Supplementary Information

Datasets

We incorporated several datasets to examine P fluxes and to calculate the net annual P inputs and P mass accumulated within the landscape, which includes soils, aquatic systems, reservoirs, and floodplains. A summary of the sources of P flux data, and calculations, is provided in Table S2, and these involved import of mineral P fertilizer, P leaving farms as agricultural products, river P export, and other human fluxes.

Framework

To conceptualize broadscale P dynamics, Haygarth et al. [1] recently proposed that long-term catchment development consists of an accumulation phase, when P gradually builds up, and a depletion phase (Fig. S1), when P outputs remain elevated despite declining P inputs.

Basin-specific Steps

Maumee River Basin

For Maumee Basin, fertilizer inputs were estimated using data from multiple sources including the International Plant Nutrition Institute (period of record 1987-2010), Baker and Richards 2002 (period of record 1976-1995)[2]; hereon we refer to these data sources as IPNI and BR02. Gap years in the IPNI fertilizer data were interpolated. Then, for each year, we used the average of the available values to estimate annual fertilizer P import (Fig. S2). For manure P, IPNI had slightly lower values than BR02 during the common years of record (1987, 1992; data ratio (IPNI:BR02)= 0.71). To address this moderate difference, we managed the time series as follows: in years when both manure data sources were available, we used the average of the two values; in years only one manure data source was available, we filled in the missing value assuming a constant data ratio, then used the average of the original and filled-in values for each year; in the few remaining gap years, we interpolated the missing values. The mass of P taken up by the dominant harvested crops ($P_{harvest}$), a precursor to $P_{food/feed,out}$, was estimated using National Agricultural Statistics Service (NASS) county data on crop-specific volumetric yields (reported as bushels) of corn, soybeans, and wheat, and mass yields of hay (reported as short tons), converted to mass units of P using crop-specific P density coefficients (Fig. S2). For volumetric yields we used the conversion factor of 0.035 m^3 per bushel. The methods for estimation of $P_{harvest}$ follow from BR02. Namely, we used the same percentages for contributing county areas in the aggregation of county data to the basin-level, and the same crop-specific P density coefficients as BR02, which in kg P m^{-3} are 2.09, 4.54, and 3.62 for corn, soybean, and wheat, respectively, and 2.38 kg Mg^{-1} for hay. $P_{harvest}$ is the sum of the crop-specific P values. $P_{food/feed,out}$ was estimated as $P_{harvest}$ - P_{manure} where P_{manure} is P from harvested crops not exported as agricultural products, but instead withheld in the basin via manure production. Here we have assumed annual food P import to Maumee is negligible (<1.0 kt) in this rural basin relative to the gross inputs. River total P export values (annual load estimates) for Maumee River are from Baker et al. [3] and were based on total P and daily discharge data from USGS station 04193500 at Waterville, OH, which is upstream of the Toledo, Ohio metropolitan area. We interpolated a gap in record for the river total P export between 1979-1981. Our estimates of net P input were highly positively correlated with those from BR02 over common period of record (1976-1995, correlation coefficient = 0.99). The data ratio for net P inputs (this study: BR02) averaged 0.85

between 1976 and 1995, and differences are explained mostly by the moderately higher manure P values of BR02.

Thames River Basin

For Thames, which has a substantial human population along with major agriculture, we incorporate several additional human P fluxes besides to fertilizer and crop export. The human population is associated with the southwest suburbs of London, UK, and some large towns like Swindon, Reading, and Oxford. Consistent with equations 2 through 5, in our approach we assume imported P from outside the basin enters the landscape P pool shown in Fig. 3 via a combination of fertilizer application, sewage effluent, and biosolids/sludge spreading, with the remainder destined for sewage treatment and landfills that are not included in the landscape pool. Consequently, the return of sewage biosolids/sludge to soils, and effluent release to rivers, are major pathways that allow internally produced food P to remain within the landscape pool, whereas much remaining P in locally-produced food/feed is ultimately destined for export to landfills or markets outside the basin. More specifically for Thames, gross P input to the landscape pool was calculated as the sum of 1) fertilizer import, 2) the subset of sewage effluent originating from imported food and detergent, and 3) the subset of sewage biosolids waste that both originated from imported food and detergent and was eventually applied to soils. Gross P output was the sum of 4) river export, 5) disposal of food waste to landfills, 6) export of internally produced food beyond the basin via trade, and 7) disposal of sewage waste originating from internally produced food at sea, landfill, or incinerator.

