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Article

Mapping the National Phosphorus Recovery Potential from Centralized Wastewater and Corn Ethanol Infrastructure

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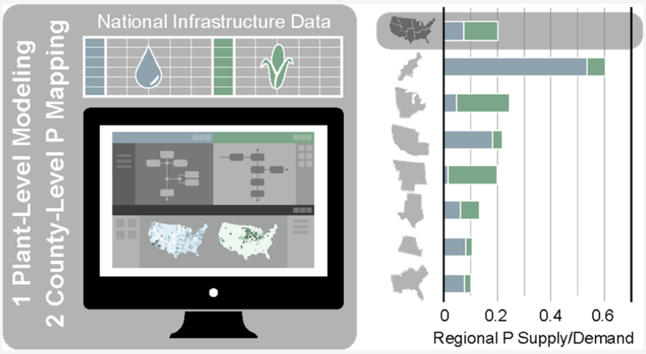
ABSTRACT: Anthropogenic discharge of excess phosphorus (P) to water bodies and increasingly stringent discharge limits have fostered interest in quantifying opportunities for P recovery and reuse. To date, geospatial estimates of P recovery potential in the United States (US) have used human and livestock population data, which do not capture the engineering constraints of P removal from centralized water resource recovery facilities (WRRFs) and corn ethanol biorefineries where P is concentrated in coproduct animal feeds. Here, renewable P (rP) estimates from plant-wide process models were used to create a geospatial inventory of recovery potential for centralized WRRFs and biorefineries, revealing that individual corn ethanol biorefineries can generate on average 3 orders of magnitude more rP than WRRFs per site, and all corn ethanol biorefineries can generate nearly double the total rP of WRRFs across the US. The Midwestern states that make up the Corn Belt have the largest potential for P recovery and reuse from both corn biorefineries and WRRFs with a high degree of co-location with agricultural P consumption, indicating the untapped potential for a circular P economy in this globally significant grain-producing region.



KEYWORDS: *wet milling, dry grind, struvite, calcium phytate, nutrient recovery, plant-wide modeling*

INTRODUCTION

Phosphorus (P) inputs as fertilizers are essential to support crop yields by replenishing P exported by biomass harvest. The United States (US) accounts for approximately 8.5% of global P fertilizer use[1] and is dependent on non-US phosphate rock reserves to meet the majority of its P needs. A major factor in the large consumption of P is inefficient use throughout agricultural application. Globally, apparent P use efficiency (PUE) has been estimated to be 16% during 1961−2013[2] and 9−12%,[3] though this may be elevated due to soil P mining. Across US croplands, apparent PUE varies drastically, from 10 to 100%, though decadal estimates suggest a US-wide average of 60% over 1987−2012 and a Midwest average of 80%.[4] Although approximately 1.71 Gg of P as the fertilizer is applied annually to US croplands, nearly 0.54 Gg of P enters water bodies due to soluble and erosive losses.[5] Recovery and agricultural reuse of P from wastewater and aqueous food processing streams can both reduce the amount of phosphate rock needed while also reducing the amount of P in livestock feed that would eventually be lost to water bodies from animal waste streams.[6−8] Outside of P recovery that is already done (e.g., land application of manure and wastewater sludge),[5] human waste and animal manure present two main untapped pathways for renewable P (rP).

Prior estimates for national P potential from waste have coupled human population and animal production data sets with estimates of P release in excreta.[9−13] Although this provides a reference point for the spatial distribution of potential rP, it does not consider the centralized nature of water resource recovery facilities (WRRFs) and in some instances assumes that up to 95% of excreted P is recovered.[11] With the development of P recovery methods for WRRFs (i.e., the precipitation of P with magnesium to form struvite), process modeling incorporating these recovery methods can generate WRRF-specific rP potentials and better national estimates. Animal waste is often used as another means for estimating rP potential from livestock production. These animal waste techniques often involve separate processing and treatment technologies that are capital and operationally intensive.[9] Additionally, large livestock hubs are not necessarily co-located with agricultural P consumption, requiring rP to

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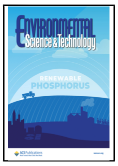

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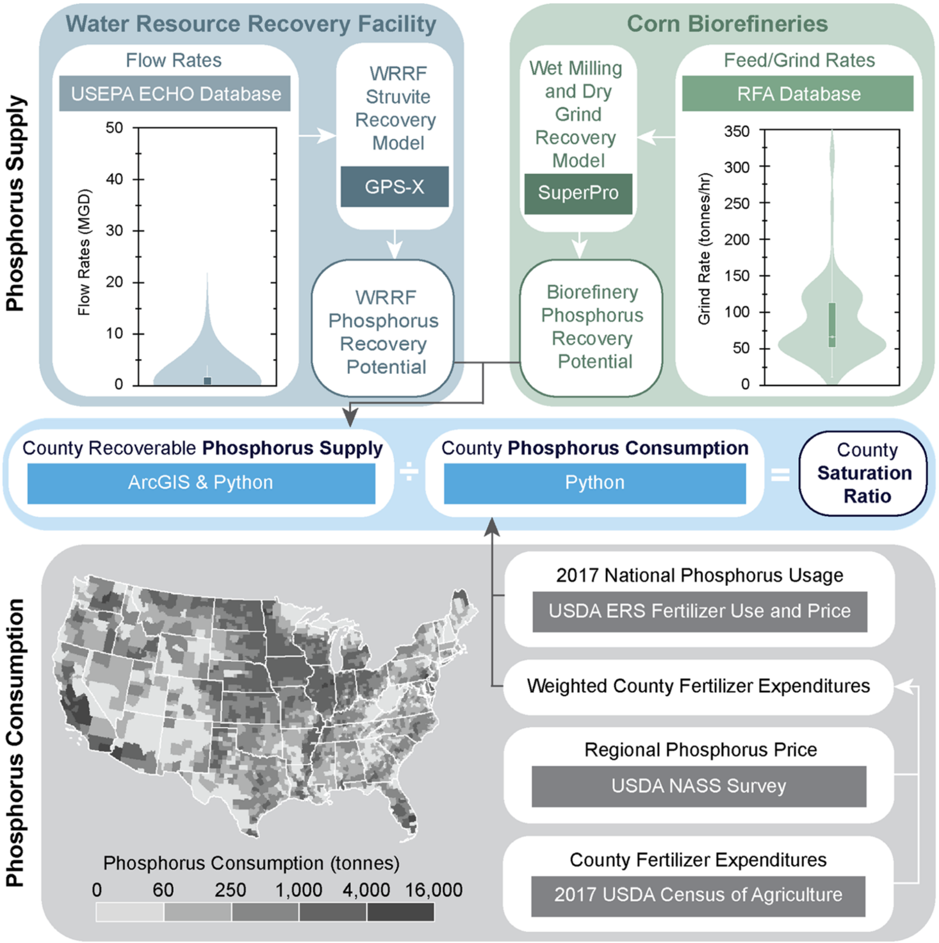

Figure 1. Schematic of the P model. Steps for determining rP potential from WRRFs and corn biorefineries, county-level P consumption, and the overall saturation ratio of P for each county in the US.

