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# Linking terrestrial phosphorus inputs to riverine export across the United States

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ABSTRACT

Humans have greatly accelerated phosphorus (P) flows from land to aquatic ecosystems, causing eutrophication, harmful algal blooms, and hypoxia. A variety of statistical and mechanistic models have been used to explore the relationship between P management on land and P losses to waterways, but our ability to predict P losses from watersheds often relies on small scale catchment studies, where detailed measurements can be made, or global scale models that that are often too coarse-scaled to be used directly in the management decision-making process. Here we constructed spatially explicit datasets of terrestrial P inputs and outputs across the conterminous U.S. (CONUS) for 2012. We use this dataset to improve understanding of P sources and balances at the national scale and to investigate whether well-standardized input data at the continental scale can be used to improve predictions of hydrologic P export from watersheds across the U.S. We estimate that in 2012 agricultural lands received 0.19 Tg more P as fertilizer and confined manure than was harvested in major crops. Approximately 0.06 Tg P was lost to waterways as sewage and detergent nationally based on per capita loads in 2012. We compared two approaches for calculating non-agricultural P waste export to waterways, and found that estimates based on per capita P loads from sewage and detergent were 50% greater than Discharge Monitoring Report Pollutant Loading Tool. This suggests that the tool is likely underestimating P export in waste the CONUS scale. TP and DIP concentrations and TP yields were generally correlated more strongly with runoff than with P inputs or P balances, but even the relationships between runoff and P export were weak. Including P inputs as independent variables increased the predictive capacity of the best-fit models by at least 20%, but together inputs and runoff explained 40% of the variance in P concentration and 46–54% of the variance in P yield. By developing and applying a high-resolution P budget for the CONUS this study confirms that both hydrology and P inputs and sinks play important roles in aquatic P loading across a wide range of environments.



## 1. Introduction

Human-accelerated phosphorus (P) transport from land to aquatic ecosystems has prompted major concern among scientists and policy makers because of its detrimental effects on water quality (Elser and Bennett, 2011). Although fertilizer use has increased crop yields, export of nutrients from agricultural fields and urban wastewater have also resulted in serious water quality issues. Algal blooms fueled by the excess nutrients can be directly toxic to humans and pets, limit fishing and recreational activities, and contaminate drinking water sources, as well as indirectly affect humans and fisheries by causing hypoxia (Anderson et al., 2002; Michalak et al., 2013; Rabalais et al., 2009). Like much of the rest of the world, United States (U.S.) waters are subject to eutrophication stress[1] and algal blooms, including blooms that produce cyanotoxins (Anderson et al., 2002).

Addressing these challenges will require quantitative understanding of P flows across the landscape and through ecosystems. An improved accounting of terrestrial P flows can also support

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[1] Information on impaired waters in the U.S. available at: https://www.epa.gov/nutrient-policy-data/waters-assessed-impaired-due-nutrient-related-causes.

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0043-1354/

effective ecosystem management and help identify opportunities for increased efficiency and recycling in natural, agricultural, urban, and industrial settings (Chowdhury et al., 2014). In the U.S. for example only 8% of P fertilizers used in 2007 were consumed in U.S. diets, and a quarter of mineral P fertilizer use was linked to agricultural exports (MacDonald et al., 2012), pointing to both high losses within the agricultural system (supported by Suh and Yee (2011)) and the importance of trade. In European Union member states, which are all dependent on P rock imports to support their food systems, national P budgets demonstrate that losses are also high in the food production/consumption system, often resulting in long-term soil P accumulation (van Dijk et al., 2016). Trade is also playing an increasing role in EU P flows, potentially because of increasing environmental regulation within the EU (Nesme et al., 2016). Still, there is significant variation among nations. For example although most soils in the EU have accumulated large stocks of P, some countries like Belgium and the Netherlands have continued to accumulate P while others like Slovakia or Austria are mining these soil resources (van Dijk et al., 2016; looking at 2005 annual P budgets). There can also be large variation within a country. For example, France reduced the magnitude of soil P budget surpluses between 1990 and 2006 across most regions, and as such increased its P use efficiency. However agricultural specialization has resulted in an uneven increase in P use efficiency between regions. More specifically animal intensive regions have not seen the same level of increased efficiency as crop-producing areas because of the local reapplication of manure (Senthilkumar et al., 2012). As international and national pressures on agricultural and waste management systems shift (e.g. by changes in fertilizer prices or pollution regulations), it is important to develop up-to-date information on P flows to ascertain what the impacts of such shifts may be on potential opportunities for changes in management (e.g., increased recycling or efficiency in use). In addition, a spatially explicit understanding of these P flows can help identify opportunities and consequences associated with changes (e.g., Metson et al., 2012 at the city scale), especially in a large country like the U.S. with large regional differences (e.g., Metson et al., 2016).

Terrestrial nutrient budgets are useful, but if we aim to target terrestrial sources to improve water quality, budgets must be paired with understanding of the relationship between sources of P and losses to waterways. A variety of statistical and mechanistic models have been used to better understand the relationship between terrestrial P management and losses to waterways. For example, at the regional scale the SPAtially Referenced Regression On Watershed attributes (SPARROW) model can account for 87% of the variance in TP loads and 68% of the variance in yields across the Mississippi River Basin (MRB), but requires 16 input parameters to achieve this level of predictive skill (Alexander et al., 2008). In contrast, a recently developed, uncalibrated, spatially-explicit, process-based global model, the Integrated Model to Assess the Global Environment–Global Nutrient Model (IMAGE-GNM), systematically underestimates Mississippi River P concentrations at 11 stations (with a root mean squared error of 51% (Beusen et al., 2015)). On the other hand smaller scale models such as the Soil and Water Assessment Tool (SWAT) (Douglas-Mankin et al., 2010; Gassman et al., 2007), the field-scale Erosion Policy Impact Climate (EPIC) model, and the multi-field extension version called Agricultural Policy Environmental eXtender (APEX, Gassman et al., 2004) have been used to predict P loading when using fine-scale location-specific data (e.g. Tripathi et al., 2003). In addition to these more complex mechanistic or statistical models, simple soil and water net budgets (e.g., Sobota et al., 2011) and net system budgets (also referred to as a mass balance approach), such as the net anthropogenic phosphorus inputs (NAPI, Russell et al., 2008; Hong et al., 2012) methods have be calculated at a variety of scales and geographical contexts and linked to water quality statistically. These budgeting approaches usually require fewer (and often more widely available) data inputs than the global or field-scale models, giving them a strong advantage in comparative work. Although there exist comparisons of such regional terrestrial budgets to riverine P, most studies focus on only one or two specific geographic areas. For example, Han et al. (2011) examined 24 watersheds around Lake Erie and Lake Michigan. Jacobson et al. (2011) looked at 113 watersheds in the MRB, while Hale et al. (2015) 42 watersheds in the North Eastern US,. In summary, although mechanistic models provide essential insights on the drivers of P riverine losses, simpler statistical models linking terrestrial budgets and riverine P are more easily implementable for comparison and targeting reductions at regional or large watershed scales.

With all of this as context we developed an updated (year 2012), spatially continuous P flux database for the entire conterminous U.S. (CONUS), and used this dataset to investigate continental-scale P dynamics. We then applied this dataset, in combination with a separate dataset containing river P export estimates from 72 watersheds distributed throughout the CONUS, to explore relationships between watershed characteristics (including P sources, sinks) and aquatic P loading across a wide range of environments. Importantly this exploration uses the same datasets across these regions as opposed to comparing previous works to eachother where methods, target years, and data-sources can confound the patterns that are identified between regions.

## 2. Methods

### *2.1. Overview*

In order to explore the relationship between anthropogenic terrestrial P sources, landscape characteristics, and water quality we quantified aquatic P fluxes as well as landscape and climate variables that have been shown to mediate the loss of P to waterways in other studies (Harrison et al., 2010, Jacobson et al., 2011, see Table 1 and supplemental information (SI)). For each of these landscape and climate characteristics a mean value for each watershed was extracted from gridded data using the “Zonal Statistics by Table” tool in ArcGIS 10.2 (ESRI, 2013), using watershed area polygons. We applied similar methods to those used in regional N studies (Boyer et al., 2002; Schaefer and Alber, 2007; Schaefer et al., 2009; Sobota et al., 2009) and one regional P study (Sobota et al., 2011) across the U.S., utilizing simple linear regressions between water quality variables: concentrations, yields, and TP fractional export (TP yield/nutrient inputs, see Eq. (6)), and potential explanatory variables. Water quantity and quality information were obtained from the USGS National Water Information System through the R dataRetrieval package (Hirsch et al., 2015) and processed with LOADEST (Runkel et al., 2004).

