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Managing Diffuse Phosphorus at the Source versus at the Sink

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Abstract

Judicious phosphorus (P) management is a global grand challenge and critical to achieving and maintaining water quality objectives while maintaining food production. The management of point sources has been successful in lowering P inputs to aquatic environments, but more difficult is reducing P discharges associated with diffuse sources, such as nonpoint runoff from agriculture and urban landscapes, as well as P accumulated in soils and sediments. Strategies for effective diffuse-P management are imperative. Many options are currently available, and the most cost-effective and practical choice depends on the local situation. This critical review describes how the metrics of P quantity in kg ha^{-1} yr^{-1} and P form can influence decision-making and implementation of diffuse-P management strategies. Quantifying the total available pool of P, and its form, in a system is necessary to inform effective decision-making. The review draws upon a number of "*current practice*" case studies that span agriculture, cities, and aquatic sectors. These diverse examples from around the world highlight different diffuse-P management approaches, delivered at the source in the catchment watershed or at the aquatic sink. They underscore workable options for achieving water quality improvement and wider P sustainability. The diffuse-P management options discussed in this critical review are transferable to other jurisdictions at the global scale. We demonstrate that P quantity is typically highest and most concentrated at the source, particularly at farm scale. The most cost-effective and practically implementable diffuse-P management options are, therefore, to reduce P use, conserve P, and mitigate P loss at the source. Sequestering and removing P from aquatic sinks involves increasing cost, but is sometimes the most effective choice. Recovery of diffuse-P, while expensive, offers opportunity for the circular economy.

1. Introduction

1.1. The Importance of P

Judicious phosphorus (P) management is a global challenge.[1,2] As a life-essential element required for all living organisms, P plays a vital role in our food chain. Agriculture is not sustainable in its absence; in fact, to sustain one person required 22.5 kg of phosphate rock per year in 2009.[3] This has since increased to 35.2 kg based on current global phosphate production and population.[4] This reliance on phosphate rock has critical implications for global food security and vulnerability to supply shocks. Reserves of phosphate rock are geographically concentrated, with 84% of the world's easily exploitable reserves located in only four countries.[5] Globally, about 62% of P output to oceans occurs from point sources, with 38% from agriculture.[6]

Losses of P occur along the food-supply chain, from mine to farm to fork, and via point and diffuse sources in the catchment watershed.[7,8] These losses are illustrated in the left-hand side of Figure 1.[7] The lost P has

significant deleterious impacts on water quality through eutrophication, formation of hypoxic zones, and deterioration of ecosystem services.(9,10) For example, over 400 hypoxic coastal "*dead zones*" exist due to pollution from P and nitrogen (N).(11) It is estimated that freshwater eutrophication in the United States costs a minimum of $2.2 billion every year, with the economic impact of harmful algal blooms alone approximated at $100 million per year.(12,13) Similarly, the cost of freshwater eutrophication in England and Wales has been valued at $100 to $160 million per year, with a further $70 million spent per annum to address this damage and meet legal obligations.(14) In China, economic losses associated with algal blooms occurring in Lake Tai and its catchment area were estimated at approximately $6.5 billion USD in 1998.(15) The economic need to protect such systems from nutrient enrichment is further evidenced by the monetary value of ecosystem services and biodiversity in Ireland (assessed at over $3 billion USD per year(16)), while environmental degradation places at risk a $12 billion USD tourist industry in New Zealand, where over 90% of tourists visit for the quality of the natural environment.(17) Although national and international strategies have been developed to manage P at agricultural, industrial, and municipal scales,(18−23) the problems associated with diffuse-P losses continue to grow.
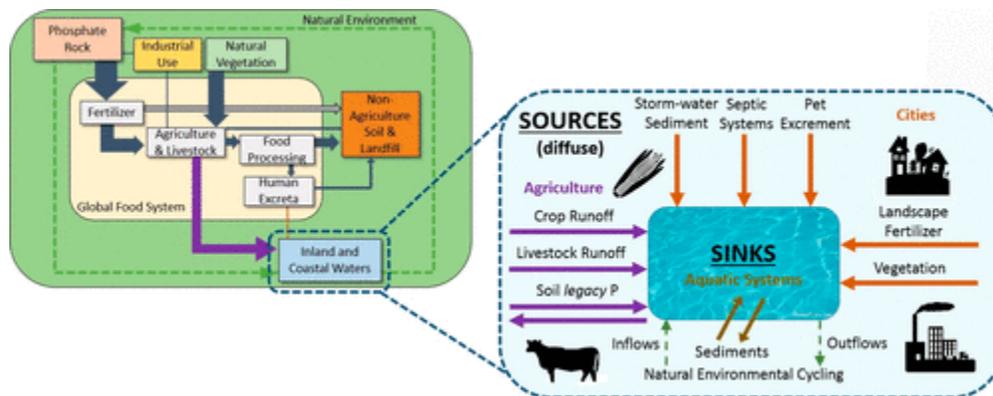

Figure 1. Where is the P? On the left, flows of P through the global food system are illustrated in a simplified schematic, where arrow size correlates to the magnitude of the flow (adapted from Cordell and White (2014)(7)). On the right, examples of diffuse-P sources entering aquatic systems (sinks) are illustrated (these examples are explored in case studies in this article). These sources are intrinsically diffuse and periodic, as discussed further in the Supporting Information, which can hamper nonpoint P-source management.

In addition to its positive impact on water quality, improved P management is an important factor to improve resource efficiency and move toward a circular economy.(24) A circular economy transforms today's wastes into resources and adopts sustainable options to recover the value, which can include P, N, organics, energy, metals, and water. Therefore, measures to lessen the negative impacts of lost P on water quality also can bring about recovery and reuse of P in some cases.(10,24) Given the importance of P removal (possibly paired with P recycling), strategies for effective management are essential, and it is important to consider an array of options, including those accounting for point-source P and diffuse-P, which are present in variable quantities across a wide range of settings.

1.2. Where is the P?

Legislation in most countries controls wastewater discharges through standards for the collection, treatment, and discharge of P in wastewater. These controls have succeeded at lowering P inputs to sensitive water environments in large part because the wastewaters are *point sources*, or waters collected in pipe networks. Point sources are amenable to *end-of-pipe treatment*, which is widespread for treatment of domestic and industrial wastewaters. For example, point-source controls in the U.S.—including P-detergent bans and advanced P removal at wastewater treatment facilities—since enactment of the EPA's Clean Water Act have effectively reduced P loads to the water environment. For instance, the annual municipal P load to Lake Erie

dropped from 14 million kg in 1972 to 2 million kg in 1990.(25) The effectiveness of point-source control has now shifted attention to improved management of diffuse sources in the catchment.

