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Article

Modeling National Embedded Phosphorus Flows of Corn Ethanol Distillers' Grains to Elucidate Nutrient Reduction Opportunities

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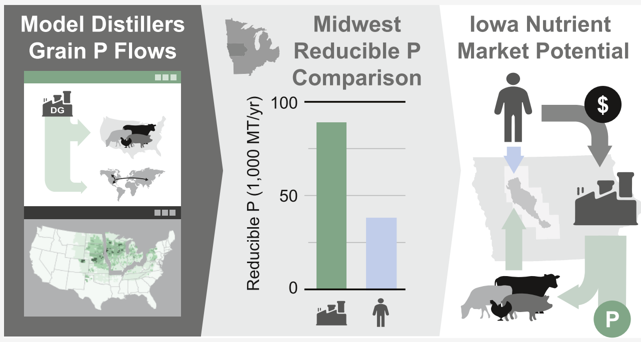
ABSTRACT: Freshwater quality and ecosystem impairment associated with excess phosphorus (P) loadings have led to federally mandated P reduction for certain organic waste streams. Phosphorus reduction from livestock and poultry feeds such as corn ethanol distillers' grains (DGs) presents a centralized strategy for reducing P loss from animal manurein agriculturally intensive states, but little is known about the actual distribution and geospatial P contributions of DGs as animal feed. Here, a county-level flow network for corn ethanol DGs was simulated in the United States to elucidate opportunities for P reduction and the potential for nutrient trading between centralized sources. Overall, the estimated P in DGs that was transferred to US animal feeding operations was nearly twice that present in all human waste prior to treatment. Simulation results suggest that Midwestern states account for an estimated 63% of domestic DG usage, with 72% utilized within the state of production. County-level data were also used to highlight the potential of using nutrient trading markets to incentivize P recovery from DGs at biorefineries within an agriculturally intensive watershed region in Iowa. In summary, corn ethanol biorefineries represent a key leverage point for sustainable P management at the national and local scales.



KEYWORDS: *animal feeding operations, resource recovery and reuse, agricultural watersheds*

■ INTRODUCTION

Phosphorus (P) pollution is a critical threat to aquatic ecosystems and freshwater resources globally, including numerous areas of the United States (US).[1−3] As of 2014, nearly 58% of assessed river and stream miles in the US were rated as poor for P pollution which was an 11% increase from a similar survey conducted in 2009.[4] In many freshwater systems, P is the limiting nutrient and a primary contributor to algal blooms,[5] which are prevalent in the agriculturally intensive US Midwest[6] region (i.e., Illinois, Indiana, Iowa, Michigan, Minnesota, Missouri, Ohio, and Wisconsin). Although P is an essential nutrient for both animals and humans, it is often consumed in amounts much greater than P requirements due to high feed or food consumption to meet energy rather than P demands, leading to excretion of excess P in human waste and animal manure.[7,8]

The contribution to nutrient pollution from the continued loss of P through waste streams coupled with an increasing demand for nonrenewable P fertilizers for food, feed, and biofuel crop production has a direct impact on the resiliency of the food−energy−water nexus.[9,10] The concerns associated with P pollution and fertilizer usage have led to an increased interest in, and implementation of, P recovery technologies from waste streams in an attempt to both reduce P pollution and generate a more renewable P source through P recycling.[11,12] However, despite P security and resiliency being a concern in the US,[13] regulations related to P use and loss to the environment are minimal. Current regulations to reduce P in the environment have targeted point sources, which are primarily made up of water resource recovery facilities (WRRFs),[14] while agricultural nonpoint sources are allowed to implement only voluntary P reduction methods[15] despite being primary contributors to P pollution in many parts of the US.[16−18] While animal feeding operations can be subjected to permitting requirements through the National Pollution Discharge Elimination System, variations in state interpretation of federal rules have reduced the ability to manage this key source of excess P.[19] This inconsistency has led to conflict and litigation between large urban WRRFs that are required to implement costly P removal technologies and rural agriculture, particularly in Midwestern states with dense livestock and poultry operations.[20] This conflict has also triggered proposals for new legislation meant to limit large

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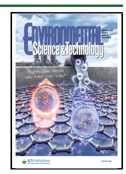

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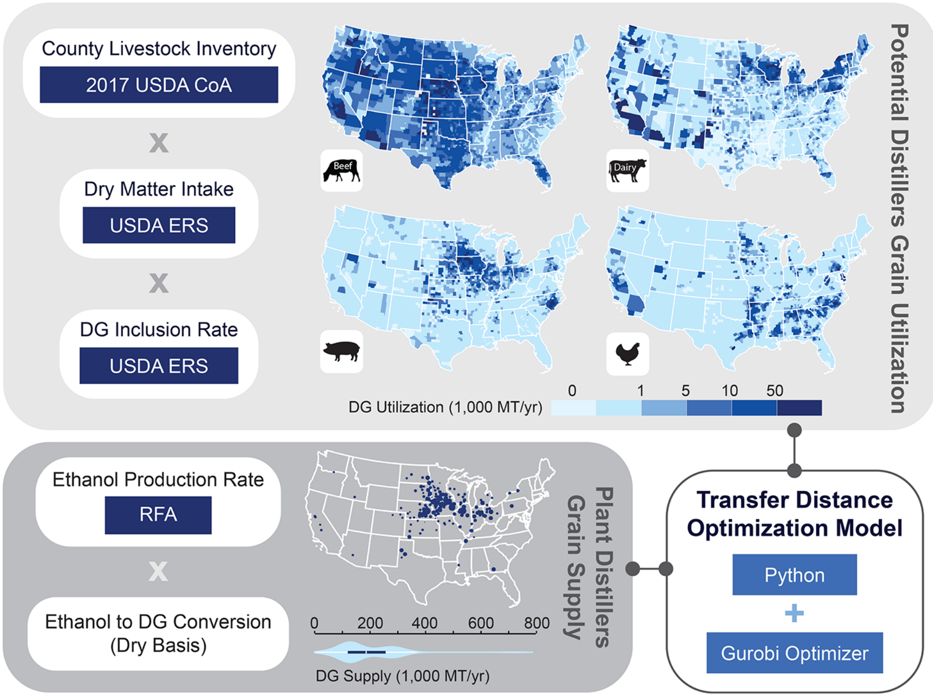

Figure 1. Schematic of DG distribution model. Steps for determining potential DG utilization, plant DG supply, and flow optimization in the US.

animal operations in these livestock and poultry-dense states in an attempt to reduce P loadings.[21]