### *2.2. Anthropogenic phosphorus sources*

#### *2.2.1. Agricultural system*

P inputs considered included inorganic P fertilizers and manure P from confined livestock. P removal in harvested crops was also considered. For each of these P flows, data were obtained from two main sources to ensure consistency. We used phosphorus fertilizer sales, P produced by animals in confined feeding operations, and P removed in crop harvest compiled by the International Plant Nutrition Institute (IPNI, 2012) at the county level, following methods described by Kellogg et al. (2000) for manure. County level P fluxes were spatially disaggregated by assuming that fertilizer and manure P were applied to NWALT (U.S. National wall-to-

**Table 1**
Terrestrial P sources, land use, climate and hydrologic variables considered in this study with corresponding calculations, data sources and assumptions used for each variable.

| P input, output, and explanatory variables | Data year | Equation | Data source(s) | Major assumptions | Units |
|---|---|---|---|---|---|
| Fertilizer | 2012 | County P fertilizer sales/Area in cultivated crop and hay/pasture land uses in the county | (IPNI, 2012)<br>(Falcone, 2015)<br>(USDA ERS, 2015) | - Capped fertilizer application rate to 6000 kg P km$^{-2}$ yr$^{-1}$<br>- Mean fertilizer application by county over agricultural lands as opposed to crop, season, or field specific application rates.<br>- Assumes fertilizer sold in one county is applied in the same county (some exceptions where IPNI used smoothing functions). | Kg P ha$^{-1}$ yr$^{-1}$ |
| Manure | 2012 | County P in manure from confined operations/Area in cultivated crop and hay/pasture land uses in the county | (IPNI, 2012)<br>(Falcone, 2015) | - Capped manure application rate to 10,000 kg P km$^{-2}$ yr$^{-1}$<br>-Only considering recoverable P from confined operations.<br>- Mean manure application by county over agricultural lands as opposed to crop, season, or field specific application rates.<br>- Assumes manure produced in one county is applied in the same county. | Kg P ha$^{-1}$ yr$^{-1}$ |
| Crop uptake | 2012 | County major crops yields x major crop P content/Area in cultivated crop and hay/pasture land uses in the county | (IPNI, 2012)<br>(Falcone, 2015) | - Capped crop P removal rate to 9000 kg P km$^{-2}$ yr$^{-1}$<br>- Only considers uptake from 24 major crops as opposed to all crops.<br>- Mean crop uptake by county over agricultural lands as opposed to crop, season, or field specific yields and P content. | Kg P ha$^{-1}$ yr$^{-1}$ |
| Human sewage | Combination of 1970, 1993, 2012, and 2015<br>2012 | Population x (P excreted + P in detergents) x proportion connected to treatment *(1-(connection to primary, secondary, tertiary, and no discharge facilities x P removal from facilities)<br>Sum of major facilities (P concentration x discharge) | (Bright et al., 2012; Chapra, 1980; Garnier et al., 2015; Litke, 1999; Morse et al., 1993; US EPA2012)<br>EPA (2015) DMR tool | - Applies the same retention rate and sewage treatment level to the whole U.S.<br>- Assumes equal P emitted for each individual for the whole U.S.<br>- Only covers major point sources that have reported P loads. State data reporting varies and as such this represents an underestimate of P from major point sources. | Kg P km$^{-2}$ yr$^{-1}$<br>Kg P km$^{-2}$ yr$^{-1}$ |
| Natural land use | 2012 | Low usage (50) + Very low usage, conservation categories (60) in NWALT | (Falcone, 2015) | | % |
| Agriculture land use | 2012 | Crop production (43) + Hay/ Pasture production categories (44) in NWALT | (Falcone, 2015) | | % |
| Developed land use | 2012 | Developed (21,22,23,24,25,26,27) + Semi-Developed (31,32,33) categories in NWALT | (Falcone, 2015) | | % |
| Water land use | 2012 | Low usage + Very low usage, conservation categories in NWALT | (Falcone, 2015) | | % |
| Wetland land use | 2012 | Wetland category in NWALT | (Falcone, 2015) | | % |
| Potential grazing land use | 2012 | Potential grazing category in NWALT | (Falcone, 2015) | | % |
| Other land use | 2012 | Mining + timber categories in NWALT | (Falcone, 2015) | | % |
| Watershed area | NA | NA | HydroSHEDS Digital Elevation Model | | Km$^2$ |
| Elevation | 2006 | NA | HydroSHEDS Digital Elevation Model (Lehner et al., 2006) | - Mean elevation is representative for the watershed<br>- 15 arc sec resolution is high enough for purposes of this analysis | m |

(*continued on next page*)

**Table 1** (*continued*)

| P input, output, and explanatory variables | Data year | Equation | Data source(s) | Major assumptions | Units |
|---|---|---|---|---|---|
| Slope | 2006 | NA | HydroSHEDS Digital Elevation Model (Lehner et al., 2006) | - Mean slope is representative for the watershed<br>- 15 arc sec resolution is high enough for purposes of this analysis | degree |
| Temperature | 2012 | NA | (PRISM Climate Group, 2013) | - Mean annual temperature is representative for the watershed<br>- 4 $km^2$ resolution is high enough for purposes of this analysis | Degree Celsius |
| Precipitation | 2012 | NA | (PRISM Climate Group, 2013) | - Mean total precipitation is representative for the watershed<br>- 4 $km^2$ resolution is high enough for purposes of this analysis | mm |
| Fournier precipitation | 2012 | (Sum of square root of monthly precipitation)/Total annual precipitation | (PRISM Climate Group, 2013) following method in (Beusen et al., 2005) | - Mean annual variability in precipitation is representative for the watershed<br>- 4 $km^2$ resolution is high enough for purposes of this analysis | mm |
| Runoff | 2012 | LOADEST annual discharge estimate for site/Area of watershed | See text for LOADEST model and watershed delineation | - 2012 discharge is based on 10 years of measured discharge and concentration from the USGS for each station which was then processed in LOADEST<br>- Raw LOADEST discharge estimates are a sum of cubic feet per second for each month and thus we converted this to liters per year, divided by area which was in $km^2$, and converted to $l/m^2$ which are mm per year | mm |
| Tile drain | 1992 | NA | (Sugg, 2007) | - The overlap of row crops and poorly drained soils indicate the presence of tile drains<br>- There has not been significant changes in tile drainage since 1992<br>- County level resolution is high enough for the purposes of this analysis | % |
| Area of dams | 1999 | NA | The U.S. Army Corps of Engineers National Inventory of Dams (U.S. Geological Survey, 1999) | - The 75,187 dams considered in the survey are representative of the surface area (water area) taken up by dams | % |

wall land use trends map) defined cultivated crop and hay/pasture lands (Falcone, 2015, see Tables 1 and SI). In order to avoid unrealistically high calculated inorganic P fertilizer application rates (in some cases caused by high fertilizer sales in a county with low agricultural land area), P application rates were capped at 60 kg of P $ha^{-1}$, a rate that is double the highest application rate reported for corn in the 2010 ARMS Farm Financial and Crop Production Practice survey (USDA ERS, 2015). Similar maxima were implemented for manure P application and crop P removal. The cap for manure P application was set at 100 kg P $ha^{-1}$, which was slightly higher than the maximum reported application rate reported for 2006 in the ARMS Farm Financial and Crop Production Practice survey (MacDonald et al., 2009). The cap for crop P removal was set at 90 kg P $ha^{-1}$, which was higher than potential removal by a high-yield corn crop (300 bushels/acre (Murrell and Childs, 2000)) and corn P removal (Heckman et al., 2001). Lands where P application and P removal caps were applied did not account for a large amount of application or removal at the national level (1% of fertilizer sales and less for manure and crop removal, see SI). Estimates of agricultural net P balances across the U.S. were calculated at 60 m resolution as were the individual inputs and outputs of P (Eq. (2)) by subtracting crop removal from the total application of fertilizer and manure on a per area basis (Eq. (1)):

$$\boldsymbol{P_{net}} = \left(\boldsymbol{P_{fert}} + \boldsymbol{P_{manure}} - \boldsymbol{P_{harv}}\right) \tag{1}$$

where $P_{net}$ is the net anthropogenic agricultural P balance, $P_{fert}$ is P input as inorganic fertilizer on agricultural lands, $P_{manure}$ is P input as confined animal manure, and $P_{harv}$ is P removed as harvested agricultural crops all expressed as kg P $ha^{-1}$ $yr^{-1}$.

The per area P input and output of type *i* (fertilizer, manure, or crop harvest) on land cover type *j* (crop and pasture land uses) in county $k$ ($P_{ijk}$, kg P ha$^{-1}$ yr$^{-1}$) was calculated as:

$$P_{ijk} = \frac{P_{ik}}{A_{jk}} \tag{2}$$

where $P_{ik}$ is annual P input or output (kg P ha$^{-1}$ yr$^{-1}$) of type $i$ to county $k$, and $A_{jk}$ is the area of the land cover type receiving P input or output in county $k$. Application and removal rates were then distributed to pixels of agricultural land use (60 m resolution) in each county to create continuous maps of annual P application and removal rates for the CONUS.

### 2.2.2. Human excreta, detergents, and point sources

We used and compared two methods for estimating the amount of P entering waterways from human waste: a population-based method and a point-source database.

Population-based sewage P ($P_{sew}$) estimate was calculated as:

$$P_{sew} = D_{conn}H(P_{det} + P_{hum})(1 - ((D_{prim}R_{p-prim}) + (D_{sec}R_{p-sec}) + (D_{ter}R_{p-ter}))) \tag{3}$$

where $D_{conn}$ is the fraction of the U.S. population connected to centralized sewage, $H$ is population density (people per km$^2$, Bright et al., 2012), $P_{det}$ is per capita detergent use (Chapra, 1980; Litke, 1999), and $P_{hum}$ is per capita P excreted (Garnier et al., 2015). $D_{prim}$, $D_{sec}$, and $D_{ter}$ are the proportion of sewered population that is covered by primary, secondary, and tertiary sewage treatment nationally (US EPA, 2012), and $R_{p\text{-}prim}$, $R_{p\text{-}sec}$, and $R_{p\text{-}ter}$ are the P removal rates associated with primary, secondary, and tertiary treatment (Morse et al., 1993). This is similar to how Van Drecht et al. (2009) calculated human N and P sewer emissions globally, and Hale et al. (2015, 2013) calculated human N loads for the northeastern U.S. (Tables 1 and SI).