What has proven much more difficult is reducing P discharges that emanate from *diffuse sources*, such as *nonpoint* runoff from agricultural and urban landscapes, as well as managing *legacy* P stored in soils and accumulated in sediments. Examples of diffuse-P sources detailed in this critical review are illustrated in the right-hand part of Figure 1.(7) Their intrinsically diffuse and periodic nature makes nonpoint sources difficult to manage by technological means, such as collection and end-of-pipe treatment. (More information on variability of diffuse sources is available in the Supporting Information (SI)). Nevertheless, diffuse-P management strategies are being implemented. For example, monitoring of catchment sites dominated by intensively grazed pasture in New Zealand showed reductions in median P concentrations at 57% of sites between 2004 and 2013. This was attributed to a range of factors from improved awareness of diffuse-P issues, to having and implementing more options to mitigate against diffuse-P losses.(26) In many situations, the magnitude of diffuse sources of P dwarfs point-source discharges. On a global scale, point sources are only about 6% of the total P load to environmental waters.(3,8)

In this review article, we evaluate management options for nonpoint, diffuse-P. We present several good "*current practice*" case studies, selected from the global literature, exemplifying settings in which P losses to surface water are dominated by diffuse agriculture and city runoff, along with the recycling of *legacy* P stores. We define good *current practices* as options that offer practicality, cost-effectiveness, and/or legislative compliance in diffuse-P management for water quality. To make informed P-management decisions related to source versus sink scenarios, the diffuse-P losses from a catchment watershed (sources of P) and P inputs to a particular water body (sinks of P) must be evaluated. The quantity and form of diffuse-P varies widely across systems, and they have major implications for effective management approaches. Diffuse-P management options discussed in this review are transferable to other jurisdictions at the global scale.

1.3. Objectives

Here, we review P management for diffuse, nonpoint sources that span agriculture, cities, and aquatic environments; the suite of diffuse sources considered is illustrated in Figure 1.(7) We address the following questions:

(1)	**How do the quantity and form of diffuse-P vary across agriculture, cities, and aquatic systems?**
(2)	**What *current practice* diffuse-P management options and technologies are available for these diverse systems?**
(3)	**When is it more appropriate to manage diffuse-P at the source versus at the sink?**
(4)	**When does it make sense to recover the diffuse-P for beneficial reuse?**

2. Metrics for P Management

To quantify and compare diffuse-P across contrasting systems requires applying appropriate standardized metrics and classifications. Here, we apply two critical dimensions—P quantity and P form—and use them to compare the potential for diffuse-P management opportunities.

2.1. P Quantity

An obviously essential metric for P management is quantity. Quantifying the total available pool of P in a system is necessary to inform effective decision-making.(27) Identifying and targeting the largest P flow in a system is the basis for a cost-effective management strategy, one that has a high return on investment.

To quantify P across widely variable systems, we used annual surface loading, i.e., kg P ha^{-1} per year. Table1(28−35) summarizes global ranges of total P loading associated with the major system types noted

in Figure 1(7) (on the right). Global estimates of P quantity demonstrate that high-density megacities can have higher P fluxes than animal- and crop-based agriculture on an areal basis (kg P ha^{-1} yr^{-1}), though collection and sewerage infrastructure vary. However, the agricultural sector occupies large surface areas, making it by far the largest user of phosphate in the form of fertilizer for crop and livestock production and the largest producer of diffuse-P. Inefficiencies and large P losses occur at many stages in the food-production system.[36] Aquatic sediments are the repository of much diffuse-P that emanates from agricultural and urban areas, and internal recycling within waterbodies can also constitute a major P source.[37−39]

Table 1. Global Ranges for Total-P Quantity

system type	description	P quantity (kg ha^{-1} yr^{-1})	data source
agriculture, animal based	**manure P production, livestock-dominated lands worldwide**	5–75	**(28)**
		3–92	**(29–31)**
agriculture, crop based	**fertilizer P application, crop-dominated lands worldwide**	5–40	**(28)**
		3–28	**(32)**
citiesa: Knoxville, TN	**low density, 5 ha^{-1}**	5	**(33)**
citiesa: Dhaka, Bangladesh	**High density, 457 ha^{-1}**	457	**(33)**
aquatic sediments	**freshwater sedimentation flux, lakes and reservoirs worldwide**	2–15	**calculation based on (34,35)**

*aEstimates are based on human P production of 1 kg P produced per person per year.

2.2. P Form

Figure 2 shows one approach for fractionation of total P (TP) into particulate and soluble fractions, which can then be further partitioned into organic and inorganic components. A number of other P fractionation approaches are commonly used around the world; all are operationally defined and cannot identify discrete P compounds.[40] (Additional descriptions of approaches to P fractionation are included in the SI). However, partitioning schemes are important, because the form of P determines whether the P is immediately available to spur photosynthesis, and it also dictates the feasibility of processes for removing or recovering P, as well as corecoverable energy and N.
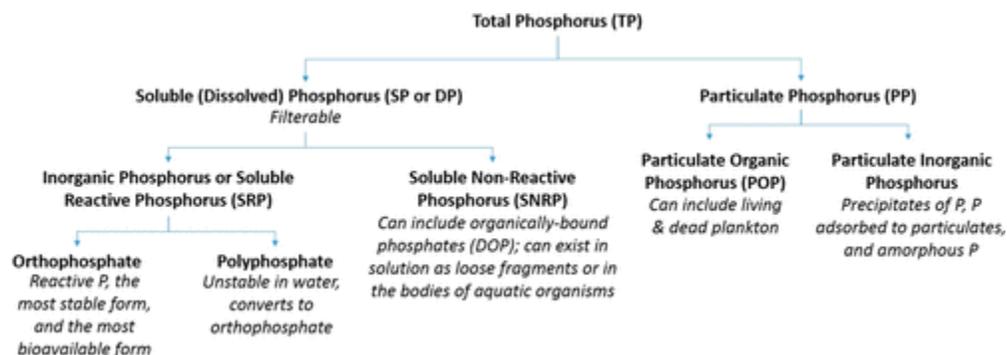
Figure 2. P forms and their characteristics.

