



Fertilizer, landscape features and climate regulate phosphorus retention and river export in diverse Midwestern watersheds

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Abstract Non-point source pollution of phosphorus (P) is a primary cause of eutrophication of aquatic ecosystems, and poses a persistent management challenge due to the dynamic and poorly understood processes controlling the transport and transformation of P at the watershed scale. We examined phosphorus inputs, retention, and riverine losses in 62 diverse watersheds that included a wide range of land cover and use (minimally disturbed to human dominated) and human P inputs in Minnesota, USA. Fertilizer inputs from row crop cultivation were the dominant source of P to agricultural watersheds. A large majority of P inputs was retained in watershed soils or removed in agricultural products. However,

fertilizer input was the most important factor associated with average annual river transport of total, dissolved, and particulate phosphorus (PP). Mean annual runoff increased total and dissolved P yields and decreased P retention. Dissolved P made up a significant portion of annual river loads at sites with high rates of P inputs and river TP export, with the ratio of dissolved to PP export increasing with crop cover and fertilizer inputs. PP export rose with larger extents of eroding bluffs near channels. Bluffs constitute only a small proportion of watershed area, but make a disproportionately high contribution to sediment loads due to their close proximity to river channels that are increasingly affected by human and climate-driven changes to river hydrology. Together, our results suggest that rising discharge and flow variability due to climate change and agricultural intensification coupled with high rates of P inputs will maintain elevated fluvial P export into the future.

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Without reductions in P inputs and reductions in both soluble P losses and stream bank erosion, reversal of water quality degradation will be difficult to achieve.

Keywords Phosphorus · Watersheds · Nutrient budget · Eutrophication · Legacy

Introduction

The global phosphorus (P) cycle has been highly modified by human activities, resulting in widespread negative impacts on aquatic ecosystems from eutrophication. Understanding the drivers of P mobilization in watersheds has thus been a major focus of eutrophication management over the past four decades (e.g. Carpenter et al. 1998; Schindler 2006; Sharpley et al. 2013). Despite this, the problem of eutrophication has stubbornly defied efforts intended to address it. Some success in reducing P loading to aquatic systems has been achieved via improvement and expansion of waste-water treatment systems (Jeppesen et al. 2005; Van Drecht et al. 2009; Worrall et al. 2016; Tong et al. 2017). In contrast, only limited success in reducing eutrophication has been achieved in areas with diffuse (i.e. non-point) agricultural and urban P sources (Dubrovsky et al. 2010; Renwick et al. 2018). As a result, eutrophic conditions persist worldwide in human dominated areas (Smith et al. 2006; Oliver et al. 2017).

While inputs from human activities are very often the main source of P affecting freshwaters, P transport within watersheds is controlled by complex processes that influence movement, mobilization, and retention across the land and land–water interface. P movement to streams and lakes is influenced by P concentrations in soils and sediments, soil properties, pH and redox sensitive reactions with metals, biotic processes, watershed flow paths and hydrology, land use practices, and climate. As a consequence, processes controlling P movement in watersheds are rarely sufficiently understood to efficiently guide management actions to control and reduce non point P losses to waterways.

Four factors likely contribute to sustained high rates of P export from human influenced landscapes to aquatic ecosystems. First, agricultural expansion and intensification have offset some progress in controlling non-point P losses. For example, in the upper

Midwest, field erosion has been reduced via improved agricultural management (Belmont et al. 2011), yet more land has entered production. Meanwhile, more cultivation has shifted to phosphorus intensive crops such as corn and soybeans, leading to increased or sustained high levels of P inputs to many watersheds (Dietz et al. 2015; Fofoula-Georgiou et al. 2015) and accelerated runoff and erosion due to agricultural drainage and crop changes (Heathcote et al. 2013; Kelly et al. 2017). Second, fertilization over many decades has led to P accumulation in soils and sediments (Bennett et al. 2001; Powers et al. 2016). This “legacy” P continues to influence watersheds and waterbodies via erosion of P rich sediments, and transport of dissolved P (Sharpley et al. 2013). A related third factor is that losses of dissolved P have not been effectively managed by standard best management practices (BMPs) for control of particulate P (PP) losses, perhaps because it has been assumed that most phosphorus is transported with particles. As a consequence, dissolved P losses represent a difficult problem that may be worsened by P accumulation in soils (Dodd and Sharpley 2015). Finally, climate change may be undoing some recent management progress. While field erosion has generally decreased, downstream erosional losses from near channel sources has accelerated (Belmont et al. 2011), leading to increases in sediment and potentially P inputs from stream banks (Schottler et al. 2013; Grundtner et al. 2014; Dolph et al. 2019). Thus, despite many years of research, the interacting drivers and processes that regulate non-point P losses are not understood thoroughly, limiting effective management to reduce freshwater eutrophication.

The upper Midwest has diverse land cover that includes areas with substantial human inputs of P, including agriculture and urban centers, as well as natural features such as forests, lakes and wetlands. Inter-watershed comparisons of river phosphorus transport across land use and climate gradients have been essential for revealing controls of nutrient movement from land to water (e.g. Howarth et al. 1996; Dillon and Molot 1997; Jordan et al. 1997), especially at scales necessary to integrate small-scale heterogeneity and to provide guidance for landscape scale evaluation of restoration and management. In this study, we sought to determine how land cover, phosphorus inputs, climate (precipitation), and hydrology (river flows) interact to influence P losses in

watersheds across diverse landscapes in Minnesota, USA. The study watersheds spanned gradients of agricultural and undeveloped land cover, anthropogenic inputs from fertilizer, food, and feed imports, annual runoff, and wetland density. We examined climate and landscape conditions to explain variation in annual total P (TP), dissolved P, and particulate P (PP) export and retention. Across a wide gradient in land use, we hypothesized that P losses would be related to human P inputs, but could be modified by landscape features such as lake and wetland cover. We integrated datasets representing P inputs and outputs to examine how watershed-scale P balances and environmental factors influence rates of river exports and watershed retention across 62 watersheds in Minnesota.

Methods

Study system

Study watersheds were distributed across major ecoregions of the state of Minnesota, ranging in size from 176 to 68,117 km² (Supplementary Table 1). These watersheds included approximately 65% of the state. Watersheds were selected for this study based on data available to estimate annual river P fluxes using the Metropolitan Council (METC) stream monitoring program and the Minnesota Pollution Control Agency (MPCA) Watershed Pollutant Load Monitoring Network (Fig. 1). Together, the study watersheds included land-use and land-cover representative of much of the upper Midwest.

Agricultural areas dominated land use in the south and western part of the state, where agricultural production has largely transitioned from hay and small grain production to corn and soy production over the past 50 years (Foufoula-Georgiou et al. 2015). Extensive areas of forests, wetlands and lakes dominate central and northeastern MN. These regions were historically heavily logged and now are lightly populated. Several watersheds intersected the seven county Twin Cities region, a major urban center, but only larger watersheds that were characterized by suburban land cover were included in this study, as they could be analyzed with methods we applied to other sites. More highly urbanized watersheds were excluded from our analysis due to their small size and

centralized waste water treatment systems. The north temperate climate of the region has warm summers and cold winters, with precipitation gradients that yield more rainfall in the eastern part of the state compared to western areas.