A separate estimate of point source P utilizing a point-source database took into consideration site-specific emissions, but data coverage was unequal and incomplete across the U.S. We used the EPA's Discharge Monitoring Report Pollutant Loading Tool[2] downloaded on July 1st, 2016 to compile annual 2012 P loads to waterways from major point sources (i.e., classified as major facilities in the tool). The tool combined National Pollution Discharge Elimination System (NPDES) permits and monitoring data as well as other federal agency data such as the Toxic Release Inventory for the whole U.S. annually (available for 2007–2016). Point sources for P included sewage treatment facilities as well as fertilizer plants, animal processing facilities, paper pulp and wood facilities, and other industries that use P. P loads for facilities in 2012 were downloaded and projected using latitude and longitude for each facility with ArcGIS. These data were the most recent and comprehensive available but likely underestimate P loaded to waterways because non-permitted facilities are not included in this database. In addition, although national in coverage, individual states may not have collected or reported the same quality of data, creating additional uncertainty.

## 2.3. Watershed selection and delineation

We selected USGS water quality monitoring stations with sufficient TP, DIP, and discharge information to run LOADEST. We used the USGS R dataRetrieval[3] package to access the National Water Information System (NWIS) database and iteratively applied the following criteria:

- At least 1 TP and 1 DIP sample per month covering 12 months between 2002 and 2012
- TP and DIP data in 2012
- Continuous, co-located water discharge data
- Drainage area >1000 km$^2$

15 arc-second flow direction maps from the HydroSHEDs project[4] (Lehner et al., 2006) were used in conjunction with the ArcGIS Watershed delineation tool to delineate watershed areas contributing to each water quality monitoring station (Fig. 1). Study watersheds averaged 201,389.79 km$^2$ in size (range: 2626 - 3,182,415 km$^2$). 44 out of the 72 watersheds were nested. Our analysis also included a wide range of total river flows (0.003–333 km$^3$ yr$^{-1}$, average 22.82 km$^3$ yr$^{-1}$, median 1.84 km$^3$ yr$^{-1}$). To normalize for basin size we used runoff (Eq. (4)), not discharge, as a potential explanatory variable for water quality variables. Runoff (*Runoff*) values ranged from 0.1 mm yr$^{-1}$ to 1494. mm yr$^{-1}$ and were calculated as:

$$Runoff = \frac{Flow}{Area_{watershed}} \tag{4}$$

where *Flow* is the total river discharge (liters per year) estimated by LOADEST at a selected USGS monitoring station and $Area_{watershed}$ is the drainage area of the watershed (km$^2$ see section 2.3), both of which are multiplied by $1.0 \times 10^{-6}$ to convert to mm per year.

## 2.4. River phosphorus export

Based on the criteria described in section 2.3 we compiled daily discharge (USGS code 00060), DIP concentration (USGS code 00665), and TP concentration (USGS code 00671) from 2002 to 2012 for 72 water quality monitoring stations. Median annual measured TP and DIP concentrations (mg TP or DIP l$^{-1}$) were used in statistical analyses. We ran the load estimator (LOADEST) program (Runkel et al., 2004) with these 10 years of data to calibrate LOADEST models for each site and calculated flow weighted TP and DIP loads (kg TP or DIP yr$^{-1}$) using the Adjusted Maximum Likelihood Estimation method. We then calculated yields (Eq. (5)) and TP fractional export (Eq. (6)). TP fractional export is expressed as a percentage of total P inputs (as in Eq. (1)) in order to be comparable to TN export studies (e.g. Boyer et al. (2002)).

P yields ($P_{yield}$, kg TP or DIP km$^{-2}$ yr$^{-1}$) were calculated as:

$$P_{yield} = \frac{P_{load}}{Area_{watershed}} \tag{5}$$

where $P_{load}$ is TP or DIP load as calculated by LOADEST (kg TP or DIP yr$^{-1}$).

Fractional TP export ($TP_{frac}$, %) was calculated as:

$$TP_{frac} = \frac{TP_{yield}}{\left(P_{fert} + P_{manure} + P_{sew}\right)} \times 100 \tag{6}$$

[2] https://cfpub.epa.gov/dmr/about-the-data.cfm.

[3] https://github.com/USGS-R/dataRetrieval dowloaded July 2015.

[4] Coordinates for each monitoring station from the USGS were shifted manually to create pour points that matched to the flow lines of HydroSHEDS flow accumulation layer. Each pour point was then run individually with the Watershed delineation tool using the HydroSHED DEM raster, transformed to a polygon, and then merged into 7 non-nested watershed layers for the extraction of mean landscape characteristics for the watersheds.

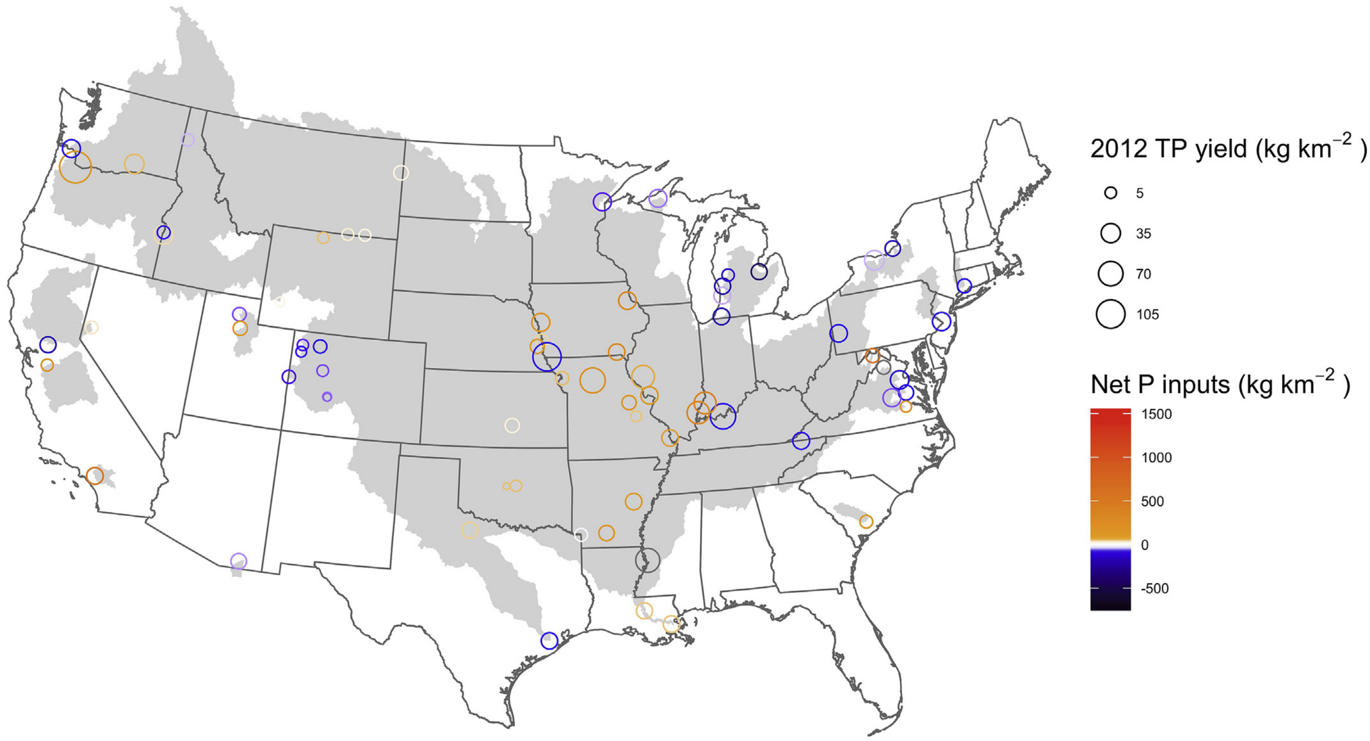


**Fig. 1.** Map of selected 72 USGS water quality monitoring stations (circles) and watershed area draining to these stations (grey area). The size of each circle represents the calculated TP yield (kg P km$^{-2}$) for the station and the color of each circle represents the net P additions (fertilizer + manure + sewage-crop uptake) to the watershed draining to the station (red indicates inputs exceed crop uptake while blue indicates crop uptake exceeds P inputs). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

### *2.5. Relationships between P inputs, P riverine exports, and watershed characteristics*

We used simple linear regression to evaluate relationships between water quality-related dependent variables (TP and DIP concentrations, yields, and TP fractional export) and independent variables likely to control P loading to surface waters, including: anthropogenic P inputs, and landscape, climate, and hydrologic variables. Most variables were natural log-transformed to improve normality, but land use percentages were arcsin square root transformed to improve normality. Because dependent variable sample size (water quality variables) was not large (72) relative to the number of explanatory variables, we chose not to use a multiple linear regression approach. We did however test multiple regressions with two independent variables: runoff plus one P source or sink at once, and single linear regressions between the residuals associated with runoff vs. water quality variables and P sources and sinks in order to more fully investigate the potential interplay between hydrology and terrestrial P (See SI Tables S5 and S6). All statistics were performed in R 3.2.2 (R Development Core Team, 2015). It is important to note that some of the linear regressions between explanatory and water quality variables were not completely independent, although previous regional fractional N and P export studies have also reported these regressions (e.g. Schaefer and Alber (2007), Sobota et al. (2011)). For example, runoff (Eq. (4)) is used to calculate yields (Eq. (5)) as they both used total flow divided by watershed area.

