Electronic Supporting Material

Online Resource 1

Title: Spatially explicit fate factors of phosphorous emissions to freshwater at the global scale

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1. Details on the calculations of river volume, retention rate, transfer fractions and net removal rates

Water volume

The total volume is the sum of the volume of the rivers in the main water system, and the volume of lakes and reservoirs. River volume is the product of river width, depth and length. Width and depth are calculated according to equations 1 and 2.

$$W_{sr,i,n,m} = aw \cdot Q_i^{bw} \tag{1}$$

$$D_{sr,i,n,m} = ad \cdot Q_i^{bd} \tag{2}$$

Where aw, bw, ad and bd are constants from Wollheim et al. (2006), given in Table 1.1, and Q_i (km^3yr^{-1}) is the discharge from Fekete et al. (2002). Length is related to grid cell surface area through a shape constant S_b (dimensionless). S_b was derived by Vörösmarty et al. (2000) by a determining the coefficient from calculated length and surface areas of the world's 522 largest rivers:

$$L_{mw,i} = S_b \cdot \sqrt{A_i} \tag{3}$$

A_i (km^2) is the surface area of land in grid cell i, as calculated in ArcGIS. The relation based on S_b applies to watersheds as a whole, whereas many grid cells are or only a part of a watershed.

The volume of large lakes in the 0.5° x 0.5° model was adjusted to compensate for the source data set configuration (Vörösmarty et al. 2000). In that hydrologic model, the flow within lakes is assigned to the centerline of the water body, while the volumes are assigned geographically. I.e., in a large lake, the cells in the center of the lake will have volume corresponding to that cell's surface area and depth, but will have a flow that represents the sum of flows in upstream cells (i.e., which flow into that centerline cell). In turn, cells on the lake edge will have a volume corresponding to their true water volume but a flow that has been reduced – as the flow has been 'transferred' to the centerline. As a result, the residence times of cells on the edge of the lake are artificially high.

To compensate for this, the volumes of cells on the edge of lakes were transferred to the centerline. This was accomplished by first identifying potential lateral cells of a large lake with volume > 25 km^3. For each of these cells i, a downstream analysis was conducted, to test for the presence of a centerline: If the flow in cell i was < 5 km^3/yr and the flow in a downstream cell j (at most 3 grid cells downstream from cell i) was > 5 km^3/yr, then the volume of the cell i was moved to this downstream cell j. These volumes and flows were chosen based on a survey of the original hydrologic model data and an iterative check of the chosen parameters to ensure many large lakes' volumes were thus corrected and that the only affected cells were on the edge of lakes. This process resulted in a modified hydrologic model that avoided cells with artificially low flows and thus high residence times.

Retention rate

The retention rates of phosphorus are determined via the following relationships:

$$\begin{aligned} k_{ret,j} &= \sum_{wb} \frac{V_{wb,j}}{V_{tot,j}} \cdot k_{ret,wb,j} = \frac{1}{V_{tot,j}} \left(V_{riv,j} \cdot k_{ret,riv,j} + V_{lak,j} \cdot \frac{\nu_f}{D_{lak,j}} + V_{res,j} \cdot \frac{\nu_f}{D_{res,j}} \right) \\ &= \frac{1}{V_{tot,j}} \left(V_{riv,j} \cdot k_{ret,riv,j} + A_{lak,j} \cdot \nu_f + A_{res,j} \cdot \nu_f \right) \\ &= \frac{1}{V_{tot,j}} \left(V_{riv,j} \cdot k_{ret,riv,j} + \nu_f \cdot (A_{lak,j} + A_{res,j}) \right) \end{aligned} \tag{4}$$

Here, $V_{wb,j}$ and $V_{tot,j}$ (both in km^3) are the volume of the water body of focus (subscript wb, indicating either river, lake or reservoir) and the total water volume in grid cell j, respectively. $k_{ret,wb,j}$ (day^{-1}) is the removal rate of phosphorus in that water body. ν_f ($km \cdot day^{-1}$) is the phosphorus uptake velocity, which is set to $3.8 \cdot 10^{-5}$ $km \cdot day^{-1}$ (Alexander et al. 2004). ν_f is divided by the depth $D_{wb,j}$ (km) to convert into a removal rate. $A_{wb,j}$ (km^2) is the surface area of a water body. The lake surface area was calculated by multiplying the lake surface density (Environmental Systems Research Institute Inc. (ESRI) 1995) with the grid cell surface area calculated in ArcGIS. Reservoir surface areas were taken from ICOLD (ICOLD International Commission on Large Dams 1984, 1988).