Estimation of P loads

Flow and water quality data for estimation of annual loads were collected from 2007 to 2011. Sampling targeted a range of flow conditions including snow-melt and large storm events. Loads of soluble reactive phosphorus, i.e. orthophosphorus (SRP), total phosphorus (TP) and total suspended sediment (TSS) were calculated with FLUX32, a pollutant load model driven by data for P concentration from an average of 35 sampling events per year and continuous discharge measurements (MPCA 2015). To avoid sensitivity to spatial and temporal variability in climate conditions (see Dolph et al. 2019), we used annual averages to examine and compare watershed mass balances of P. We calculated average annual SRP, TP, and TSS loads for sites with at least two years of complete data between 2007 and 2011 (mean = 3.96 years of data per site). Sampling encompassed substantial variation in climate and runoff, representative of the typical range of conditions over the past decade, including wet and dry years, with no multi-year droughts or major flooding events. We estimated PP by subtracting SRP from TP. This approach slightly overestimates PP due to the presence of dissolved organic phosphorus (DOP) not detected in SRP analyses. The lack of DOP data for our study sites causes small overestimation of PP concentrations and slight underestimation of the concentration of total dissolved (SRP + DOP) P concentrations. Loads were normalized by watershed area to estimate average annual yields.

Land cover and bluff area analyses

Watershed delineations used in our analyses were provided by the MPCA and METC. The 2011 National Land Cover Database (NLCD) was used to determine the proportion of each watershed in land cover categories such as crops, pasture or hay, urban, and open water (Homer et al. 2015). We used the National Wetlands Inventory (NWI) to determine connected wetland cover in watersheds, and the USGS National Hydrography Dataset (NHD) to determine the

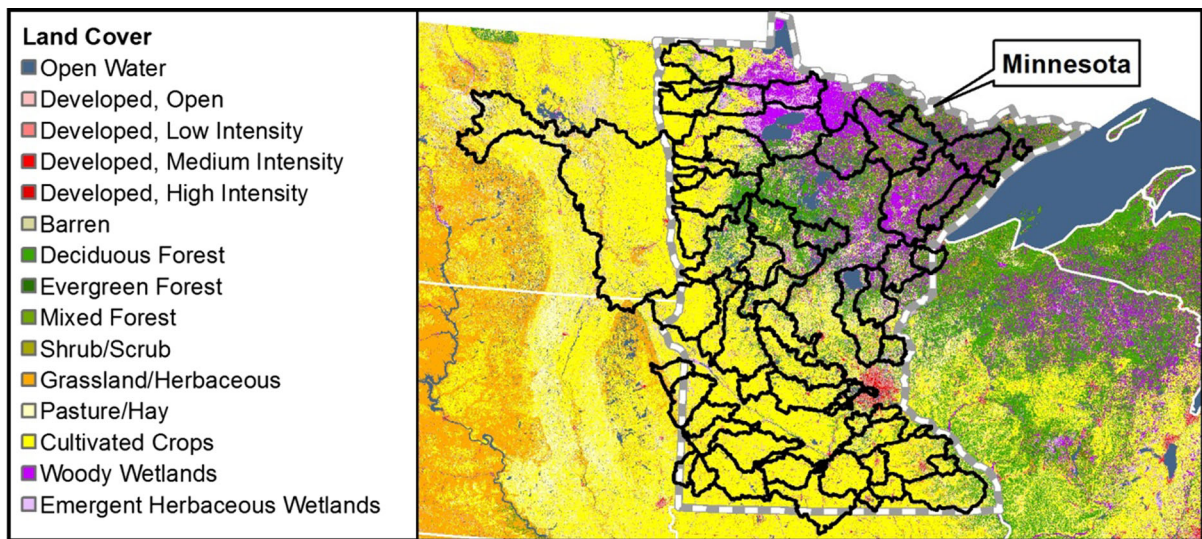


Fig. 1 Study watersheds, outlined in black, and land cover information from NLCD (2011)

connected lake and pond cover in watersheds. The NWI has recently been updated for much of our study region, including southern and east-central Minnesota (MNDNR 2016). An updated NWI was not available for northern Minnesota, so the original NWI was used to estimate wetland cover in this region (USFWS 2015). Wetlands and water bodies within 100 m of the center line of a NHD perennial stream were considered connected to the channel (Powers et al. 2014). One kilometer gridded precipitation data for 2007–2011 compiled by the PRISM Climate Group (<http://prism.oregonstate.edu/>) were used to determine mean annual precipitation for each watershed.

High bluffs (i.e. banks 4–40 m tall) are important topographic features of some watersheds, especially in tributaries to the Minnesota and southern Mississippi Rivers. These bluffs are the results of Pleistocene down-cutting of the Minnesota River following rapid drainage of Lake Agassiz, and the ensuing incision of tributaries (Gran et al. 2011). We estimated the area of bluffs along river channels for each watershed using LiDAR elevation data from the Minnesota Geospatial Information Office (<http://www.mngeo.state.mn.us/chouse/elevation/lidar.html>). Within a moving 12 m by 12 m window, pixels with elevation differences greater than 4 m were initially designated as bluffs. Then, only those pixels falling within a buffer extending 50 m from a channel centerline were

included as near channel bluff area for the purpose of our analysis.

Phosphorus inputs and retention

We used the Net Anthropogenic Phosphorus Input (NAPI) toolbox (Version 3.0.1β, available from Cornell University (<http://www.eeb.cornell.edu/biogeo/nanc/nani/nani.htm>)) to estimate net P inputs for study sites completely within the boundaries of the United States (Figs. 1, S1). We did not estimate inputs to watersheds smaller than 150 km² because of the coarse scale of input data (Howarth et al. 2012), hence eliminating small watersheds from our analyses and leaving 62 watersheds for inclusion in this study. Input data consisted of: (1) USGS mineral fertilizer input estimations (Gronberg and Spahr 2012) (2) the USDA agricultural census of crops and animals (USDA 2009), and (3) the USA Census (Howarth et al. 1996; Boyer et al. 2002). These county-level data were area weighted based on the proportion of each county intersecting the watershed. Crop import and export data were translated into estimates of P using crop specific P content. Livestock data were converted into P using rates of intake and excretion and P content of edible portions and literature values for P excretion. Finally, human population data were translated to P using values for P in food consumed, food waste disposal, and detergent use (Landler et al. 1998; Russell

et al. 2008; Han et al. 2011; Swaney and Hong 2017). P outputs from each watershed included P associated with food, feed or biofuel exports. We used NAPI estimates to determine agricultural and non-agricultural fertilizer imports and net food and feed import or export.