## 3. Results

We developed a national U.S. P budget in order to explore the relationship between anthropogenic terrestrial P inputs and outputs, landscape and climate characteristics, and water quality for the year 2012. We first present national P input and output results (Sections 3.1.1 and 3.1.2) as they both contextualize the P inputs to watersheds (Section 3.1.3), and provide insight into P dynamics across the U.S.. We then present water quality data (Section 3.2), and our efforts to relate P inputs and landscape characteristics to riverine P exports (Section 3.3).

### *3.1. Patterns of anthropogenic terrestrial phosphorus inputs and outputs*

#### *3.1.1. Agricultural P*

In 2012, the CONUS experienced a net P accumulation rate of 7.5% yr$^{-1}$ on agricultural lands: a surplus of 0.19 Tg P (a surplus here refers to a positive net P balance as described in Eq. (1), Fig. 2). Nationally, fertilizer was the most important P input, accounting for 74% of P inputs to these systems. Approximately 1.85 Tg of P was applied as inorganic fertilizer and 0.65 Tg of P was applied as manure. Of the 2.50 Tg of P applied to agricultural lands, 92.4% was removed with crop harvest (2.31 Tg P).

P application and removal rates across the U.S. varied widely, with large P surpluses most strongly mirroring confined manure P application (Figs. 3 and 4). On average, croplands had a 2.48 kg P ha$^{-2}$ yr$^{-1}$ surplus, but net P balances (Eq. (1)) varied substantially (median −0.49, max +142.82 and min −75.91 kg P ha$^{-1}$ yr$^{-1}$). Net agricultural P surpluses were fairly strongly and positively correlated with confined manure P application rates ($r^2 = 0.61$), and more weakly, although statistically significantly, correlated with P throughput (absolute value of inputs + outputs as a measure of P flow activity on the landscape; $r^2 = 0.35$), fertilizer application rates ($r^2 = 0.21$), and agricultural area ($r^2 = 0.01$). Net agricultural P surpluses were not correlated with crop P removal. In summary,

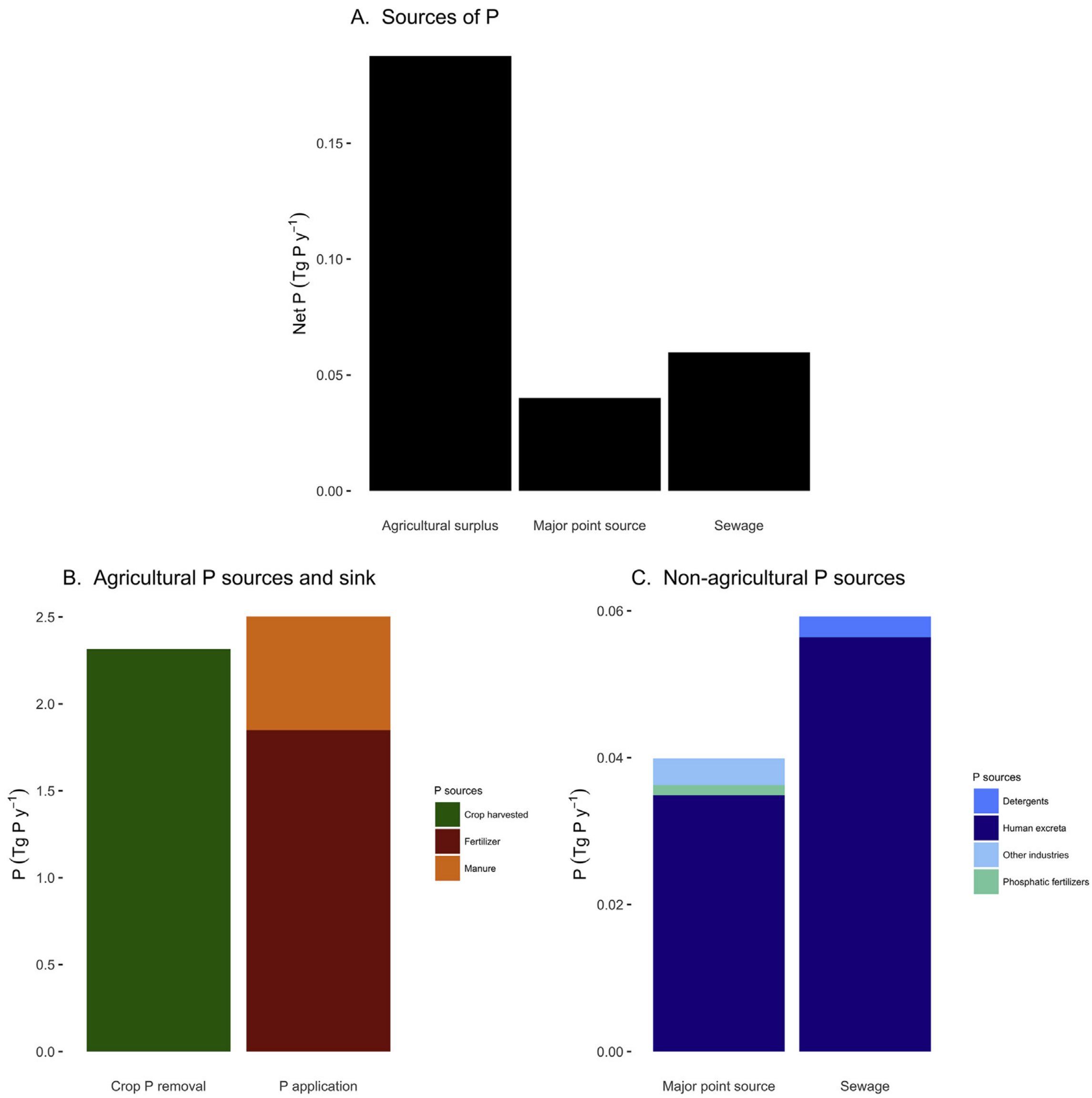


**Fig. 2.** Total 2012 terrestrial inputs and outputs of P for the conterminous U.S., showing the relative importance and magnitude of agricultural and non-agricultural inputs to the landscape and waterways as net sources (A), agricultural total inputs and outputs (B), and a comparison of non-agricultural P source estimates (C).

although U.S. agricultural lands may have had an overall P surplus in 2012, there were large variations across counties, which were mostly strongly correlated with confined manure applications.

Hotspots for confined manure P are clearly visible (Fig. 4B) and include: beef production in California's central valley, pork production in North Carolina, and poultry production in Delaware, Virginia, and West Virginia. Although patterns of P surplus were most strongly driven by manure P application; fertilizer was the largest single agricultural P input in most counties (78%) and states (34 out of 48). The average P fertilizer application rate was 8.72 kg P ha$^{-2}$ yr$^{-1}$ (median 5.68 kg P ha$^{-2}$ yr$^{-1}$). Application rates were highest in Nebraska, Arizona, Wyoming, and Indiana at the state level, but counties in other states also exhibited high rates, especially in California and Texas. High P fertilizer application rates correlated broadly with high crop P removal rates ($r^2 = 0.32$). The average application of P as confined manure was 5.12 kg P ha$^{-2}$ yr$^{-1}$ (median 1.37 kg P ha$^{-2}$ yr$^{-1}$) in 2012. Average P removal by major crops on agricultural land via harvest was 11.41 kg P ha$^{-2}$ yr$^{-1}$ (median 9.41 kg P ha$^{-2}$ yr$^{-1}$), and Arizona, California, Delaware, and Nebraska exhibited the highest state level removal rates (noting that Iowa, Minnesota, and Illinois had larger total P harvests, however), with the highest county level rates located in Texas and California. In summary, P fertilizer was the largest source of P in most counties, and patterns of confined feeding operations and the manure they produce were clearly visible in maps of agricultural P sources and in P surpluses.