Thames fertilizer P import and river P export (annual load estimates of total P) are from Haygarth et al. [1]. Calculations for crop/livestock P leaving farms, and foodwaste P to landfills, involved multiple steps. First, P in harvested crops ($P_{harvest}$) was calculated using the average crop yields for England [4] and P content of grains [5]. In England, between 2001 to 2010, 44% of crop production went to livestock [4], so we estimated crop P export from the farm as $P_{harvest} \times (1.0 - 0.44)$, where we also assume the flux of animal feed P across the basin boundary is net zero. Food export from farms was calculated based on product-specific values for milk, egg, livestock, and wool production [6] and product-specific P content [5]. Recognizing losses of meat/bone P that occur between the farm and table, dressed carcass to live weight ratios were taken from Lord et al. [7], and the ratio of meat:fat:bone was assumed to be 67.5: 7.5: 22.5. The proportion of meat production supplied to consumers, as meat on the bone, is from the UK Family Food Report [8], which indicates 26.2% of the dressed carcass meat, by weight in year 2013, was supplied to consumers as meat on the bone. To calculate the bone P supplied to consumers, in the form of meat on bone, we therefore multiplied meat production by 26.2%. We then assumed that all bone P supplied to consumers was disposed of in landfills, whereas all P in food waste produced during intermediate manufacturing, and during dressing of the carcass, was rendered, incinerated, and returned to soils. Because the Thames crop, livestock, and bone P fluxes are based partly on coefficients from the 2000's, these calculations are likely most robust for the recent period of record. For the historical net P inputs to Thames, potential biases from the component P fluxes are partly countered by the very large fertilizer P flux.

Detergent P inputs up to 1998 are from the Foundation for Water Research [9] and for the remainder of the period of record, we used the values from Comber et al. [10]. Sewage production (sewerage influent to treatment works) for the years 2000-2010 was estimated by multiplying Thames population data (Table S4, Table S6) by the per capita rate of waste-P generation of Comber et al. [10], reported as 2.3 g person^{-1} day^{-1} (0.84 kg person^{-1} yr^{-1}). The population values were from county-level census data, and projected county population for the 2000's (http://www.ons.gov.uk). More specifically, for each year, we used the area-weighted sum $\sum P_i \times F_i$ where P_i is the population of county i and F_i is the fraction of county i that falls within the catchment boundaries. Gaps in record were linearly interpolated. The food fraction of sewage P production for 2000-2010 was calculated by subtracting the detergent contributions from Comber et al. For 1936-2000, the non-detergent contribution to sewage P production was assumed to scale in direct proportion to the P footprint for UK[11] (Table 6) using the ratio of non-detergent sewage: footprint from 2000-2010 (this ratio was 0.126). Trade fluxes were calculated as total human consumption -livestock production-crop production*(0.56)-livestock consumption, where 0.56 is the fraction of crop production not fed to livestock [6]. For sewage biosolids, we assumed 50% of biosolids production was applied to soils (constant over entire period of record), although there are reports that biosolids returns to soils exceeded 65% [12], and we assume the remaining 50% is exported to sea, landfills, or incinerator [13].