Dissolved inorganic P, also known as soluble reactive P (SRP), is the pool most amenable for chemical reactions and biological uptake by plants and microorganisms.[41] Soluble nonreactive P (SNRP) includes dissolved organic P (DOP), which originates from excretion, decomposition, death, or autolysis of biomolecules. It is primarily present in esters, polyphosphates, phosphonates, and nucleic acids.[42,43] DOP is generally less bioavailable

than SRP, but phytoplankton and bacteria are able to uptake DOP to some extent.[44–48] Particulate P (PP) may be present in inorganic forms, for example, mineral phases; adsorbed to biotic or abiotic particles; or as intracellular components (orthophosphate, pyrophosphate, or polyphosphate).[49] PP also may be in the organic form, comprised of P incorporated in living and detrital organic solids.[49]

3. Examples of *Current Practice* Diffuse-P Management at Source and at Sink

In this section, we explore examples of good *current practices* for diffuse-P management, drawing upon case studies that span agriculture, cities, and aquatic sectors. Using our standardized dimensions of P quantity and form, we examine diffuse-P management options currently implemented or being considered for the near future. These *current practice* examples, selected from around the world, highlight instances where different approaches and actions, coupled with robust monitoring data at the source in the catchment watershed or at the aquatic sink, are of particular relevance to achieving water quality management goals.

3.1. Agriculture As a Diffuse Source of P

Because diffuse-P from agriculture is a principal driver of freshwater eutrophication,[50,51] national and regional policies are aimed at the farm source.[18] Policy must be developed and implemented in conjunction with other factors influencing on-farm decision making, such as profitability and practicality. Here, we focus on *current practice* examples of P management at source in crop- and livestock-dominated agriculture. Examples from the Island of Ireland and Albert Lea Lake watershed in south-central Minnesota demonstrate P-use efficiency (PUE) in livestock- and crop-dominated systems, respectively. The New Zealand system illustrates P loss reduction via targeted farm-level mitigation strategies. Livestock-based agriculture dominates global land use, and this is reflected in our case study selection.[23]

3.1.1. Island of Ireland: Diffuse-P Management in a Livestock-Dominated System

Against a backdrop of intensification to increase production, agriculture in Ireland (North and South) faces challenges for managing a *P-legacy* surplus in soil, while improving water quality to meet European Union Water Framework Directive targets. One approach is to maintain soil P at its agronomic optimum, but soil-P levels are generally well above the optimum. The farm-gate P balance for dairy farms in Ireland was 6 kg P ha^{-1} in 2012, a 50% decline from 2006 levels, but still highlighting a net P surplus.[52,53] During the same period, TP imports on-farm declined by 30%; this was primarily driven by a reduction in chemical P fertilizer imports of 50%; TP exports off-farm remained constant, and the PUE improved by 18% even though milk solids output increased.[52,53] In Northern Ireland, the dairy sector's national farm-P balance was 11 kg P ha^{-1} in 2014, down from 18 kg P ha^{-1} in 2003,[54] but still high and representing accumulation. The soil's P surplus stems from postwar application of chemical fertilizer, compounded by the application of organic P-rich livestock manures to land, particularly from intensive dairy systems using concentrated feedstocks to increase milk production.

On-farm P management aims to reduce losses to surface water, while lowering production costs, thus improving efficiency for sustainable and profitable growth. Options to reduce P input at the source include prohibiting P fertilizer application on high-P soils, reducing the use of P-rich feedstock through improved forage utilization, and the redistribution of organic P-rich manures to areas of requirement. In southwest Ireland, a study of dairy-dominated grassland catchments reported an average P balance of 2.4 kg ha^{-1} yr^{-1}, down from 20 kg ha^{-1} yr^{-1}.[55,56] A reduction in imported inorganic P fertilizer, from 24 kg P ha^{-1} to 5.2 kg P ha^{-1}, was identified as the primary cause for declining farm-gate P surpluses.[55,56] Prohibiting organic manure spreading during the winter period also was linked to a decline in P runoff.

3.1.2. New Zealand: Implementation of Targeted on-Farm Diffuse-P Mitigation Strategies

New Zealand agriculture is not subsidized, but has stringent policies to protect water quality. P losses from grazed pasture can range from 0.2–12 kg P ha^{-1} yr^{-1}, averaging 1.2 kg P ha^{-1} yr^{-1}.[57] For a range of cropping

systems, P losses via leaching are estimated at approximately 0.3–0.5 kg P ha^{-1} yr^{-1}.[58] Table 2 [59−103,162] categorizes mitigation strategies based on cost-effectiveness and the form of P being mitigated from across a diverse range of farm enterprises. As shown, the cost to remove or remediate the effects of P generally increases with distance from the source of loss,[104] such that the cost-effectiveness of in-field strategies is greater than those applied at the field boundary or beyond.

Table 2. Summary of Efficacy and Cost of Diffuse-P Mitigation Strategies for Different Farming Enterprises

enterprise type	strategy		main targeted P form(s)	relative effectiveness	relative cost	references
all farming enterprises	stream fencing	in-field management	dissolved and particulate	high	low	(72,76,89)
all farming enterprises	vegetated buffer strips		dissolved and particulate	high	high	(90,91,95,97)
all farming enterprises	precision agriculture		dissolved and particulate	very high	low	(73)
all farming enterprises	low water-soluble P fertilizer		dissolved and particulate	medium	low	(85,88,96)
all farming enterprises	optimum soil test P concentration		dissolved and particulate	low	low	(62,81,164)
all farming enterprises	refurbishing and widening flood irrigation bays		dissolved and particulate	very high	high	(67,98)
all farming enterprises with forage crops	restricted grazing of winter forage crops		dissolved and particulate	high	medium	(78,82,83)
cropping	bunds to prevent runoff from leaving field		dissolved and particulate	very high	high	(61,66)
cropping	contour cultivation		dissolved and particulate	very high	low	(61,66)
cropping	cover crop		dissolved and particulate	medium	high	(61,66)
cropping	minimum tillage		particulate	high	low	(61,66)
cropping	tillage of wheel track to improve infiltration		dissolved and particulate	medium	high	(61,66)
dairy	greater effluent pond storage and deferred irrigation		dissolved and particulate	medium	low	(68)
dairy	low rate effluent application to land		dissolved and particulate	high	low	(69,93)
red deer	alternative wallowing		particulate	very high	medium	(77,79)
red deer	preventing fence-line pacing		particulate	low	high	(84,89,165)
all farming enterprises	sorbents in and near streams	amendment	dissolved and particulate	medium	very high	(74,75)
all farming enterprises	tile drain amendments		dissolved and particulate	very high	medium	(63,87)
all farming enterprises	applying alum to forage cropland		dissolved	medium	high	(78)
all farming enterprises	applying alum to pasture		dissolved	low	very high	(80)
all farming enterprises	red mud (bauxite) to land		dissolved	very high	medium	(99−101,103)