Table 1. Data Sources for Determining P Supply and Consumption

source	time scale	data description	spatial scale
P Consumption			
USDA Census of Agriculture	2017	total fertilizer expenses	county
USDA National Agricultural Statistics Service Survey	2009−2014	price of phosphate fertilizer by multi-state region	regional
USDA Fertilizer Use and Price	annually, 1964−2018	total and crop-based fertilizer used by type	national
WRRF P Supply			
EPA Enforcement and Compliance History Online	2017	average wastewater influent flow rates (MGD) for plants across the US	point
Corn Biorefinery P Supply			
Renewable Fuels Association Ethanol Plant Locations	present	ethanol production capacity (MGY) for plants across the US	point

travel longer distances to get to P-intensive regions.[14] Animal waste has also been directly land applied as a means of nutrient recycling but is done to primarily meet nitrogen (N) needs, which can lead to overapplication of P. The environmental impacts associated with transportation are also higher for manure than concentrated rP products due to the excess weight of the organic matrix.[15] An alternative to P recovery from manure is to recover P from highly concentrated biorefinery coproduct streams that are sold as animal feeds such as corn (*Zea mays*) gluten feed and distillers dried grains with solubles (DDGS),[8] especially considering that the magnitude of P in maize and soybean milling coproducts is nearly equivalent to that of P in excess manure.[5] Currently, this animal feed is fed to livestock and excreted as manure which may be overapplied and cause excessive P losses.[5] Recovering and concentrating P upstream at biorefineries allow for application of rP that meets, but does not exceed, crop needs. In contrast, most manures—though highly variable in N/P based on species, diet, and manure storage[16]—generally have a lower N/P than crop needs,[17] meaning that the typical use of manure to meet crop N needs will entail overapplication of P.[18] Although there is work outlining the rP potential from corn biorefineries[6,7] and the technology for recovery does exist, they are often overlooked as a source of rP since they are upstream of P loss through animal waste, and the magnitude of recovery potential at the national scale is yet to be elucidated.

This work had two primary objectives: (i) to quantify US potential rP supply from centralized grain processing and waste handling infrastructure, specifically corn ethanol biorefineries and WRRFs, and map co-location with P consumption and (ii) to assess the impacts of rP transport and blending with synthetic P fertilizer on rP supply co-location with crop P consumption. To determine potential rP supply from both WRRFs and corn biorefineries, publicly available databases of wastewater flow and ethanol production rates were used as inputs to plant-wide process model simulators (Figure 1 and Table 1). Corn biorefineries were limited to ethanol plants due to the availability of capacity data in public databases. To assess co-location, saturation ratios were developed and mapped on a county-level geospatial scale based on the ratio of rP supply to P consumption (Figure 1). Saturation ratios are the primary technique used in this work to quantify the relationship of rP supply to P consumption on a county-level geospatial scale. Co-location of supply and consumption for each rP source was determined by fitting a logistic curve to saturation ratios and cumulative rP supply. The rP supply on a county level was combined for both WRRFs and corn biorefineries, and transfer distances were calculated for individual plants to transfer excess rP to nearby undersaturated counties. Due to low water solubility of rP from WRRFs and bioavailability challenges of rP (i.e., P present in organic bonds that must be hydrolyzed) from corn biorefineries, the impact of blending of rP and highly water-soluble P fertilizers [e.g., monoammonium phosphate (MAP), diammonium phosphate] was also considered to determine the change in rP geographical spread and transfer distance. Specifically, blending with highly water-soluble P can provide greater early-season P availability for crops.[5] Potential rP was then compared across regions under these various blends.

■ METHODS

Data Sources. Data sources were primarily limited to governmental agencies and organizations that collect fertilizer and plant operating data. To calculate P consumption, the USDA Quick Stats program was used to obtain data from the 2017 Census of Agriculture[19] for fertilizer expenditures and the National Agricultural Statistics Service survey (NASS)[20] for fertilizer regional prices. The USDA Census of Agriculture is a census done through a questionnaire every 5 years which provided county-level fertilizer expenditures used to proportion national-level P consumption to a county level. The USDA also generates numerous surveys on a more frequent basis through NASS that polls US farms and ranches and provides regional price data to weight county expenditure data to account for regional variation.[21]

Individual plant-level data were acquired and used for both WRRFs and corn biorefineries to input into process models. Public databases, such as the Renewable Fuels Association (RFA) and US Environmental Protection Agency's (EPA's) Enforcement Compliance and History Online (ECHO), were used to gather influent data for corn biorefineries and WRRFs throughout the US (Table 1). These influent data were input into process models developed in GPS-X (Hydromatis EES, Inc., Ontario, Canada) for WRRFs and SuperPro Designer (Intelligen, Inc., Scotch Plains, NJ) for corn biorefineries to determine rP supply. The EPA ECHO database, which contains compliance data for regulated point sources in the US, was used to generate a listing of average daily wastewater flow rates for each reporting WRRF in the US.[22] The data in ECHO is self-reported by the regulated entity and is as accurate as their reporting. To estimate P recovery from corn biorefineries, production capacity data for each biorefinery were obtained from the RFA database that contains ethanol production capacities.[23]