Yangtze River Basin

Yangtze River is Asia's longest river, and Yangtze Basin was by far the largest of our analysis, draining more than 1.8×10^6 km^2 (about 20% of China's land) en route to the East China Sea. Our estimates of fertilizer P input are revised from that of Haygarth et al. [1], and these integrate multiple data sources: 1) for 1979-2010, estimates are from National Bureau of Statistics of China for each province of Yangtze River Basin; 2) for 1970-1978, before provincial data were available, the estimates are from the International Fertilizer Industry Association (IFA) database, assuming that 45% of the IFA value is used in Yangtze Basin. These fertilizer P estimates fall intermediately between the P data from two other sources (Fig. S4) during the shared period of record (1970-1997): Liu et al. [14] P application data; food P demanded by the average national diet of China (Table S7) from Metson et al. [11], re-aggregated to Yangtze Basin based on Yangtze population data from Luo and Huang [15]. Unlike Maumee and Thames, Yangtze has substantial internal production of P fertilizer from mining. In our approach we exclude un-mined P from the landscape pool, and thus internally produced and applied fertilizer P is considered a new input. In the Yangtze basin, most food produced is also consumed internally, meaning that $P_{sewage,in}$ and $P_{food/feed,out}$ are small relative to the chemical fertilizer P input. We therefore simplified the calculation from equation 5 for Yangtze Basin, assuming $P_{sewage,in} = P_{food/feed,out}$. More specifically, this means that sewage effluent P produced from imported food P is assumed to equal the food/feed P exported through trade + waste fluxes (e.g., food waste or sewage waste transported to landfills). The assumption is justified by the large basin size and large human population (high P demand) which limit the escape routes for P. For example, since the 1980s the Yangtze fertilizer P input has clearly exceeded the human P footprint, a measure of the P demanded by total food consumption (domestic + imports) [11]. Also, compared to the very large fertilizer P flux, other fluxes were rather modest, such as the flux of sewage effluent P from imported food (<3.1% of fertilizer P input), removal via sewage treatment (<0.5 % of fertilizer P input), and food/feed export via trade (<4.5% of fertilizer P input). Thus, the pattern of P accumulation and depletion in Yangtze Basin was predominantly controlled by fertilizer P input. Applications of manure or human excrement to soils (night soil) originate mainly from food/feed produced within the basin, so do not represent a new P input, and also this practice is becoming less prevalent in rural areas of China, though once common. Of course the above P fluxes still vary among different provinces and cities within the basin. Because anthropogenic P also accumulated prior to the onset of Yangtze River P monitoring, our estimate of cumulative net P input represents a conservative estimate of the actual P stored currently. We also caution that an unknown and potentially large quantity of anthropogenic P may currently reside within landfills of Yangtze basin, and it is not yet clear how landfills may have directly or indirectly influenced the other P pools and fluxes.

While river total P has been monitored in the upper Yangtze River [16], currently there are limited published observations of total P near the river mouth. In response to this limitation, we estimated Yangtze River P export by taking the sum of annual dissolved P export and particulate P export (Fig. S5). The values for dissolved P export are directly from Dai et al. [17], with additional values from provided by multiple sources [18-21]. To estimate particulate P, we multiplied the suspended sediment export values from Dai et al. [17] by a P density coefficient of 0.5 g P per kg sediment as reported in Zhou et al. [22]. Thus our estimates of particulate P are based on the assumption of a constant sediment P density over time. But overall, we remind that the pattern of P accumulation in Yangtze Basin is strongly controlled by fertilizer P input.

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Table S1. Features of the three basins.

	Maumee R.	Thames R.	Yangtze R.
Region	midwestern USA	southern England	central China
Basin area	16,000 km^2	12,000 km^2	1,800,000 km^2
Relief	low	low	High
Climate	north temperate	north temperate	semi-arid
Major human impacts	rowcrop and animal agriculture	rowcrop and animal agriculture, urban development	rowcrop and animal agriculture, urban development, large dams
Human population (2010)	< 1 million	3.8 million	492 million
Human population density (2010)	< 60 per km^2	320 per km^2	270 per km^2
Basin P phase	late accumulation or early depletion (?)	late accumulation or early depletion (?)	early accumulation

Table S2. Key data sources and methods for estimating basin P inputs and outputs.