### 3.1.2. *Human excreta and detergent and major point sources*

P fluxes from sewage were much smaller than P fluxes in the agricultural system (Fig. 2). Estimates of P losses based on national wastewater treatment coverage, population density, and per capita excreta and detergent use indicated that 0.06 Tg of P were lost to waterways, which is less than a third of our estimated net agricultural P surplus (0.19 Tg of P). At the state level, only Massachusetts was dominated by non-agricultural P sources. At the county scale however non-agricultural P inputs dominated in 157 counties in 35 states; the largest proportions of counties dominated by sewage P inputs occurred in Georgia, New York, New Jersey,

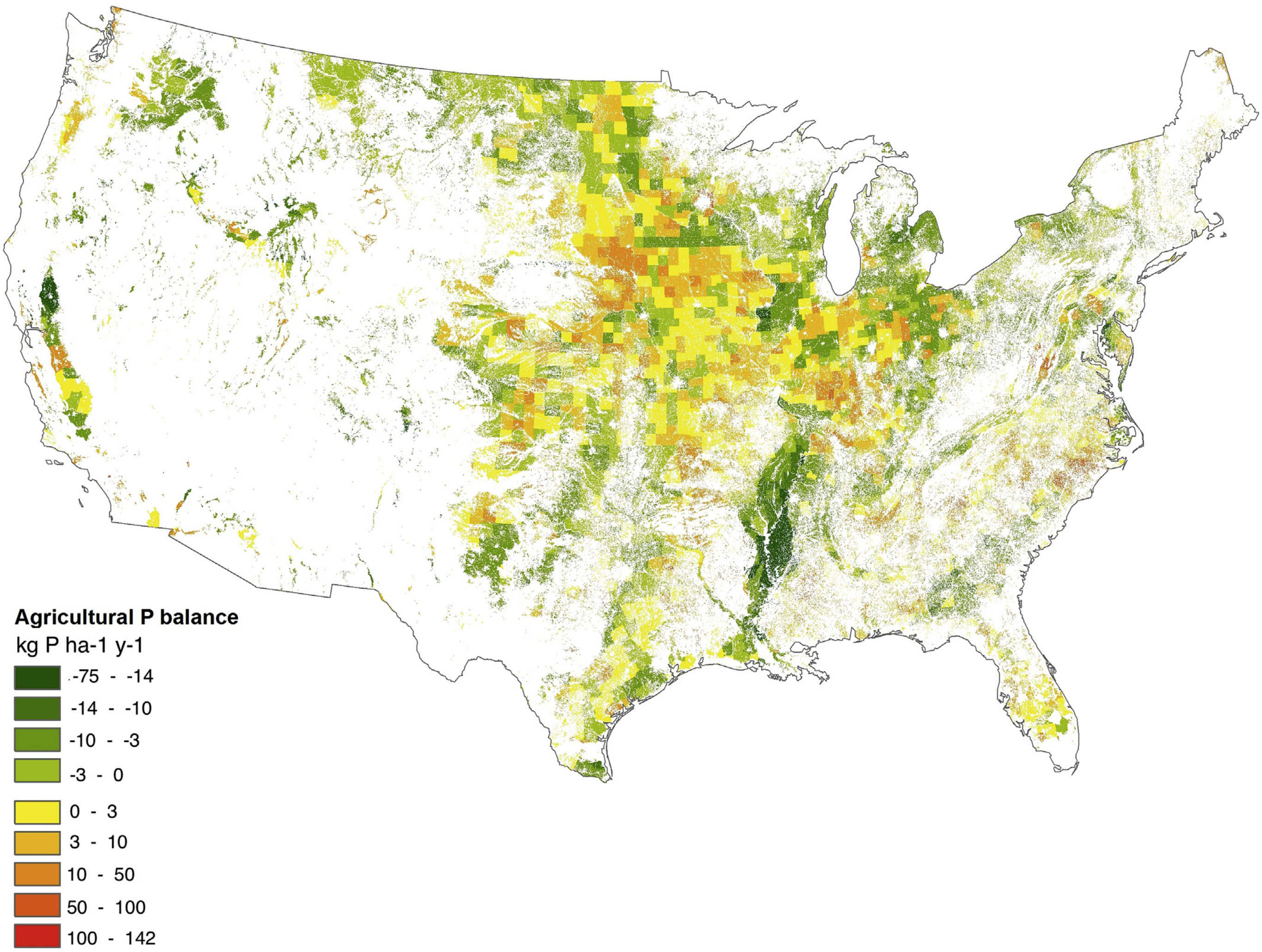


**Fig. 3.** Net agricultural P balance across the U.S. in 2012.

Colorado, Florida, and Connecticut.

Using two methods to calculate non-agricultural P inputs resulted in differences in total P input for the U.S. as well as differences in the spatial patterns of these sources (Figs. 2C and 4D). Estimates of P losses based on population density were 1.5 times greater than major wastewater treatment plant and industrial point sources reported to the EPA. Population density based estimates suggest that 0.06 Tg of P was lost to waterways while summing reported major point sources indicates that 0.04 Tg of P were lost to waterways. The 0.02 Tg discrepancy between these estimates suggests that non-major sources may be important or that state governments are underreporting point sources to federal agencies. Discrepancies between approaches are particularly pronounced west of the Mississippi river, and especially in Iowa (where no point sources were reported, Fig. 4D). In summary, although EPA's NPDES reporting provides important information on point sources of P to waterways, it appears to be under-reporting P delivery to aquatic systems by wastewater treatment systems. Consequently, population density estimates may be a superior estimate of point source P loading to US surface waters.

### 3.1.3. Watershed P balances

P inputs and outputs varied greatly across this study's 72 focus watersheds. The average net agricultural P balance (Eq. (1)) of our 72 focus watersheds (mean 2.23 kg P ha$^{-1}$ yr$^{-1}$) was similar to the national average (mean of 2.47 kg P ha$^{-1}$ yr$^{-1}$), although the watershed net agricultural P balances had a smaller range than all U.S. counties (Table 2). Average watershed P application rates and crop P removal on agricultural land were similar to national averages (mean fertilizer application: 8.62 kg P ha$^{-1}$ yr$^{-1}$, mean manure: 5.60 kg P ha$^{-1}$ yr$^{-1}$, mean crop removal: 11.98 kg P ha$^{-1}$ yr$^{-1}$ for watersheds and national averages in section 3.1.1 and 3.1.2). Crop removal amounted to 12–565% of P applied to fields as fertilizer and manure (average: 135%, median: 97%). Slightly more than half of the watersheds (38 out of 72) had a P surplus, meaning that P accumulated within the watershed during 2012 (fertilizer + manure + sewage-crop uptake). Values of P accumulation ranged from −7.82 − 15.2 kg P ha yr$^{-1}$ in 2012 (Fig. 1, Table S3). Fertilizer was the largest P input to the majority of watersheds, on average accounting for 64% of P inputs across watersheds. Manure inputs dominated in just two small, East Coast watersheds and, on average, sewage accounted for just 5% of P sources entering individual watersheds (mean sewage inputs: 6.32 kg P km$^{-2}$ yr$^{-1}$ min: 0.07 kg P km$^{-2}$ yr$^{-1}$ max: 91.38 kg P km$^{-2}$ yr$^{-1}$). Only one watershed in our study had non-agricultural P sources greater than agricultural P sources (the Santa Ana River below Prado Dam CA station, located in the densely populated Los Angeles-Riverside area). The negative P balances of watersheds

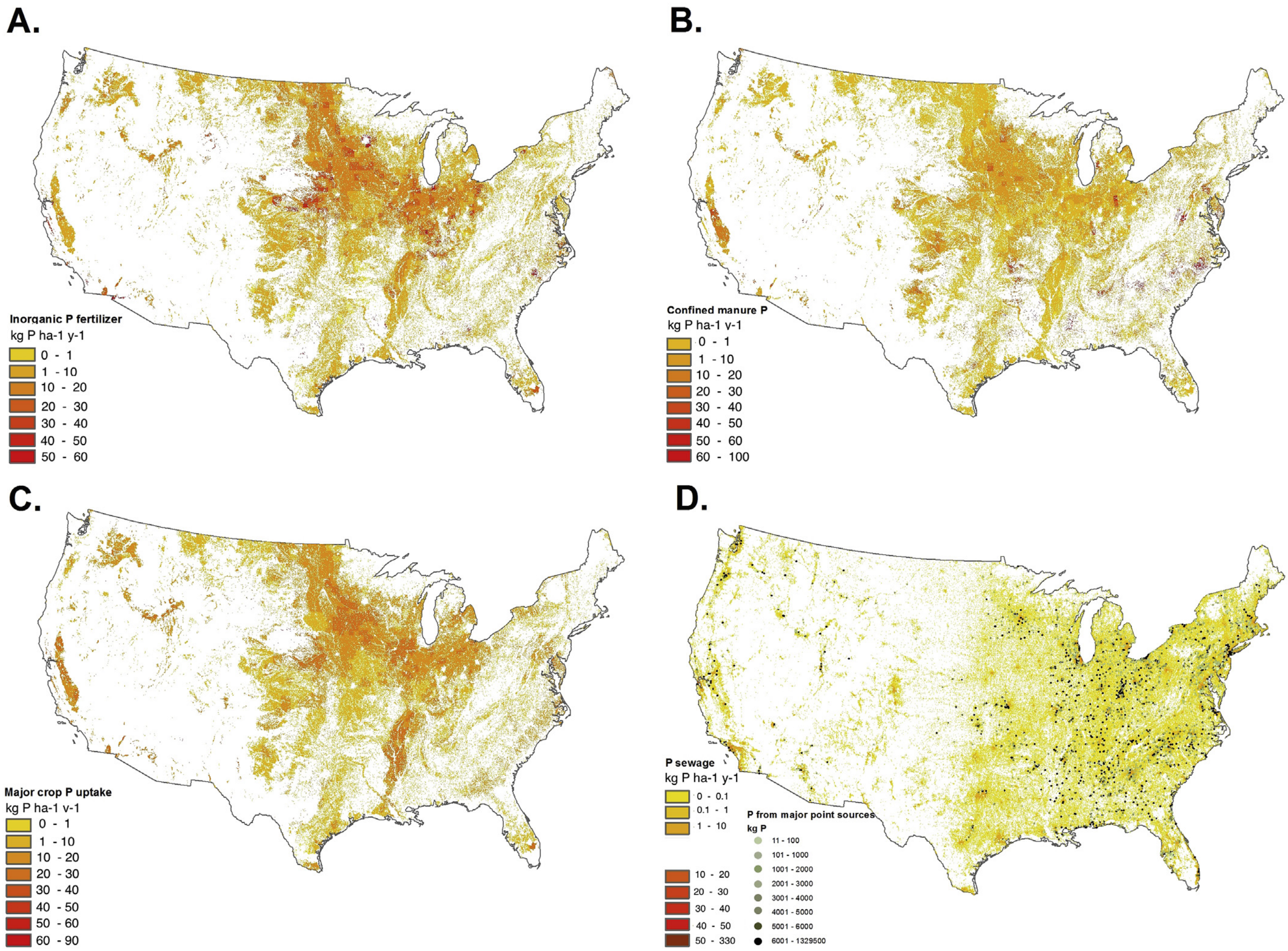


**Fig. 4.** Map of agricultural and non-agricultural P inputs and outputs across the U.S. in 2012 as fertilizer (A), manure (B), crop harvest (C), and human excreta and detergents and known point source emissions (green-black coloring) (D). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

indicated that crop removal was larger than manure and fertilizer applications and sewage combined in 2012. These negative balances may indicate that crops are utilizing P already present in soils naturally from rock weathering and plant decomposition or from previous fertilization practices, and/or that sources of P such as atmospheric deposition are also fertilizing crops. In summary, slightly more than half of our study watersheds exhibited P surpluses and, for 81% of watersheds mineral fertilizer was the single largest P source.

