Variable impacts of contemporary versus legacy agricultural phosphorus on US river water quality

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Phosphorus (P) fertilizer has contributed to the eutrophication of freshwater ecosystems. Watershed-based conservation programs aiming to reduce external P loading to surface waters have not resulted in significant water-quality improvements. One factor that can help explain the lack of water-quality response is remobilization of accumulated legacy (historical) P within the terrestrial-aquatic continuum, which can obscure the beneficial impacts of current conservation efforts. We examined how contemporary river P trends (between 1992 and 2012) responded to estimated changes in contemporary agricultural P balances [(fertilizer + manure inputs)—crop uptake and harvest removal] for 143 watersheds in the conterminous United States, while also developing a proxy estimate of legacy P contribution, which refers to anthropogenic P inputs before 1992. We concluded that legacy sources contributed to river export in 49 watersheds because mean contemporary river P export exceeded mean contemporary agricultural P balances. For the other 94 watersheds, agricultural P balances exceeded river P export, and our proxy estimate of legacy P was inconclusive. If legacy contributions occurred in these locations, they were likely small and dwarfed by contemporary P sources. Our continental-scale P mass balance results indicated that improved incentives and strategies are needed to promote the adoption of nutrient-conserving practices and reduce widespread contemporary P surpluses. However, a P surplus reduction is only 1 component of an effective nutrient plan as we found agricultural balances decreased in 91 watersheds with no consistent water-quality improvements, and balances increased in 52 watersheds with no consistent water-quality degradation.

legacy phosphorus | mass balance | eutrophication | phosphorus runoff | water-quality trend

The link among agriculture, excess phosphorus (P), and eutrophication in freshwater ecosystems is well established (1, 2). For several decades, watershed-based conservation programs have targeted reductions in P moving to surface waters either through erosion control or through reductions in P fertilizer application rates, but water-quality improvement has been limited. One factor that can help explain the lack of water-quality response after reductions in external P loading to freshwater ecosystems is the remobilization of legacy P, which can obscure the beneficial impacts of contemporary management efforts (3–6). Legacy P that has accumulated from historical fertilizer and manure applications can be remobilized at any point between the agricultural field and the watershed outlet and contribute to elevated surface water P concentrations and export (6). Additionally, as the buffering capacity of agricultural soils is reduced because of legacy P, freshly applied P is more susceptible to surface runoff (3). Global agricultural P balances, which compare the magnitude of manure and fertilizer inputs to crop uptake and harvest removal, have proven to be effective in assessing agronomic imbalances and the magnitude of legacy P stocks (7, 8), but studies that systematically identify where legacy P is a factor affecting water quality are limited. This is because of the high costs associated with sample collection and analysis of freshwater P concentrations (9, 10) as well as the challenges associated with connecting water-quality records to spatially and temporally consistent estimates of agricultural P balances.

Studies that have identified legacy contributions to river P export used 20–100-y P flux datasets and focused on 1 to 3 river basins (11–14). Generalized phases which characterize long-term watershed P input and output fluxes are as follows: equilibrium, accumulation, and depletion (15). In the United States, the equilibrium phase (inputs equal outputs) characterizes fertilizer and manure P inputs that are balanced by outputs, such as crop P uptake and harvest removal. Watersheds with more specialized crop and animal production, which are less integrated and require increased anthropogenic P inputs, are often characterized by an accumulation phase (inputs exceed outputs) (16, 17). The magnitude of legacy P in a watershed has been estimated by accounting for the cumulative agricultural P surplus during an accumulation phase (13, 15). Conservation efforts to reduce nonpoint source pollution and legislation to reduce point source pollution (1972 Clean Water Act) have promoted some watersheds to enter a depletion phase (outputs exceed inputs) (18). When watersheds enter this phase after a period of P accumulation, legacy P can be identified as a source to river export (13, 15).

The unique contribution of this study is the coupling of agricultural P balances ([fertilizer + manure inputs]—crop uptake and harvest removal) across a wide physiographic gradient with an analysis of river fluxes from both contemporary and legacy P sources. For the purposes of this study, the time frame for contemporary sources spanned from 1992 to 2012 and legacy sources were defined as inputs to the watershed prior to 1992. We

Significance

Management of agricultural phosphorus (P) has been in effect for decades with limited improvements in downstream water quality. Accumulated legacy (historical) P sources can mobilize, serve as a continual nutrient source, and mask the effects of conservation efforts to improve water quality through reductions in contemporary agricultural inputs to surface waters. We used a proxy estimate of legacy sources and assessed if P lingering in soils long after application was a major contributor to river export. For most watersheds, contributions of legacy P to river export were small in comparison to contributions from contemporary surpluses (fertilizer + manure > crop uptake). Estimating the magnitude of contemporary versus legacy P sources provides critical information to support effective implementation of management plans.