Flux direction	Basin	Flux type	Data sources	Period of record
Inputs	Maumee	Fertilizer-P import	Annual fertilizer-P imports from IPNI 2015, aggregated from county level to basin level	1987-2010
			Annual fertilizer-P imports from Baker and Richards 2002	1975-1995
	Thames	Fertilizer-P import	Annual fertilizer-P imports from Haygarth et al. 2014	1936-2010
		Sewage-P	Annual sewage-P effluent from Haygarth et al. 2014	1940, 1950, 1960, 1970, 1980, 1990, 2000, 2010
	Yangtze	Fertilizer-P import*	Annual fertilizer-P application rates, revised from Haygarth et al. 2014	1970-2010
Outputs	Maumee	Food/feed-P	P in agricultural products, based on crop yield data from National Agricultural Statistics Service, and crop-specific P density coefficients from Baker and Richards 2002, aggregated from county level to basin level	1976-2010
		River-P export*	Annual total P export from Baker et al. 2014, based on total P concentration and daily discharge data for US Geological Survey station 04193500 (Waterville, OH)	1976-1978, 1982-2010
	Thames	River-P export	Annual total P export from Haygarth et al. 2014	1936-2010
		Food/feed-P	Calculation based on multiple sources	see text
	Yangtze	River-P export*	Calculation based on dissolved P export + particulate P export, using data from Dai et al. 2011, Zhang et al. 1999, Shen and Liu 2009, Gao et al. 2012, Qu and Kroeze 2012, Zhou et al. 2013	1970-1990, 1997, 2000-2008

* Gap years interpolated or extrapolated in net P accumulation calculations (Fig. 3).

Supplementary Figures

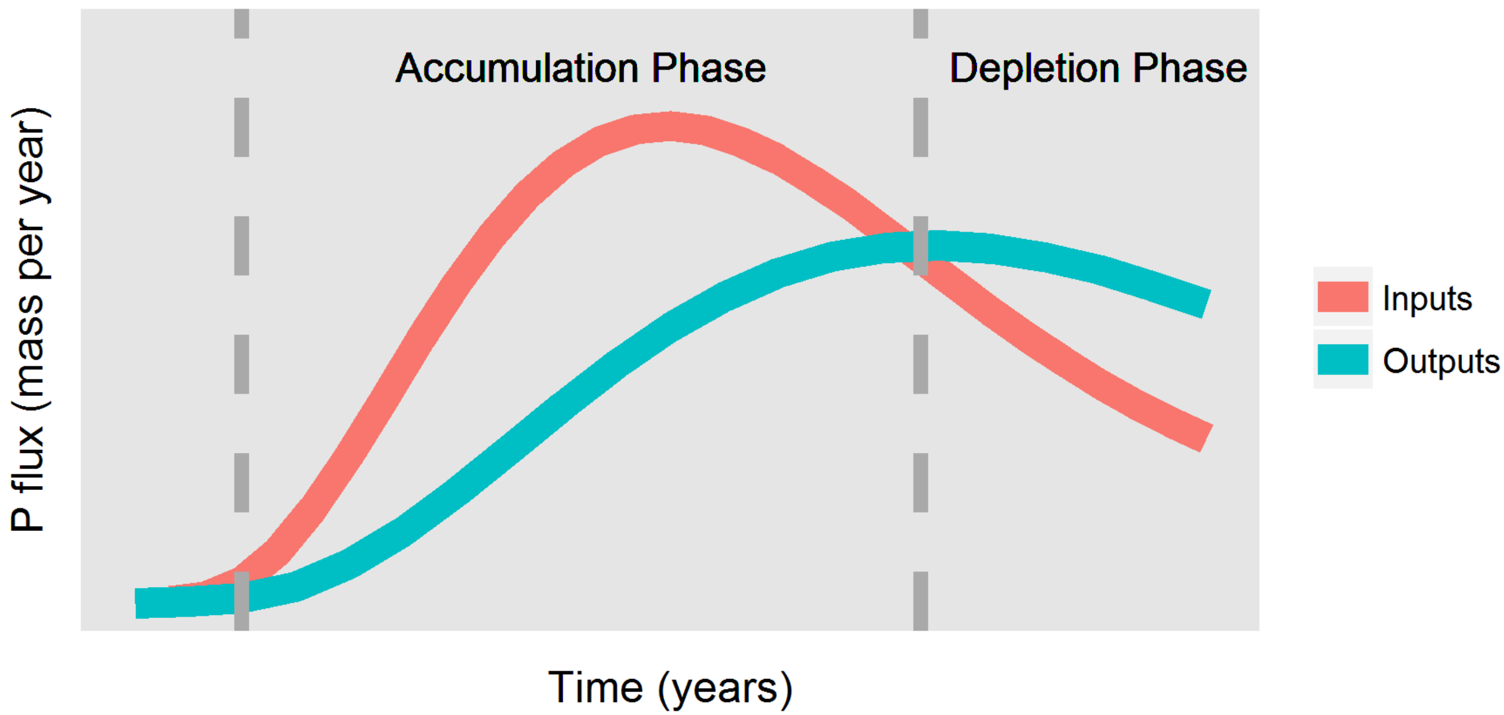
Figure S1. Accumulation-depletion framework for understanding landscape P dynamics over the long-term (decades to centuries). During accumulation phase, input exceeds output, and P builds up. During depletion phase, human P inputs decline and mobilization of accumulated P potentially causes outputs to exceed inputs. Adapted from Haygarth et al., 2014.