Discharges of P from wastewater treatment plants (WWTPs) released within the watershed were not considered in NAPI estimates since inputs were represented by food and feed fluxes. WWTP effluent loads may strongly influence river P dynamics, however, so P point sources were assessed in our analyses of river exports (see below). Data for point discharges arising from industrial facilities and domestic wastewater treatment plants requiring a permit from the National Pollutant Discharge Elimination System or State Disposal System (hereafter ‘permitted discharges’) were obtained from MPCA (<https://www.pca.state.mn.us/water/phosphorus-wastewater>). Several suburban watersheds had substantial exports of sewage to centralized plants outside of their watersheds. These transfers represent an uncharacterized loss of P from the watershed that could not be accounted for in our study, as indicated in Table 1.

Inputs of P from mineral weathering and atmospheric deposition were not represented as new inputs in our P balance estimates. Weathering transforms mineral P to more bioavailable forms (Hartmann et al. 2014) but was not considered a new input since most rock sources were buried under deep glacial sediments in most areas of the state. Weathering rates of soil minerals is likely less than $10 \text{ kg km}^2 \text{ year}^{-1}$ (Hartmann et al. 2014). Atmospheric deposition is not monitored systematically across the study region, but statewide estimates from previous measurements indicate atmospheric deposition of 20 to $40 \text{ kg km}^2 \text{ year}^{-1}$ (MPCA 2004). The majority of this deposition originates from local sources, and it is likely that most deposited P does not reflect new inputs of P (Eimers et al. 2018). However, the specific amount of deposition that represents new P input from long range transport is unknown, and important to P balances of watersheds with low rates of direct human inputs. This issue is addressed in the discussion of watershed P balances, especially with reference to northern watersheds with low anthropogenic P inputs.

The balance of direct human inputs and outputs of P from watersheds determines NAPI (Supplementary

Fig. 1). We calculated the proportion of P retained (R_p) by the watersheds as follows, in units of kg/km^2 :

$$R_p = \frac{\text{NAPI} - \text{Hydrologic P Export}}{\text{NAPI}}$$

NAPI estimates were based on the most recent data available representing human inputs and outputs of P across the study watersheds. To understand the trends in P accumulation over time and the potential effects of historical inputs on current P losses, we also examined historical P fertilizer inputs to the study watersheds. We estimated historic fertilizer inputs using USDA agricultural survey data and methods modified from Dietz et al. (2015). Briefly, we used historic county-level areas of corn, wheat, and soy multiplied by fertilization rate data for each crop type to estimate historic fertilizer inputs. We then summed cumulative fertilizer inputs over 5 year increments starting from 1972 and ending in 2006, the period directly preceding the 2007 to 2011 period for which average river yields were determined.

Data analysis

Our statistical analyses were designed to disentangle the relative strengths of P inputs, land use, and environmental factors in determining river P losses and watershed retention. Candidate explanatory variables to represent key controls included human P inputs (which are related to watershed accrual and losses to rivers), land use (associated with P inputs, as well as erosion and drainage effects), land cover (which influences erosion rates, and includes land features such as wetlands and lakes that may trap P), and annual precipitation and runoff (that affect P transport and mobility). Although soil properties vary across the state, we did not include soil chemistry in our models due to uncertainty in soil coverage within the study watersheds, and an anticipated large effect of human P inputs. Multiple linear regression was conducted using JMP Pro 12 (SAS Institute, NC, USA). P export, NAPI, connected lake and wetland cover, agricultural (crops + pasture) land cover, bluff area and manure inputs were log-transformed to meet normality requirements for statistical analyses. Forward stepwise multiple linear regression was used by adding the independent variables that explain the most residual variance in the dependent variable (Burnham and Anderson 2002). Variables were selected for the

Table 1 Averages and ranges of values for 62 watersheds examined in this study

Parameter	Units	Average	Minimum	Maximum
Anthropogenic phosphorus inputs				
Inorganic fertilizer P	kg/km ²	626	0	1350
Net food and feed P	kg/km ²	– 319	– 1002	587
Manure P ^a	kg/km ²	332	0	983
NAPI	kg/km ²	307	2	1131
Hydrologic export				
Average TP export	kg/km ²	43	3	121
Average SRP export	kg/km ²	22	0	82
Average estimated PP export	kg/km ²	19	2	69
P retention (Rp)	–	0.64	– 1.75	0.99
SRP:PP	–	1.1	0.2	3.1
Average TSS export	kg/km ²	16,843	243	75,140
Watershed area, runoff, and precipitation				
Area	km ²	3742	176	68,117
Average runoff	m	0.20	0.10	0.34
Average precipitation	m	0.75	0.62	0.94
Runoff:precipitation		0.27	0.14	0.48
Land cover characteristics				
Bluff Area	km ²	4	0	57
Cropland	%	48	0	91
Pasture or hay	%	8	0	24
Urban	%	6	1	47
Forested	%	15	0	61
Connected lakes	%	3	0	18
Connected wetlands	%	6	0	56
Additional characteristics for watersheds completely within MN				
NPDES discharges	kg/km ²	5	0	67
Number of active feedlots	–	421	0	1903
Number of active CAFOs	–	14	0	87

All watersheds had an area greater than 175 km², and at least two years of water quality data

^aManure does not represent a new input but was considered as an independent variable due to its potential to influence watershed P losses

final models based on evaluation of Akaike Information Criterion (AIC), which describes the variance explained while accounting for the number of variables included in the final model (Burnham and Anderson 2002).

Results

Patterns of phosphorus transport

Study watersheds showed wide variation in land cover, land use and anthropogenic phosphorus inputs, and moderate variation in annual precipitation and runoff (Table 1, Fig. 1). Crop cover ranged from 0 to

91% (average 48%) and urban cover ranged from 1 to 47% (average 6%). Connected lake and wetland cover ranged from 0 to 18% (average 3%) and 0–56% (average 6%) of watershed area, respectively. Annual average precipitation and runoff ranged from 0.62 to 0.94 m (average 0.75 m) and 0.10–0.34 m (average 0.21 m), respectively, during the studied five-year period (Supplementary Fig. 2), with less precipitation and runoff in western regions.

Watershed P inputs, retention (Rp), and river P export were variable across the study area, with the highest inputs, Rp, and export occurring in agricultural watersheds in southern and western regions (Fig. 2). Annual river P exports were moderate in central compared to southern Minnesota but were elevated

compared to northern watersheds. Rivers of watersheds dominated by agricultural land use exported between 10 to 120 kg km⁻² year⁻¹ (mean 58 kg km⁻² year⁻¹) while largely forested areas in north central and northeastern regions exported 2 to 17 kg km⁻² year⁻¹ (mean of 15 kg km⁻² year⁻¹). P export was comparatively low in watersheds with low to moderate suburban land uses in the Twin Cities region.