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leveraged newly developed empirical datasets of P inputs (fertilizer, manure, and wastewater treatment facility [WWTF] effluent), P outputs (crop uptake and harvest removal), and river P export for 173 watersheds in the United States. We assigned watersheds to 1 of 3 status categories (accumulation, depletion, or equilibrium) and used a proxy estimate of legacy P to determine if historical P inputs contributed to contemporary river export. This information provides knowledge that helps explain the variability in river P export trends in response to changes in agricultural P balances across watersheds in the conterminous United States.

Results and Discussion

Elevated P Concentrations in US Rivers. Classification of stream trophic status is based on nutrient concentration and producer biomass data collected from a large number of distinct river systems. In general, a total P (TP) concentration of 76 μg P L^{-1} or greater indicates a highly productive, or eutrophic system (19). Using this concentration of 76 μg P L^{-1} as a threshold for comparison, we found that 60% of the 173 watersheds examined as part of this study had TP concentrations that exceeded this value in 2012 (Fig. 1), suggesting that there is a high potential for eutrophication in most US rivers.

Agricultural P Surpluses Are Widespread Across the Landscape. Agricultural nonpoint P sources were the dominant signal in river export across the 173 watersheds. Manure and fertilizer fluxes were roughly 2 orders of magnitude larger than WWTF fluxes, which represented on average 10% of river P export (*SI Appendix*, Fig. S1). Using the manure, fertilizer, crop uptake, and harvest removal data, we estimated agricultural P balances for 5, 5-y time steps between 1992 and 2012. We averaged the 5, 5-y values to derive a mean watershed agricultural P balance and used a *t* test to assign each watershed to an agricultural P balance category. A mean balance significantly greater than 0 indicated accumulation, significantly less than 0 indicated depletion, and not significantly different from 0 indicated equilibrium (Fig. 2 *A*–*C*). Across all 173 watersheds, 68% consistently had P surpluses ranging from 3 to 1,950 kg km^{-2} (Fig. 2*D*). The accumulation phase watersheds had the highest animal production rates per unit area and had the highest manure inputs (*SI Appendix*, Table S1). These results indicate that there are opportunities to better manage agricultural P in relation to animal production by accounting for the manure application rate, the amount of land area available for spreading, and the distance from the manure source (18, 20, 21). In contrast, only 7% of the watersheds were deemed to be in a depletion phase with mean agricultural P balances ranging between −287 and −21 kg km^{-2}.

The combined mean values of fertilizer and manure application rates were higher for the 44 watersheds in the equilibrium phase as compared to the 117 watersheds in an accumulation phase. However, the mean agricultural P balance was less for those in an equilibrium phase because of higher crop P uptake and harvest removal rates. These watersheds had the highest corn and soybean production rates out of the 3 agricultural P status categories (*SI Appendix*, Table S1). Since our method for classifying P balances relied on the results of a *t* test, many of the study watersheds in equilibrium showed large fluctuations in P balances above and below 0 between 1992 and 2012 without having a mean P balance significantly different from 0. Of the 44 total watersheds in this P status category, 19 had mean P balances that were relatively close to 0 (±30 kg km^{-2}), but 25 watersheds had much a much wider range (between −140 and 350 kg km^{-2}). Assuming that equilibrium is desired (15), the variability of agricultural P balances across equilibrium watersheds (Fig. 2*D*) suggests that balancing P inputs with crop P uptake and harvest removal is challenging at this scale.

Many of the equilibrium and depletion-phase watersheds were located in basins targeted for nonpoint source P reductions, while the accumulation phase watersheds were distributed more widely (Fig. 1*B*). Equilibrium watersheds were located on the mid-Atlantic coast, including some of the smaller tributaries to the Chesapeake Bay as well as in the Upper Midwest, including rivers draining to the Lake Erie Basin. Many of the depletion-phase watersheds were located in the Susquehanna River Basin, which is a tributary to the Chesapeake Bay. Both the Lake Erie and the Chesapeake Bay basins have been the targets of efforts to reduce nonpoint sources of P since the early 1980s and 1990s (11, 18, 22).