Figure S2. Fertilizer P import and manure production of Maumee basin. Values are from Baker and Richards (2002) and basin-aggregated data from IPNI.

Figure S3. Agricultural P fluxes of Maumee basin. Food/feed P export ($P_{food/feed,out}$ in Eq. 5) was estimated by taking the sum of P in annual harvest (corn+soy+wheat+hay) minus manure P production from Figure S2. Values for wheat and hay were unavailable for recent years (2008, 2009, and 2010), and we substituted the crop-specific means during these years.

Figure S4. Fertilizer P input ($P_{fert,in}$ from Eq. 5) to Yangtze River basin between 1970 and 2010, and two related P data sources (fertilizer P applied Liu et al. 2003; food P demand, Metson et al. 2012).

Figure S5. River export of P from Yangtze basin. Because total P has not been frequently reported near the Yangtze River mouth, we estimated total P for each year as the sum of dissolved P export and particulate P export. Dissolved P data are directly from Dai et al. 2011. Particulate P data are from Dai et al. 2011 suspended sediment data multiplied by the P density coefficient from Zhou et al. 2013 (0.5 g P per kg sediment).



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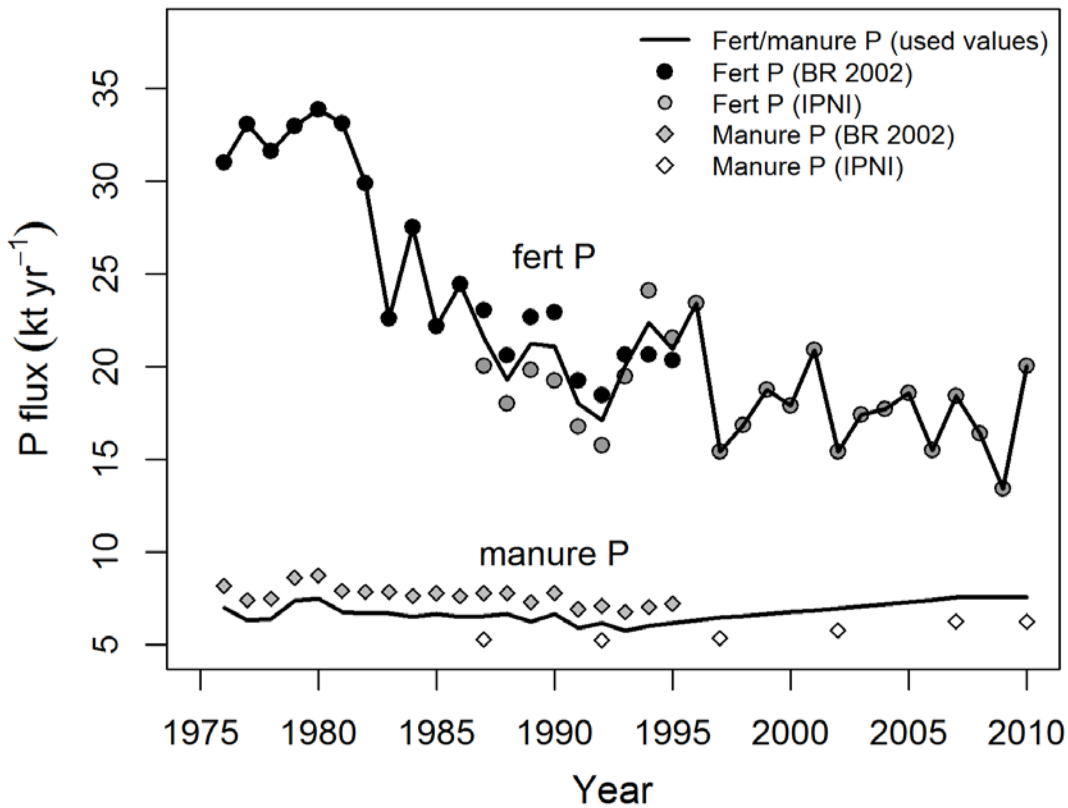