Table 1.1: Constants used for the calculations of grid hydrological parameters and phosphorus persistence

Parameter	Symbol (Unit)	Value	Source
Width constant	aw ($km^{-0.56}yr^{0.52}$)	5.01E-2	(Wollheim et al. 2006)
Width exponent	bw	0.52	(Wollheim et al. 2006)
Depth constant	ad ($km^{-0.11}yr^{0.37}$)	1.04E-3	(Wollheim et al. 2006)
Depth exponent	bd	0.37	(Wollheim et al. 2006)
Removal rate	$k_{riv,i}$ (yr^{-1})	71.2 if Q_i<0.0882 km^3yr^{-1} 25 if 0.4473<Q_i<0.0882 km^3yr^{-1} 4.4 if Q_i>0.4473 km^3yr^{-1}	(Alexander et al. 2004)

Transfer fraction from soil to freshwater

$f_{DIP,i}$ and $f_{DOP,i}$ (dimensionless) were estimated with the method and parameterization of Mayorga et al. (Mayorga et al. 2010) using R, runoff ($m\ yr^{-1}$) (Fekete et al. 2002) as input, as shown in Online Resource 2 and 3, respectively:

$$f_{DIP,i} = \frac{0.29}{(1 + (R/0.85)^{-2})} \quad (8)$$

$$f_{DOP,i} = 0.01 \cdot R^{0.95} \quad (9)$$

Following Klepper et al. (Klepper et al. 1995), the fraction of particulate phosphorus transported from agricultural soils into freshwater via erosion of topsoil can be derived by:

$$P_{out,PP} = P_{input,TP} \cdot b \cdot TSSY \quad (8)$$

$$f_{PP} = \frac{P_{out,PP}}{P_{input,TP}} = b \cdot TSSY \quad (9)$$

Here, $P_{out,PP}$ (ton km^{-2} yr^{-1}) is the amount of particulate phosphorus transported into freshwater, $P_{in,TP}$ (ton km^{-2} yr^{-1}) is the amount of total phosphorus emission applied to the soil. b (1.18E-4 ton^{-1} km^{2} yr) is an empirical factor derived for Europe in Klepper et al. (Klepper et al. 1995) and TSSY (ton km^{-2} yr^{-1}) is the total suspended solids yield from Beusen et al. (Beusen et al. 2005). f_{pp} was derived for European watersheds, as shown in Online Resource 4 .

Relative importance of removal processes

The influence of the separate removal processes described above was determined by calculating the net removal rate for each process via the following:

$$k_{adv} = \frac{1}{FF_{adv}} \tag{10}$$

$$k_{ret} = k_{fw} - k_{no\_ret} = \frac{1}{FF_{fw}} - \frac{1}{FF_{no\_ret}} \tag{11}$$

$$k_{use} = k_{fw} - k_{no\_use} = \frac{1}{FF_{fw}} - \frac{1}{FF_{no\_use}} \tag{12}$$

k_{adv} (the net removal rate by advection, day^{-1}) was calculated directly by omitting retention and water use during the derivation of the fate factor. Because advection cannot be omitted, k_{ret} and k_{use} were calculated indirectly. The overall net removal rate (k_{fw}, day^{-1}) was calculated, and the net removal rate of the process except the process of focus (k_{no_ret}, and k_{no_use}, both in day^{-1}) as well. Subtraction yields k_{ret} and k_{use}. The relative importance of these processes was determined for each grid cell.