Discharge from wastewater treatment plants was a minor component of river P transport and watershed P balance for most sites. However, two watersheds (Shell Rock and Cedar Rivers) were exceptions, having relatively high WWTP losses that represented more than 50% of the observed annual TP yield (Supplementary Table 2). Sewage losses could not be

estimated for several of the suburban sites where domestic and industrial sewage was transferred across watershed boundaries to centralized treatment plants.

Watershed P retention was high for most sites in terms of mass (arithmetic mean of 30.3 kg km⁻² year⁻¹) and proportion of inputs (60% of inputs). However, substantial variability was observed, ranging between small apparent net losses of P to retention of up to > 500 kg km⁻² year⁻¹ (Supplementary Table 2). In other words, some watersheds retained nearly all NAPI, while a small number of northern sites showed little net P retention.

Low watershed Rp, and several cases of apparent net P loss, occurred in the least developed region of the state characterized by the highest coverage of wetlands, lakes and forests, and lowest total river P export

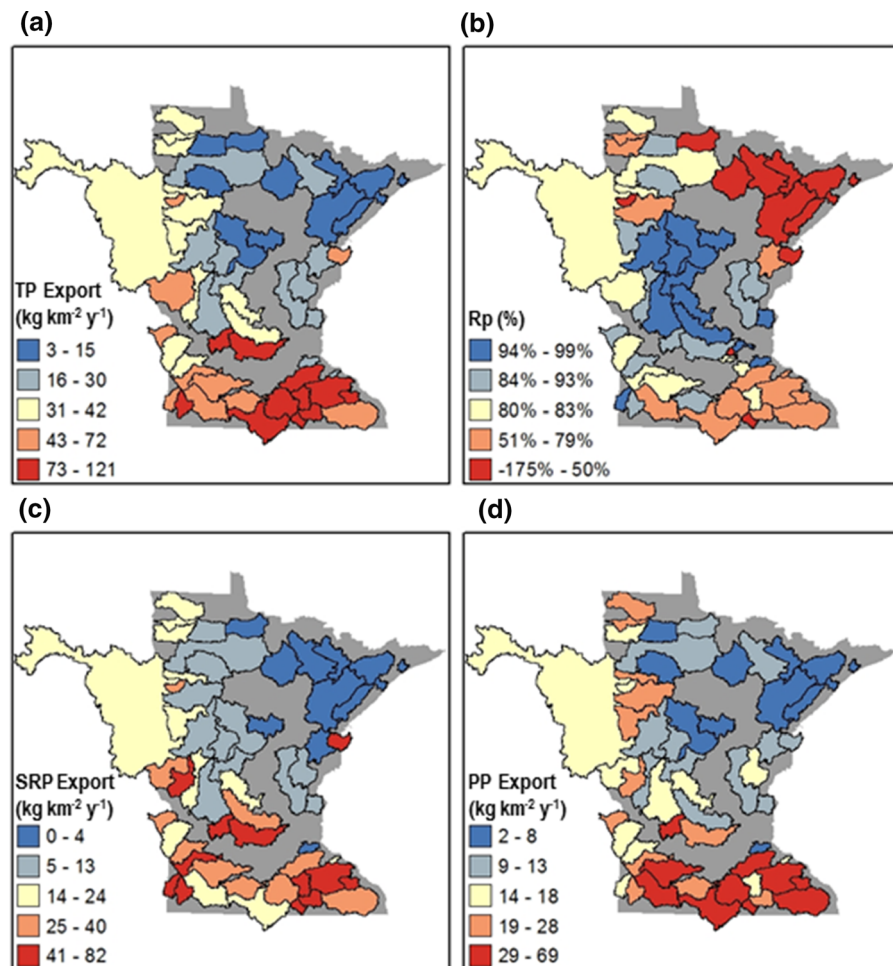


Fig. 2 Total phosphorus export (a), phosphorus retention, Rp (b), SRP export (c), and estimated particulate phosphorus export (d) for study watersheds. Export reported in kilograms per square kilometer (kg km⁻² year⁻¹)

(Fig. 2). Since human inputs were low at these sites, retention estimates were likely sensitive to lack of information about background P inputs, especially atmospheric sources. Atmospheric P inputs were not included in NAPI since it is unclear how much of atmospheric P inputs originates locally versus longer distance transport, with only the latter source representing new P inputs to watersheds. Inclusion of a small amount of atmospheric deposition of P at these sites would substantially increase estimated P input, and shift watershed R_p from very low or negative values to slightly positive R_p (see “Discussion”). Thus, apparent low R_p for northern sites is likely related to underestimation of TP inputs in areas of low human input.

In contrast, the greatest R_p in terms of both mass and proportion of human inputs was observed in watersheds of west central Minnesota, where agricultural intensity, P inputs and runoff were moderate (Fig. 2). R_p was also high for agricultural watersheds in southern Minnesota (Fig. 2). Although rates of annual river P export were highest in this region, a large proportion of human inputs was retained, similar to findings in other watersheds in the Midwest (e.g. Bennett et al. 1999).

The dominant form of P in rivers shifted spatially in response to human inputs and the presence of actively eroding bluffs (Figs. 3, 4). A substantial portion of river annual TP export occurred as dissolved P (Supplementary Fig. 3), with contribution to annual TP loads ranging from 7 up to nearly 100% in agricultural watersheds (average 46%) for all sites during the study period (2007 to 2011). Our estimates of dissolved P losses are conservative, because dissolved organic phosphorus, which averaged about 17% of total dissolved phosphorus in our agricultural sites (Baker 2018), was not included in our estimates of dissolved P export. In agricultural watersheds, where TP input and export were highest, dissolved phosphorus loads ranged from about half to up to 150% of the PP load. Lightly developed watersheds in northern MN had the lowest dissolved P and TP export, and lowest ratio of dissolved to particulate P of annual loads.

Controls of watershed phosphorus retention, yield, and form

Multiple regression analysis highlighted the dominant roles of agricultural P inputs (Fig. 3) and runoff (Supplementary Fig. 2) in determining P yields. TP export was best explained by inorganic fertilizer inputs, runoff, and the proportion of combined crop and pasture land within watersheds (Table 2). Thus, agricultural intensity and runoff were the most important factors associated with high P export.

While river P yield tended to increase with greater anthropogenic inputs, R_p exhibited complex non-linear relationships with P inputs (Fig. 3b, d). These non-linear relationships were driven largely by very low R_p in a small number of northern forested sites with the lowest anthropogenic P inputs (i.e. sites with NAPI of $< 14 \text{ kg km}^2 \text{ year}^{-1}$). As discussed above, forested wetlands dominated land cover in NE Minnesota, and watersheds in this area showed very low R_p even though estimated P inputs to these sites were also the lowest observed in our study. Given the possibility that negative and very low R_p values for these nine sites may have arisen from lack of accurate characterization of atmospheric deposition, regression analysis that included these sites required careful interpretation. When we included these sites in regression analysis, R_p was positively associated with inputs of manure and fertilizer, and negatively associated with runoff (Table 2). When these sites were excluded, the regression analysis indicated that R_p among the remaining 47 sites was positively associated with food and feed inputs, fertilizer inputs, and % connected wetlands and negatively associated with runoff (Table 2).