As one potential mechanism for reducing P at animal operations, nutrient trading markets allow regulated point sources to trade P reductions with other point sources and, in certain nutrient markets, nonpoint sources such as livestock and poultry operations to minimize the overall cost of P reduction within a watershed.[22] Despite the appeal of reducing the overall cost of P management within a watershed, little activity has occurred between point and nonpoint sources in existing nutrient trading markets. Lack of participation is associated with the difficulty in securing commitments to nutrient management obligations from unregulated nonpoint sources and the uncertainty in actual reductions from agricultural best management practices.[23] While nutrient trading remains infrequent, the untapped potential for P reduction from manure generated from animal operations has led to a large body of research focused on both technologies for direct P extraction from manure[24] and comparative estimates of P in manure and human waste throughout the US.[9,25,26]

Several feed formulation and feeding strategies are being used to reduce P excretion in animal manure but additional efforts are needed.[27−30] A significant portion of P in grains and oilseed meals is in the form of phytate which is indigestible for monogastric animals (i.e., swine and poultry[31]) but is more digestible for ruminants (i.e., beef and dairy cattle).[32] As a result, the overall P utilization efficiency from animal consumption to edible meat, milk, and eggs is less than 60%.[33] Distillers' grains (DGs) are the co-product produced by corn ethanol biorefineries (CBs), which have become a major alternative feed source to partially replace corn, soybean meal, and supplemental inorganic P in animal diets in the US.[34] DGs contain greater concentrations of P (0.9% P)[35] than traditional feeds like corn grain (0.3% P) and soybean meal (0.7% P).[36] The practice of adding phytase enzymes during the fermentation process in some dry grind ethanol production facilities to degrade phytate in corn and improve ethanol yield further enhances the digestibility of P in DGs for swine and poultry.[37,38]

Due to the large annual production of DGs from the US CBs [~37 million metric tons (Mt) per year],[39] there is great potential for this co-product to serve as a centralized source of P recovery and reduction in the Midwest region.[35,40] Recent research has shown that P can be precipitated as calcium phytate from the thin stillage fraction after ethanol distillation and before it is concentrated in DG. Initial estimates indicate that agricultural reuse of calcium phytate recovered from biorefineries could potentially displace 12.5% of national P fertilizer consumption and up to 37% of fertilizer use in agriculturally intensive Iowa.[40] Removal of P from DGs also represents a pathway to reduce a large amount of P from animal feed making it a potential centralized source of P reduction credits that could more accurately and effectively participate in nutrient trading markets within the Midwest. However, the current lack of understanding of the potential distribution of DGs and their embedded P flow in the US makes it difficult to fully elucidate the potential geospatial benefits of low-P DGs as a feed ingredient, and the role that CBs could play by participating in localized nutrient trading programs. While previous work focused on quantifying and discussing the potential for producing renewable P from biorefineries in the US,[40] this study focuses on the implications of removing P from DG. Although this study is focused on DGs from corn ethanol production in the US, it is also possible that this approach can be used for similar types of processes that generate co-product feeds outside of the US, such as the increasing production of corn ethanol in Brazil.[41] There were three primary objectives of this study: (i) develop an estimate of DG flow across the US, (ii) compare embedded P in human waste, animal manure, and DGs across the US, and (iii) apply these findings at the county-scale to estimate nutrient trading

Table 1. Data Sources for Determining DG Supply and Utilization and Human and Livestock P Excretion Rates

source	time scale	data description	spatial scale
		DG Supply	
RFA Ethanol Plant Locations	2017	ethanol production capacity (MGY) for plants across the US	point
		DG Potential Utilization	
USDA COA	2017	animal inventory numbers	county
		Human P Excretion	
US Census Bureau	2017	human population	county
		Animal Manure P Excretion	
USDA COA	2017	animal inventory numbers	county

potential for an agricultural watershed in the Midwest (Northern Raccoon River, IA).

■ METHODS

To meet the research objectives of this study, a transportation optimization model was used to determine the distribution of DGs throughout the US by optimizing travel distances between biorefinery suppliers and livestock and poultry utilization at the county level. Supply of DGs was determined using biorefinery data from the Renewable Fuels Association (RFA), and potential utilization in livestock and poultry feeds using information from the 2017 US Department of Agriculture (USDA) Census of Agriculture (COA) as shown in Figure 1. Embedded P in DGs was then compared to estimates of P excretion from both human waste and animal manure mapped across the US on a county level. Estimates of P from human waste and animal manure excretion were determined using existing methodologies that utilize population counts and average excretion rates of P. Phosphorus in DGs and human waste was then assessed for nutrient trading potential in a specific watershed region in Iowa.

Data Sources. Data sources were restricted to publicly available resources from governmental and other organizational databases (Table 1). Individual CB capacities were obtained from the RFA to calculate DG supply.[42] Livestock and poultry inventory data were obtained from the USDA National Agricultural Statistics Service (NASS), which provides data collected as part of the 2017 USDA COA.[43] Human population data estimates for each county were obtained from the US Census Bureau.[44] All maps and geographical outlines were generated in ArcGIS (Esri, Redlands, CA) using shapefiles for states and counties from the US Census Bureau.[45]

DG Supply. The quantity of DGs produced by CBs was estimated using the ethanol production capacities of individual biorefineries. The RFA data included both geospatial coordinates and maximum ethanol production capacities in million gallons per year (MGY), which were converted to million liters per year, for 182 dry grind corn biorefineries.[42] Although biorefineries may sometimes produce ethanol exceeding their rated capacity, particularly when profitable, the RFA capacities were utilized in this study because actual annual operating data for individual facilities are not available. For dry grind ethanol production facilities, DGs were estimated for 2017 by using the maximum ethanol capacity of each biorefinery, an average 2017 conversion of 0.4 L ethanol per kilogram of corn, and 0.3 kg dried DGs with solubles (DDGS) per kilogram of corn[46] to calculate annual DDGS production. DDGS is the predominant type of corn co-product of the multiple forms of DGs (i.e. dried, wet, and modified) produced by CBs, and was used as a proxy to determine overall facility-level DG production because facility-level production data of different DG types was not available. To determine facility-level DG, DDGS production was converted to a dry basis (i.e., 0% moisture) by assuming an average of 10% moisture by weight in DDGS.[35] Conversion to a dry basis allows for easier attribution to animal dry matter consumption and reduces the uncertainty of total mass depending on the actual form of DGs produced.

Dry grind corn ethanol biorefineries produced an estimated 33.8 Mt of DG per year on a dry basis in 2017, assuming maximum supply by all plants as used in this study. The RFA estimated approximately 33.3 Mt DGs per year on a dry basis in 2017 (i.e., 37 Mt of DGs produced per year at 10% moisture).[39] The difference between estimates was likely due to not all plants operating at full capacity and variations in the moisture content of the various types of DGs used in determining the RFA estimate. Individual estimates of biorefinery DG supplies are shown in Supporting Information Table S1. The total state supply of DG amounts are presented in Supporting Information Figure S1.