Legacy P Contributions to River Export. Our proxy estimate of legacy P was based on the ratio of contemporary watershed mean river P export to mean agricultural P balance. If the ratio was greater than 1, we assumed legacy P contributed to river P export and the source was an external P input to the watershed before 1992. If the mean P balance was less than 0, legacy P was assumed to be the entire source of P exported by rivers. We excluded 30 watersheds from the original 173 for this analysis because estimated P export from upstream WWTF accounted for more than 50% of the total river P export (*SI Appendix*, Table S2). Considering the 143 remaining watersheds, our proxy estimate of historical P sources showed that legacy P contributed to 100% of river P export for the 11 depletion watersheds, 54% for the 39 equilibrium watersheds, and 3% for the 93 accumulation watersheds (Fig. 1*C*). Across all 143 watersheds, we identified 49 watersheds where river export was supported by contributions from legacy P.

Remobilization of accumulated legacy P can occur anywhere along the terrestrial-aquatic continuum and contribute to elevated river P export. P can desorb from agricultural soils long after fertilizer and manure applications and move via surface or subsurface flow paths into freshwater ecosystems (6). Additionally, legacy P can desorb from sediments in riparian and river bed environments and be released directly into the water column (23). Our results reflect an integration of legacy P sources that may occur in the field, riparian wetland, and river bed environments within each watershed. The average river export for all 49 watersheds where legacy P was a river source was 86 kg km^{-2}, and the mean agricultural P balance was 5 kg km^{-2}. Interestingly, these watersheds were distributed across all P status categories. For the other 94 watersheds, agricultural P balances were large relative to river P export, and the ratio of mean river export to agricultural P balance for each of these watersheds was always positive, but less than 1. The average P balance across these watersheds was 370 kg km^{-2}, and the average river export was 56 kg km^{-2}. In these cases, legacy P may be contributing to the river P export, but it was not possible to conclusively establish the presence or magnitude of those legacy inputs because contemporary P surpluses were so large. It is important to bear in mind that our results do not preclude a scenario of historical P accumulation in these locations, and once these watersheds enter a depletion phase, the relative magnitude of legacy sources to river export may be quantified.

A limitation to our approach is that we assumed manure and fertilizer P always fed a crop, and only after crop needs were met, did surplus P contribute to river export. Similar to other P balance studies (24), we depended on historical P balance data that are only available at coarser spatial and temporal scales than is required to capture events where fertilizer and manure may unintentionally be transported to the river instead of being taken up by a plant. This includes events, such as runoff following a livestock manure application to frozen fields or fertilizer and manure applications just prior to an irrigation or rain event. Additional research and modeling of stochastic field-scale P balances are required before we can account for these contributions to the long-term watershed scale P balances that were estimated in this study.

Impacts of Agricultural P Management on Water-Quality Trends. This study provides a comprehensive analysis of how river P trends respond to changes in agricultural P balances, while also accounting for legacy P contributions (Fig. 3). The synthesis of these 20-y trend datasets showed that agricultural P balances decreased in 91 watersheds, but those reductions did not consistently translate