As for TP, annual river SRP yield was positively related to fertilizer inputs, with influences of agricultural land use, and watershed runoff also evident (Table 2). PP export was strongly related to TSS export (linear regression $r^2 = 0.81$; Fig. 4), as expected. Multiple regression models for PP subsequently included factors that influenced sediment export but excluded sediment export as an independent variable. These analyses showed a strong positive relationship between P fertilizer input and river PP losses, with significant positive effects of bluff areas near channels and mean annual runoff (Table 2). PP export was negatively related to lakes and wetland cover within watersheds likely due to trapping of P.

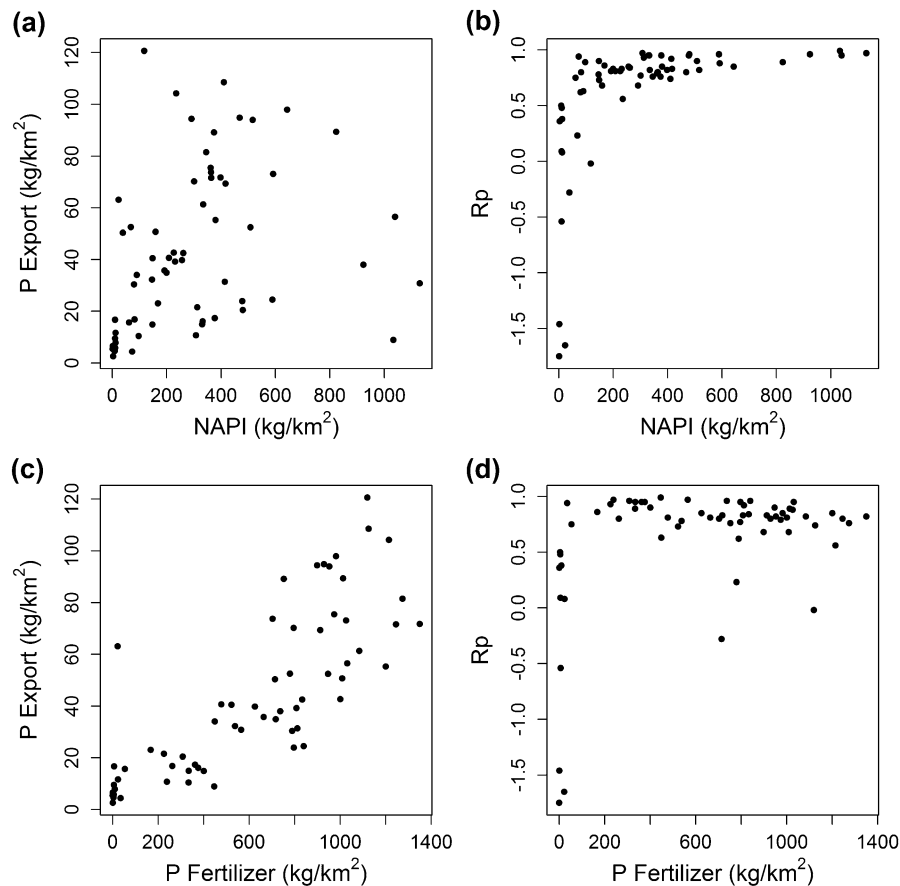


Fig. 3 Phosphorus export (a) and R_p (b), vs. NAPI and P export (d) and R_p (e), vs P fertilizer inputs. Units of R_p are proportions (e.g. 1 indicates 100%)

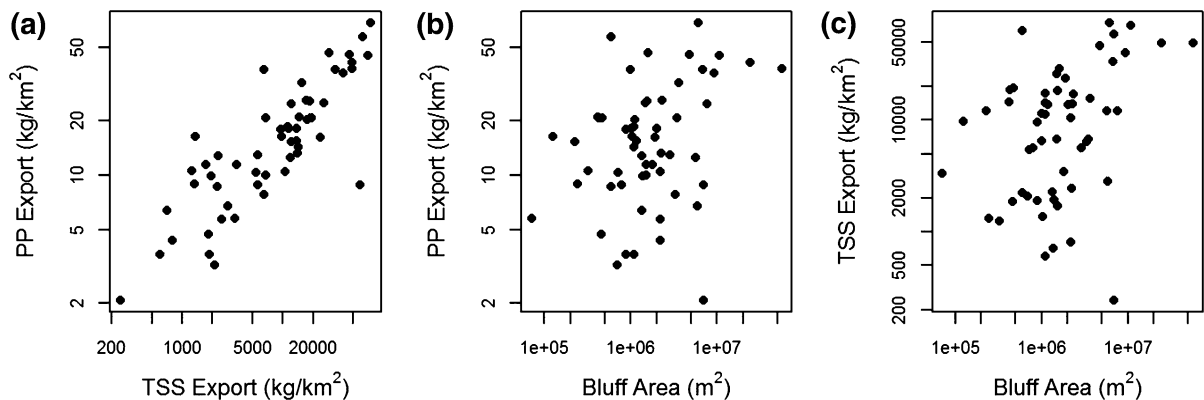


Fig. 4 Particulate phosphorus export vs. TSS export (a) and watershed bluff area (b), and TSS vs bluff area (c)

Area of near channel bluffs (as a proportion of total watershed area) was also associated with greater TSS yields (Fig. 4).

The form of P transported by rivers was strongly affected by human P inputs. Fertilizer inputs were strongly associated with dissolved:particulate P ratio

Table 2 Multiple regression analyses results for total P export and retention

Dependent variable	<i>N</i>	<i>R</i> ²	Term	Coefficient	<i>P</i> value	Corrected AIC
ln (total P export)	56	0.91	Inorganic P fertilizer	0.001	< 0.001	56.5
			ln (crop + pasture cover)	0.33	< 0.001	48.8
			Runoff	3.94	< 0.001	24.0
			Intercept	0.80		
Total P retention	56	0.82	ln (manure P ^a)	0.34	< 0.001	34.4
			Runoff	− 2.24	< 0.001	12.1
			Inorganic P fertilizer	− 0.001	< 0.001	− 2.6
			Intercept	− 0.28		
Total P retention (9 forest wetland-dominated sites excluded)	47	0.52	Net food and feed P	0.0004	< 0.001	− 43.0
			Inorganic P fertilizer	0.0004	0.002	− 44.7
			Runoff	− 0.86	0.013	− 47.1
			ln (connected wetlands)	0.07	0.016	− 51.6
			Intercept	0.83	< 0.001	
Dependent variable	<i>N</i>	<i>R</i> ²	Term	Coefficient	<i>P</i> value	Cumulative AIC ^a
ln (SRP Export)	56	0.91	Inorganic P fertilizer	0.001	< 0.001	87.5
			ln (crop + pasture cover)	0.49	< 0.001	68.7
			Runoff	3.81	< 0.001	55.0
			Intercept	− 0.64		
ln (PP export)	56	0.73	Inorganic P fertilizer	0.001	< 0.001	67.8
			ln (water)	− 0.22	0.004	63.2
			ln (bluff area)	0.08	0.02	59.8
			Intercept	1.04		
ln (SRP:PP)	56	0.58	ln (crop cover)	0.30	< 0.001	74.5
			Bluff area normalized by watershed area	− 9.6	0.02	71.4
			Intercept	− 0.87		

