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Journal of Environmental Quality SPECIAL SECTION

EMERGING TECHNOLOGIES TO REMOVE NONPOINT P SOURCES FROM SURFACE WATER AND GROUNDWATER

Emerging Technologies for Removing Nonpoint Phosphorus from Surface Water and Groundwater: Introduction

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Coastal and freshwater eutrophication continues to accelerate at sites around the world despite intense efforts to control agricultural P loss using traditional conservation and nutrient management strategies. To achieve required reductions in nonpoint P over the next decade, new tools will be needed to address P transfers from soils and applied P sources. Innovative remediation practices are being developed to remove nonpoint P sources from surface water and groundwater using P sorbing materials (PSMs) derived from natural, synthetic, and industrial sources. A wide array of technologies has been conceived, ranging from amendments that immobilize P in soils and manures to filters that remove P from agricultural drainage waters. This collection of papers summarizes theoretical modeling, laboratory, field, and economic assessments of P removal technologies. Modeling and laboratory studies demonstrate the importance of evaluating P removal technologies under controlled conditions before field deployment, and field studies highlight several challenges to P removal that may be unanticipated in the laboratory, including limited P retention by filters during storms, as well as clogging of filters due to sedimentation. Despite the potential of P removal technologies to improve water quality, gaps in our knowledge remain, and additional studies are needed to characterize the long-term performance of these technologies, as well as to more fully understand their costs and benefits in the context of whole-farm- and watershed-scale P management.

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The accelerated eutrophication of freshwater and coastal ecosystems in many regions of the world is largely driven by nonpoint source phosphorus (P) pollution from agriculture (Carpenter, 2008). Despite the widespread implementation of conservation and nutrient management strategies to control agricultural P losses, a lack of water quality improvement in estuaries such as the Chesapeake Bay in the United States (Sharpley et al., 2011) and the Baltic Sea in Europe (Conley et al., 2009) has prompted a reexamination of potential approaches to mitigate eutrophication. Indeed, the European Union's (EU) Water Framework Directive (European Commission, 2000) and the U.S. Environmental Protection Agency's total maximum daily load for the Chesapeake Bay (Executive Order 13508; Federal Leadership Committee for the Chesapeake Bay, 2009) will require significant curtailments in nonpoint P pollution from agricultural sources over the next decade. To meet these strict water quality goals, new tools will be needed that address acute P sources, such as incidental transfers of applied P from fertilizer and manure to runoff waters (Preedy et al., 2001), as well as chronic P sources such as dissolved P transfers from high P (i.e., legacy P) soils to surface water (Kleinman et al., 2007) and groundwater (Koopmans et al., 2007).

An emerging set of technologies is being developed to reduce nonpoint P pollution using reactive elements as low-cost P sorbing materials (PSMs). A number of substrates have been evaluated, including natural materials, synthetic filtration products, and by-products from industrial activities (see Table 1 and recent reviews by Douglas et al., 2004; Westholm, 2006; Ballantine and Tanner, 2010; Vohla et al., 2011). Phosphorus sorbing materials provide a metal cation (Table 1) to react with dissolved phosphorus to create an insoluble compound by sorption processes, including adsorption or precipitation or both. Because these processes are finite, one of the main considerations in choosing a PSM to treat nonpoint P pollution is P-sorption capacity

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Abbreviations: CD-MUSIC, Charge Distribution- MUlti SIte Complexation; DOC, dissolved organic carbon; EU, European Union; FGD, flue gas desulfurization; NOM, natural organic matter; PSM, phosphorus sorbing material; TDP, total dissolved phosphorus.

(Drizo et al., 2002; Westholm, 2006), which varies depending on solution pH, ionic composition, and the concentration of natural organic matter (NOM) in the influent water (Weng et al., 2008; Miller et al., 2011). Other characteristics of an ideal PSM include low cost, high availability, low toxicity to soil and water resources, and potential for reuse as a soil amendment once it has been fully saturated with P (Leader et al., 2008; Ballantine and Tanner, 2010). Although the release of toxic metals tends to be a potential concern when using industrial by-products as PSMs (Fenton et al., 2009), they are often the substrate of choice due to their widespread availability and low cost compared with natural materials and synthetic filtration products (McDowell et al., 2008).