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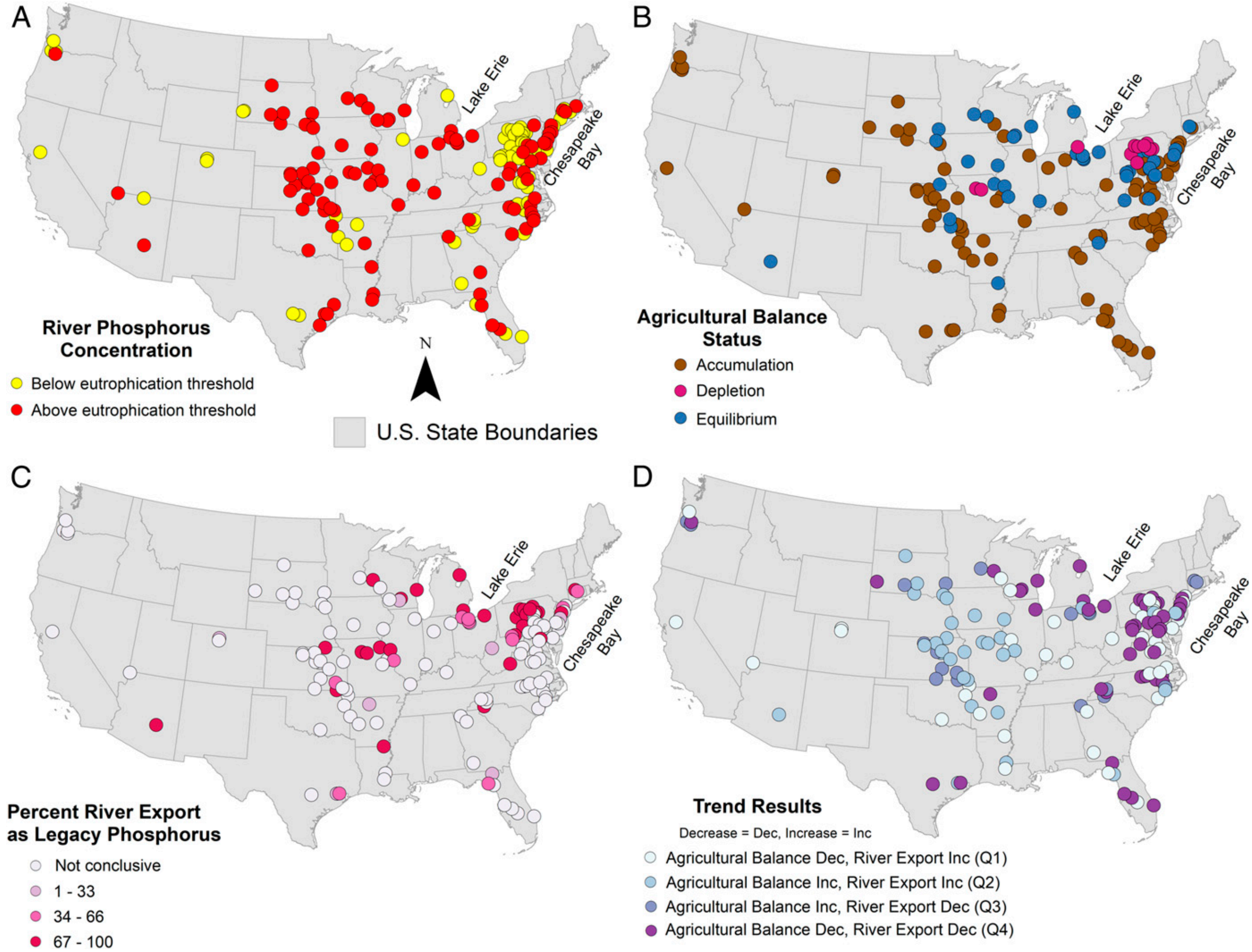

Fig. 1. (*A*) River total P concentrations in 2012. A 76-μg L^{-1} eutrophication threshold was used. (*B*) Watershed agricultural P balance status. (*C*) Percentage of river P export from legacy P sources. The 94 watersheds had contemporary mean agricultural P balances that exceeded mean river P export (white circles), so the proxy estimates of legacy contributions to river export were inconclusive at these locations. (*D*) Agricultural P balance and river P export trends. Q1–Q4 refer to the 4 quadrants in Fig. 3 decrease (Dec) and increase (Inc). Symbols in all 4 panels indicate the locations of streamgages in our water-quality data set that were used to estimate river P export and represent the most downstream point in the watershed.

into improved water-quality conditions. For the 43 watersheds in Q1, river P export increased despite reductions in agricultural P balances. Legacy P contributions to river export did not help explain this apparent paradox as only 8 of the 43 watersheds were identified as being affected by legacy P (*SI Appendix*, Table S3). Even though these watersheds showed decreases in agricultural P balances over time, fertilizer and manure inputs still exceeded crop uptake and harvest removal in these locations, and 80% of the watersheds in Q1 were in an accumulation phase. For the 48 watersheds in Q4, reductions in agricultural P balances were associated with decreases in river P export. The average agricultural P balance of the watersheds in Q4 where river P export decreased was 76 kg km^{-2}. In contrast, the average balance for Q1 where river P export increased was twice as large at 160 kg km^{-2}. Agricultural balances increased in 52 watersheds (Q2 and Q3), but those increases did not consistently translate into increasing P export. Thirty-three watersheds showed river P export increases (Q2), and 19 watersheds showed decreases (Q3). The mean agricultural P balance for the watersheds in Q2 and Q3 were 189 and 182 kg km^{-2}, respectively. Overall, there was no significant relationship between the change in P balance and the river P export (slope = 0.02, intercept = −1.9, R^2 = 0.02, and P value = 0.05), indicating that changes in agricultural P management aimed at decreasing P surpluses did not consistently result in improved downstream water quality.