Potential Utilization of DGs in Livestock and Poultry Feeds. The potential DG utilization for each county was calculated based on its inventory of various livestock and poultry categories and exports. The method for calculating potential DG utilization in animal feed was adapted from a method used by USDA for a similar national-scale estimate.[47] The USDA 2017 COA was used to determine the number of each animal type per county based on four main categories (i.e., beef, dairy, hogs, and poultry) that included a series of subcategories (i.e., beef: beef cows, cattle on feed, and other beef including heifers, steers, bulls, and calves; dairy: dairy cows, dairy heifers; hogs: breeding and market swine; and poultry: broilers, turkeys, layers, and pullets). The number of animals in each category was used to calculate the total dry matter intake of feed based on the average dry matter intake per COA animal subcategory (Supporting Information Table S2). Potential DG utilization was determined for each county based on converting total dry matter intake by diet inclusion rates of DGs for each animal category (Supporting Information Table S3). A more detailed explanation of the determination of potential DG utilization as well as limitations are found in Supporting Information Method S1.

The estimated national potential utilization of DGs (64 Mt per year in 2017), based on the methods described, is nearly twice as much as what is currently produced in the US. This

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estimate is consistent with the USDA's estimate of 62 Mt in 2006/2007.[47] Based on the national estimates, beef cattle represent the majority of potential DG utilization, with an estimated 52% (33 Mt per year), while dairy cattle consumed 23% (15 Mt per year), swine utilized 14% (9 Mt per year), and poultry utilized 11% (7 Mt per year). However, these national estimates do not reflect the actual domestic DG utilization of 47, 31, 14, and 7% for beef cattle, dairy cattle, swine, and poultry, respectively, in 2017 because the amount of DGs produced is only consumed by a fraction of total livestock and poultry.[39] The distribution of potential DG utilization by animal category is shown in Figure 1, with potential beef cattle utilization widespread across the US, but other types of livestock and poultry utilization concentrated in particular regions. Beef cattle in feedlots have the largest potential DG utilization due to their overall greater dry matter consumption per head and their ability to utilize high (i.e., up to 40% of dry matter intake) dietary inclusion rates of DGs. DGs are a preferred energy source for beef feedlot cattle because they can be fed in wet or dry form, contain 120−130% of the energy value of corn, and the high fiber content minimizes the risk of rumen acidosis compared with feeding high amounts of corn containing rapidly fermentable starch.[34] However, diet inclusion rates of DGs containing high sulfur content should be limited to prevent polioencephalomalacia, which has historically been an occasional problem.[34] Potential utilization of DGs in beef cattle diets was determined based on all cattle being housed in confinement feedlot facilities, but there is a portion of these cattle that are in rangelands which was not accounted for due to the lack of national data. Although this does lead to an overestimation of potential utilization by cattle, range-fed cattle are likely more common in states outside of the Midwest where the lower density of corn biorefineries exists. Potential utilization of DGs by county for all livestock and poultry types and exports is provided in Supporting Information Table S4.

Besides livestock and poultry consumption in the US, nearly 30% of DGs produced in 2017 were exported to animal feeding operations in other countries, primarily in the form of DDGS. Approximately, 9.9 Mt per year were exported on a dry basis (i.e., 11 Mt per year at 10% moisture) based on the USDA Foreign Agricultural Service Global Agricultural Trade System (GATS) database.[48] Exports were distributed to counties based on exporting ports that were reported in USDA GATS, where nearly 60% of DDGS were shipped from the two major ports of Orleans Parish, LA, and Los Angeles County, CA.

DG Allocation. The allocation of DG supply to county animal production and exports was done using a transportation optimization model. Data on DG movement in the US are sparse, leading to the need to use limited information on DG use to define an appropriate allocation model. A Python extension for Gurobi Optimizer (Gurobi Optimization, LLC), which is a mathematical programming solver, was used to develop an optimization model for DG based on minimizing transfer distances for flows between CBs that supply DGs and centroids of counties. All potential DG supplies from biorefineries were assumed to be used in animal feed either domestically or in exports. Although DGs can move through intermediary storage facilities or feed distributors, these intermediary pathways were not considered in this study. The primary constraints included in the model were based on national- and county-level utilization estimates of DGs allocated to each animal category. Total national DG flows to each animal category were limited to 47, 31, 14, and 7% of total domestic DG usage for beef, dairy, swine, and poultry, respectively, based on 2017 RFA estimates.[39] Although this distribution of national DG utilization by animal category slightly varies from year to year, values were limited to 2017 to maintain consistency with available 2017 USDA livestock and poultry inventory and ethanol production data. DG flows to counties were limited to the potential utilization by each animal category. The model also included a constraint that required DG exports to be fully satisfied in counties where ports were located. A detailed description of the model is found in Supporting Information Method S2.

DG-Embedded P. The P levels in DGs were estimated based on the supplied DGs in each county. Embedded P of DGs supplied to each county was calculated using an average P concentration of 9.26 mg P per gram of DGs on a dry basis[35] while also considering a potential uncertainty range of P concentration of 7.0−9.9 mg P per gram of DGs.[49] Feeds containing low concentrations of P were also considered for each county using supplied DGs based on modeled estimates for precipitation of P at biorefineries, leading to a concentration of 3.25 mg P per gram of DGs on a dry basis or a 65% reduction in P.[35] An uncertainty range of 60−65% reduction[35] in P concentration of DGs was also considered for all P reduction calculations. This embedded P was then compared to results associated with both human P excretion and total livestock and poultry P excretion in each county and state.

Human P Excretion. To compare the quantity of P embedded in human excreta with that in DGs used in animal feeds on a county basis, the total amount of P in human excreta in a county was estimated. For a more direct comparison with P excretion associated with animal manure, human population data were used to determine county-level human P excretion rates. The human population per county for 2017 was determined using estimates from the US Census Bureau.[44] The excretion rate of P from humans in the US was estimated using an approach that considered total food and plant protein estimates. The amount of P excreted by an individual (grams P/capita/day) was determined by eq 1.[50]

$$\text{Phosphorus} = 0.011 \times (\text{total food protein} + \text{plant food protein}) \quad (1)$$

In the US, in 2017, the average total food protein was approximately 113.73 g per capita per day and plant food protein was 39.86 g per capita per day[51] leading to an estimated P excretion of 0.62 kg P per capita per year. This P excretion rate was then used along with the county human population estimates to determine county-level human P excretion rates. To account for uncertainty in the amount of human P excretion, available yearly protein data were used to develop a range of human P excretion amounts in the US of 0.54−0.64 kg P per capita per year.