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2. Transfer fractions for dissolved inorganic phosphorus and dissolved organic phosphorus (f_{DIP} and f_{DOP} respectively, dimensionless)

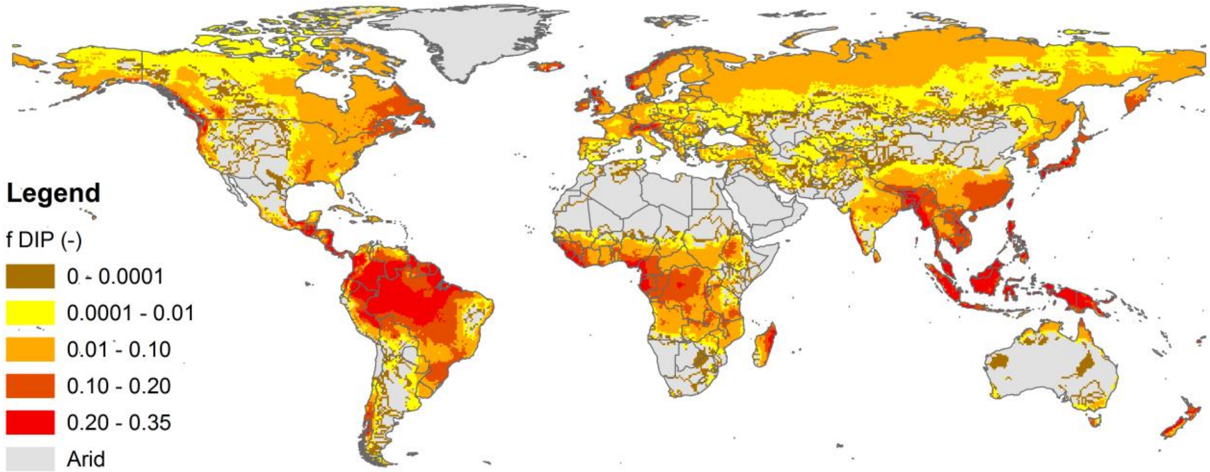

Fig. 2.1: Dissolved Inorganic Phosphorus transfer fraction (f_{DIP}, dimensionless)

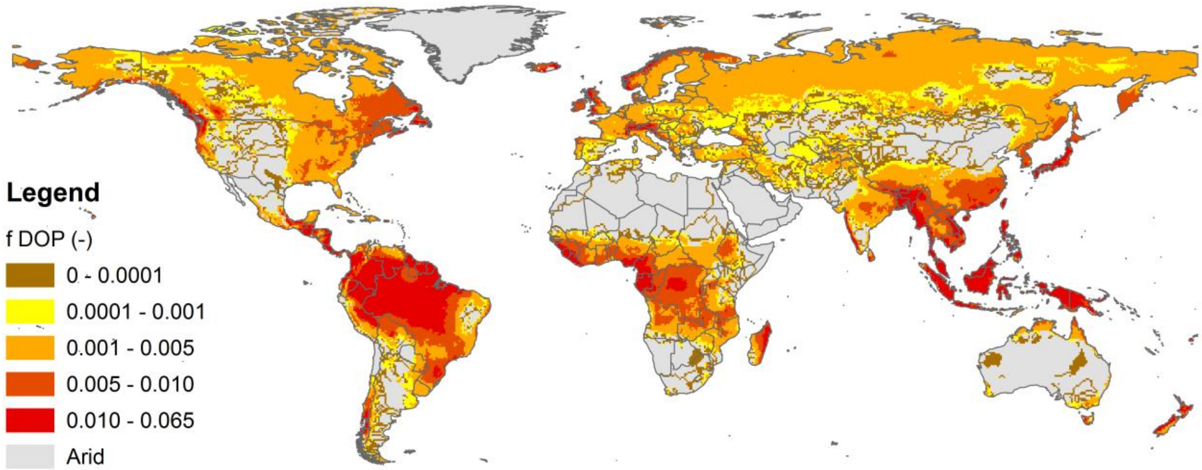

Fig. 2.2: Dissolved Organic Phosphorus the transfer fraction (f_{DOP}, dimensionless)

3. Additional analyses of phosphorus fate factors

The cumulative distribution of the fate factors is presented in Fig. 3.1.