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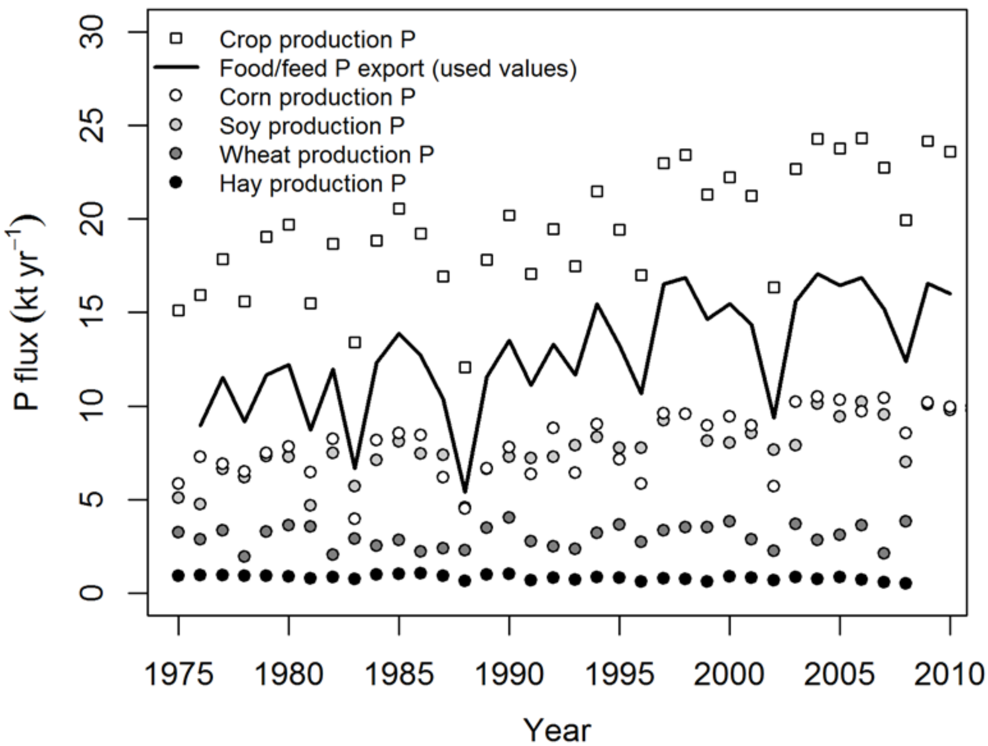

