54	-0.59	-0.62	-0.10	-0.10	-0.13	-0.28	-0.31	-0.36
C_{sro}	0.02	0.04	0.13				0.01	0.03	0.10
f_{qsro}	0.02	0.04	0.13				0.02	0.03	0.10
P_{soil}	0.55	0.59	0.63	0.09	0.10	0.13	0.28	0.32	0.36
$F_{leach,crop}$									
$F_{leach,grass}$									
$F_{leach,nat}$									
$P_{weathering}$	0.27	0.23	0.17	-0.05	-0.05	-0.04	0.21	0.19	0.15
$A_{flooding}$	0.23	0.19	0.13	-0.02	-0.02	-0.02	0.16	0.14	0.11
$AOMI$	0.24	0.20	0.14	-0.02	-0.02	-0.02	0.17	0.15	0.12
CP_{aomi}	-0.24	-0.20	-0.14	0.02	0.02	0.02	-0.16	-0.15	-0.11
F_{aomi}	0.24	0.20	0.14	-0.02	-0.02	-0.02	0.17	0.15	0.12
A				0.24	0.24	0.22	-0.19	-0.19	-0.17
A_1				-0.14	-0.14	-0.13	0.11	0.11	0.10
B				-0.01	-0.02		0.01	0.02	0.01
$D_{flooding}$				-0.01	-0.01	-0.01			
L_1				0.31	0.31	0.28	-0.25	-0.25	-0.22
R_a				-0.27	-0.26	-0.24	0.21	0.21	0.19
R_b				0.17	0.17	0.16	-0.13	-0.13	-0.12
R_L				0.53	0.53	0.49	-0.42	-0.42	-0.38
$Temp$	0.19	0.17	0.12	0.21	0.22	0.27	-0.04	-0.06	-0.12
$v_{f,lake,P}$				0.05	0.05	0.06	-0.03	-0.03	-0.04
$v_{f,reservoir,P}$					0.02	0.10		-0.01	-0.08
$v_{f,river,P}$				0.43	0.43	0.40	-0.33	-0.33	-0.30
$v_{f,wetland,P}$									
V_{water}				0.01	0.01	0.01			

P_{aqua}			0.01						0.02
P_{point}	0.04	0.07	0.14	-0.02	-0.03	-0.06	0.04	0.07	0.15

[a] See footnote Table SI3.

SI5. Figures

Figure captions

Figure SI1. Calculated relative change in soil P content in world agricultural soils during the 20th century.

Figure SI2. N delivered to surface water from different sources (surface runoff from natural ecosystems and agriculture, groundwater, wastewater, atmospheric deposition and aquaculture) for rivers discharging in the Arctic ocean, Atlantic Ocean, Indian Ocean, Mediterranean Sea and Black Sea, Pacific Ocean, endorheic systems, and global for the 20th century.

Figure SI3. P delivered to surface water from different sources (surface runoff from natural ecosystems and agriculture, wastewater, aquaculture and weathering) for rivers discharging in the Arctic ocean, Atlantic Ocean, Indian Ocean, Mediterranean Sea and Black Sea, Pacific Ocean, endorheic systems, and global for the 20th century.

Figure SI4. Molar N:P ratio of water delivered to surface water for rivers debouching in the Arctic ocean, Atlantic Ocean, Indian Ocean, Pacific Ocean and Mediterranean Sea and Black Sea for the 20th century.

Figure SI5. Molar N:P ratio of water exported to coastal marine ecosystems for rivers debouching in the Arctic ocean, Atlantic Ocean, Indian Ocean, Pacific Ocean and Mediterranean Sea and Black Sea for the 20th century.

SI6. Movies

Movie captions

Movie SI1. Nitrogen (N) delivery in kg for all grid cells of the world, presented with 5-year interals for the period 1900-2000.

Movie SI2. Phosphorus (P) delivery in kg for all grid cells of the world, presented with 5-year interals for the period 1900-2000.

Movie SI3. Dominant source of nitrogen (N) for all gridcells of the world, obtained by accounting for all N delivery and in-stream retention in upstream gridcells, presented with 5-year interals for the period 1900-2000. Dominant sources are not presented for amounts < 1000 kg per grid cell (see e.g. desert areas).

Movie SI4. Dominant source of phosphorus (P) for all gridcells of the world, obtained by accounting for all P delivery and in-stream retention in upstream gridcells, presented with 5-year interals for the period 1900-2000. Dominant sources are not presented for amounts < 1000 kg per grid cell (see e.g. desert areas).