### 3.2. River P export

Concentrations and yields of TP and DIP, and fractional watershed export of TP varied considerably across watersheds (Table 2). Median measured concentrations of TP and DIP ranged over 3 orders of magnitude in 2012 (TP: 0.006–1.54 mg $^{-1}$ and DIP: 0.004–1.27 mg $l^{-1}$), with most concentrations elevated substantially above U.S. reference rivers, which have a median concentration of 0.023 mg P $l^{-1}$ (from 0.006 TP mg $l^{-1}$ in West to TP 0.08 mg $l^{-1}$ in the Great Plains, Smith et al., 2003). As such our studied watersheds seem to be human influenced. DIP made up 33% of TP concentrations on average and more than 50% of TP in 14 sites (total range: 3.2–97%). P yields (Eq. (5)) spanned five orders of magnitude (0.03–138.01 kg TP km$^{-2}$ yr$^{-1}$ and 0.02–35.90 kg DIP km$^{-2}$ yr$^{-1}$), which was similar to the variation in runoff (Eq. (4)) across watersheds (0.10 mm yr$^{-1}$ to 1494.5 mm yr$^{-1}$). The lowest TP and DIP yields were at the North Canadian River, Oklahoma station, and the highest were at the Willamette River station near Portland, Oregon. Both of these sites are outliers with regard to runoff; the Oklahoma site had the lowest runoff values in our dataset by an order of magnitude, and the Oregon site had the highest runoff values by more than an order of magnitude. The fractional export for TP (Eq. (6)) also varied considerably (Table 2), ranging from 0% in the medium-sized (35,359 km$^2$) North Canadian River, Oklahoma site to 442% in the small (3478.01 km$^2$) Ontonagon River Near Rockland, Michigan site. The average fractional export of watersheds was 25% (median 6%); in other words annual TP yields were often less than a quarter of the amount of terrestrial P added to a watershed. There was a weak (but statistically significant), positive relationship between the natural log transformed estimates of mean annual TP yields and total P inputs to watersheds (slope = 0.26, $r^2$ = 0.197, $p$ < 0.001). In summary, ranges of TP and DIP concentrations across watersheds are consistent with systems where nutrients are significantly impacted by human activities, and TP and DIP yield estimates vary with hydrology, as expected.

**Table 2**
Range of county and watershed land use, biophysical, and P river export characteristics *expressed as application and uptake on agricultural land and not on total area of county or watershed.

| | | County values | | | | watershed values | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | min | max | average | median | min | max | average | median |
| Area | ($km^2$) | 4.48 | 52,252.33 | 2505.13 | 1611.84 | 2626.00 | 3,182,415.00 | 201,390.00 | 14,098.00 |
| Elevation | (m) | 1.99 | 3464.73 | 447.58 | 282.29 | 75.96 | 3121.09 | 927.93 | 548.13 |
| Mean slope | (degree) | 0.05 | 14.27 | 1.51 | 0.76 | 0.24 | 11.30 | 2.91 | 1.91 |
| Mean temp | (C) | 2.09 | 24.98 | 13.87 | 13.86 | 3.18 | 19.23 | 10.99 | 10.35 |
| Precip | (mm $year^{-1}$) | 66.11 | 3621.52 | 890.44 | 889.74 | 223.40 | 2177.30 | 718.30 | 701.60 |
| Fournier precip | | 14.21 | 562.58 | 104.83 | 99.24 | 28.10 | 286.31 | 86.38 | 81.33 |
| Land use (% of watershed) | Natural | – | 99.81 | 48.53 | 49.05 | 5.00 | 97.97 | 61.08 | 65.04 |
| | Agriculture | – | 97.02 | 32.67 | 24.61 | 0.37 | 91.98 | 27.59 | 21.09 |
| | Urban | 0.08 | 97.39 | 11.48 | 3.72 | 0.28 | 43.86 | 3.64 | 6.87 |
| | Water | – | 60.66 | 2.29 | 1.14 | 0.03 | 13.69 | 1.46 | 1.02 |
| | Wetland | – | 76.46 | 3.43 | 0.31 | – | 36.15 | 1.86 | 0.28 |
| | Potential grazing | – | 20.86 | 1.13 | 0.53 | 0.01 | 3.54 | 0.84 | 0.30 |
| | Dam | – | 129.08 | 1.23 | 0.16 | 0.01 | 13.30 | 1.13 | 0.56 |
| | Tile drain | – | 81.73 | 3.51 | – | – | 43.36 | 3.61 | 0.09 |
| Population | (ind $km^{-2}$) | 0.03 | 29,741.03 | 88.26 | 14.42 | 0.37 | 474.51 | 33.00 | 16.51 |
| Fertilizer* | (kg $ha^{-1}$ $yr^{-1}$) | – | 60.00 | 8.72 | 5.67 | 0.66 | 30.14 | 8.62 | 6.47 |
| Manure* | (kg $ha^{-1}$ $yr^{-1}$) | – | 10.00 | 5.12 | 1.36 | 0.03 | 78.93 | 5.59 | 2.75 |
| Crop uptake* | (kg $ha^{-1}$ $yr^{-1}$) | – | 90.00 | 1.14 | 9.40 | 2.19 | 20.6 | 11.99 | 11.34 |
| Human sewage | (kg $km^{-2}$ $yr^{-1}$) | 0.00 | 6598.55 | 19.24 | 3.14 | 0.07 | 91.38 | 6.32 | 3.05 |
| Watershed net P inputs | (kg $km^{-2}$ $yr^{-1}$) | −1961.26 | 6598.55 | 64.82 | −49.64 | −788.40 | 1516.43 | 48.67 | 9.39 |
| Major point source | (kg $km^{-2}$ $yr^{-1}$) | – | 9418.55 | 15.50 | – | – | 38.66 | 2.83 | 0.49 |
| Runoff | (mm $yr^{-1}$) | – | – | – | – | 0.10 | 1494.47 | 194.42 | 121.99 |
| TP conc. | (mg $L^{-1}$) | – | – | – | – | 0.01 | 1.54 | 0.17 | 0.08 |
| DIP conc. | (mg $L^{-1}$) | – | – | – | – | 0.00 | 1.27 | 0.09 | 0.02 |
| TP yield | (kg P $km^{-2}$ $yr^{-1}$) | – | – | – | – | 0.03 | 138.01 | 22.32 | 17.14 |
| DIP yield | (kg P $km^{-2}$ $yr^{-1}$) | – | – | – | – | 0.02 | 35.90 | 6.13 | 3.73 |
| Fractional export | (%) | – | – | – | – | 0.01 | 441.87 | 25.79 | 6.03 |

### *3.3. Relationship between P inputs, watershed characteristics, and river P export*

Of the potential explanatory variables considered, TP concentrations were most strongly and negatively correlated with runoff, which explained 22% of the variance in TP concentration (Fig. 5). Inorganic fertilizer and manure application were also positively, although weakly, correlated with TP concentrations ($r^2 = 0.12$ for fertilizer and $r^2 = 0.12$ for manure, Table 3). DIP concentrations were also negatively correlated with runoff ($r^2 = 0.18$), with the second strongest correlation being a positive relationship with sewage P inputs ($r^2 = 0.10$). TP yields were most strongly positively correlated with runoff ($r^2 = 0.33$, Table 3, Fig. 5), followed by precipitation ($r^2 = 0.30$, Fig. S2), and although there was no significant relationship between TP yields and net P inputs, TP yields were positively correlated with P inputs to watersheds (Table 3, Fig. S1). The percent agricultural area and tile drained area in a watershed also correlated positively with TP yields, explaining 26% and 24% of the variance respectively. DIP yields, on the other hand, were most strongly positively correlated with estimates of major P point sources ($r^2 = 0.43$, Fig. S5) and P inputs from sewage ($r^2 = 0.42$, Fig. 1), followed by a positive relationship with crop P removal rates ($r^2 = 0.32$, Fig. S1). Doing multiple regressions with runoff and one P source or sink at a time increased the amount of variance we could explain. Combining runoff and sewage inputs resulted in the best-fit models for TP and DIP concentrations, and using runoff and manure as independent variables produced the best-fit model for TP and DIP yields (see SI Tables 5 and 6).