Candidate variables included NAPI as well as its components, runoff and precipitation, and watershed land cover. A total of 56 sites had complete information for analyses. NAPI was not included as a predictor of retention as it directly contributed to its calculation; however, components of NAPI were included as potential explanatory variables

^aManure does not represent a new input but was considered as an independent variable due to its potential to influence watershed P losses

of annual yields (Fig. 5) with ratios increasing from values much less than one in forested sites to above three in heavily fertilized agricultural watersheds. This relationship was modified by high sediment inputs from erosion in watersheds with extensive bluff areas near channels (Table 2). High sediment inputs from bluff erosion (Belmont et al. 2011) and P associated with this sediment (Grundtner et al. 2014) decreased dissolved:particulate P ratios at high levels of fertilizer inputs (Fig. 5).

Annual fertilizer P input was the single most important factor for river P yields in our analyses. However, these relationships likely reflect the

cumulative impact of fertilization and accumulation of P in watershed soils, especially since intensification of farming following World War II. While we lack information to assess the impact of fertilization on historic river P export, we examined trends in P fertilizer use for previous decades. For 57 watersheds with adequate crop data, we found similar significant relationships between average annual TP yield and both historic and current P inputs (Supplementary Fig. 4). Recent fertilizer inputs were strongly correlated with inputs during previous periods and similar relationships were observed between historical fertilizer inputs and recent river export (Supplementary

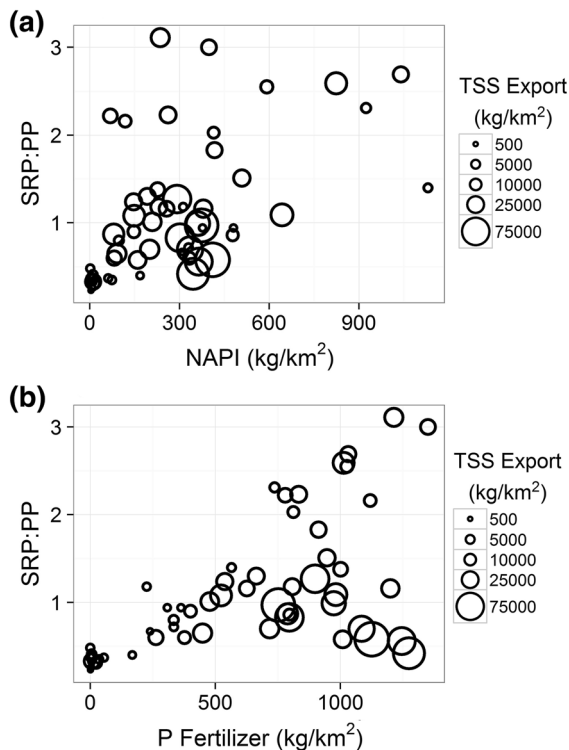


Fig. 5 Relationship between **a** NAPI and **b** fertilizer input and the ratio between dissolved and particulate annual phosphorus yields. The size of each circle is proportional to the annual TSS yield at each site

Fig. 4). While the rate of P input per unit farmland has slightly declined in recent years, little deviation in relationships between most recent and historical inputs was observed in most cases. Thus, while fertilizer inputs have fluctuated over time, they have remained high over the previous four decades in most watersheds included in this study, with some exceptions that depended on location and land use (Fig. 6). For example, increases were observed in western MN where agricultural land use has more recently intensified (i.e. cropping shifts from small grains to row crops; Foufoula-Georgiou et al. 2015). Watersheds near the Twin Cities metropolitan area especially show decreases in fertilizer inputs as land use has transitioned from agricultural to suburban and urban land use (e.g. Fig. 6c, f), where residential P fertilizer use is banned. Overall, fertilization and P accumulation rates, particularly in southern and western Minnesota, showed few signs of major declines necessary to begin depleting P stores in the landscape (Fig. 6).

Discussion

Across gradients in human impacts and natural landscape features, P transport and retention were driven primarily by anthropogenic factors, especially P inputs (both food/feed and fertilizer inputs), and secondarily by landscape and climate factors. Environmental variables, including precipitation and runoff and variation in physiographic features, such as extent of watershed area as lakes, wetlands, and unstable bluffs near channels, modified the relationship between P inputs and river export and watershed P retention. Both dissolved and particulate P contributed strongly to annual watershed losses. While fertilizer P inputs were the best predictor of overall P losses and the dominant form of P transported by rivers, the contrasts in the controls of dissolved vs. particulate P export provide insight into current nutrient management activities and future challenges in the face of climate and land cover change as discussed below.

River Phosphorus Yields

Watershed P losses are expected to increase with greater concentrations of bioavailable soil P, as plant and microbial demand and geochemical stabilization processes are saturated (Menge et al. 2012; Sharpley et al. 2013). However, variation in processes that immobilize, store or mobilize P in soils and sediments often leads to weak relationships between watershed inputs and riverine losses (Haygarth et al. 2012, 2014; Powers et al. 2016). Across a wide gradient of land use and land cover, P export was most strongly dependent on agricultural P inputs and intensity. Among the land use and cover variables examined, inputs of P from fertilizer, agricultural land cover and runoff explained the majority of variation in average annual TP yields. These relationships were likely driven by long-term buildup of P in watershed soils since intensive fertilization began over 60 years ago (Dietz et al. 2015). P losses occurred primarily as particulate and dissolved organic P in less human impacted watersheds, and increasingly as SRP as P inputs increased, except for watersheds with high rates of bluff erosion and sediment loss. For most sites, wastewater inputs represented a minor fraction of river P export, with several exceptions for monitoring sites that were