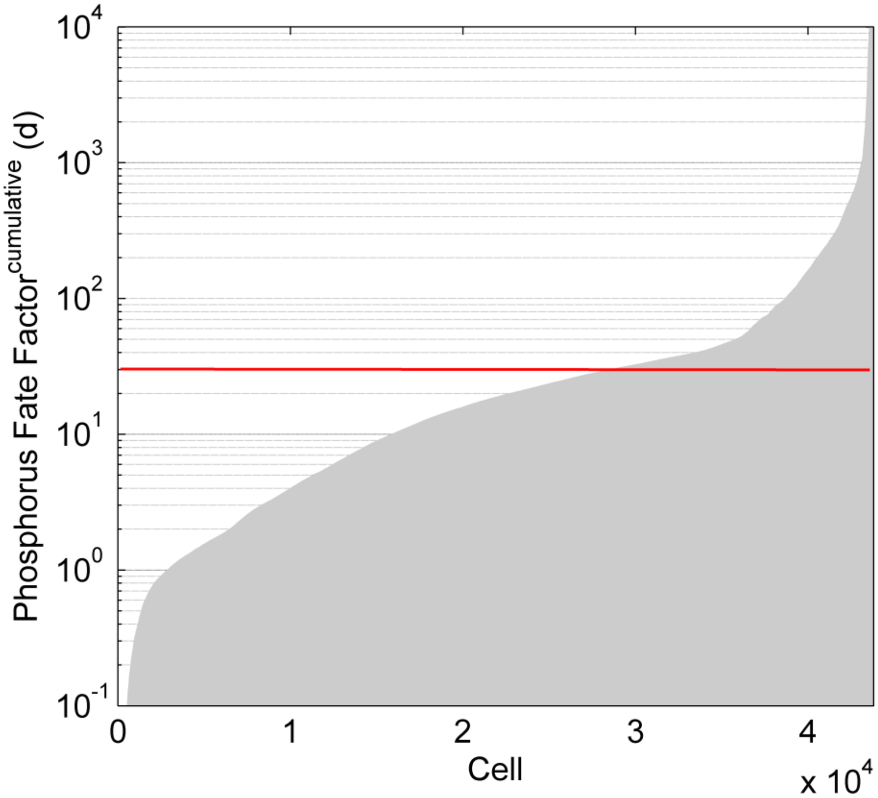

Fig. 3.1: Ranked cumulative fate factors ($FF^{cumulative}$) for phosphorus. $FF^{cumulative}$ = 30 days is indicated; this value was chosen as a cutoff for separating emission locations with short and long travel times to the sea.

Phosphorus removal mechanisms are further detailed in Fig. 3.2, where the cumulative fate factors are plotted against the cumulative residence time of water for the same grid cell. The data points are colored and symbolized according to their dominant removal mechanism. When advection is the only removal process on the route from emission location to ocean, phosphorus resides in the freshwater compartment exactly as long as the water itself, so that the fate factor of phosphorus is equal to the cumulative residence time of water. When the processes of retention or water use start to play a role, they cause the cumulative fate factor to be lower than the cumulative residence time of water and the data points are located below the diagonal. Fig. 3.2 shows that this can lead to a reduction of typically two orders of magnitude. Water use tends to be dominant mainly at low water residence times in the lower half of the distribution.