County-Level P Consumption. To obtain county-level P fertilizer consumption, fertilizer use on a national scale was proportioned to a county scale using county-level fertilizer expenditures adapted from a previous state-level USGS method as shown in eq 1[24]

$$P_c = \frac{\mathrm{FE}_c/\mathrm{RPP}_c}{\sum_{c=1}^{n} \mathrm{FE}_c/\mathrm{RPP}_c} \times P_{\mathrm{nat}} \tag{1}$$

where P_c is total county-level fertilizer consumption in tonnes P per year, FE_c is county-level fertilizer expenditures in 2017 from the USDA 2017 Census of Agriculture, RPP_c is the regional price of the P fertilizer for county *c*, n is the total number of counties, and P_{nat} is the total P consumption in the US in tonnes P per year. Due to regional variation of fertilizer costs, the fraction of expenditures by a county were weighted by the regional phosphate price in the region the county is located in. Regional phosphate prices for 2009−2014 were converted to those of 2017 prior to use (Table S1). P_{nat} for all crops was determined using a previously developed methodology by forming a regression model of national P fertilizer use (1994 to 2015) and national P fertilizer use on corn (1994 to 2018) and interpolating to extend total national P fertilizer use to 2017.[14] A Python program was developed that then converted the national phosphate fertilizer use to P and apportioned it to the county level by multiplying by the proportion of the weighted county fertilizer expenditure and total weighted county fertilizer expenditures. Phosphate (as P_2O_5) was converted to total P by dividing by a conversion factor of 2.29. Other estimates for county-level P consumption have been executed using data from the Association of American Plant Food Control Officials (AAPFCO),[25] but these data are limited to 2016 which is why the more recent 2017 Census of Agriculture data was utilized in this study. A comparison between approaches, which showed over 80% of total P consumption located in similar counties, is explored in detail in the Supporting Information (Method S4).

Corn Biorefinery P Recovery. P recovery potential from corn biorefineries was estimated using process models for dry grind and wet milling biorefineries with feed grind rates as the primary variable parameter. The RFA database of corn biorefineries was used to find geospatial coordinates and ethanol production capacity in million gallons per year (MGY) of 196 corn biorefineries. Ethanol production was converted to the grind rate by dividing by 2.8 gallons per bushel and then multiplying by 56 pounds per shelled bushel (15.5% moisture) and converting to tonnes per hour. The process models for P recovery in wet milling and dry grind processes were developed using SuperPro Designer (Intelligen Inc., Scotch Plains, NJ, USA) and were based on previously developed models.[26,27] Models were run at varying grind rates to develop a linear relationship between the grind rate and rP potential for both wet milling and dry grind processes (Figure S1). A detailed description of the models is available in the existing literature.[6,7] For additional details on wet milling and dry grind process simulations, refer to the Supporting Information (SI) (Methods S1 and S2, respectively).

WRRF P Recovery. P recovery potential from individual WRRFs was estimated using process models with the average flowrate as the primary variable parameter. Plant flow rates and geospatial coordinates for 13,882 WRRFs were obtained through the USEPA ECHO database in million gallons per day (MGD).[22] Based on the WRRF flow data in ECHO, it was determined that process models across a range of flow rates—0 to 5 MGD, 5 to 20 MGD, 20 to 50 MGD, and 50+ MGD—were necessary to estimate rP potential as struvite (Figure S2) where process models optimized at the high end of each flow range—5, 20, and 50 MGD—can be used to estimate rP potential within the bin.

Individual wastewater treatment layouts were designed using GPS-X (Hydromantis, Inc.) and included modified enhanced biological phosphorus removal (EBPR) as a mainline treatment process and phosphorus recovery through struvite precipitation from sludge handling liquids (Figure S3). Influent flow composition, unit process sizing and dimensions, and operating conditions were matched with reported values in the literature[28] (Table S2). For additional details on the WRRF process simulation, refer to the Supporting Information (Method S3).

To account for the impact of influent flow variations on the treatment process efficacy, the design capacity of the layouts was fixed at 5, 20, and 50 MGD. However, the 5 MGD layout did not properly capture P recovery under lower flow conditions, so layouts were also fixed at 1, 2, 3, and 4 MGD. Each layout was simulated under steady-state conditions and low- (3.7 mg-P/L), medium- (5.6 mg-P/L), and high- (11.0 mg-P/L) strength influent P characteristics to measure the extent of biological phosphorus removal and recovery.[28] The average performance of each layout under low-, medium-, and high-strength influent wastewater stream conditions was considered as the overall treatment/recovery potential in each flowrate (Table S3). The low influent P concentration (3.7 mg/L) most closely aligned with reported effluent P concentrations in the USEPA ECHO database, and this concentration was used for further analysis (Figure S4). P recovery was estimated as a function of average plant flowrate based on the linear relationship between flow- and side-stream struvite precipitation in the process model outputs (Figure S5). P recovery from biosolids and biosolid application was not considered in this study as the resulting Class B biosolids would not be applicable for food production.

The plant-level P recovery approach was compared with the population estimate approach for recovery potential. Since the population estimate approach has not been done for 2017, it was recreated for the sake of comparison. Population data for individual counties came from the US Census Bureau.[29] Based on previous estimates, 1.61 g of P per capita per day is excreted in both urine and feces, and this amount was adjusted based on a phosphorus recovery efficiency of 95% (1.53 g P/capita/day) to maintain consistency with other studies.[11] Since the focus of this study is concentrated on rP fertilizer products, the population-based P recovery estimate was approximated based on only the P excreted in urine which could be precipitated as struvite (0.93 g P/capita/day).[30,31] County-level P recovery estimates are determined by multiplying the county population by the yearly P recovery potential per capita (0.76 lb P/capita/year). A map of estimated rP from urine is included in the Supporting Information (Figure S6).

Mapping Co-location of rP Supply with Consumption. To aggregate total rP supply by the county, WRRFs and corn biorefineries were mapped in ArcGIS (Esri, Redlands, CA) using their geospatial coordinates and individual recovery rates for each plant in tonnes per year. County polygons were added, and recovery rates for WRRF and corn biorefinery points located inside of a county polygon were separately aggregated.[32] Counties' saturation ratios were determined based on the ratio of supply to consumption separately for WRRFs and corn biorefineries. There were WRRFs located in counties with no consumption, termed nutrient islands, where a saturation value of >100,000 was assigned since it is over an order of magnitude greater than the highest calculated saturation ratio of 2300. The saturation ratios were mapped using ArcGIS, and counties were characterized as undersaturated if the ratio was less than 1 and oversaturated if greater than 1.