Total P Excretion from Livestock and Poultry. Total P excretion rates from livestock and poultry were determined to make a more direct comparison of animal manure P contributions to overall human P contributions. Total P excretion per county was determined using an existing US Geological Survey (USGS) approach which uses livestock and poultry inventory numbers by category, and P excretion rates from the literature, to calculate P excretion rates on a per-head basis in each animal category.[52] This approach is also similar to

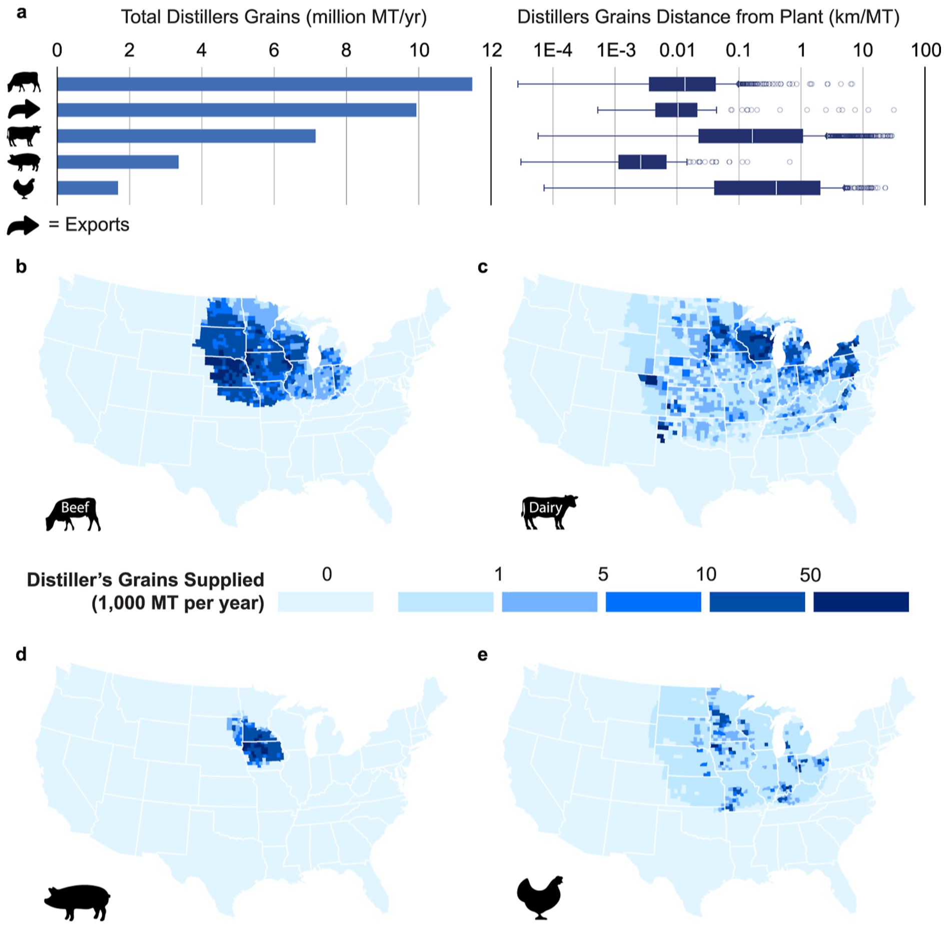

Figure 2. Modeled distribution of DGs among counties. (a) Total DGs supplied and the range of distances traveled per metric ton for each DG flow by animal category in 2017. Supplied DGs are shown for each county for (b) beef cattle, (c) dairy cattle, (d) swine, and (e) poultry. Beef cattle and swine have the most concentrated areas of DGs supplied leading to shorter distances per MT supplied, whereas dairy cattle and poultry have lower concentrated utilization leading to a longer distance versus supply ratio.

a method used by the nutrient use geographic information system (NuGIS) database for livestock and poultry P, which is a database that estimates county and watershed-level nutrient balances on croplands.[53] The animal inventory values for each county that were obtained from the 2017 USDA COA were used for multiple animal categories.[43] Animal categories were similar to those used to determine potential DG utilization, and P excretion rate data per animal category (Supporting Information Table S5) were based on an earlier USGS report.[54] Total livestock and poultry numbers in each county were then multiplied by their associated P excretion rates to determine overall animal P excretion per county. Although there is uncertainty in the animal P excretion rates, a range of excretion rates were not considered in our analysis because of the wide variability in estimates based on the type of animal and diet composition fed, which makes it difficult to develop a defensible range for the amount of P excreted. Additionally, total livestock and poultry P excretion estimates were only used as a reference and did not influence the comparison between P embedded in DGs and human P excretion.

Watershed Region Analysis. Nutrient trading markets provide a potential economic mechanism to incentivize corn biorefineries to perform P recovery by trading nutrient reduction credits with existing point sources, like WRRFs. To assess the localized potential for a nutrient market, the embedded P in DGs utilized by livestock and poultry and the P in human wastes were assessed in a watershed region. Although markets are frequently based on a watershed geospatial scale, data in agricultural databases are not reported on this scale, and attempts to proportion county-level statistics based on the area of inclusion in the watershed can lead to large inaccuracies in estimates due to unequal distributions of animals in a county.[55] Therefore, this study maintained the use of county-level embedded P in DG distribution data and human waste P estimates to provide some context on how a biorefinery may contribute to a nutrient market in the North Raccoon Watershed (NRW), which is a priority watershed for nutrient pollution in Iowa.

Uncertainty and Sensitivity Analyses. The lack of national and local data involving DG usage and quality creates uncertainty in the distribution model. To better understand how uncertain parameters impact embedded P transport to animal feeding operations, a sensitivity analysis was performed on a state and regional level on variability of input parameters (Supporting Information Table S6) to the DG distribution model (i.e., DG production and P concentration, national DG utilization by animal category, and diet inclusion rates of DGs). A triangular distribution was used based on the minimum, maximum, and mode for each uncertain parameter for Latin hypercube sampling to generate 1,000 samples for Monte Carlo simulation based on parameter uncertainty. Each parameter was then assigned a Spearman's rank coefficient to

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Table 2. Ranking of Top 10 Intrastate and Interstate Flows of Domestic DG Feed (Mt Per Year) and Embedded P (1000 MT Per Year) with Uncertainty Ranges

intrastate flows			interstate flows				
state	DG	P	states			DG	P
Iowa	5.2	48 [36–51]	Iowa	→	Wisconsin	0.66	6.1 [4.6–6.5]
Nebraska	2.9	27 [20–29]	Iowa	→	Missouri	0.53	4.9 [3.7–5.2]
Minnesota	2.3	21 [16–23]	Minnesota	→	Wisconsin	0.49	4.5 [3.4–4.8]
South Dakota	1.6	15 [11–16]	Iowa	→	Nebraska	0.48	4.4 [3.3–4.7]
Wisconsin	1.5	12 [9–13]	South Dakota	→	Nebraska	0.38	3.5 [2.6–3.7]
total	**17.2**	**160 [121–171]**	**total**			**6.66**	**62 [47–66]**

determine the greatest contributors to sensitivity in embedded P of the distribution model. From the sensitivity analysis, the most sensitive parameters were then utilized to determine uncertainty ranges in results. A more detailed description of the uncertainty and sensitivity analyses is found in Supporting Information Method S3.