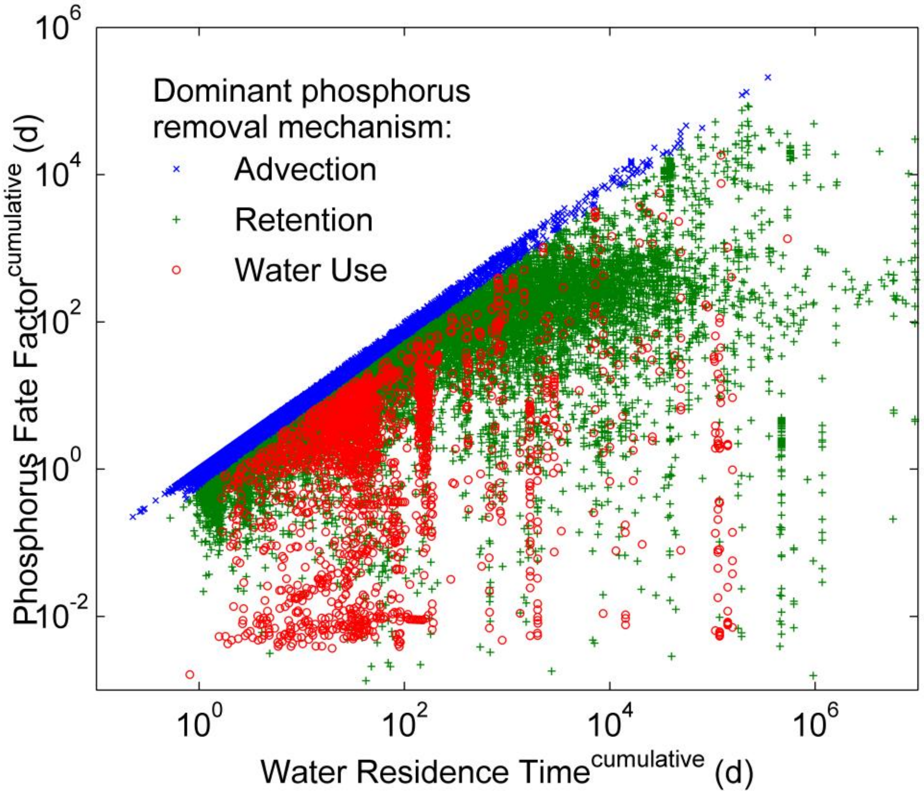

Fig. 3.2: Scatter plot of the cumulative fate factors for phosphorus emissions against the cumulative residence time of water for the same grid cell

4. Particulate phosphorus transfer fraction for European watersheds (f_{PP}, dimensionless)

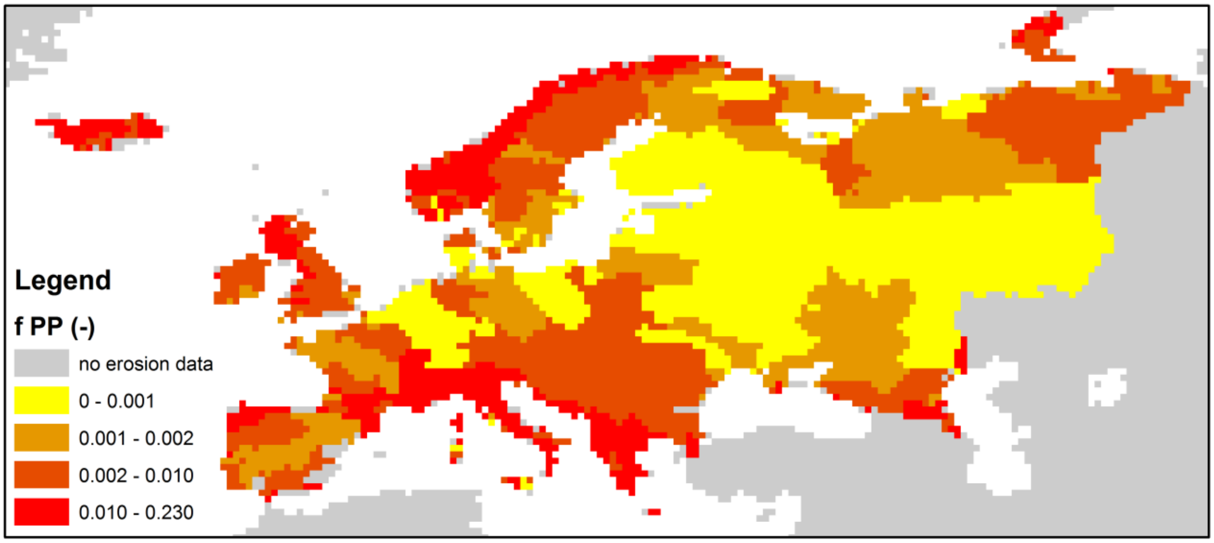

Figure 4.1: Particulate Phosphorus transfer fraction for European watersheds (f_{PP}, dimensionless)