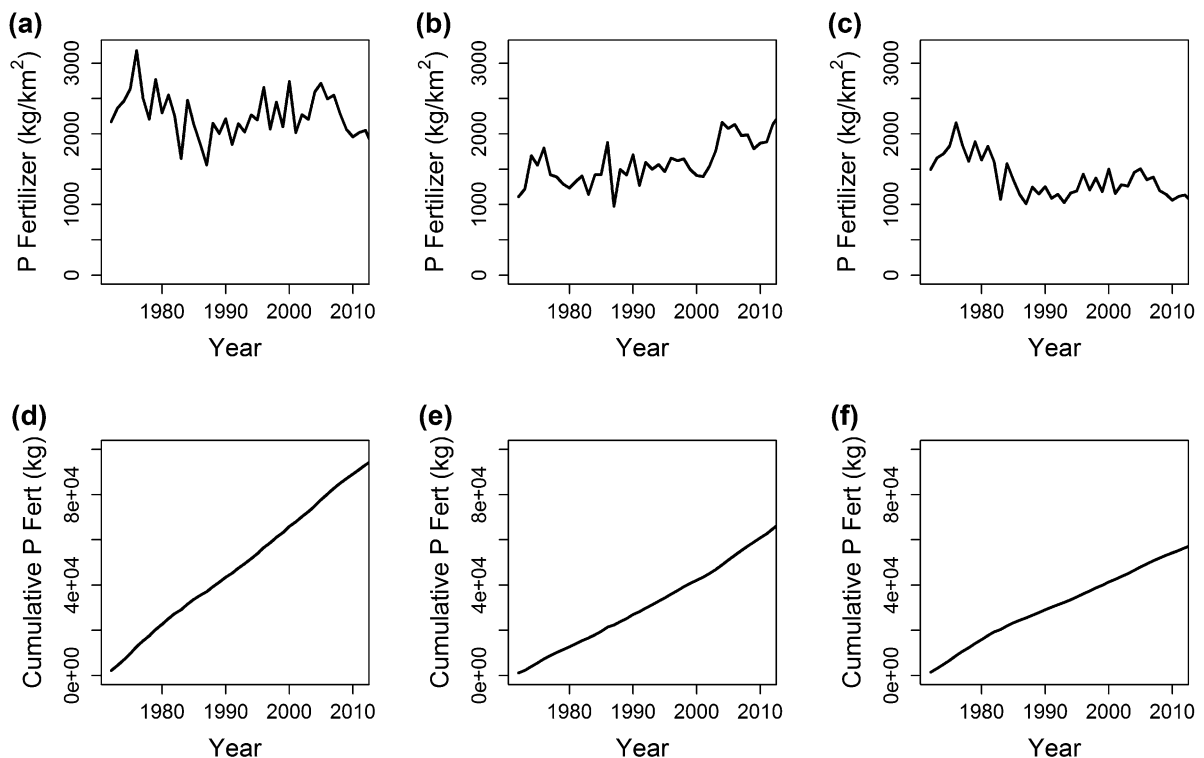


Fig. 6 Representative plots showing watersheds with fluctuating fertilizer P inputs that remain relatively high with stable agricultural land cover (left panel), increase over time due to agricultural intensification (middle), and decrease over

time due to urbanization (right). Plots show annual estimates of inorganic P fertilizer inputs (**a–c**) and cumulative P fertilizer inputs (**d–f**) for the Le Sueur River (**a, d**), Marsh River (**b, e**), and Sand Creek (**c, f**) from 1972 to 2011

downstream of large point source inputs (Supplementary Table 2; see “[Discussion](#)” below).

In addition to P inputs, hydrology and natural watershed features played significant roles in mediating river P transport. Higher annual precipitation and runoff mobilized more P and increased export, especially in highly fertilized watersheds. Regional climate changes coupled with hydrologic modifications of agricultural watersheds may have increased P export by increasing and intensifying runoff. Recent increases in precipitation amount and intensity have increased the erosive capacity of Midwestern rivers (Kelly et al. 2017). Ditching and especially subsurface tile drainage has increased in agricultural areas during recent decades promoting the efficient transport of water away from fields (Danesh-Yazdi et al. 2016), further increasing erosion downstream and counteracting conservation efforts to stabilize farm field erosion rates (Schottler et al. 2013). Tile drainage may also stimulate dissolved P losses (Jarvie et al. 2017;

Smith et al. 2014; Dolph et al. 2019) although limited observations suggest that direct tile losses are low in soils of southern MN (Randall et al. 2000). Together, increased precipitation and agricultural drainage accelerate P loss from watersheds and channels, primarily from erosion of P enriched soils and mobilization of dissolved P.

Phosphorus retention

Phosphorus retention was high in all watersheds except for nine in northern MN with very low anthropogenic P inputs and four outliers in human-dominated watersheds. Among sites with agricultural and/or urban influence (i.e. excluding the 9 forested wetland-dominated sites with very low P retention, see below), R_p averaged $\sim 83\%$. This result is broadly consistent with previous studies which found high P retention in human-dominated watersheds. In Chesapeake Bay, Lake Michigan, Lake Erie, and Central

California watersheds, P losses equivalent to 5–10% of NAPI were exported via rivers (Russell et al. 2008; Han et al. 2011; Sobota et al. 2011). R_p in most human dominated watersheds was positively associated with both food and feed P inputs and fertilizer inputs. This finding reflects the high capacity of many watersheds to retain a large proportion of added human P inputs, a feature common to many intensively cultivated landscapes. Higher river losses also resulted in these watersheds, likely as a result of mobilization of P from sensitive, near channel areas as discussed further below. Notably, our study sites did not include watersheds with the most intensive urban land uses, where watershed P retention is very low due to high rates of P losses via storm sewers (Hobbie et al. 2017) and additional losses from waste water treatment plant releases.

Phosphorus retention decreased with higher runoff and increased for watersheds with more wetlands connected to the river network, indicating the important roles of hydrology, watershed connectivity and storage in P retention and export. Our findings indicate that connected wetlands (i.e., those wetlands intercepting flow directly from river networks) may be particularly important in increasing watershed-scale P retention in agricultural and suburban landscapes. Likewise, Hansen et al. (2018) recently demonstrated the importance of connected wetlands in modulating the export of nitrogen in our study region.

Although most sites dominated by agricultural or suburban land use exhibited high R_p , there were four such sites that were characterized by $R_p < 60\%$. Two of these sites were highly affected by large permitted TP discharge from wastewater treatment plants (i.e. Shell Rock and Cedar Rivers). Higher river P losses (35% of net inputs) have previously been observed in the Illinois River where wastewater treatment plant effluent was a major source of P (David and Gentry 2000). This and other studies indicate that wastewater plays an important role in watershed scale P balances for some watersheds, and is consistent with the low R_p observed at sites downstream from wastewater point sources in this study. Permitted discharges are not explicitly considered in NAPI calculations as these exports ultimately originate as, and are represented by, food and feed inputs (Chen et al. 2015). However, direct proximity of point sources to river channels likely enhances their influence on watershed P export. Comparisons of total permitted discharges to annual

river export (Supplementary Table 2), show that they were a small proportion of annual river export (9.5% of TP yield), on average across all study sites. The Shell Rock and Cedar Rivers stood out as outliers with permitted discharges at 55% and 59% of annual P export, respectively, which likely contributed to lower R_p at these sites compared to other similar agricultural watersheds. Point source discharges from WWTP of small cities in these two watersheds occurred just upstream (within several km) from the water quality monitoring stations, likely enhancing their impact on river P yields. In addition to known WWTP inputs, three of the four low retention sites (Cedar River, Shell Rock River and Carver Creek) were located in relatively urbanized watersheds, suggesting that low retention could arise from storm water runoff from impervious surfaces, and/or contribution of human sewage to river P export via sanitary waste systems (via leakage). For the majority of agricultural and urban-influenced watersheds in our study, however, a small proportion of NAPI was exported each year and a small fraction of this was derived from permitted sources, showing that non-point phosphorus sources dominated P export for most watersheds we examined.