RESULTS AND DISCUSSION

Although the reduction of P from human wastes at WRRFs is often prioritized and regulated, the total estimated P in animal manure is nearly an order of magnitude larger nationally, and represents the primary source of P pollution in the Midwest.[18,56] The decentralized nature of animal manure makes it difficult to regulate and control loss of P, but the production and use of DGs in animal feeds represents a potential centralized approach to reduce P contributions to animal manure in areas where it is utilized as a feed ingredient. While animal manure is widespread across the US, modeled DG utilization in animal feeds is centralized in the Midwest where transport distance between biorefineries and animal feeding operations is minimized (Figure 2). The geospatial mapping of DGs and associated P flows is shown in subsequent sections, as well as a comparison to P in animal manure and human waste. Based on transport optimization modeling, reduction of P from DGs in complete feeds transported to livestock and poultry farms was estimated to be significantly greater than human-associated P excretion in several Midwestern states, indicating that CBs represent a major potential source of P reduction credits in the region.

DG Supplied. Estimated amounts of DGs transported between CBs and livestock and poultry operations were concentrated in the Midwest due to the intentional synergy between grain production, bioethanol refining, and livestock and poultry farms (Figure 2b–e). Although there are a small number (i.e., 12%) of CBs located outside of the Midwest region and agriculturally intensive neighboring states (i.e., South Dakota, North Dakota, Kansas, and Nebraska), these biorefineries were estimated to primarily supply their DGs to more remote port counties for exports based on the optimization model constraint requiring full supply of exports and the relative proximity of these biorefineries to port counties. However, it is likely that some of these biorefineries outside of these states supply DGs in feed to local livestock and poultry operations to some degree, particularly, if wet DGs are produced, but the lack of biorefinery-specific information makes it impossible to account for this possibility. The variation in geospatial DG utilization between animal categories is due largely to the constraint on the optimization model that only allows a specified portion of DGs to be supplied to each livestock and poultry category. Cattle are the primary users of DGs (Figure 2a), utilizing 78% of total domestically consumed DG, which is due to their ability to use both dry and wet DGs as feed and their relatively greater nutritional value compared with swine and poultry. Estimated DG consumption associated with beef cattle was largely in the Midwest and Northern Plains (Figure 2b). The high density of potential utilization for beef cattle led to a smaller spatial distribution of DG use estimates than dairy cattle or poultry (Figure 2a). Dairy cattle had the broadest spatial distribution of DG usage (Figure 2c) due to their larger share of nationally reported DG usage (i.e., 31% of DG) but smaller individual county-level utilization with higher utilization densities focused in specific states like Wisconsin, Michigan, Minnesota, and Pennsylvania. All modeled DG flows are provided in Supporting Information Table S7.

In comparison to cattle, DG usage estimates were significantly less for swine and poultry (Figure 2a). DG use in swine and poultry diets is comparatively limited due to its suboptimal nutritional characteristics including (1) high polyunsaturated fatty acid content of corn oil in DDGS that can reduce carcass pork fat quality,[57−59] (2) high fiber content which reduces carcass yield in pigs[60] and reduced metabolizable energy content for poultry,[61] (3) amino acid imbalances relative to requirements for both swine[62] and poultry,[63] which requires supplementation of multiple crystalline amino acids, (4) relatively low and variable amino acid digestibility for swine[64] and poultry,[65] and (5) concerns of potential mycotoxin contamination which could adversely affect animal health and performance.[66] Although higher dietary inclusion rates of DGs are being used in swine diets compared with broiler diets, the variability in energy and digestible amino acid content has led to conservative usage rates due to uncertainty of actual metabolizable energy,[67] net energy,[68] and digestible amino acids[64] despite the development and availability of accurate prediction equations based on actual nutrient composition of the DDGS source being fed. Similarly, variability in metabolizable energy content[61] and amino acid content and digestibility[65] among DG sources creates uncertainty regarding the accuracy of nutrient loading values to use in precision feed formulation in poultry diets and results in conservative dietary inclusion rates to minimize the risk of reduced performance.

Swine had the most limited distribution of modeled P, which was confined mainly to Iowa, southern Minnesota, and eastern South Dakota (Figure 2d) due to the low overall national use of DGs, but high swine population density in those states. Although there is DG utilization in swine diets in other Midwestern states such as Illinois and Indiana, where DGs were estimated to be supplied in diets fed to other types of animals, the limitation of the total national DGs allocated to swine caused the DG allocation model to concentrate most allocated supply in areas with concentrated DG supplies, such

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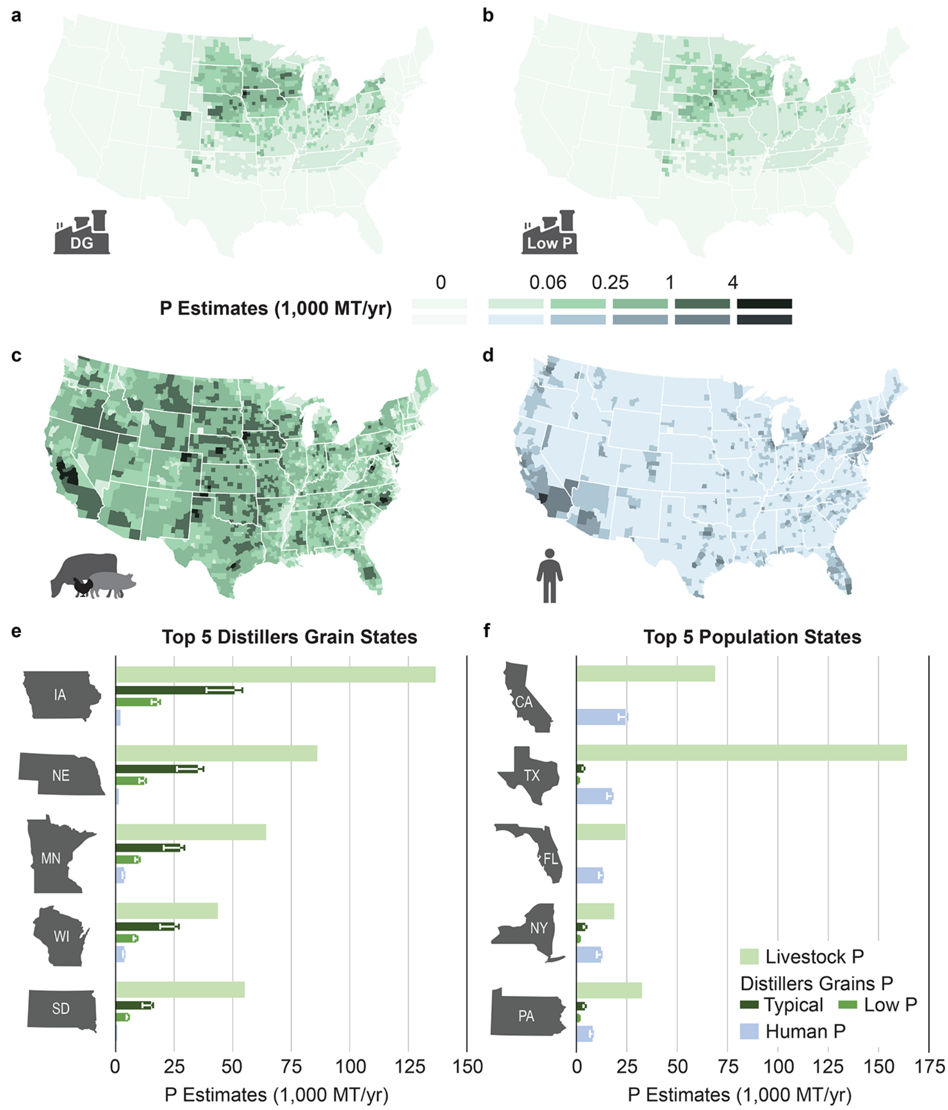