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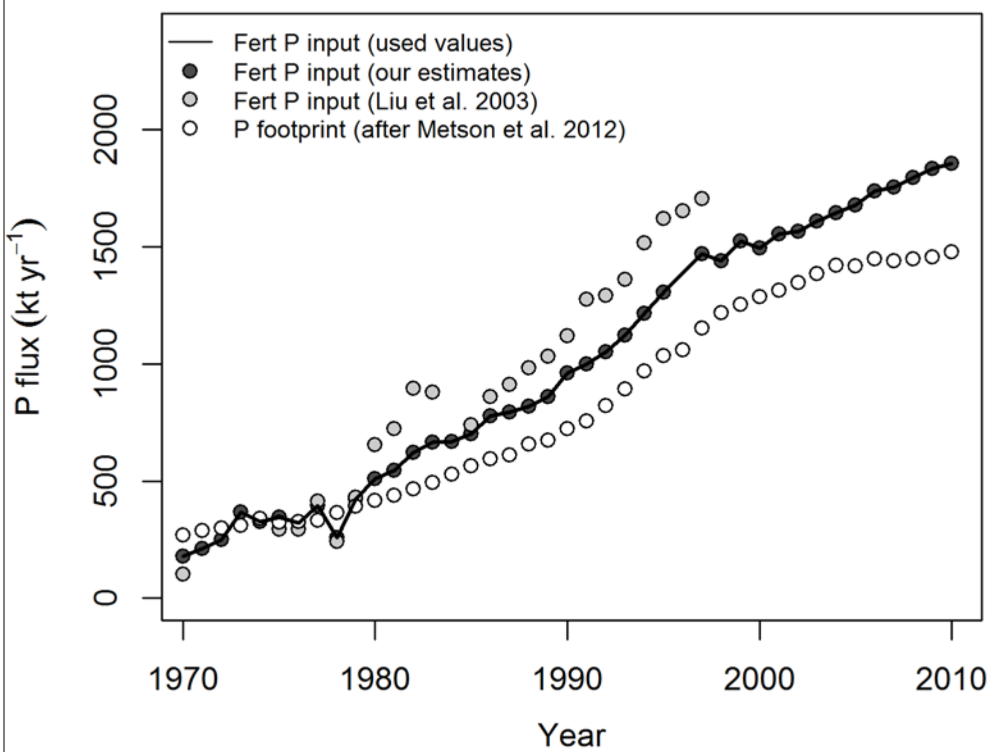

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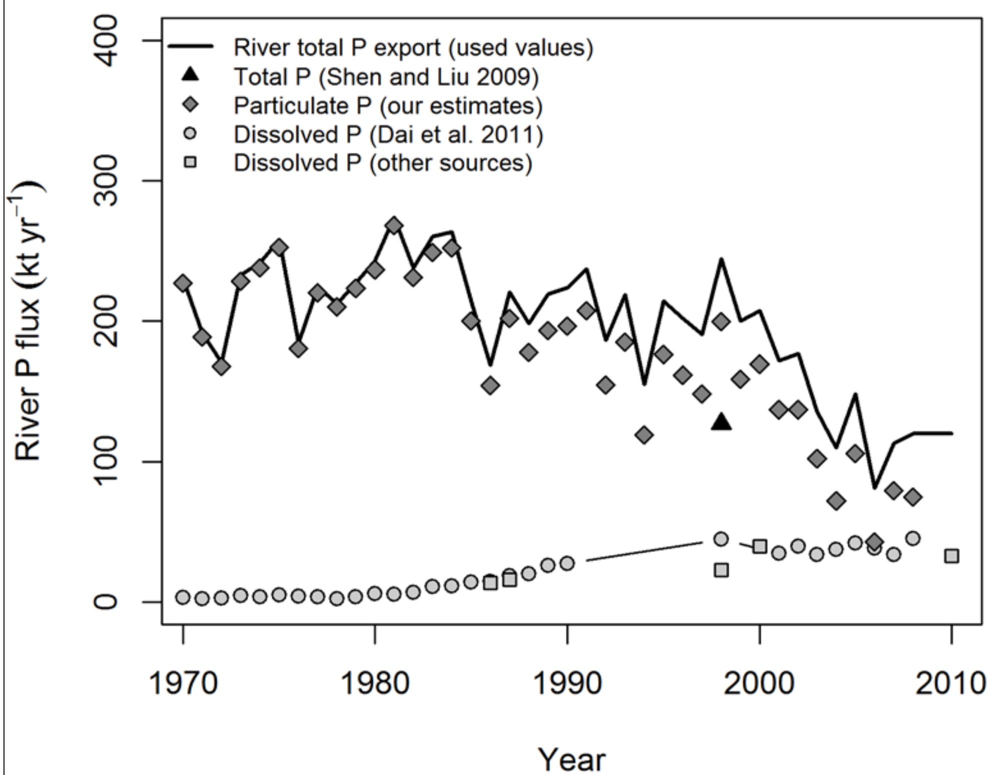

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