A wide array of P sorbing technologies has been conceived to date, ranging from preventive measures that focus on P immobilization in soils and manures (Fig. 1a) to remedial measures that utilize filters to remove dissolved P from runoff waters (Fig. 1b,c). For instance, early work on P immobilization by Moore and Miller (1994) showed that amending poultry litter with alum could reduce water soluble P by up to 60% compared with unamended controls. A number of subsequent studies also demonstrated that various PSMs, including alum, water treatment residuals, and coal-fired combustion by-products, were effective at reducing P solubility in animal manures (Smith et al., 2001; Dou et al., 2003), biosolids (Elliott et al., 2002), and high P soils (Peters and Basta, 1996; Callahan et al., 2002; Novak and Watts, 2005; Agyin-Birikorang et al., 2007). Research has also revealed that dissolved P in surface runoff is reduced substantially by amending high P soils with PSMs (Stout et al., 2000), as well as by blending manures with PSMs before land application (Shreve et al., 1995; Smith et al., 2001; Buda et al., 2010; Brennan et al., 2011), although there may be some limitations to these practices in calcareous soils (Leytem and Bjorneberg, 2009). While the majority of P immobilization studies have been conducted on pasture and cropland soils, recent work by Penn and Bryant (2006) also documented the potential for PSMs to control dissolved P losses from barnyards and cattle loafing areas.

In contrast to preventive measures, which control P loss at the source, remedial technologies seek to address dissolved P after it has been mobilized in agricultural drainage waters. In general, two main strategies are typically used. The first strategy involves using PSMs in filters to treat concentrated agricultural drainage water from a variety of nonpoint P sources, including barnyard runoff (Weber et al., 2007), golf course runoff (Agrawal et al., 2011; Penn et al., 2012), and tile drain outlets (McDowell et al., 2008). In addition, others have trialed filtration systems in small streams (McDowell et al., 2007) and agricultural drainage ditches (Penn et al., 2007; Bryant et al., 2012) (Fig. 1c). A common finding in many of these studies is that P removal efficiency tends to be high (>60%) at low flow rates but decreases dramatically (<25%) at much higher flow rates seen during storm events (McDowell et al., 2007; Bryant et al., 2012; Penn et al., 2012). Essentially, high flows can overwhelm the saturated hydraulic conductivity of the filter material, resulting in decreased retention times and reduced treatment efficiencies. In addition, a portion of the flow may actually bypass the filter altogether during large storms, which is a serious drawback given that these events can be responsible for the majority of P loss on an annual basis (Pionke et al., 2000). As a result, the application of filtration technologies for concentrated flows may be limited to treating lower flow rates or treating flows that can be adequately regulated before entering the filter system.

Alternative strategies have been sought that focus on filtering P from diffuse flows, which exhibit less variability than concentrated surface flows. One potential technique is the use of permeable reactive barriers to remove P from shallow lateral groundwater flows before their discharge to surface water (Fig. 1b). A key consideration in the design of permeable reactive barriers is to match the saturated hydraulic conductivity of the filter material with that of the surrounding soil, which ensures the hydrological integrity of the system remains unchanged (Miller et al., 2011; Chardon et al., 2012). Not many field trials of permeable reactive barriers to remove dissolved P from groundwater have been performed, but some studies have shown promising results for treating wastewater effluent from septic drain fields using silica sand and crushed limestone (Baker et al., 1997; 1998), as well as for treating nonpoint P leached from high P soils using iron oxides (Kronvang et al., 2005). Borrowing from

Table 1. Types of P sorbing materials used to remove P sources from water, with main reactive compounds and examples of application.