Much lower than average R_p was observed for nine northern watersheds (Fig. 2) compared to sites that had much higher levels of disturbance and P input. Most of the land cover (76–95%) in these watersheds was forest, lakes and wetlands, and these sites had the lowest human population density of all sites. The low apparent retention is at least in part related to underestimation of P inputs because atmospheric P inputs were not included in our estimated human P inputs. A recent study in an adjacent region (Eimers et al. 2018) suggested that winter P inputs are representative of long-range new inputs of P to forested watersheds. These authors estimated long-range P transport into forested watersheds in Ontario, Canada of around $5 \text{ kg km}^{-2} \text{ year}^{-1}$. Adding this amount of P input to our NE Minnesota sites would increase P inputs by 60%, increasing R_p substantially to values ranging from neutral (total P input = river output) up to 70% retention. These retention values are consistent with comparable minimally developed landscapes in Ontario (Crossman et al. 2016) but would still be lower than R_p observed at most other sites in this study, which were much more intensively managed or modified. Many of the low retention northern watersheds in Minnesota have extensive lake

coverage, and P inputs from seasonal residences, such as lakeshore cabins with older septic systems, may not have been adequately represented in our input methods, possibly further underestimating P inputs at these sites. In addition, P losses were dominated by a combination of particulate and dissolved organic P at these northern sites, as observed elsewhere in boreal regions (Palviainen et al. 2016). Poor retention of P bound with organic matter may thus have also contributed to low Rp. Overall, our results for northern forested watersheds were similar to those from nearby watersheds in Ontario, and require further study to identify the causes of apparent low P retention (Eimers et al. 2009; Crossman et al. 2016; Eimers et al. 2018).

Our large scale analyses of phosphorus balance in major watersheds has advantages in enabling comparisons across a wide range of environmental conditions and gradients, but also some limitations. In particular, our estimates of P inputs were made at coarser scale relative to more finely scaled, data intensive approaches (e.g. Peterson et al. 2017) and representative data for soil P pools and specific farming practices could not be considered in our analyses. Such approaches are much more feasible at smaller scales. Estimates of P retention are most influenced by error particularly when flux terms are low, such as in northern, less modified watersheds. Ideally, areas of uncertainty identified via large scale approaches can be explored further with more detailed mechanistic studies.

Contrasts in controls on dissolved and particulate P

Although agricultural management often focuses on reducing particulate P loads, we found dissolved P was a significant portion of annual P export, especially in P enriched watersheds. Both PP export and SRP export increased with increased fertilizer inputs. While best management practices emphasize control of field-scale losses of PP, but these approaches may not effectively reduce dissolved P export or in-channel losses of particulate P. A review by Dodd and Sharpley (2015) found conservation practices implemented to reduce PP, such as vegetated buffers and wetlands, often increased organic and inorganic dissolved P. While vegetated buffers have been shown to trap particulate P transported via overland flow, some studies have shown buffers are net exporters of dissolved P due to conditions that promote microbial

processing and desorption processes (Roberts et al. 2012; Dupas et al. 2015). Flushing of dissolved reactive P was found to occur when soils were rewetted after dry periods in the fall and when anoxic conditions released bound P in late winter (Dupas et al. 2015). Additionally, conservation practices to promote soil health in some cases contribute to phosphorus build-up at the soil surface, which may then be released via desorption and microbial transformations (Dodd and Sharpley 2015). Thus, while buffers may trap some PP from upland erosion of fields, they may increase dissolved P export.

Implications for management of phosphorus

Results of this study and others underscore the importance of reducing P inputs to achieve lower P export. Inputs of phosphorus from fertilizer was the strongest factor associated with P loss from agricultural watersheds. This relationship likely arose not from direct losses of new P inputs but rather from an accumulation of P over time in soils, leading to higher loss rates via solubilization along flow paths near and adjacent to channels (Duncan et al. 2017; Haygarth et al. 2014; Powers et al. 2016) and losses during erosion of P enriched soils and sediments (Chen et al. 2015; Fox et al. 2016; Records et al. 2016). Long-term accrual of P over many decades of fertilization in excess of crop demand leads to storage of legacy P (Haygarth et al. 2014; Powers et al. 2016; Goyette et al. 2018) which may be mobilized during many years following reduction in watershed P inputs (Goyette et al. 2018). Conversely, there may be a lag between the timing of increased P inputs or changes to the landscape and water quality deterioration.

Lag effects in the relationship between P inputs and river export should be taken into consideration when evaluating P management and assessing the effect of land cover change. Research from a watershed in China found a strong relationship between NAPI and river TP export that may be due to P saturation from past inputs. Such inputs were estimated to account for a substantial portion of annual TP export (Chen et al. 2015). Our estimates of historical fertilizer inputs suggest in most study watersheds, fertilizer P has fluctuated over time but has not significantly decreased. Sustained high rates of P fertilization in intensively managed agricultural lands will likely lead to continued P accumulation in soils and persistent

high rates of losses of both dissolved and PP from non-point sources. Strategies to minimize or eliminate fertilizer application and maximize P removal in crops or storage in perennial vegetation would increase P retention and draw down excess soil P that enhances both soluble and particulate PP losses to aquatic ecosystems (e.g., Dodd and Sharpley 2015; Duncan et al. 2017; Chen et al. 2019). In addition to reductions of inputs, treatment or reduction of water released by tile drains and control of near channel P mobilization processes could help mitigate P losses.

Conclusions

Anthropogenic inputs, especially of fertilizer, coupled with hydrologic drivers are important to determining P losses from agricultural watersheds. While retention was high in most agricultural watersheds we examined across the Midwest, proportionally small losses of P inputs from these watersheds nevertheless resulted in high P export by mass, given the high mass of current and legacy inputs. Losses were highest in the most intensively managed agricultural areas, with high levels of dissolved and particulate P export in response to fertilizer applications. Our results point to the need for agricultural approaches that minimize application of P inputs, while managing for retention of legacy sources of dissolved P. Although soils in our region are not yet as P enriched as in other parts of the Midwest, the influence of legacy soil P was still apparent and represents a challenge to water quality improvement in agricultural lands. Further exploration of the factors affecting phosphorus retention can aid in directing management strategies toward an effective combination of phosphorus input reduction and watershed management to reduce channel erosion and soluble P mobilization from near channel areas.

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