Watershed buffering capacity and implementation of best management practices (BMPs) may explain the variable water-quality response to changes in agricultural P balances. From a water-quality perspective, buffering capacity on a field scale refers to the ability of soil to absorb additional P without contributing to river P export, and factors that affect this include a soil's mineral composition, organic matter content, and redox condition (25, 26). The buffering capacity concept can also be applied on a larger scale to indicate an entire watershed's ability to absorb excess P in the soil, groundwater, or riparian areas (27, 28). Estimation of a watershed buffering capacity requires P fluxes that capture the onset of the P accumulation phase and the abrupt shift in river P export relative to total P stocks, which indicates the loss of the watershed's ability to absorb additional P (29). For the purposes of this study, BMPs refer to in-field management (e.g., vegetated buffer strips, cover crops, and reduced tillage) or edge-of-field practices (e.g., sediment traps or constructed wetlands) implemented to improve downstream water quality. There are a wide range of BMPs available to reduce physical transfers of P from soils to surface waters (20), and, in the United States, these practices are funded by multiple entities on the

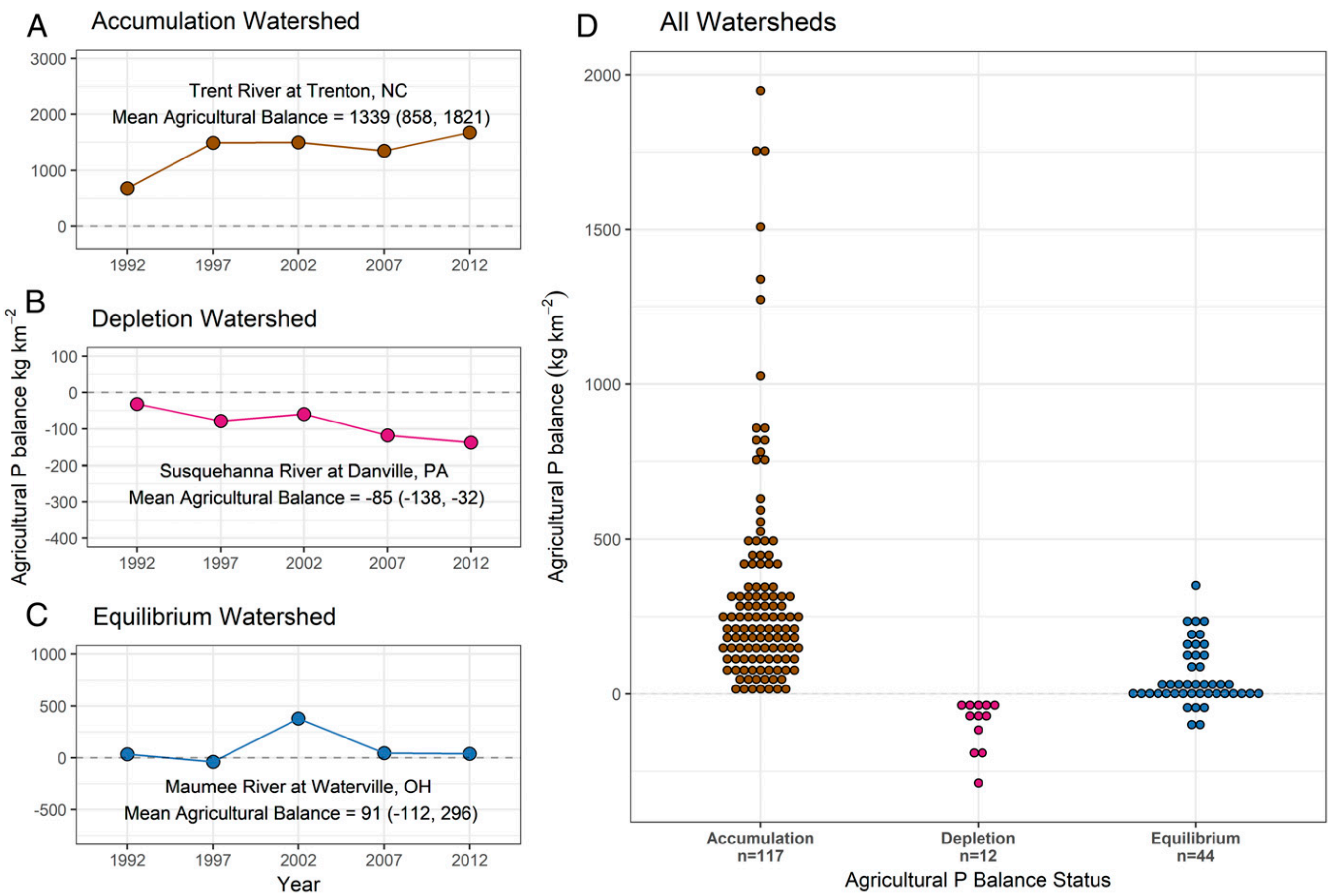

Fig. 2. Examples of watersheds in an (*A*) accumulation, (*B*) depletion, and (*C*) equilibrium status category with the mean of the 5 data points and lower and upper confidence intervals. (*D*) Mean agricultural P balances for 173 study watersheds by P status category. Each point represents a watershed. N = the number of watersheds within each status category. See Fig. 1*B* for the geographic locations of the watersheds in each category.

federal, state, and local scales. To date, this information has not been assimilated into a national-scale database.

Given the wide geographic distribution of our study, there is likely a range of BMPs implemented across the different watersheds. Additionally, our study watersheds reflect differences in agricultural pressures and conservation histories, so it is likely that the amount of P stored in the soils, sediments, and groundwater at the start of this study period in 1992 varied. We were not able to address watershed buffering capacity or BMPs directly in our analysis, but consideration of their effects on water quality, in general, could help explain the different trajectories in river P export trends in response to changes in contemporary P balances. We hypothesize that implementation of BMPs along with overall reductions in agricultural P balances could reduce the transport of surplus P from