all farming enterprises	constructed wetlands	edge of field	particulate	medium	very high	(64,90, 102)
all farming enterprises	natural seepage wetlands		particulate	low	very high	(71,90, 92,94)
all farming enterprises	sediment traps		particulate	low	very high	(65,70, 86)
all farming enterprises	dams and water recycling		dissolved and particulate	very high	medium	(59,68)
dairy	enhanced pond systems		dissolved	high	very high	(60)

ªRelative effectiveness measured in quartiles.

ᵇRelative cost breakdowns for each quarter were (low, medium, high, and very high): < 35, 36–85, 86–200, and >200 USD $ per kg P retained per year.

As part of a management response to improve water quality, mitigation plans tailor strategies to a particular enterprise (e.g., red deer, dairy, or cropping farm). Mitigations are then implemented to critical source areas (CSAs), which account for the majority of P loss, but comprise small areas of the catchment.(105−107) Diffuse-P mitigations are targeted to CSAs to improve their cost-effectiveness, as opposed to blanket implementation across the entire farm. Research to identify CSAs of diffuse pollution is ongoing in other counties, such as Ireland, as part of efforts to target P mitigation management strategies.(108,109) McDowell (2014)(110) showed that targeting mitigations to CSAs enhanced cost-effectiveness, on average, seven times over untargeted implementation across 14 catchments. For example, the broadcast application of in-field alum amendment to reduce P losses in surface runoff from grazed pastures can cost US $160 to $940 per kg P mitigated.(111) When applied to a CSA in the same field, such as a laneway used for daily traffic to the milking shed, the cost decreased to US $51 to $75 per kg P.(112)

3.1.3. Albert Lea Lake Watershed: Diffuse-P Management in a Crop-Dominated System

The Albert Lea Lake watershed in south-central Minnesota is an example of a high production, crop-dominated system, where 64% of land is cultivated for corn and soybeans.(113) P was added to crops as fertilizer (85% of total) and manure (15%), for a total of 17 kg P ha^{-1}.(114) An agricultural P-balance calculator was developed to enable watershed-scale P balance estimation.(114) PUE was calculated as deliberate outputs expressed as a ratio of deliberate inputs: a PUE of approximately one indicates a balanced system, where exports equal imports. In 2010, PUE in the Albert Lea Lake watershed was 1.7, indicating that deliberate P exports from crops (19.8 kg P ha^{-1} of watershed) exceeded deliberate imports from fertilizer (11.7 kg P ha^{-1} of watershed), suggesting that crops were utilizing soil P stores.(114) Total agricultural stream P load to Albert Lea Lake during the same period was 0.58 kg P ha^{-1}, equivalent to only 5% of deliberate P inputs.

This study highlights the benefits of detailed watershed-level P mass balance and soil-P testing for effective source reduction by clearly identifying where efficiency modifications can be prioritized in an effort to meet water quality targets, without jeopardizing yield. It also emphasizes the need for regular soil-P testing to maintain optimal soil-P fertility as a consequence of nondeliberate losses, such as leaching and erosion, plus the benefits of mixed agriculture via manure application to cropland, which reduces fertilizer requirement and increases profitability. Table2(59−103,162) includes key best management practices (BMPs) for cropping enterprises. Examples such as minimum tillage, cover crops and contour cultivation, have been shown to reduce both soluble and particulate-P losses from crop- dominated watersheds.(73,115) Furthermore, cropping mitigation strategies, like minimum tillage, offer cost-effectiveness in terms of the quantity of P retained.

3.2. Cities As a Diffuse Source of P

Although urban runoff contributes a small portion of TP inputs to major regional watersheds, diffuse sources of urban P can be important contributors within or near cities. Major sources of P to urban landscapes include fertilizer, pet food (which enters landscapes via excrement), atmospheric deposition, and imported compost. In some cities, an additional P input to landscapes comes from septic systems. Unlike agricultural systems, where P is removed by crops and livestock, deliberate export from urban watersheds is small; hence, small P inputs may translate into relatively high runoff P concentrations. We focus on the associated approaches for effectively managing these urban diffuse-P sources in a case study of the Minneapolis-St. Paul region of the U.S., and septic-system drainage in the U.S. and Ireland. Case studies have been selected based on their richness of data, and the Minneapolis-St. Paul region is one of the most intensively studies cities with respect to P budgets.

3.2.1. Landscape Fertilizer

A key input in most cities is P fertilizer applied to vegetated landscapes, such as residential lawns, parks, and golf courses. Soil P correlates with P runoff from lawns across a broad range of soil P concentrations.[116] Recommended fertilization rates are usually based on tested soil-P levels. For example, the University of Minnesota Extension Service recommends zero P application to soils with high soil P, and 22 kg P ha^{-1} for lawn with low-P soils.[117] Bans on lawn fertilizing, as happened in Minnesota in 2003, presumably reduce P application rates to near zero, although P fertilization is allowed if soil test P levels are low. With no new inputs of P, the P stored in soils is gradually "*mined*" and enters lawn runoff through decomposition and release of P in mowed grass or through senescence at the end of the growing season.[118] Limited evidence suggests that the Minnesota P law has reduced P levels in lakes within the Minneapolis-St. Paul region.[119]

3.2.2. Pet Excrement

After lawn fertilizers were banned in Minneapolis-St. Paul, the major input of P to landscapes shifted to pet excrement. For the entire watershed, the input of P from pet food was 1.43 kg ha^{-1} yr^{-1}, of which 0.82 kg P ha^{-1} yr^{-1} entered the landscape either as urine or as feces not picked up by owners, and another 0.61 kg P ha^{-1} yr^{-1} was exported to landfills as waste.[120]