## 4. Discussion

### *4.1. Comparison of national terrestrial P budget to the literature*

To our knowledge, this 2012 inventory of terrestrial P sources and P concentrations and yields presented here is the most up-to-date yet developed for the CONUS, an important advancement since most global and national models of P losses are calibrated for the 1990's and early 2000's (e.g. Harrison et al., 2010; Alexander et al., 2008). We found that agricultural lands in 2012 received 0.19 Tg P more as fertilizer and confined manure than was harvested in major crops, and as such had a net surplus of P. Previous efforts to develop national P balances have found similar (albeit somewhat higher) agricultural P surpluses. For example, MacDonald et al. (2012) found a net agricultural surplus on agricultural lands in the U.S. for 2007 almost 3 times higher than our estimates for 2012 (0.53 Tg in 2007 compared to 0.18 Tg in 2012). They used different methods to calculate P flows, which could explain part of the discrepancy. MacDonald et al. (2012) used crop application rates as opposed to fertilizer sales data, resulting in a 1.4-fold higher estimate of per-area P balance for 2007 conditions. Similarly, summing the global half-degree resolution P balance data used recently by Beusen et al. (2016) for the continental U.S. suggests a year 2000 annual surplus 60% higher than our 2012 estimate (0.39 Tg of P in 2000). Again, the methodology and data sources were different from those used in our study. For instance, these year 2000 estimates were based on coarse land use data and incorporated pasture P application and grass uptake. In other words methodological differences certainly play a role in the discrepancy between this study and previous ones. However, part of the discrepancy could also be due to changes in management over time. U.S. fertilizer sales steadily increased from 1945, when total P fertilizer sales were 0.53 Tg P, to a maximum of 2.22 Tg of P in 1977 (Alexander and Smith, 1990), and then fluctuated between 1.9 and 1.5 Tg P $yr^{-1}$ between 1987 and the early 2000's (Gronberg and Spahr, 2012; IPNI, 2012; Ruddy et al., 2006); years 2000 and 2007 do have 2–3.4% higher P sales than 2012, which could also explain part of the variability observed between these studies and ours.

Across the U.S., there was a large amount of spatial variation in P surpluses and deficits (Fig. 3). More specifically, we found that areas with larger amounts of confined manure correlated more strongly

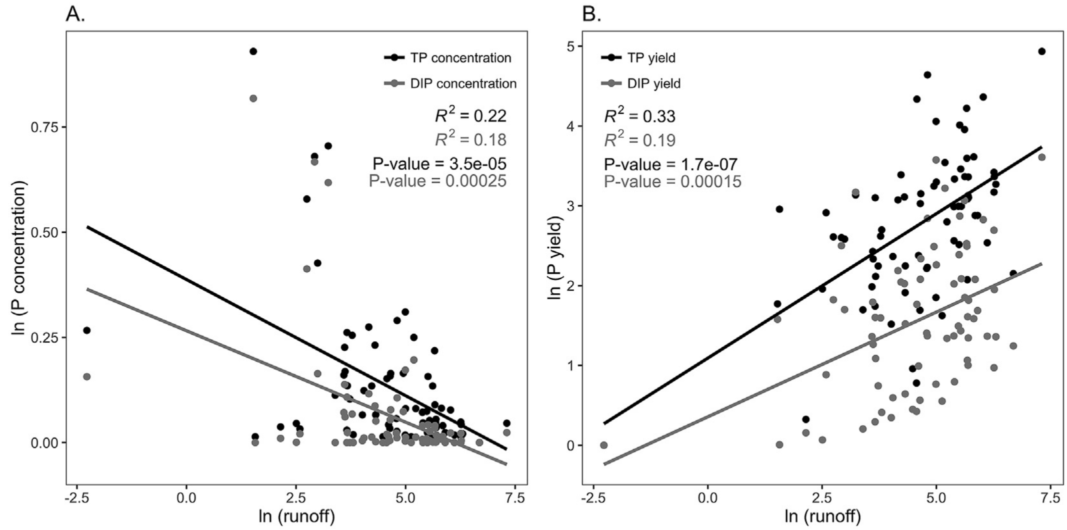


**Fig. 5.** Regressions of natural log transformed runoff with natural log transformed TP (black) and DIP (grey) concentrations (A) and natural log transformed (black) and DIP (grey) yields (B).

with P surpluses across the US, as has been shown in previous studies using older data. For example, concentrated pig production often results in soil P surpluses regionally as manure is often not transported very far (Jongbloed and Lenis, 1998), and countries with high animal production rates, such as the Netherlands, often have net nutrient surpluses (De Buck et al., 2012; Schipanski and Bennett, 2012).

The spatial segregation of recyclable P sources and needs has been identified as barrier to increasing recycling. For example in 2010, Jarvie et al. (2015) found that recyclable P sources (livestock and human) in the U.S. were 1.3 times larger than crop P needs and highlight how this discrepancy and lack of co-location of sources and needs prevents easy recycling. Although required transport distances vary, in order to meet 2002 U.S. corn demand for P from the Midwest US, recyclable P sources including manure not used within counties would have had to travel an average of 302 km (Metson et al., 2016).

Finally, non-agricultural P sources were much smaller than agricultural ones across the CONUS and were more spatially concentrated (Fig. 2), and were not consistent across data sources. We estimate that 0.06 Tg P were lost to waterways as sewage and detergent in 2012, a figure 1.5 times larger than that estimated using EPA records of major point sources of P. This discrepancy suggests that EPA's NPDES underestimates P delivery to aquatic systems in sewage by as much as 50% across the US, possibly more in western states. This is the first study to compare these two approaches to estimating non-agricultural nutrient sources to waterways in 2012. In summary this study provides updated spatially explicitly agricultural and non-agricultural P data for future use, in line with previous studies, and highlights data gaps.

### 4.2. Comparison with other studies linking terrestrial P budgets to aquatic P loading

Linking our national spatially-explicitly terrestrial P budget to riverine P export confirms many observations of links between P sources and losses in local studies, and does so across a wide range of environments with a consistent methodology and datasets: hydrology is often the best predictor of P losses. This result is consistent with observations for example from the Northeastern U.S. and Ecuador (e.g., Hale et al. (2015), Han et al. (2012), Borbor-Cordova et al. (2006)). Looking at the MRB, with averaged data from 1997 to 2006, Jacobson et al. (2011) used multiple linear regression to infer that TP yields were best predicted by the area planted in crops, human P consumption and precipitation. We also found that TP yields were correlated with precipitation ($r^2 = 0.30$), agricultural land use ($r^2 = 0.26$), and sewage discharge ($r^2 = 0.1$), although the strongest relationship in our study was with runoff (Table 3, Figs. S1, S2 and S4). Russell et al. (2008), like us, did not find a statistically significant relationship between riverine P loads and net terrestrial inputs to the Chesapeake Bay. In contrast, Hong et al. (2012) found a very strong ($R^2$ 0.94) between net P inputs (NAPI approach) and riverine P yields in the Baltic Sea, and so did Zhang et al. (2015) in the Huai River Basin of China. The combination of hydrology and P sources did increase our capacity to explain variance but together inputs and runoff explained just 40% and 42% of the variance in TP and DIP concentrations respectively and 46 and 54% of the variance in TP and DIP yields respectively (SI Tables 5 and 6). Runoff[5] was significantly ($p < 0.001$) correlated with all water quality variables examined, and agricultural P sources explained more variance in TP concentrations and TP yields than non-agricultural sources, while non-agricultural sources (sewage) explained comparatively more variance for DIP concentrations and yields than agricultural P sources. However no single explanatory variable (terrestrial P input, climate, hydrology or land use variable) accounted for more than 56% of the observed variance in riverine P variables.

Our results for DIP were consistent with insights from global modeling efforts which found that urban centers, and their associated human excreta and other urban waste inputs (e.g. detergents), were the most important source of DIP to the coastal ocean globally (Harrison et al., 2010). Wastes from point sources are often directly discharged to waterways, as opposed to agricultural P inputs, which are applied to soils and as such may be retained in soils and vegetation (Boynton et al., 1995; Carstensen et al., 2006). In contrast to prior studies (e.g., Beusen et al., 2016 Alexander et al., 2008) we did not find reservoirs, dams, or pasture and rangelands

[5] It is important to note that we expect correlations between runoff and TP and DIP yields because runoff (Eq. (4)) is used to calculate yields (Eq. (5)). As noted in the methods however, this regression is still commonly run in studies looking at drivers of nutrient losses towards waterways.