Material	Compounds	Example applications	References†
Natural materials			
Minerals (e.g., iron oxide, shale, limestone)	Fe, Al, Ca	Filter for wastewater, agricultural runoff	1, 2
Polymineralic soils or sands	Fe, Al, Ca	Constructed wetlands	2
Synthetic filtration products			
Expanded calcinated clay aggregates	Ca	Constructed wetlands	2
Lanthanum modified clay (Phoslock)	La	Trap P in sediments	3
Synthetic analogs of natural minerals (e.g., zeolite)	Al	Wastewater treatment	4
By-products from industrial activities			
Blast furnace steel slag	Ca, Al	Golf course runoff	5
Red mud	Fe, Al, Ca	Septic tank effluent	6
Fly ash	Al, Fe	Constructed wetlands	2
Melter slag + basic slag	Ca	Drainage backfill, filter sock	7
Flue gas desulfurization gypsum	Ca	Agricultural drainage water	8
Drinking water treatment residuals	Fe, Al	Immobilizing P in soils	9
Oxygenation products of anaerobic groundwater	Fe	Tile drain envelope	10

† 1. Dobbie et al. (2009); 2. Drizo et al. (1999); 3. Robb et al. (2003); 4. Dao (2003); 5. Penn et al. (2012); 6. Cheung and Venkitachalam (2006); 7. McDowell et al. (2008); 8. Bryant et al. (2012); 9. Elliott et al. (2002); 10. Chardon et al. (2012).

the permeable reactive barrier concept and work by McDowell et al. (2008), a new study by Chardon et al. (2012) highlights the potential of using iron oxides as an envelope for tile drains, which would remove dissolved P from shallow groundwater before it enters the drain (Fig. 1). In addition to targeting diffuse groundwater flow pathways, recent studies have shown that burnt lime and spent lime filters can be placed in the riparian zones of small streams to successfully remove dissolved P from overland flows (Kirkkala et al., 2012).

Presently, the majority of studies on preventive and remedial technologies to remove nonpoint P from agricultural drainage waters have been conducted in the laboratory, with slightly fewer field-scale examples. Laboratory studies represent a logical first step in determining the suitability of PSMs for field-scale experimentation, which explains their prevalence in the literature. Although laboratory studies are important from a developmental standpoint, their results may be difficult to compare with long-term field studies (Westholm, 2006) and may not effectively simulate the varying climatic and hydrologic conditions observed in a field situation. Some evidence indicates that theoretical P sorption models (e.g., Charge Distribution–MUlti SIte Complexation [CD-MUSIC] model) may soon bridge the gap between laboratory data and results obtained from field experimentation (Weng et al., 2012), but these studies are still preliminary in nature. As a result, field-scale trials are rapidly increasing in importance (e.g., Weber et al., 2007; McDowell et al., 2007; 2008; Bird and Drizo, 2010) as the demand for information on the long-term performance of PSMs under field conditions increases (e.g., Shilton et al., 2006; Turtola et al., 2010; Kirkkala et al., 2012). Indeed, a number of new research initiatives across the EU are now focusing on widespread implementation and testing of P removal technologies in field situations (Turtola et al., 2010). Despite the increased focus on field-scale research and long-term monitoring, little information is available on how emerging P removal technologies might fit into the larger P management scheme at farm and watershed scales, particularly in terms of evaluating their economic and environmental benefits against existing and well-established P management strategies.

This special collection of papers presents the latest research being conducted on removing nonpoint sources of P from surface water and groundwater. It is a product of a special symposium that was sponsored by the American Society of Agronomy's A-05 division (Environmental Quality), and held at the ASA, CSSA, and SSSA 2010 International Annual Meetings in Long Beach, CA, on 3 Nov. 2010. The collection highlights several important aspects of P removal technologies, including the theoretical principles of P sorption, insight gained from laboratory and field-scale studies of P removal potential, and consideration of the environmental and economic benefits of these technologies in whole-farm systems.

Emerging Phosphorus Removal Technologies Paper Summaries

Modeling to Understand the Principles of Phosphorus Sorption

Understanding the principles of P sorption represents a critical first step in the development of P removal technologies.


a. Preventive strategy for high P soils

b. Remedial strategies to address diffuse P flow pathways

c. Remedial strategy to address concentrated P flow pathways

Fig. 1. Examples of different P sorbing technologies for addressing nonpoint P sources from agriculture, including (a) a preventive strategy whereby flue gas desulfurization (FGD) gypsum is applied to high P soils to immobilize water-soluble P and prevent P loss in surface runoff (photo credit: Ray Bryant); (b) two remedial strategies to remove dissolved P from soil water and shallow groundwater flow paths using a permeable reactive barrier filled with iron-coated sand (left) and enveloping a tile drain with iron-coated sand (middle and right) (photo credits: Wim Chardon and Gerwin Koopmans; see Chardon et al., 2012); and (c) a remedial strategy to remove dissolved P from concentrated surface flows in an agricultural drainage ditch using a FGD gypsum filter (photo credits: Ray Bryant; see Bryant et al., 2012).