fields to streams and decrease river P export. This would help explain the watersheds that were in an accumulation phase but responded positively to decreased agricultural P balance (i.e., Q4 in Fig. 3). The river P export increases for watersheds in Q1 could indicate an exceedance of the watershed buffering capacity. Since the majority of these watersheds were in an accumulation phase, surplus P was available, and we hypothesize that the limit for absorbing additional P had been reached in these locations, so river P export increased despite improvements in the agricultural P balance. Absence of BMPs in these watersheds could also have further limited the ability to absorb additional P. Some watersheds in Q2 and Q3 had large increases in agricultural P balances with little response in river export (increases or decreases were less than 10 kg km^{-2}). We hypothesize that landscape management could have decreased overland erosion and runoff of P into rivers for these watersheds. These results could also reflect available buffering capacity such that surplus P accumulated in the watershed but was not reflected by increases in river P export.

Our results highlight the pressing need to understand how watersheds may respond to decreasing agricultural P balances, while also accounting for temporal lags in water-quality improvements and interactions with BMPs. Furthermore, additional strategies are needed to promote the adoption of nutrient-conserving practices without compromising agricultural yields. These practices need to be developed in the context of climate change as heavy precipitation has been linked to enhanced transfer of nutrients from terrestrial to aquatic systems (30, 31). Observed increases in heavy rainfall events across most of the country are projected to continue (32). Therefore, the strategies to address P management at its source, the field, are urgently needed now as only a small amount of the P available in the soil solution needs to be transferred to freshwater ecosystems to cause a significant increase in stream water concentrations and promote eutrophic conditions (26). Additionally, the cost to remove or remediate excess P generally increases with the distance from the source of loss (20). Further research to integrate information about watershed buffering capacities and BMPs into a mass balance framework that is system specific is needed. This would contribute to the achievement of more consistent water-quality improvements across the United States.

Methods

Selection of River P Export Study Watersheds. River P concentrations and export were estimated using the Weighted Regression on Time, Discharge, and Season (WRTDS) model (33), and input data were empirical discharge and TP