**Table 3**
Coefficients of determination ($r^2$) for all tested explanatory variables (rows) vs. dependent water quality variables (columns). These values are for the natural log transformed values of variables, except temperature which was not transformed, and land use variables which are arcsine-square root transformed. Bold values are for a significance of $p < 0.001$, and non-bold $r^2$ are for a significance of $p < 0.05$.

| Variables | TP conc. | DIP conc | TP yield | DIP yield | TP fractional export |
|---|---|---|---|---|---|
| Area | NS | NS | NS | NS | 0.058 |
| Elevation | NS | NS | **0.27** | **0.14** | NS |
| Mean slope | NS | NS | 0.075 | NS | **0.14** |
| Mean temp | 0.066 | NS | 0.072 | NS | 0.1 |
| Precip | **0.11** | 0.078 | **0.3** | **0.24** | NS |
| Fournier precip | 0.067 | NS | **0.3** | **0.26** | 0.057 |
| Natural land use | 0.058 | NS | **0.33** | **0.34** | **0.19** |
| Agriculture land use | 0.057 | NS | **0.26** | **0.26** | **0.3** |
| Urban land use | NS | NS | 0.12 | **0.22** | NS |
| Water land use | NS | NS | NS | 0.064 | NS |
| Wetland land use | NS | NS | NS | NS | NS |
| Potential grazing land use | NS | NS | NS | NS | **0.17** |
| Dam area | NS | NS | NS | NS | NS |
| Tile drain area | NS | NS | **0.24** | **0.24** | **0.12** |
| Population density | 0.074 | 0.097 | **0.21** | **0.46** | NS |
| Fertilizer | **0.12** | NS | **0.23** | **0.28** | **0.46** |
| Manure | **0.12** | 0.065 | **0.1** | **0.28** | **0.56** |
| Crop uptake | 0.089 | NS | **0.24** | **0.32** | **0.39** |
| Human sewage | 0.066 | **0.1** | **0.1** | **0.42** | NS |
| Watershed net P inputs | NS | NS | NS | 0.083 | 0.11 |
| Major point source | NS | NS | **0.2** | **0.43** | NS |
| Runoff | **0.22** | **0.18** | **0.33** | **0.19** | **0.12** |
| Flow | NS | 0.064 | **0.24** | **0.21** | NS |

to be an important explanatory variables for P export in our study watersheds.

Our calculated TP watershed fractional exports are reasonably similar to other studies of watershed P balances. In 2012, we found that riverine TP yields represented a small fraction of annual inputs for most watersheds but exhibited a very wide range of values across the U.S. (median of 6% ranging from 0.01% to 441%) even though at the national scale we found P to be used quite efficiently in agriculture (Fig. 2). The fractional P export values that we report have a similar median value, but larger range, than P fractional export reported in previous studies using the NAPI approach. Estimates of fractional export were −287% to 63% in California (using 100 × TP yield/net soil balance Sobota et al. (2011)), 0.7–8.7% in the Chesapeake Bay (using 100 × TP yield/net P balance for the NAPI approach, which is different than our definition: Eq. (6), Russell et al. (2008)), 3.6–25.3% in the Lake Erie and Lake Michigan watersheds (also using the NAPI approach in Han et al. (2011)). International examples using the NAPI approach include fractional exports of 1.6–14.2% in China (Chen et al., 2015), 0.6–71.4% in Ecuador (Borbor-Cordova et al., 2006), and 3–23% in the Baltic Sea (Hong et al., 2012). The larger range of fractional P export we report for U.S. watersheds may be related to the larger range of land-use and hydrology characteristics of watersheds in this study compared to the others cited above.

### 4.3. Other P sources and cycling mechanisms to consider

Although our analysis brings together recent and spatially resolved data on terrestrial and riverine P in the U.S., interpretations must take into consideration a number of limitations. For example we did not consider atmospheric P deposition because of the lack of national reporting on this flux, although P deposition may be an important driver of P concentrations, particularly in less disturbed watersheds (Stoddard et al., 2016). We also did not consider soil characteristics that mediate P loss such as clay content and soil bulk density (Jacobson et al., 2011), soil P content and soil porosity (Beusen et al., 2015), or soil P sorption and desorption properties (Frossard et al., 2000). These soil characteristics vary throughout CONUS and may help explain some of the large variations in TP export factions, as different soils vary in their capacity to retain new P inputs. Similarly, we did not take into consideration P processing and retention within aquatic ecosystems, which could alter the relationship between terrestrial P inputs and measured riverine P downstream (Powers et al., 2012). We also did not take into account within-watershed spatial variability of P inputs, and used annual averages for both terrestrial and aquatic P variables. Using an annual time step may have masked important seasonal patterns affecting P losses. Finer temporal resolution patterns may be particularly important as the timing of P applications and precipitation has been shown to affect P runoff and erosion (Sharpley et al., 2001; Haygarth et al., 1999, 2004). Inter- and intra-annual precipitation patterns, although affecting both N and P losses (Kleinman et al., 2006), may disproportionately affect P as particulate forms are a larger contribution of TP than for TN and particulates are more sensitive to storm-induced erosion (Boynton et al., 1995; Eyre and Pont, 2003; Yang et al., 2009), and such losses are predicted to increase in the face of climate change (Ockenden et al., 2016).

Finally, our focus on a single year precluded us from examining the role of soil P accumulation. Our findings indicate that on average, soil P stocks can be used by crops over time without the application of new P, but these stocks can also contribute to P losses to waterways even after terrestrial inputs have ceased (Jarvie et al., 2013). In fact, we posit that these soil P stocks may be part of the reason relationships between annual terrestrial P inputs and annual riverine P exports identified in this study were not as strong as those found in similar studies looking at N dynamics. Soil P status due to past management, also called legacy P, may explain why some watersheds in this study exported more P than was applied in 2012 (soil P stocks released), while others with high P application did not have high P yields (soil P stocks building up). Taking into account legacy P will be an important and challenging component of managing agricultural P, including the utilization of soil P stocks that have built up over time to keep yields high without additional P inputs (Haygarth et al., 2014; Sprague and Gronberg, 2012). As such legacy P will be an important area for future research (Jarvie et al., 2013; Sharpley et al., 2013). Continuing and expanded (time and space) soil P testing to allow for updated fertilizer recommendations should also be a topic of additional investigation (Kleinman et al., 2011).

### 4.4. Comparison of P and N dynamics

Interestingly the relationship between regional terrestrial N inputs and riverine N export in other studies is often stronger than is the case for P in this study. We found that runoff was the strongest predictor of TP and DIP concentrations but only accounted for 22 and 18% of variance for the two response variables, respectively. In contrast, regional studies of N find stronger statistical relationships. In California's Central Valley manure application was the best predictor of TN concentrations, explaining 62% of the variance (Sobota et al., 2009). Similarly, while we found that runoff accounted for 33% of the TP yield variance, the best fits for N yields explained 53% (total N inputs along the South Eastern U.S. (Schaefer and Alber, 2007)), 60% (total N inputs in California (Sobota et al., 2009)), 66% (streamflow along the U.S. West Coast (Schaefer et al., 2009)), and 80% (total N inputs along the North Eastern U.S. (Boyer et al., 2002)). In summary, P export to waterways seems to be more difficult to model than N to waterways which has also been shown in work that has explicitly looked at both nutrients such as the NEWS models globally (Seitzinger et al., 2005).

Our analysis suggests that P use efficiency in 2012 is generally higher (although more variable) than N use efficiency. We estimate that 92.4% of P applied to agricultural soils was harvested in crops, whereas only 74.6% of N applied as fertilizer, recovered manure, or fixed by legumes is incorporated into crops across the conterminous U.S. (IPNI, 2012). Watershed P balances, as well as TP fractional export, exhibited a wider range than previous regional watershed N studies in the US. Net inputs of Total N (TN) to East and West Coast watersheds in the U.S. have been reported as consistently positive (Boyer et al., 2002; Schaefer and Alber, 2007; Schaefer et al., 2009; Sobota et al., 2009) while the watershed P balances we report are roughly half negative and half positive (ranging from −788.4–1516 kg P $km^{-2}$ $yr^{-1}$). N fractional export in the studies mentioned above varied between 1 and 45%, with the majority of watersheds exporting less than 10% of the total N inputs to watersheds. Looking across the U.S. and Europe Howarth et al. (2012) found that on average 25% of TN is exported from watersheds (using the NANI approach). In the Baltic Sea region watersheds, there is higher fractional N export (15–47%) than P export (3–23%) using the NANI and NAPI approaches (Hong et al., 2012), which is generally consistent with our 2012 U.S. findings.

In summary although the median TP fractional export (6%) in this study is lower than for TN in the majority of studies, the range is much larger; some watersheds export more P than was applied in 2012 and others exporting less than 1% of terrestrial P inputs.

## 5. Conclusion

Eutrophication remains a major concern for U.S. waterways and the communities, farms, and fisheries that depend on this water for their health and livelihoods. At the national level we identified a net positive agricultural P balance for 2012, where areas of high livestock concentration exhibited even larger surpluses, which is consistent with other studies of previous years in the US. Our exploration of relationships between TP and DIP concentrations, yields, and TP fractional exports, and terrestrial P sources and landscape characteristics suggests that P losses are often mediated by runoff. In addition, considering P sources and runoff together increased the amount of variance that could be explained in water quality parameters. Our findings support previous studies that have demonstrated the importance of hydrology in mediating P losses in waterways. The relationship between annual terrestrial P management practices and losses to waterways is complex, requiring an understanding across scales, from local catchment-appropriate interventions, up to the importance of national and global trade and flows. Our 2012 spatially-explicit annual P balance approach and use of riverine P across CONUS confirms the importance of hydrology and difficulty of explaining the majority of the variance in riverine P across a wide range of environments with a consistent methodology and datasets. We suggest that future work should include more complete assessment of P inputs, including atmospheric deposition. It may also be fruitful to include soil characteristics explicitly in future continental-scale analyses. Finally, because of the strong P retention observed in most of our study watersheds in 2012, future work should also consider temporal P legacies and carryover from previous years as a driver of water quality responses.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.watres.2017.07.037.

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