Weng et al. (2012) present a summary of the key factors that influence P sorption by iron oxides, a commonly used material to remove P from surface water and groundwater (Chardon et al., 2012). The authors used the CD-MUSIC model (Hiemstra and van Riemsdijk, 2006) to simulate P adsorption to goethite and compared the results to a series of batch adsorption experiments. The CD-MUSIC model is a surface complexation model that describes the adsorption of charged compounds such as P on charged surfaces of metal oxides. Results showed that under typical soil conditions encountered in the field, solution pH, Ca concentration, and the presence of NOM were the most important factors controlling the adsorption of P to iron oxides such as goethite. In particular, knowledge of NOM levels in solution were especially critical to predicting long-term performance of the filter material because NOM actively competes with P for adsorption sites. Weng et al. (2012) demonstrated the importance of the NOM effect by increasing dissolved organic carbon (DOC) from 0.5 to 50 mg L^{-1}, holding all other constituents constant. The increase in DOC resulted in a 50% decrease in P adsorption to goethite. This study demonstrates the utility of applying theoretical P sorption models to predict the potential for P sorbing materials to remove P from agricultural drainage waters.

Batch and Flow-Through Experiments to Characterize Phosphorus Sorption Properties

Batch and column studies are common tools used to characterize the P sorption and hydraulic properties of reactive materials under controlled conditions, as exemplified by three studies in this collection. Chardon et al. (2012) conducted a laboratory study using iron sludge and iron-coated sand to evaluate and compare their potential to remove dissolved P from soil water discharged by tile drains in The Netherlands. Iron sludge and iron-coated sand are by-products of the process of purifying anaerobic groundwater, which typically contains dissolved iron in the reduced form (Fe^{2+}) that must be removed to prevent colored or bad-tasting drinking water. After the oxidation of Fe^{2+} in the raw water, amorphous $Fe(OH)_3$ forms as a sludge or as a coating on filter sand. The iron in the sludge and coated sand is of natural origin and is not added during raw water treatment. Iron sludge and iron-coated sand contained low amounts of As and heavy metals that had a low potential for release to a dilute $CaCl_2$ extract solution. A series of batch and column studies confirmed the importance of kinetics for P sorption and saturated hydraulic conductivity as key factors dictating the P removal potential of the two iron-based materials in the field. Results showed that iron-coated sand had much higher P sorptive capacity, stability of the iron during leaching, and saturated hydraulic conductivity than iron sludge, making it the most promising material for field-scale testing.

Egemose et al. (2012) also used a sequence of batch and column experiments to assess the P retention capacity of crushed concrete, a relatively inexpensive industrial by-product derived from the demolition of construction sites. Five different types of crushed concrete were tested, including three pure concrete types and two that were derived from a commercial crushing company. The most abundant element in crushed concrete was Ca (24–55% by dry weight) followed by minor amounts of Fe and Al (1.8–2.4%), and even lower heavy metal (Cu, Pb, Cr, and Cd) contents (<0.01%). In general, P retention by the five types of crushed concrete ranged from 40 to nearly 100%, with higher retention capacities related more to physical properties of the concrete (e.g., structure, hydraulic conductivity, and contact time) than to Ca content. Equally important, P release remained low throughout the experiment for all five concrete types, suggesting that the P removed from solution remained strongly fixed even after the sorption capacity of the crushed concrete was exhausted. All five types of crushed concrete had high amounts of $CaCO_3$ and $Ca(OH)_2$, which led to effluent pH values that ranged from 9.8 to 12.2. Egemose et al. (2012) conclude that crushed concrete holds promise as a P filtration technology in most agricultural and urban settings. However, they also caution that elevated pH and alkalinity levels in the effluent, a concern for receiving waters, may ultimately limit its widespread application.