To better understand how rP supply aligned with country-level P fertilizer consumption for both corn biorefineries and WRRFs, the co-location was characterized by performing a log transformation on the saturation ratio and creating a cumulative histogram to show the total rP fraction across saturation ratios. The cumulative histogram was fitted to a generalized logistic function (i.e., Richard's curve) as shown in eq 2[33]

$$Y(r_x) = \frac{1 - X}{\left(1 + e^{-B(r_x - M)}\right)^{1/\mu}} \qquad (2)$$

where Y is the fraction of total P supply, r_x is the independent variable that is the log-transformed saturation ratio, X is the fraction of nutrients in consumption-limited counties (or nutrient islands) where there is supply but no or minimal consumption, B is the growth rate which demonstrates the uniformity of saturation ratios for each source across the US, μ is the shape parameter which affects near where asymptote maximum growth occurs, and M is the positioning parameter which positions the curve on the x-axis relative to μ.[33] The resulting logistic functions were then plotted side-by-side for WRRFs and corn biorefineries. For rP supply to be better co-located with P consumption, the resulting curve would be characterized by (1) a higher fraction of rP in undersaturated counties, (2) a lower X value or lower fraction of rP in nutrient islands, and (3) a higher μ or higher curve growth rate.

rP Distance Analysis. A transport model was developed in Python which distributed excess rP from WRRFs and corn biorefineries in oversaturated counties to nearby undersaturated counties. ArcGIS was used to determine centroids of each county to use as its geospatial coordinates. A Python program was developed that created a list of all plants with excess rP and developed a matrix with the distance in km between the plant and each county's centroid coordinates using the great circle distance. It then individually assigned excess rP to the closest county to each plant up to a saturation ratio of 1; any additional P in excess of saturation was then assigned to the next nearest county; this was repeated until all excess P was accounted for (Figure S7). All transfers made were recorded by using the plant ID, source county, destination county, amount transported (tonne), and travel distance (km). New saturation ratios were generated for each county after all transfers were made and the distance traveled per mass of all transfers (km/tonne) calculated. The travel distances were compiled and plotted for major regions in the

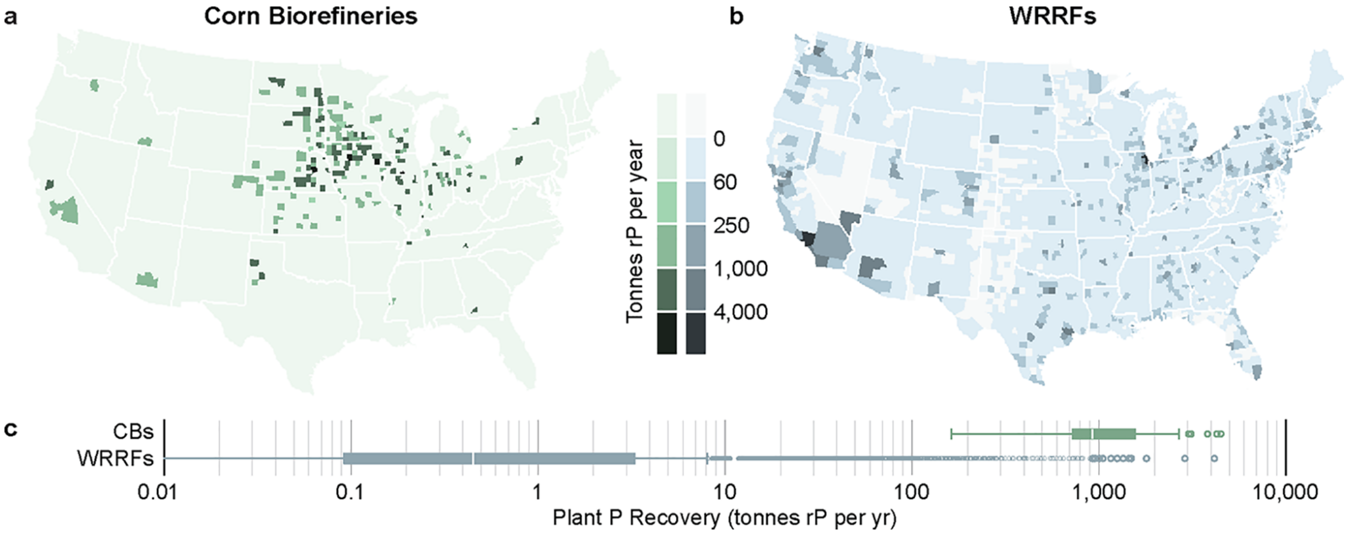

Figure 2. County-level rP supply from WRRFs and corn biorefineries. rP supply of each county by the source based on aggregation of individual plants with a range of plant rP potentials. (a) rP supply based on 14 wet milling and 182 dry grind corn biorefineries. (b) rP supply based on 13,882 national WRRFs. (c) Range of P recovery potentials for WRRFs and corn biorefineries on a plant level. The WRRF plant-level recovery range goes below 0.01 tonnes/yr. Although more widespread, WRRFs have a much lower median rP supply potential on a plant level than corn biorefineries.

Table 2. Ranking of Top 10 States by P Consumption and rP Supply Potential (1000 tonnes P per Year)

rank	state	total P consumption	state	WRRF rP supply	state	biorefinery rP supply
1	Iowa	158.3	California	17.5	Iowa	58.0
2	Illinois	156.0	Illinois	12.1	Nebraska	31.1
3	Minnesota	117.9	Texas	11.7	Illinois	24.6
4	Nebraska	106.9	New York	11.0	Minnesota	20.2
5	California	105.1	Pennsylvania	8.0	Indiana	16.7
6	Texas	90.1	Ohio	6.6	South Dakota	16.6
7	Indiana	86.8	Michigan	5.6	Ohio	9.0
8	Kansas	83.7	Indiana	4.4	Wisconsin	8.3
9	North Dakota	80.3	Washington	3.6	North Dakota	6.7
10	Missouri	72.9	Massachusetts	3.3	Kansas	6.5

US—the Northeast, Southeast, Midwest, Northern Plains, Southern Plains, Pacific Northwest, and the Southwest. The final rP in each county was also aggregated after transfers for both WRRFs and corn biorefineries by the major US regions. Total rP was reported in each region, and the amount of rP in a region that was used directly in the county it was supplied to was reported—transfers were excluded to show the direct co-located rP amount.