Figure 3. DG, human waste, and animal manure embedded P. Embedded P estimates with uncertainty ranges are shown across the US for (a) DG, (b) low-P DG, (c) total animal manure excretion, and (d) total human waste excretion with the legend indicating a range of values for each county color. State-level data is also compared for these four categories and ranked based on estimated (d) P embedded in DGs and (e) human (population) P excreted. Overall, estimated P excretion from animal manure is often significantly greater than estimated human P excretion even in the most populous states. There are also numerous states in which estimated DG-embedded P is greater than the amount of P excreted by humans.

as Iowa and southern Minnesota. This trend was also observed based on the shorter distances of estimated transfers between CBs and swine farms (Figure 2a). Poultry have a broad distribution of DGs supplied due to their generally low utilization (Figure 2e). Distributions of county DG utilization density by animal category are shown in Supporting Information Figure S2.

Based on total intra- and interstate flows, an estimated 72% of all DGs used in animal feed were supplied in the same state where the DGs were produced. As shown for the top five states in each category, modeled intrastate DG flows were much greater than interstate flows (Table 2), with the greatest intrastate flows observed in the Midwest due to the abundance of CBs and livestock and poultry DG utilization. Also, an estimated 57% of the total DGs and 81% of domestically used DGs were supplied within 100 miles of production (Supporting Information Figure S3). Wet DGs are typically sold to beef cattle feedlots within 100 miles of a biorefinery due to storage and transportation limitations.[69] When only considering those DGs supplied to domestic cattle within 100 miles, only about 43% of total DGs can potentially be in the form of wet DGs. This estimate is consistent with the estimated 41% of DGs that were classified as a type of wet DGs in 2017.[39] The large proportion of the localized supply of DGs further demonstrates the synergistic relationship between ethanol production and consumption of DGs by livestock and poultry production operations. A distribution of DGs in the US based on maximum diet inclusion rates for each type of livestock and poultry was also considered (Supporting Information Figure S4), where the use of higher dietary inclusion rates resulted in an even greater utilization density of DGs in the Midwest, with more supply to local livestock and poultry farms. One particular benefit of the proximity between DG production and supply to livestock and poultry farms is the potential for localized P reduction through the reduction of P in DGs at biorefineries using precipitation technologies[35] prior to animal consumption.

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Estimates for exported DGs were largely based on sourcing from CBs in the Midwest and accounted for approximately 30% of the total potential DGs produced in 2017 (Supporting Information Figure S5). About 75% of modeled exported DGs were produced in Midwestern states, with Illinois, Nebraska, Indiana, and Iowa estimated to contribute nearly 50% of the exported DGs. Ethanol plants located in Illinois were estimated to be the primary contributors to exports due to the limited use of DGs in animal feed and proximity to the large Port of New Orleans, which accounts for nearly 40% of the total exported DGs. The transport optimization model indicated that ethanol facilities in Illinois alone export more DGs than Texas, California, Georgia, and Tennessee combined. This is significant because the export of DGs affects the loss of embedded P in those grains, particularly from the agriculturally P-intensive Midwestern states. Only a quarter of exported DGs are exported to North American countries (i.e., 20% imported by Mexico and 6% by Canada) while most of the remaining DGs are exported to markets in Asia.[39] DG distribution simulations were developed in the absence of exports (Supporting Information Figure S6a) and any national limitations on distribution (Supporting Information Figure S6b) showing the potential for larger distributions of DGs or for greater usage potential for other animal categories in the Midwest.

The sensitivity analysis showed that the DG distribution model was highly sensitive to diet inclusion rates, particularly for beef and dairy cattle diets in the Midwest and Northern Plains (Supporting Information Figure S7), with low to moderate sensitivity to national DG utilization rates by animal category and overall DG production (Supporting Information Figure S8 and Table S8). Although the model was sensitive to diet inclusion rates, there can be wide variability in DG usage in diets between individual animal feeding operations, making it difficult to determine localized county-level dietary DG inclusion rates without additional data. Therefore, DG distribution estimates, therefore, do not include uncertainty ranges. The sensitivity analysis of embedded P also showed high sensitivity to P concentration in DGs (Supporting Information Figure S9 and Table S9). Because the P concentration in DGs is well documented and easily verified at individual biorefineries, the range of uncertainty was included when considering embedded P estimates in the next section.

Comparing Phosphorus Flows in DG, Animal Manure, and Human Waste. An estimated 63% of total P embedded in DGs fed to livestock and poultry flows both within and to the Midwest. The greatest density of estimated embedded P from DGs is in the upper Midwest and Northern Plains states (Figure 3a) with an estimated 153,500 [116,000−164,000] metric tons (MT) of P per year concentrated in the top five states: Iowa, Nebraska, Minnesota, Wisconsin, and South Dakota (Figure 3e). An estimated 75% of P in these states was from DGs utilized by beef and dairy cattle. The large share of DG usage by ruminants has more significant P pollution implications than feeding DGs to swine and poultry due to the greater excretion rates of P in cattle caused by dietary inclusion rates and excess feed of P from DGs[70−72] compared with swine and poultry which have less P excretion from DGs due to lower diet inclusion rates, high P digestibility, and formulating diets on a digestible P basis.[73−75] The P concentration of DGs is also generally greater than other major types of feed ingredients[76] leading to a higher overall potential for excess P excretion at higher dietary inclusion rates. As a result, because DGs are a preferred energy source for finishing beef cattle and are fed at diet inclusion rates up to 40% of dry matter intake, the amount of protein (N) and P consumed in the total diet greatly exceeds the requirements and leads to excess excretion in manure. Outside of the Midwest region, estimated DG usage is primarily dominated by dairy cattle, but because of less overall density, there are less embedded P contributions.