3.2.3. Removing and Recycling of Vegetation P

Source reduction from vegetated landscapes occurs by removal of grass clippings, tree leaves, and vegetative debris, which can be composted and exported. Most lawn-management advice calls for mulching grass clippings (returning clippings to the soil) and mulching light deposition of tree leaves, removing only thicker leaf layers to composting sites. Fissore et al. (2011)[120] reported that 85% of households in the St. Paul region left grass clippings in place, while 15% removed them; 57% removed leaves from all or part of their property, and 43% left leaves in situ. At the watershed scale in the same region, P removal by grass clippings was 0.33 kg ha^{-1} yr^{-1} (19% of watershed output), and P removal by leaves was 0.25 kg ha^{-1} yr^{-1} (14% of watershed output), or 4660 kg P yr^{-1}, equivalent to P excretion from about 5830 residents.[120] The export of such organic residues, with no new inputs of fertilizer P, would eventually lead to a decline in soil P to below levels required to sustain aesthetically pleasing lawns.

Another way to reduce P from vegetation is street cleaning. Tree leaves are an important input of nutrients to streets, and cleaning can remove substantial quantities of nutrients and reduce stormwater P concentrations (reviewed by[121,122]). With canopy levels greater than 30%, the potential input of coarse organic P to streets may approach 50% of total watershed P yield, suggesting that street sweeping could be highly effective at reducing stormwater P. Street cleaning at critical times (e.g., late spring and fall) and locations (high tree canopies) can be highly cost efficient, with costs often less than US $200 per kg P removed,[123] compared with costs greater than US $1,000 per kg for many structural stormwater BMPs.[124]

3.2.4. Urban Stormwater Best Management Practices

Urban stormwater BMPs typically are pond-type structures, where sedimentation is a dominant mechanism of pollutant removal, or infiltration-type structures, wherein stormwater infiltrates through soil or artificial media to remove pollutants by straining, adsorption, and other mechanisms. Periodic removal of pond sediments is required, and sediments are generally disposed to landfills or used for fill. For infiltration basins, most P is removed in the surface layers, where P input per meter of filtered water (i.e., m^3 filtered per m^2 of surface) is approximately 2.5 kg P ha^{-1} yr^{-1} (using 0.25 mg P L^{-1}, as cited by Janke et al. (2014)[125]). For moderate water loads, this would be sufficient to support many crops, which has several benefits: utilizing the trapped P; preventing buildup of soil P, which eventually could lead to soil saturation and subsequent leaching; and maintaining the soil's infiltration capacity.

3.2.5. Septic-System Drainage

In catchments with dispersed populations, domestic wastewater often is treated via septic tank systems.[126] At the household scale, wastewater consists of black water (urine, feces, and flush water) and greywater. It may also include kitchen waste when an in-sink grinder is present. P-load estimates are 1.5 g P $person^{-1}$ d^{-1} for black water, 0.5 g P $person^{-1}$ d^{-1} for greywater, and <0.3 g P $person^{-1}$ d^{-1} for kitchen waste.[127,128] The form of P in black water includes SRP in urine, inorganic PP from feces,[127] and SRP and inorganic PP in greywater, depending upon the products used in the household.[129]

In a study of P losses to aquatic sinks in Ireland (seven regions ranging in size from 846 000 to 7 080 000 ha), 1–3% of emissions emanated from septic systems, with agriculture (8–47%) and wastewater effluent from centralized point sources (8–78%) accounting for the greatest P loads to water bodies.[130] At the catchment scale in rural Ireland, where all study homes were on septic systems, the potential human P load to the environment was estimated at 39 kg P y^{-1}.[131,132] Macintosh et al. (2011)[126] reported P loads from septic systems to be 0.26 kg ha^{-1} yr^{-1}, 0.90 kg ha^{-1} yr^{-1}, and 0.49 kg ha^{-1} yr^{-1} for Tyrone, Armagh, and Monaghan subcatchments, respectively, in 2006. Mechtensimer and Toor (2017)[133] investigated P transport from two conventional septic systems and observed no significant increase in TP concentration in groundwater. The mean TP concentration in the septic tank effluent was 9.8 mg P L^{-1} orthophosphate and 3.3 mg P L^{-1} for other dissolved P compounds, with the orthophosphate concentration 300 cm below the septic tank drain field not statistically different from the background groundwater (0.033 mg P L^{-1}). This was attributed to P precipitation and adsorption in the drain field media and pore water. Human urine accounts for 50% or more of the P load in septic tank effluent, yet only approximately 1% by volume.[127,128] Accordingly, urine diversion offers a novel approach to P recovery, thereby reducing the P load to septic systems.[134,135] Septic systems are a source of diffuse P to the environment, but their impacts vary depending on areal density (i.e., potential human P load), design and operational performance (e.g., favorable conditions to sequester P in the subsurface). They generally are at least 1 order of magnitude lower than P loads arising from agriculture and urban wastewater. Septic systems and their household inputs are considered more confined or "*point-like*" along the spectrum of diffuse P sources.

3.3. Aquatic Systems: Freshwater as a P Sink

We discuss distinct *current practice* examples of P management in aquatic sinks, including (1) the Dixie Drain project in Idaho, U.S., where diffuse-P is being removed from an agricultural return drain at lower cost compared to upgraded point source treatment; (2) the Florida Everglades in the U.S., where cost-effective innovations in P removal to low levels in environmental waters is being investigated; and (3) lakes of the Midwest U.S. and Europe, which demonstrate the significance of internal loading from sediment *legacy* P stores.

3.3.1. Dixie Drain, ID: P Removal from Aquatic Sinks As a Cost-Effective Alternative to Point Source Treatment Upgrades

The city of Boise, Idaho, is constructing the Dixie Drain Facility to remove P from the Dixie Slough, an agricultural return drain downstream of one of the city's wastewater treatment facilities. This plan was enacted to comply with stricter Environmental Protection Agency regulations to reduce wastewater effluent P from 6 mg L^{-1} to 0.07 mg L^{-1}, a 98% reduction. While the city could economically achieve 93–94% P reduction, the increase to 98% was projected to cost millions of U.S. dollars. The Dixie Drain project offsets P contributions to the Boise River from the wastewater treatment facility at a 1.5:1 ratio, that is, for each kg of P the city discharges in excess of the effluent standard, the city will remove 1.5 kg of P at the Dixie Drain Facility. This ratio was determined based on the break-even point where the costs of upgrading the wastewater treatment facility would equal the cost of the Dixie Drain Facility.(136) Thus, treatment at the sink is believed to be more cost-effective (approximately US $1,050 kg^{-1} P removed at the Dixie Drain) than direct treatment at the point source (approximately US $1,580 kg^{-1} P removed at the water renewal facility). (Additional detail is in the SI).