Due to low solubility of rP as struvite and low bioavailability of organic rP as Ca-Phytate since P is trapped in organic bonds that must be hydrolyzed, blending of rP with existing P fertilizers can assist in providing readily available P particularly in early growing seasons.[5] Varying blend ratios were considered in our analysis including 25 and 50% rP in addition to our original 100% rP scenario. These blend ratios were chosen because when blended with MAP, >50% struvite blends constrained vegetative growth of maize, and >25% struvite blends constrained vegetative growth of soybean.[34] To develop new saturation ratios, county-level consumption was proportioned by the fraction of rP (i.e., for a 25% blend, county-level consumption was reduced to 25% of the total). A transfer distance analysis was also performed from oversaturated to undersaturated counties for each blend ratio which was considered when determining saturation ratios. Similar to the no blending scenario, rP was aggregated at a 25% blend for each major US region, which was used to meet consumption in the county it was supplied to.

RESULTS AND DISCUSSION

It is estimated that rP from corn biorefineries and WRRFs can potentially displace 20% of national consumption for P fertilizer. Despite being far fewer in number (196 vs 13,882), corn biorefineries have close to twice the rP potential of WRRFs and a much larger average P recovery rate than WRRFs (Figure 2c). Corn biorefineries are also heavily concentrated in the Midwest (Figure S8), where P consumption is the greatest due to high yields and thus high harvest removal rates.[5] A large fraction of wastewater rP supply is centralized in the large, often urban, WRRFs throughout the US, but overall rP supply is more widespread than that for biorefineries (Figure 2). In the subsections below, estimates for county-level P consumption and rP supply from corn biorefineries and WRRFs are discussed further.

County-Level P Demand. Of the estimated 1.8 million tonnes of P consumption in the US in 2017, over half is largely concentrated in the North Central or Midwestern region known as the Corn Belt[35] (i.e., composed of Ohio, Indiana, Illinois, Iowa, Missouri, Kansas, and Nebraska and parts of Michigan, Wisconsin, Minnesota, South Dakota, and Kentucky) and the irrigated corridor of the Mississippi River Valley (i.e., in addition to some Corn Belt states that are parts of

Arkansas, Mississippi, Louisiana, and Tennessee) (Figure 1). This is consistent with most other national-scale nutrient consumption analyses due to intensified agriculture in these regions[10,14] engendering high crop production and thus P export with harvest that is replenished by P fertilization. The top 10 state-level P consumptions were dominated by Corn Belt states due to the large-scale production of corn and soybean (*Glycine max*) but also included California—a major producer of the nuts, fruit, and vegetables[36]—and Texas—a major producer of cotton and hay[37] (Table 2). Due to the lack of state-level fertilizer consumption data, the price variation in fertilizer across the US was accounted for by using multi-state regional price averages (Table S1). This enabled weighting of county-level expenditures for these price variations to proportion national fertilizer use more accurately. Although this allowed for estimates of more recent P consumption data, it likely introduced uncertainty in actual state fertilizer use. Consistent nitrogen−phosphorus−potassium (NPK) ratios of fertilizer application were assumed across the US since county-level expenditures were recorded for total fertilizer and not split based on the fertilizer type.

Corn Biorefineries' rP Supply. Corn biorefineries can provide nearly twice as much rP nationally as WRRFs and provide greater co-location of rP with P consumption than WRRFs. The 14 wet milling and 182 dry grind biorefineries can provide an estimated 229,000 tonnes of rP per year, accounting for over 12.5% of national P consumption in 2017. However, this percentage is even higher at the state scale, particularly for Corn Belt states in which corn biorefineries can meet from 19% (Indiana) to 37% (Iowa) of P consumption (Figure 2a). The rP estimate from corn biorefineries in Iowa alone was greater than the cumulative potential from WRRFs in California, Illinois, Texas, and New York (Tables 2 and S4). Potential rP production from individual plants can range from 162 to 4501 tonnes of P per year (Figure 2c and Table S5) with grind rates ranging from 12 to 325 tonnes of corn per hour (Figure S2). The median rP production of corn biorefineries is 925 tonnes of P per year, far greater than that for WRRFs for which only 16 out of 13,882 plants surpassed this median (Figure 2c). Wet milling plants tended to have higher grind rates than dry grind plants and therefore could on average generate approximately 40% more rP. There are a number of additional non-ethanol corn wet milling operations that were not included in this study due to a lack of publicly available data, which would even further contribute to rP potential.

WRRF rP Supply. WRRFs offer a sizeable amount of rP over a broad geospatial scale but with a high degree of dislocation from P consumption. The 13,882 WRRFs analyzed in this study account for an estimated 136,600 tonnes of rP as struvite per year which can meet over 5% of total US P consumption. With plant influent flow rates ranging from 1.1×10^{-9} MGD to 1876 MGD (Figure S2), the rP potential from individual plants ranges from near zero to 4366 tonnes of struvite per year (Figure 2c and Table S6). Nearly 96.4% of total rP potential for WRRFs came from major (>1 MGD) WRRFs which only account for 30% of plants (Figure S13). High levels of rP supply are more concentrated around urban centers where WRRFs service large populations (Figure 2b). Due to the economic difficulty of recovery at minor facilities (<1 MGD), this is promising since a vast majority of recovery potential is at major plants where rP recovery as struvite can provide operational benefits to mainstream Bardenpho EBPR processes by reducing intra-plant cycling of P.[38,39] Additionally, one of the smallest-sized WRRFs with an existing rP recovery process is approximately 13 MGD, and WRRFs larger than 13 MGD account for over 75% of total rP from WRRFs.