The estimated amounts of locally produced DGs supplied in agriculturally intensive regions highlight the potential opportunity for CBs to improve P use efficiency through production of low-P DGs and renewable fertilizers. Estimates of embedded P contributions for DGs containing low concentrations of P (i.e., 3.25 mg of P per gram of DGs) provide an additional comparison between P estimates (Figure 3b). Overall, production and use of low-P DGs would result in an estimated reduction of 143,600 [116,000−164,000] MT P per year in P fed to livestock and poultry out of an estimated 221,000 [167,000−236,000] MT P per year embedded in typical DGs in the US. Additionally, the production of low-P DGs could generate a renewable P fertilizer through P recovery that could also create localized benefits for meeting P requirements of nearby farms for crop production as demonstrated in previous work.[40] The greatest density change of P from the use of low-P DGs is in the upper Midwest and Northern Plains where the estimated DG usage was heavy. However, there is currently no regulatory or financial incentive for CBs to produce low-P DGs for animal feed due to a lack of state-level P nutrient management strategies and the lack of potential profit from the sale of DGs as renewable P fertilizer. Therefore, a potential mechanism for biorefineries to participate in nutrient management plans and potentially gain a financial incentive is through a nutrient trading market, which would require biorefineries to be capable of reducing P at or beyond the capacity of other sources, such as human waste, to allow for credit trading.

For comparison to embedded P in DGs, P excretion was estimated from both total animal and human populations throughout the US (Figure 3c,d). Although embedded P in DGs was not directly related to P excretion in animal manure because only a fraction of the dietary P consumed by animals would be excreted, the P excreted in animal manure and human waste provides a reference point for the implications of recovering embedded P in DG. When considering all counties where DGs are supplied, P in animal manure and human waste was estimated at 1.1 million and 70,400 [61,300−72,700] MT P per year, respectively, while an estimated 143,000 [116,000−164,000] MT P per year could potentially be saved by using a low-P DGs in animal feed. Of the 1666 counties supplied with DG, based on the optimization model, an estimated 54% [51−57%] of DGs would have greater embedded P in DGs than P in human waste, and 49% [44−52%] of DGs would have greater potential recovery of P than from embedded P in human waste. Of those counties with greater potential for recovered P from DGs, an estimated 82% [79−84%] of them would have the potential for more than twice the amount of recovered P relative to the amount of P embedded in human waste, with one county having more than 1000 times that amount. This potential P recovery from DGs was also observed in the top five states where DGs are supplied, with the potential P reduction estimated to be much greater than the human waste P contributions in those states (Figure 3e). However, this same trend was not observed among the most

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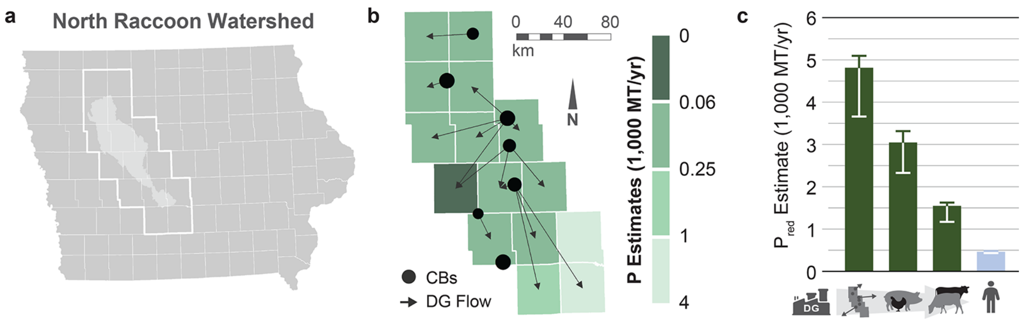

Figure 4. Phosphorus assessment of counties containing the NRW region in Iowa. (a) Map of the watershed region and associated counties. Geospatial mapping of (b) CBs with DG flows. (c) Total facility-level P reduction (P_{red}) estimates from WRRFs and CBs with consideration of DG flows to exterior counties, nonruminants (swine and poultry), and ruminants (beef and dairy cattle). P reduction potential from CBs within the NRW region is much greater overall than WRRFs even when considering only consumption of DGs by ruminants.

populous states where there was a comparatively lower supply of DGs and therefore, less embedded P (Figure 3f). Embedded P estimates by state for DGs, human waste, and total animal manure are presented in Supporting Information Table S10.

The efficient use of P throughout agricultural systems is necessary to maintain resource security in the US.[13] In addition to inefficient localized P use, an estimated 91,900 [69,500−98,300] MT per year of embedded P in DGs is also lost from the US as exports. Of the total exported embedded P in DG, more than 59,600 [45,000−63,700] MT P per year could potentially be recovered prior to export. Based on the modeled DG distribution, Illinois was the greatest contributor to exports, with an estimated 19,100 [14,400−20,400] MT P per year embedded in DG. CBs in Indiana and Nebraska were estimated to represent the second and third greatest contributors to DG exports, respectively, with an estimated 8,900 [6,700−9,500] and 8,400 [6,300−9,000] MT P per year, respectively. The Midwestern states where large amounts of DGs are produced and exported are also major crop-producing states that require large amounts of P fertilizer. Illinois alone has an estimated P fertilizer usage of more than 156,000 MT P per year, with Indiana and Nebraska using over 86,000 and 106,000 MT P per year, respectively.[40] Also, with the price of P-based fertilizer nearly doubling in the past two years,[77] the need to manufacture and purchase additional P fertilizer only further exacerbates the already fragile P security in the US.

Exploring Opportunities for Biorefinery Participation in Nutrient Trading Markets. The proximity of corn biorefineries to livestock and poultry operations where DGs are supplied presents a potential opportunity for biorefinery participation in nutrient trading markets. While estimated DG flows suggest a large potential for localized reductions of P through DGs rather than human wastes, an additional consideration is that a nutrient trading market requires some level of geographic proximity of the trading partners.

Iowa is one of the main contributors of P to the Gulf of Mexico,[17] which led to the development of the Iowa Nutrient Reduction Strategy (Iowa NRS).[78] The Iowa NRS includes the goal of reducing P discharge from WRRFs by over 1,970 MT P per year, with nutrient markets listed as a potential mechanism to accomplish this. When considering counties partially contained within the North Racoon Watershed region (Figure 4a), the total estimated embedded P in human waste was only 7% [5−9%] of that attributed to DG, and 10% [8−15%] of potential estimated P that could be extracted from the NRW region through the use of low-P DGs. Approximately 97% of the modeled DG supply in the NRW region was estimated to be from local CBs (Figure 4b). While there are numerous WRRFs in the NRW region, they are primarily small plants servicing rural communities with only one large urban-based WRRF, the Des Moines Metropolitan Wastewater Reclamation Authority (DMMWRA), located in the southern part of the region that is finalizing plans to install a P removal system.