Dixie Drain treatment will consist of an enhanced constructed wetland system including use of presedimentation, a constructed wetland, and aluminum-based coagulation followed by sedimentation. The facility is projected to remove over 2000 kg P yr^{-1} from the Drain (0.0068 kg ha^{-1} yr^{-1} from the watershed), most of which is contributed by agricultural loading, as shown in Figure 3a.(137) This approach to P management—point-diffuse pollution trading, which is the first of its kind in the U.S.—enables the city to maintain regulatory compliance at its sewage treatment plant by treating an unregulated diffuse source.(136) Application to other P-sensitive areas, including the Chesapeake Bay and Mississippi Delta, offers considerable potential.

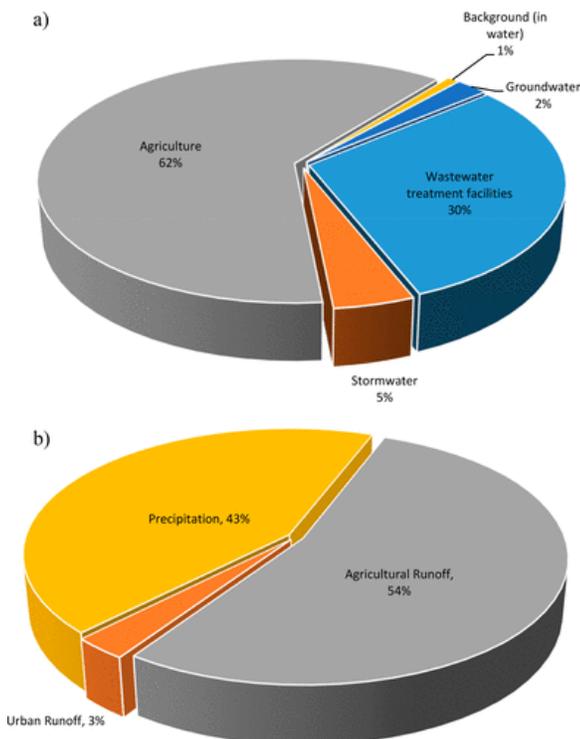Figure 3. Relative contributions to P loading grouped by source for (a) the Boise River near the Dixie Drain Facility(137) and (b) the Southern Everglades tributary areas. Additional details on values are provided in the SI.

3.3.2. Everglades, Florida: Integrated Source and Sink Approaches for Diffuse-P Management

The Florida Everglades is a unique P-limited ecosystem that historically survived on low influxes of nutrients prior to development, 90% of which came from rainfall.(138–140) However, agricultural and urban development

significantly altered historic nutrient inputs in the Everglades, shifting the balance of influent P to the distribution illustrated in Figure 3b.(137)

To counteract the increased runoff and higher P associated with development, P management strategies have been enacted at sources (BMP program targeting primarily agriculture, but also urban stormwater) and in the sink itself (stormwater treatment area wetlands). Figure 4(141) shows inflow, outflow, and interior TP concentrations before and after BMP and stormwater treatment area (STA) implementation. In spite of their combined success reducing P loads, the low targeted concentrations—less than 10 µg L^{-1}—are not consistently achieved. One difficulty is that wetlands are not capable of significantly reducing DOP and PP.(142) As incoming TP loadings dropped, these nonorthophosphate fractions have become increasingly important, as shown in Figure 4.(141) This presents a major obstacle for meeting ultralow P goals.(143,144) Additionally, internal processes can play a considerable role in aquatic systems,(145) as illustrated by the more consistent interior TP levels in spite of more dramatic variation in inflows and outflows.
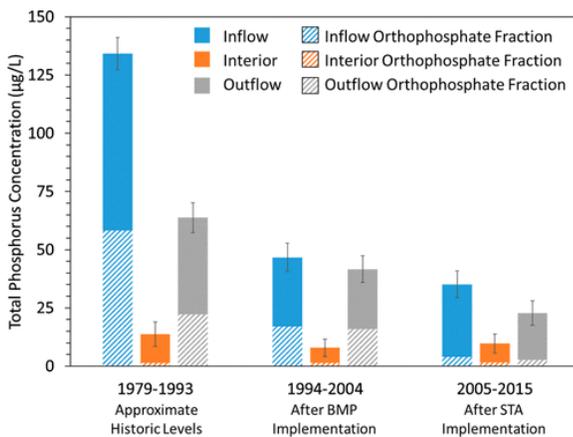

Figure 4. P concentrations in the inflows, outflows, and interior of Water Conservation Area 1 of the Everglades Protection Area. Best management practices (BMPs) were implemented at the farm-scale source, while stormwater treatment areas (STAs) were implemented in the environmental waters. Data are from Davison et al. (2017)(141) and show mean (±1 standard deviation) TP concentrations as well as the mean orthophosphate fraction.

In a unique approach to tackling the ongoing challenge of achieving ultralow P levels in the Everglades, the US $10 million George Barley Water Prize was launched in 2016. This competition intentionally seeks technologies that remove excess P from freshwater (management at the sink), not at the source (e.g., farms), an approach that has historically been considered prohibitively expensive and logistically difficult. It targets cost-effective technologies (≤US $120 kg^{-1} TP removed) capable of removing initially low P to very low levels (≤10 µg L^{-1}) in aquatic systems.(146) This approach recognizes that, (1) considerable amounts of P enter the Everglades from nonregulated or difficult-to-manage diffuse sources, (2) organic P, which is not effectively removed, plays a significant role in TP loads, and (3) *legacy P* stored in soils and sediments can continue to leach and cause negative water quality impacts for years to come.