The modeling approach using recovery of P as struvite in individual WRRFs was compared to the standard population approach for estimating P recovery from human urine, and it was found that our approach yielded approximately 22% more potential rP across the US. The rP estimate from urine based on the population was approximately 112,000 tonnes per year. There was consistency in that those states with the highest populations also tended to have the highest potential rP from WRRFs (Table 2). Urban centers also present a common concentrated source of potential rP (Figure S6), but the plant-level approach does show more centralized rP potential than the population approach, particularly for larger counties (Figure S9). The larger estimate of rP potential from the plant-level approach is partly the result of wastewater flows containing gray water and industrial wastewaters in their average daily flows. Another variation between the approaches is in how P is distributed across WRRF outflows and how efficiently it is captured. The population approach assumes that 61% of total P is captured in urine with a 95% recovery rate (approximately 58% recovery of total P), and the WRRFs modeled in this study are estimated to recover on average 33−48% of total influent P as struvite from nutrient-rich side streams at WRRFs (Table S3). One contributor to this difference in recovery of total influent P is the more dilute nature of wastewater entering a WRRF than direct human waste, which requires uptake during aerobic mainstream treatment and release during anaerobic solid handling to concentrate the P prior to recovery in a crystallization reactor.

P recovery from large WRRFs in the Midwest has a direct influence on the reduction of P discharge, while P recovery from the large corn biorefineries could reduce embedded P in livestock feed. In states like Iowa, Wisconsin, Minnesota, and Missouri with high livestock populations, the reduction of P at biorefineries could also decrease the risk of manure P losses to surface water due to the lower P content in biorefinery coproducts used in animal feeds.[6−8] The mismatch of manure N/P with crop N/P needs means that application of manure to meet crop N needs entails excessive application of P[40] and thus aggravated P loss risk to surface waters.[41] Decreasing the manure P content by reducing the amount of P fed to animals thereby stands to mitigate manure-based contributions to non-point source P losses, which in some watersheds of the US Corn Belt are thought to be the major driver.[5]

Comparing the rP potential presented from corn biorefineries in this work to a recent geospatial analysis of P recovery from Concentrated Animal Feeding Operations (CAFOs) in the Great Lakes region clearly illustrates synergistic opportunities to disrupt overfeeding of P.[42] For example, in Minnesota, where there are reportedly 1487 CAFOs and roughly 7000 tonnes rP available in excess manure, an estimated 20,000 more tonnes of rP could be extracted from corn biorefineries before it is transmitted to animal feeding operations as corn gluten feed and DDGS.

Another indirect benefit of rP generation for contributing to P loss reductions from non-point sources of agriculture may be realized for struvite-based P removal. The low water solubility of struvite means that when re-used in agricultural fields as a full or even partial substitute for highly water-soluble P fertilizers, soluble P loss risk is decreased.[5,34] In the form of lowly soluble forms such as struvite, rP generation and re-use thereby stand to indirectly improve P loss reduction goals held by the 12 states in the Mississippi River basin and involved in the Gulf Hypoxia Task Force:[43] these states also span much of the Corn Belt counties with high potential for rP production and re-use identified here. Thus, rP should be considered in states' P reduction strategies in the region toward a comprehensive, regional P management strategy.

Co-location of rP Supply with Consumption. Corn biorefineries had a narrow logistic saturation curve, indicating higher co-location with P consumption (Figures 3c and S10). Nearly 50% of potential rP was in both under- and oversaturated counties. However, the degree of over- or undersaturation was less than in WRRFs. This co-location is likely intentional since a majority of the corn production in the US is within 50 miles of a biorefinery,[44] creating a dense area of rP supply in consumption-intensive regions throughout the Midwest and Corn Belt (Figure 3a).

In comparison to corn biorefineries, WRRF rP supply appears to be less co-located with fertilizer consumption (Figure 3c). This is also evident by the small growth rate of the WRRF logistic curve relative to biorefineries (Table S7). Nearly 40% of rP is in undersaturated counties with 60% in oversaturated counties. Over 90% of minor plants and 73% of major plants are in undersaturated counties, but most plants over 15 MGD, which make up over 72% of total rP potential from WRRFs, are located in oversaturated counties. The large portion of rP in oversaturated counties means that the supply is dislocated with the agricultural consumption centers. It is also clear that highly oversaturated counties typically contain urban centers with large WRRFs but minimal agriculture (Figure 3b). This creates the need to transport large quantities of excess P from urban centers to nearby undersaturated counties for use, which would increase the costs and carbon footprint associated with rP re-use.

Overall P recovery potential and transfer distance analysis. While rP from corn biorefineries and WRRFs can potentially meet 20% of national P consumption, rP supply and P consumption varied by the geographic region (Figure 4 and Table S8). The Midwest alone can generate nearly 146,000 tonnes of rP per year from corn biorefineries and 36,900 tonnes rP per year from WRRFs which account for 64 and 27% of national estimates for each source, respectively. In aggregate, rP from WRRFs and corn biorefineries could meet 24% of P fertilizer consumption in the Midwest. The Midwest is also shown to provide a unique region for synergy between WRRFs and corn biorefineries to reduce P loading to surface waters via point source discharges while providing a regional supplement or full substitute for P fertilizers.

When incorporating the transport of rP from oversaturated to undersaturated counties, larger plants tend to provide the most noticeable increases in saturation around neighboring counties (Figure 4a). The oversaturation in urban counties due to large WRRFs is evidenced by the heightened saturation of rural counties surrounding major population centers (e.g., Chicago in Cook County, IL; Detroit in Wayne County, MI; Atlanta in Fulton County, GA, etc.). There is some noticeable increase in saturation around corn biorefineries in the Midwestern states, but the consumption-intensive counties surrounding most biorefineries could absorb rP before it can be disbursed as far as with WRRFs (Figure S11). A comprehensive list of counties with P consumption, rP supply, and saturation ratios is included in the Supporting Information (Table S9).