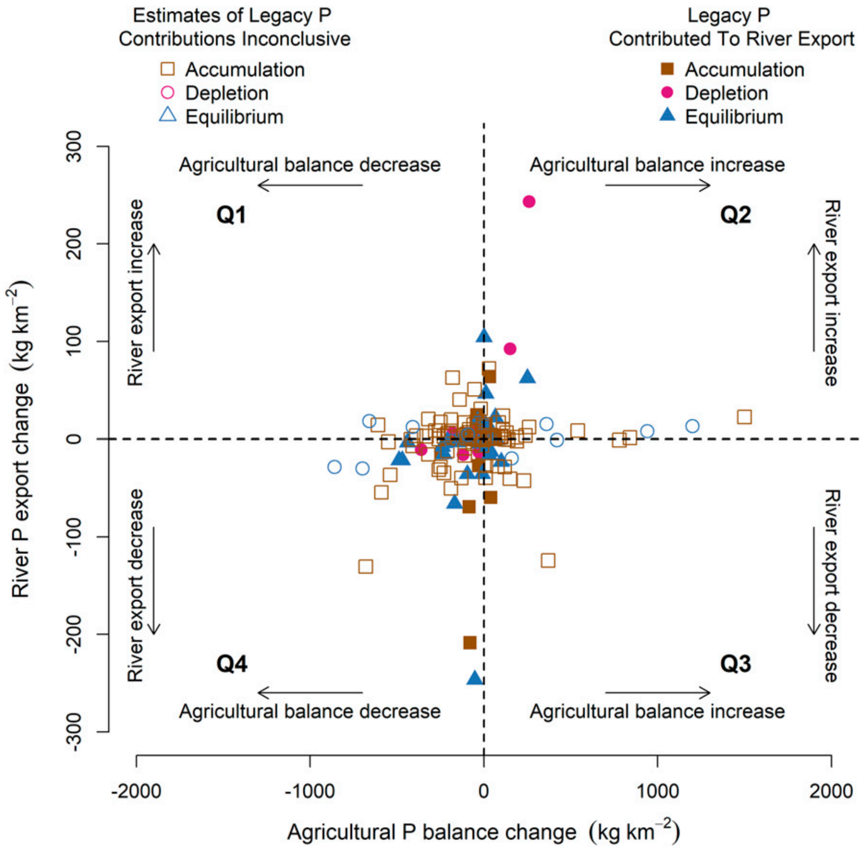

Fig. 3. Comparison of estimated change in river P export and agricultural P balances between 1992 and 2012 for 143 watersheds. The Thiel Sen Slope was used to estimate trends in agricultural P balances. The weighted regressions on time discharge and season model were used to estimate trends in river export. Symbol colors and shapes vary based on assigned watershed agricultural P balance category: accumulation, depletion, or equilibrium. Filled symbols represent watersheds where river export exceeded agricultural balances and legacy P contributions to river export were estimated. Open symbols represent 94 watersheds where it was not possible to conclusively establish the presence or magnitude of legacy input. There was no significant relationship between the change in P balance and river P export (R^2 = 0.02, P value = 0.05). See Fig. 1*D* for the geographic locations of watersheds in each of the 4 quadrants.

concentration data collected at streamgage locations throughout the United States. An expanded description of the WRTDS model is presented in *SI Appendix*, *SI Materials and Methods*. In our study, the streamgage location was used to define the watershed outlet, so river P export represented 1 of 2 main ways that P could leave the watershed; the other major P output defined in this study was crop uptake and harvest removal. We examined the finalized river P export dataset (34), which had river P export trend estimates for 551 watersheds (*SI Appendix*, Table S4) to find the maximum number of watersheds with the longest period of record, keeping in mind that all study watersheds needed to have consistent time frames for the trend analysis. Additionally, we considered that the 28 federal, state, and academic monitoring agencies which provided the data used to estimate river P export were often interested in monitoring flow and constituent transport, so multiple gages were often situated along the channel of a single river. We removed an upstream watershed if more than 50% of its area overlapped with a downstream watershed. A total of 173 watersheds with a 20-y period of river P export results between 1992 and 2012 met our requirements, and the magnitude of the fluxes for the study watersheds was representative of those included in the full river P export trend dataset (*SI Appendix*, Fig. S2).