3.3.3. Lakes of Midwest U.S. and Europe: Eutrophication and Internal Loading

Eutrophication is a widespread issue in the upper Midwest region of the U.S., which is comprised of lake-rich landscapes, along with intense crop and range lands. Over many decades, considerable quantities of P have accumulated in lake sediments, often with P concentrations in excess of 1000 mg P kg^{-1} sediment. Once P has accumulated in lake sediments, internal P loading can cause a eutrophic state to persist for decades or longer.(147–150) The relative contribution of internal P to TP loading varies widely among lakes, but can often exceed external loads during individual years or seasons.(37,38)

Figure 5(37,38,151–154) documents that the internal load can dominate or be minimal, depending on lake morphometry, hydrology, and catchment practices. For example, by far the greatest P load was from internal sediment for Pond Dongen and Lake De Kuil in The Netherlands; by comparison, the internal load was roughly equal to the external load of Lake Mendota in the U.S. Midwest. The importance of internal loading can vary with interannual differences in rainfall, runoff, and lake mixing dynamics. Soranno et al. (1997)(37) found that, during a wet summer, seasonal internal loading was similar to external loading, whereas during a dry summer, internal loading was considerably larger than external. In watersheds that contain lakes with P-rich sediments, strictly source-based management strategies provide poor returns in terms of water quality. Consequently, excavation of P-rich aquatic sediments has been demonstrated in the Lake Mendota watershed,(155) and efforts are underway to up-scale such projects.(156) In the Fox River and Green Bay, Wisconsin, industrial pollution has led to dredging to remove contaminated sediments,(157) with uncertain effects on sediment P pools. Internal lake-P levels also can be reduced by additions of lanthanum-modified clay, alum, metal-salt coagulants or mineral adsorbents,(39) although this transfers the P to the sediment pool.
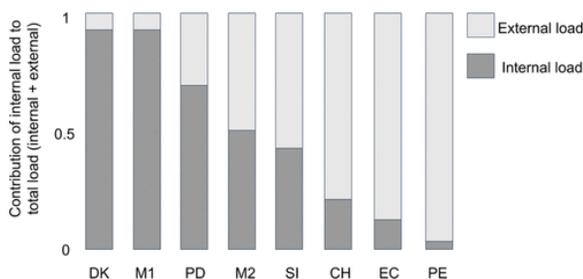

Figure 5. Examples of the relative contribution of internal P loading to previously studied lakes. DK = Lake De Kuil,(151) M1 = Lake Mendota 1992,(37) PD = Pond Dongen,(151) M2 = Lake Mendota 1993,(151) SI = Lake Simcoe,(38) CH = Lake Champlain-Missisquoi Bay,(152) EC = Lake Erie-central basin,(153) and PE = Pond Eindhoven.(151) Internal and external loading rates are annual gross rates, except in M1 and M2, which are for the summer period. Internal loading rates in SI, CH, and EC are from incubations (excludes sedimentation) reported in Orihel et al. (2017).(38) Internal loading rates for DK, PD, and PE are from the PClake ecosystem model(92) and reported in Lürling et al. (2016).(151) Internal loading rates in M1 and M2 were estimated by calculating P transported into the epilimnion after the thermocline deepened following storms.(37)

4. The Influence of Diffuse-P Form on Removal and Recovery Strategies

Our case studies illustrate some of the differences across systems and management approaches. Now, we focus on how this information can be synthesized to more broadly inform decision-making, for example, how does P form influence management strategy? Figure 6 illustrates how P-containing diffuse sources and sinks from the case studies predominantly align within the classification depicted in Figure 2. The figure's four quadrants define the spectra from particulate to soluble (horizontal axis) and organic versus inorganic (vertical axis). One important observation in Figure 6 is that the quadrant for soluble organic P is unoccupied, although lower magnitude sources, for example, some industrial wastes or organophosphorus pesticides, may contribute here. However, the other three quadrants are well populated.
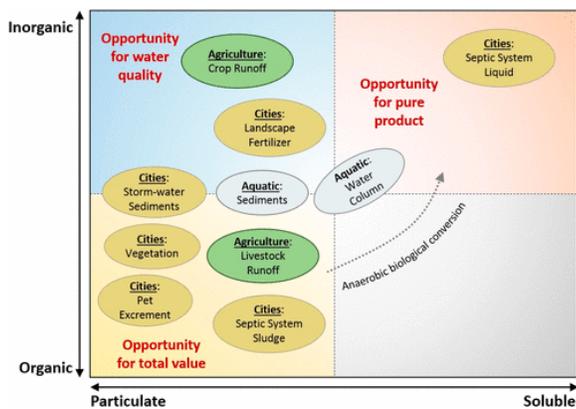
Figure 6. P-form matrix mapped to prominent sources and sinks, illustrating key opportunities in terms of diffuse-P management. Note that bubble size does not correlate to magnitude of P flow.

The upper-left quadrant features inorganic P associated with solids, and it is dominated by crop-land runoff and the waters that receive the runoff. Management in this quadrant provides the most direct opportunity to improve water quality by addressing the aquatic systems (sinks) themselves. The lower-left quadrant (particulate and organic P) is dominated by solids from waste residues, and it also contains P found in urban stormwater sediment. The high organic content of these P sources offers considerable opportunity for total value recovery, for example, P can be removed and possibly recovered along with other valuable products, particularly energy and N.(24) The upper-right quadrant (soluble and inorganic P) includes P that has been solubilized due to some form of biological treatment of the sources in the lower-left quadrant. The soluble inorganic P is the P most readily available for P recovery as a pure P product for reuse scenarios. The P in aquatic sediments spans the boundaries because it naturally undergoes processes that lead to P release from particulate forms, for example, hydrolysis and dissolution.

PP (organic and inorganic) (left side of Figure 6) can be physically separated from the water via settling or filtration, which can be enhanced by flocculation and additional precipitation using alum coagulant. When carried out in situ, physical separation normally is by sedimentation, which typically sequesters the PP into the sediments. Since sedimentation concentrates P, removal of the sediments by dredging theoretically offers an avenue for P recovery and reuse in agricultural applications. However, physical separation of particles is not selective for P removal, and many other contaminants are present in the settled organic and inorganic solids (e.g., metals and hydrophobic organic micropollutants). Thus, the value of the solids themselves for direct agricultural use or for further treatment to release P, via biological digestion or chemical oxidation, depends on the composition of the solids. Additionally, the P content of these solids may be low, imposing economic and technical constraints for recovery.