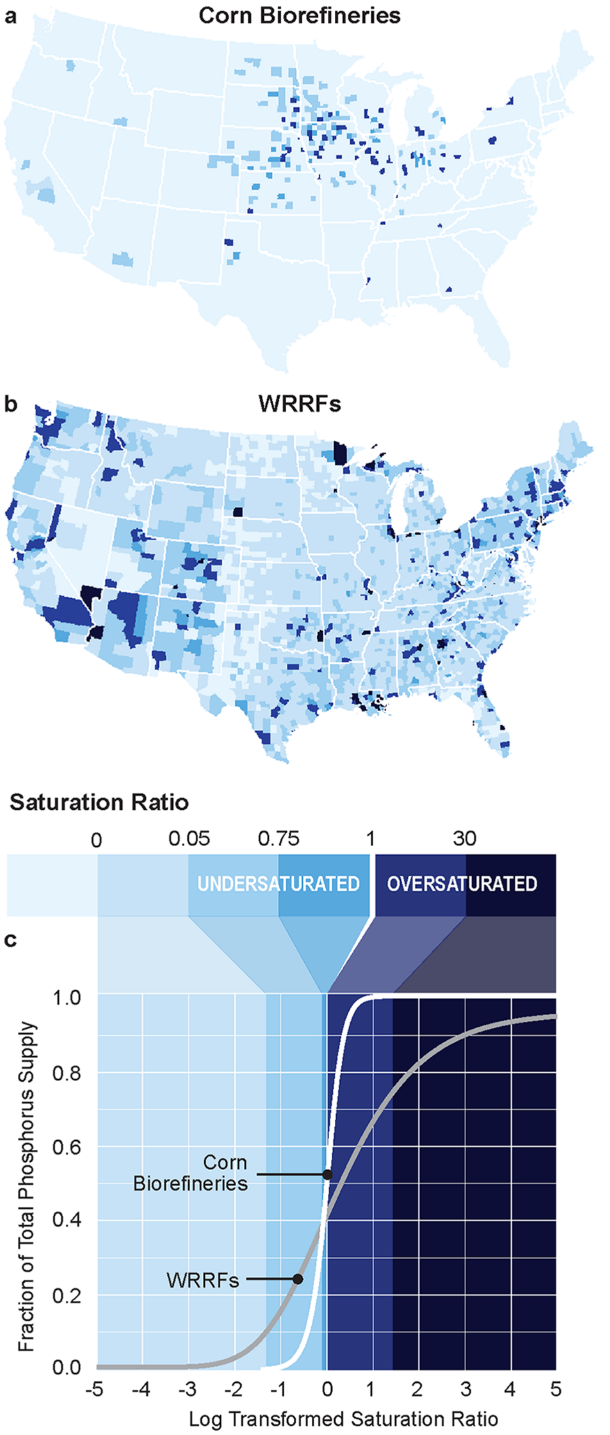

Figure 3. County rP saturation ratios for WRRFs and corn biorefineries. County-level saturation ratios of rP supply from WRRFs and corn biorefineries to county-level P consumption in fertilizer. (a) rP saturation ratios on a county level for national WRRFs. (b) rP saturation ratios on a county level for wet milling and dry grind corn biorefineries. (c) Logistic saturation ratio curves for WRRFs and corn biorefineries. WRRF potential rP supply is densely concentrated in urban centers where P consumption is smaller and is not as well localized, whereas corn biorefineries are mostly concentrated in the central United States and well localized with consumption as seen by the narrower logistic saturation curve.