Agricultural P Balances. The watershed agricultural P balances account for fertilizer, manure inputs, crop uptake, and harvest removal on the terrestrial landscape for the drainage area upstream from the streamgage location associated with the river P export estimates. Agricultural P balances for 173 watersheds were estimated as: agricultural P balance = (fertilizer + manure)—crop uptake and harvest removal, using data from Falcone (35). Watershed fertilizer and manure (kg km^{-2}) estimates for the conterminous United States used in this study can be found in Datasets 14 and 15, respectively. The Association of American Plant Food Control Officials provided fertilizer sales data to derive a state-level total P (tons) for each fertilizer product applied in both "farm" and "nonfarm" settings, and county-level P rates were derived using fertilizer expenditure data from the Census of Agriculture (CoA) and human population data from the US Census Bureau (36). Farm P rates were apportioned based on the dollar amount spent for fertilizer at the county relative to the state level, and nonfarm P rates were apportioned based on human population density. Estimates of total manure P excreted by animals were based on animal life span (days), animal population inventories (animals), and estimates of manure P content (kg animal^{-1} day^{-1}). The county-level fertilizer and manure P input data were converted to national grids by allocating the tabular county data to areas where inputs were likely to occur. Raster datasets of manure and fertilizer P (kg km^{-2}) were produced by assigning manure and farm fertilizer P to agricultural (crops and pasture/hay) pixels and nonfarm fertilizer to urban pixels. The agricultural and urban land cover data were derived from the National Water-Quality Assessment Wall-to-Wall Anthropogenic Land Use Trends (NWALT) dataset (37), which is a series of decadal raster datasets from 1974 to 2012 depicting land use at 60-m resolution for the conterminous United States. The final watershed-scale fertilizer and manure P input values (kg km^{-2}) were derived by estimating the mean value for pixels found within watershed boundaries (38).

Crop uptake and harvest removal were estimated as watershed crop P uptake and harvest removal = $\sum$ (crop nutrient content$_i$ × crop yield$_i$), where i stands for crop type. *Crop nutrient content* (kg P crop unit^{-1}) values (39) were constant throughout the study period (1992–2012). Crop yields were based on CoA data found in Dataset 3 (35), which has yields per unit crop area for 7 major crop types: corn grain (bushels km^{-2}), corn silage (bushels km^{-2}), cotton (bales km^{-2}), rice (hundredweight km^{-2}), soybeans (bushels km^{-2}), wheat (bushels km^{-2}), and alfalfa hay (dry tons km^{-2}). Pasture area (km^2) was derived from NWALT data. Watershed-scale animal production (number of animals km^{-2}) for chickens, cows, and hogs was also derived from the CoA data.

Assigning Agricultural P Balance Status Categories to Watersheds. The CoA crop and annual production values were available every 5 y between 1992 and 2012, providing 5 estimates of agricultural P balances within the study time frame (40). Contemporary mean agricultural P balances were derived by averaging data across 5, 5-y time steps for the 20-y study period. We used the results of a *t* test to determine if each watershed mean agricultural P balance was in an accumulation (significantly greater than 0), depletion (significantly less than 0) or equilibrium phase (not significantly different from 0).

Proxy Estimate of Legacy P Contributions to River Export. Contemporary mean watershed river P export estimates were derived from average annual WRTDS estimated values from five 5-y time steps between 1992 and 2012. If the ratio of watershed mean river P export to the mean agricultural P balance was greater than 1, we assumed that contemporary river P export in excess of the contemporary agricultural P balances was supported by legacy P sources. Since inputs from 1992 to 2012 were accounted for in the contemporary agricultural P balance, we determined that the source was an anthropogenic P input from an earlier time frame, prior to 1992. If the mean agricultural P balance was less than 0, which was the case for 20 watersheds in either the equilibrium or depletion P status categories, contributions from P balances were set to 0 and legacy P was assumed to be the entire source of river export. If the ratio was positive, but less than 1, we concluded that river P export was largely supported by the agricultural P balance, and the contribution of legacy inputs to river export was inconclusive. Our proxy estimate of the contribution of legacy P to river export was: percent legacy P contribution to river export = ([mean river P export − mean P balance]/mean river TP export) × 100.

The contribution of P from WWTFs to river P export can invalidate this comparison. Therefore, we excluded 30 watersheds where estimated P export from upstream WWTFs accounted for more than 50% of the total river P export (*SI Appendix*, Table S2), leaving us with 143 study watersheds. We acknowledge that river P export may also be supported naturally by P sourced as a product of weathering, the breakdown of glyphosate, or atmospheric deposition (41–43). None of these sources was included in this study as they were unlikely to have a significant impact on the results.

Agricultural P Balance and River Export Trends. The Thiel Sen Slope was used to estimate watershed changes in agricultural P balances between 1992 and 2012 (44), and the change in river P export was estimated in the WRTDS model. To determine if changes in agricultural P management over time had a significant effect on improving river water quality, a simple linear model using agricultural P balance change as the independent variable and river P export change as the dependent variable was developed using the *lm* function in R (45).

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