When considering potential reductions in embedded P in DGs from biorefineries and WRRFs located in the NRW region, biorefineries could reduce an estimated 20 times [14−25 times] more P from all of the DGs produced than from WRRFs. When considering only intraregional-modeled DG flows (i.e., those produced by biorefineries and estimated to be supplied to all animals within the NRW region), biorefineries could recover and reduce an estimated 10 times more P than WRRFs from DGs they supply in the region or 2 to 3 times more P when only considering DGs fed to ruminants in the region (Figure 4c). When considering single facility-level P reduction and recovery capacities, an estimated 310 [290−320] MT P per year[40] could be recovered by the DMMWRA, which makes up the majority of P reduction capacity in the NRW region. In comparison, the smallest biorefinery in the region could potentially extract an estimated 760 [570−810] MT P per year from the local animal feed supply chain, which is nearly twice as much as the largest WRRF, with other biorefineries representing over 1,900 [1,400−2,000] MT of potential P reductions per year. Furthermore, the generation of renewable P from biorefineries and its use in croplands could offset an estimated 37% [28−40%] of P fertilizer consumption, which could potentially reduce soluble P losses from croplands in the NRW region.

Although there is significant potential for P reductions from biorefineries in the NRW region, it is important to recognize that P reductions in DGs do not directly lead to less P loading in water bodies from manure because every MT of P reduction from a biorefinery has a different impact than P from a WRRF. The uncertainty in P contributions from DGs fed to livestock and poultry diets is primarily the result of differences in diet inclusion rates and associated P excretion rates of manure from animals fed these diets, along with the actual P loss to receiving water bodies. Alternatively, there is a limited uncertainty in P contributions from WRRFs, since the P discharged to water bodies can be directly monitored. When considering nutrient trading between trading partners where one has uncertainty in P reductions (i.e., reductions from DGs with low P), a trading ratio is often used to normalize P credits and account for the uncertainty (i.e., a trading ratio of 4:1 requires the partner with uncertainty in P reductions to reduce 4 MT P for every 1 MT

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P the regulated facility must reduce).[22] The large magnitude of potential P reductions from biorefineries would assist in overcoming the ratio that would be placed on such a trade. A summary of P data in the NRW region is shown in Supporting Information Table S11.

Study Limitations. Results from this study represent the first national-scale assessment of specific embedded P in livestock and poultry manure from feeding DGs compared with P excreted from human waste at the county and state levels. However, limitations in publicly available data created a level of uncertainty in the results. A major limitation was the lack of data related to the actual utilization of DGs in various types of food-producing animals and production phases. Due to the sensitivity of the DG distribution model to diet inclusion rates, additional data on localized diet inclusion rates would improve estimates of DG utilization in animal feed. Although certain parameters (i.e., national DG utilization by animal category, dietary DG inclusion rates, and P concentration of DG) and their uncertainty were considered in the DG distribution model, other potential parameters that could have been considered but adequate data were not available including land use (i.e., cropland versus range land versus pasture), animal life stage, and adoption rates of DGs for each animal species to develop baselines or uncertainty ranges. There were also limited data on the types of DGs (i.e., wet, dry, and modified) supplied by individual ethanol plants that prevented more accurate estimations of transportation costs and actual exports from individual biorefineries. If more information was available, the use of cost instead of transport distance could improve the DG allocation model. An additional limitation involved the comparison between embedded P in DGs and overall P excretion from animals and humans. Although embedded P in DGs was estimated throughout the US, it does not directly result in excretion of P in animal manure because a significant portion of P in DGs is digestible, utilized to meet the P requirement of animals, and is not excreted in manure. Furthermore, routine use of commercially available phytase enzymes in swine and poultry diets improves dietary P digestibility and reduces manure P excretion, but there are no data available to quantify these effects in commercial livestock and poultry operations. There are limited studies that estimate P excretion in animal manure—particularly from beef and dairy cattle[70,79]—based on a range of DG inclusion rates, but there is not a direct method to determine P contributions solely from DGs or the effects of feeding diets containing low-P DGs on manure P excretion rates. Also, the incorporation of new technologies at corn biorefineries—such as the addition of phytase prior to fermentation—can change the form, amount, and digestibility of P[38], creating variability in actual P excretion based on where DGs were sourced.

■ OUTLOOK

Phosphorus is a finite and essential nutrient required by all food-producing animals and is the third most expensive nutritional component of livestock and poultry diets beyond energy, protein, and amino acids. Unfortunately, the inefficient use of P and losses from agricultural activities[80] have disrupted biogeochemical flows,[1] contributed to eutrophication of water systems,[81] and created an urgency to develop and implement measurable and meaningful interventions. Animal agriculture contributes to P losses associated with agriculture because less than 60% of dietary P is converted to edible meat, milk, and eggs.[33] As a result, nutritional interventions to improve dietary P utilization efficiency and reduce P excretion in manure in beef, dairy, swine, and poultry production systems are needed.

The large difference between P excreted in animal manure and human waste presents an incentive for prioritizing P reduction in animal manure, particularly in the Midwest. In numerous states, embedded P in DGs and corresponding P reduction potential was estimated to be greater than P present in all human wastes. This presents a potential method for regulatory agencies developing nutrient management plans to consider and quantify the benefits of incentivizing feed producers to add P recovery or treatment technologies to reduce P to nutritionally necessary levels—or a 65% reduction[35] as utilized in this study. This new type of low-P DGs could be utilized as an alternative animal feed to provide beef and dairy cattle producers with a potentially cost-effective[35] method for reducing manure P rather than removal of P directly from manure. This study provides an inventory of P attributed to DG production as well as comparative P excretion rates from both humans and animals to assist in this assessment by regulatory authorities. It also provides a compelling argument for the production of low-P feed from DGs in order to better address localized excess P generation from beef and dairy operations to offset P from human wastes. Currently, no corn biorefinery has implemented P reduction and recovery due to the costs exceeding potential revenue from renewable P production. Further work is necessary to elucidate potential incentives that could overcome this deficit which may include participation in nutrient trading markets or direct economic incentives for P recovery or reductions. Additionally, more robust and localized techno-economic analyses are necessary for biorefineries and WRRFs to more accurately compare cost and benefits for P reduction between them, particularly due to the large variation in chemical costs which are major contributors to these cost differences.[35,82] Corn biorefineries in the US are the primary focus of this study because they are already well characterized, but the increased usage of grain processing for protein extraction as well as other ethanol production methods internationally present additional opportunities for potential P recovery and reduction in biofuels co-products both within and outside the US.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.3c02228.

Plant-level DG production (XLSX)

County-level potential DG utilization (XLSX)

Simulated DG flows (XLSX)

Detailed information on determining DG utilization and the DG allocation model; input tables to DG utilization; animal manure excretion factors; state and county-level data on DG supply and embedded P in human waste and animal manure P excretion; county-level simulation data for varying scenarios; and county-level NRW data (PDF)

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Notes

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