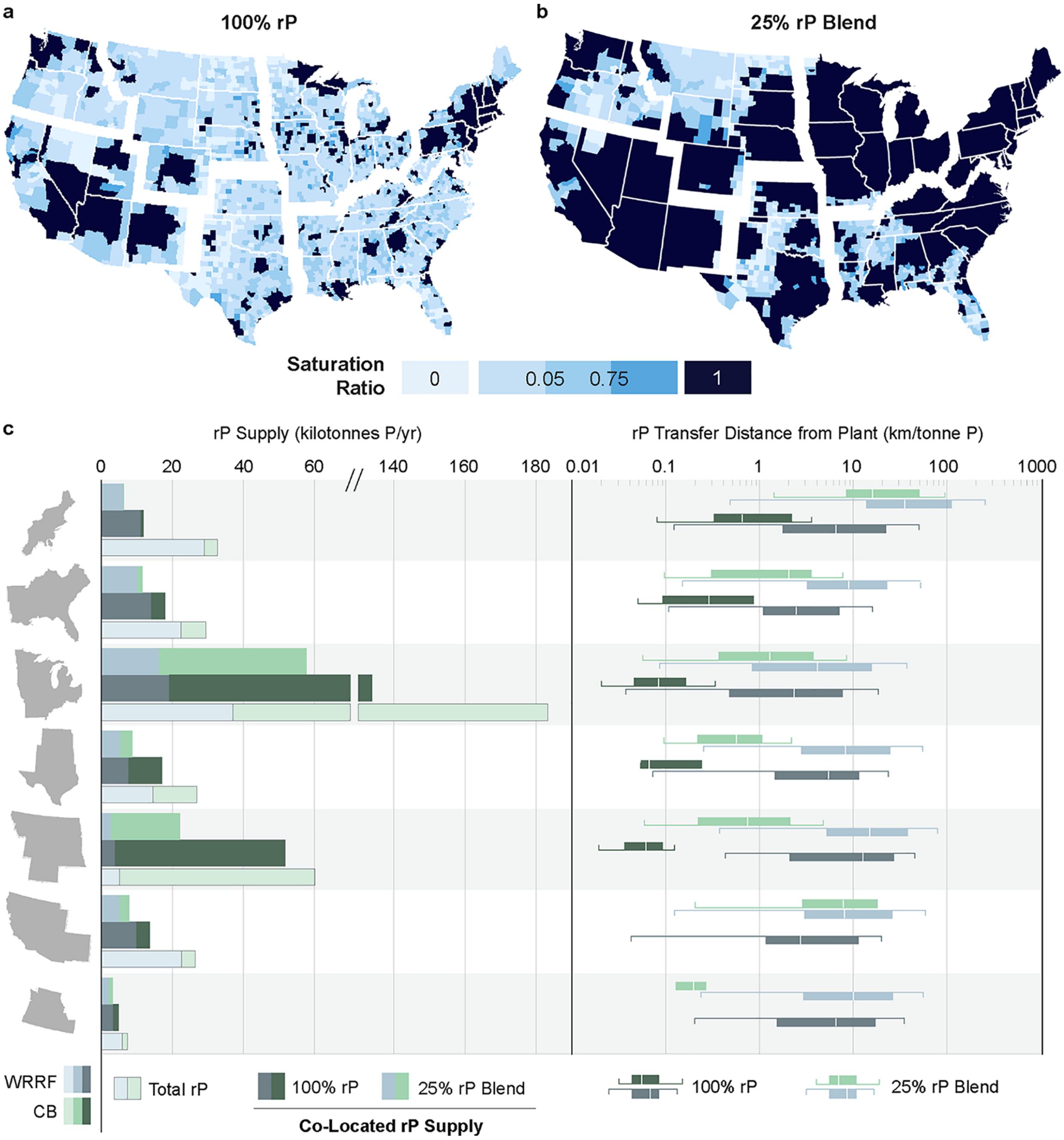

Figure 4. Saturation ratio of total rP supply including transfers of P from oversaturated to undersaturated counties. County-level saturation ratios are shown for (a) non-blended or strictly rP fertilizer and (b) 25% blended or one part of rP and three parts of synthetic P fertilizer. (c) Bars on the left represent total rP potential and co-located rP potential based on the region (the Northeast, Southeast, Midwest, Southern Plains, Northern Plains, Southwest, and Pacific Northwest) split by WRRFs and corn biorefineries. The lightest bars represent the total rP supply in the region, and the two darker sets of bars represent only co-located rP supply for 100% rP and a 25% rP blend. The box and whisker plots on the right show the range of distances that a tonne of P from each plant in that region would need to travel to an undersaturated county. The Midwest has by far the largest potential rP due to corn biorefineries, and generally, corn biorefineries have the least distance that P would need to travel to nearby undersaturated counties due to the region also having the largest P consumption.

The range of transfer distances per tonne of excess rP is on average shorter for corn biorefineries relative to WRRFs (Figure 4c). The transfer distance for WRRFs is approximately 100 times further per tonne of rP on average than that of corn biorefineries in more P consumption-intensive regions like the Midwest, Northern Plains, and Southern Plains while still nearly 10 times further in less P consumption-intensive regions such as the Northeast and Southeast. The median travel distance in the US for WRRFs is 3.8 km (km) per tonne rP and 0.09 km per tonne rP for corn biorefineries. These longer travel distances for a large portion of WRRFs provide further evidence of the general dislocation between human rP supply and fertilizer consumption. A listing of transfers is included in the Supporting Information (Table S10).

Recovered P Blend Ratio Analysis. Limited bioavailability of rP as water-soluble orthophosphate could hinder P availability to crops and may require blending with highly water-soluble P fertilizers to avoid yield losses.[5] For rP in the form of phosphate minerals such as struvite produced at WRRFs, lower bioavailability reflects low water solubility, and for the organic matter-based rP such as Ca-phytate produced from corn biorefineries, lower bioavailability is due to P being present in organic bonds that must first undergo hydrolysis.[5] On the other hand, the slow release of bioavailable orthophosphate from rP forms via dissolution (e.g., struvite) or mineralization (e.g., phytate) may present agronomic benefits by avoiding fixation of P by soil colloids[45] and synchronizing with P consumption by crops later in the season,[46,47] particularly in the reproductive growth stages for maize and soybean.[48,49] Greater synchronization can increase P use efficiency and thereby decrease net P inputs to maintain yields.[50]

In addition to avoiding agronomic penalties, blending of rP with highly soluble P fertilizer greatly expands the spatial coverage of rP contribution to crop P needs. At a rP blend ratio of 25%, the Northeast can meet all its P consumption from large WRRFs in urban centers. In the Midwest, where corn biorefineries are primarily located along with high-capacity urban WRRFs, over 95% of P consumption can be met with a 25% blend with rP (Figure 4b). Blending rP with synthetic fertilizer would also result in the need to transport rP a much greater distance to reach undersaturated counties and impact the degree of co-location of P supply to consumption in all regions, particularly for corn biorefineries (Figures 4c and S11). All transfers with a 25% blend rP are included in the Supporting Information (Table S11). All regions except the Pacific Northwest and Southwest saw a decrease of over 60% in co-location of rP supply from corn biorefineries. The rP supply from WRRFs saw at most a 45% decrease in co-location with only the Northeast and Southwest above a 40% decrease; this is likely due to the nature of WRRFs as more dispersed and a larger quantity of smaller facilities.

OUTLOOK

This study provides the first geospatial inventory of rP from centralized infrastructure in the US, revealing the untapped potential and regional synergies of recovery and reuse from corn biorefineries, particularly in the Midwest and Northern Plains. The high rP potential from corn biorefineries also can translate to promising levels of rP potential from other types of bio-facilities with high P coproducts such as soy and pea protein isolate processes. Although rP potential at WRRFs is lower than that at corn biorefineries, incentives for P recovery are growing as numeric water quality criteria for P discharge (promulgated on a federal level, as provisions in the Clean Water Act) are incorporated and implemented on the state level.[51] Although the small size of most WRRFs will restrict the use of P removal strategies that include recovery, this work generates an inventory of rP potential that can be used to further assist regions that are making the transition to numeric P criteria and identify candidate WRRFs for P recovery. To promote P recovery from corn biorefineries, incentives must be created to overcome the economic cost of Ca-phytate precipitation. Quantifying the benefits of the reduced P content in animal feed and elucidating connections between corn biorefineries and animal feeding operations could foster P extraction through nutrient credit trading or higher sale prices for low-P feed.

This study presents a novel methodology for estimating rP from centralized infrastructure using plantwide process models; however, limitations introduced some degree of uncertainty in the results. A primary limitation was the use of non-calibrated plantwide models to generalize rP potential from both WRRFs and corn biorefineries. Individual plants have unique characteristics that may hinder or enhance rP potential that is not fully captured in these models. The influent WRRF P concentration was generalized to discrete standard values, while the influent flow rate was based on self-reported values. Another limitation was the lack of county-level P consumption data; instead, P consumption had to be estimated based on extrapolated national P use and limited county-level fertilizer data. Also, transport distances were estimated using the direct distance from plants to county centroids without considering actual transfer infrastructure, likely leading to underestimates of actual travel distance. These estimates assumed that excess rP would be transferred to the nearest county, while actual optimization of the supply chain would prove more complex with multiple stakeholders and centralized rP processing including a series of potential solutions.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c07881.

Detailed information used to model wet milling and dry grind corn biorefineries and WRRFs; P consumption generated; models including layout, parameters, and inputs; full plant and county information including rP supply and saturation ratios; and transfers and blending results (PDF)

WRRF process unit parameters (XLSX)

Corn ethanol biorefinery plant information (XLSX)

WRRF plant information (XLSX)

Complete table of county-level data (XLSX)

No blending rP transfers (XLSX)

25% Blend rP transfers (XLSX)

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Notes

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