If water containing PP is intercepted before reaching surface waters or is extracted from surface water, it can undergo ex situ treatment that intensifies the in situ separation mechanisms. In particular, the water can be filtered after flocculation to provide a higher degree of P separation and concentration. Especially for intercepted water, ex situ processing may produce a solid phase that has a higher P content and has fewer of the potentially problematic materials that lower its value as a recycled P source for agriculture.

When SRP is present in waters without significant organic matter (e.g., the upper right quadrant of Figure 6), the inorganic P can be concentrated using phosphate-selective ion exchangers or other P-selective adsorbents.(7) Such adsorbents offer great potential for subsequent recovery of P in outputs suitable for agricultural reuse, as their selectivity facilitates recovery of P-rich, contaminant-free products. Direct uptake of bioavailable SRP by microalgae is another feasible P-removal strategy. Algae sequester soluble P at levels of up to 1–2% P on a dry biomass basis.(158–160)

For effective removal or recovery of nonreactive DOP, it must first be converted to a reactive form, e.g., using microbiological activity to hydrolyze complex organic molecules and release the P as SRP.(161) Following conversion, the P can be readily extracted from wastes using the approaches suitable for SRP.(8,144)

Certain bacteria are capable of *luxury P uptake* as polyphosphate, to levels of 3–8% P on a dry biomass basis or even higher.(158,159,162) Agricultural and city waste residues, which contain significant biodegradable organic matter, may be amenable to enhanced biological P uptake if the organic matter is biodegraded aerobically. Once concentrated in the biomass, P may be further recovered for reuse applications by harvesting and applying the biomass directly to crops. However, aerobic treatment of high-organic streams is energy intensive and costly; anaerobic treatment (the dashed arrow in Figure 6) is more logical as a means to recover value from these streams.(8)

5. Perspectives and outlook

P quantity and form vary widely across agriculture, cities, and aquatic systems. This variability profoundly influences the applicability of P management options in such settings. Figure 7 shows a range of options for diffuse-P management. Generally, the most cost-effective and practically implementable options are located at the top of the inverted pyramid: reduce P use, conserve P, and mitigate P loss at the source. P quantity is typically highest and most concentrated at the source, becoming more dilute downstream, which means mitigating P losses is most cost-effective at the source of loss. Despite the inherent challenges of controlling diffuse P, progress has been achieved, particularly at the farm scale, where great strides have been made via judicious nutrient management and BMPs, such as the 4-R Nutrient Stewardship Strategy in the U.S. (Right Rate, Right Time, Right Place, and Right Form)(163) and the wider-reaching 5R approach in Europe (Realign P inputs; Reduce P loss to water; Recycle P; Recover P in wastes; and Redefine P in food systems).(10) Efficient P use in agricultural settings most directly avoids excess inputs of diffuse-P into the environment, and the implementation of P mitigation strategies, specifically targeted to CSAs, further reduces losses while minimizing costs.
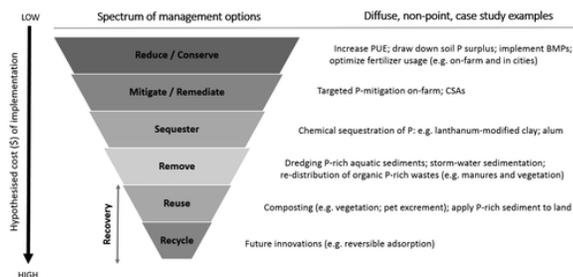

Figure 7. Tiered system of options for diffuse, nonpoint P management.

Sequestering and removing P from diffuse sinks are the next levels of management, usually at an increased cost. Such methods are widely applicable, as in the case of chemical sequestration and dredging in lakes, but technologically and economically challenging in terms of P recovery, due to the presence of other contaminants (e.g., heavy metals). However, as exemplified in the case studies, management at the sink (in combination with the mitigation of losses at the source) is an indispensable aspect of an integrated P management strategy, especially where water quality must be improved rapidly. For example, internal P loading accounts for the majority of P in Lake De Kuil, Lake Mendota, and Pond Dongen (Figure 5(37,38,151–154)), thus necessitating in-sink P management strategies to yield substantial improvements in water quality in relatively short timeframes. It also is possible that, in some situations, the challenges of retrofitting existing infrastructure to mitigate or remove P at the source may exceed the costs of downstream P removal at the sink. Accordingly, pollution trading among point and diffuse sources may play an increasingly important role in more widely distributing the equity of P management across sources (i.e., Dixie Drain).

At the bottom of the pyramid are methods to reuse and recover P that has been removed from the diffuse sources or sinks. Recovery can be technically and economically feasible in specialized cases, particularly when the P is in a concentrated form, as in animal wastes. Nevertheless, when dispersed and integrated with contaminants, as is the case with diffuse-P, recovery may be prohibitively expensive using currently available technologies. Furthermore, while the current price of phosphate rock generally dis-incentivizes P recovery, broadening the singular focus on P removal to more fully account for the total value of P recovery is needed as part of a wider circular economy.

In summary, several diffuse-P management strategies currently are available, and metrics such as P quantity, form, and cost to mitigate or remove P dictate when different strategies are more or less well suited to differing scenarios within agriculture, urban, and aquatic settings. This highlights the need for P mass balance and flow analyses using standardized metrics for comparison at the catchment scale. This kind of research is critical to inform P-policy decision-making based on realistic targets and objectives, as well as informing society about who pays. Furthermore, improved P-removal and -recovery technologies, data capture, and monitoring are necessary to refine the cost-effectiveness of management approaches implemented. In general, the effective management of diffuse-P at its source, particularly at the farm-scale, makes strong economic and environmental sense, because the concentration of P is generally at its highest, making potential for cost-effectiveness greatest. Diffuse-P management is more costly at the aquatic sink; however, it may be needed if source-based management alone is not sufficient to meet water-quality goals and rapid time scales for improvement.

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Acknowledgments

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Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b01143.

- Additional details on variability in diffuse, nonpoint sources and P form in terms of approaches to fractionation. Calculations, and embedded assumptions, performed as part of the assessment of the Dixie Drain and Everglades case studies are also included. Additional examples of diffuse-P management strategies are listed in Table S2 (PDF)

  ○ Pdf es8b01143_si_001.pdf (380.13 kb)

Managing Diffuse Phosphorus at the Source versus at the Sink