The study by Stoner et al. (2012) highlights the potential of flow-through experiments to provide additional insight into the P removal potential of reactive materials. Using a flow-through system approach, Stoner et al. (2012) tested 12 different industrial by-products, including acid mine drainage residuals, steel slag, drinking water treatment residuals, and flue gas desulfurization (FGD) gypsum. For all 12 materials, they constructed P removal curves, or "design curves," describing P removal from solution by a PSM as a function of the amount of P added to this material in a flow-through situation. By varying the retention time between 0.5 and 10 min and influent P concentration between 0.5 and 15 mg L^{-1}, the authors were able to demonstrate that the impact of these parameters on P removal depended on the chemical properties of the material, mainly Al and Fe oxides and water-soluble Ca contents. Screening for these properties allowed for comparisons between PSMs based on their potential to remove dissolved P from agricultural waste water in the field. The most promising PSMs tested by Stoner et al. (2012) were acid mine drainage residuals and steel slag. Ultimately, this approach represents a useful new tool for predicting the P sorption potential and life expectancy of potential P removal technologies.

Field-Scale Trials to Document Performance of Phosphorus Removal Technologies

Although numerous batch and column studies have been conducted to estimate the potential for reactive materials to remove P from water, slightly fewer field-scale trials have been reported in the literature. In a study conducted on the University of Maryland Eastern Shore Research and Teaching Farm (USA), Bryant et al. (2012) constructed a filter within a field ditch to remediate P-laden runoff water from a 17-ha catchment with high P soils. The filter contained FGD gypsum, a by-product of coal-fired power generation. More than 3 yr of monitoring showed that for storm-induced flow, the filter removed about 65% of the total dissolved P (TDP) load for water passing through the filter. However, this efficiency decreased to 22% when bypass flow and baseflow were taken into account. The authors conclude that FGD gypsum was chemically effective at removing P, but that the setup tested is unlikely to be practical at the whole-farm scale due to low P removal efficiencies, particularly during storms when large P loads mostly bypassed the filter. In addition, long-term field testing of the ditch filter demonstrated a need to regularly till the surface of the gypsum filter bed to maintain satisfactory flow rates for adequate P removal. The study by Bryant et al. (2012) highlights the potential for field studies to elucidate maintenance requirements for P removal technologies that would have been difficult to foresee in a laboratory setting. Future testing will focus on using FGD gypsum in a permeable reactive barrier to remove P from groundwater, which would have lower flow rates conducive to more efficient P removal.

In another field study, Penn et al. (2012) report on the development, installation, and monitoring of a P removal structure to treat golf course runoff from a 320-ha catchment near Stillwater, OK (USA). The P removal structure was filled with steel slag, a by-product of the steel industry, and was located in a drainage ditch at the outlet of the catchment. Penn et al. (2012) monitored a total of 54 runoff events over a period of 5 mo in 2010, including 20 resulting from natural rainfall and 34 resulting from irrigation of the golf greens located 130 to 150 m upgradient of the P removal structure. During that period, the structure removed about 25% of the TDP load that was delivered to it. Indeed, P removal efficiencies were higher for irrigation runoff than for natural runoff (62 vs. 21%), owing to lower retention times during large flow events produced by heavy rainfall. On the basis of a set of flow-through equations developed by Penn and McGrath (2011) for steel slag, a lifetime of the P removal structure of 16.8 mo was predicted, which corresponded reasonably well with the projected lifetime of 15.4 mo based on field measurements. The study by Penn et al. (2012) demonstrates the value of using predictive modeling to determine the life expectancy of P removal structures in field settings, as well as for providing information that can be used to make adjustments such as expanding the treatment capabilities to accommodate large runoff events.

Economic and Environmental Assessment of Phosphorus Sorption Strategies

The P removal technologies reported in the literature are often assessed in isolation, with limited consideration of their costs and benefits in comparison to other P management approaches for reducing P losses that may be utilized at whole-farm and watershed scales. McDowell and Nash (2012) provide a thorough review of the cost-effectiveness and suitability of various P mitigation strategies used to prevent P losses from dairy farms in New Zealand and Australia. They grouped strategies into three different categories, including management (e.g., decreasing soil test P, fencing streams), amendments (e.g., alum, red mud), and edge-of-field mitigations (e.g., natural or constructed wetlands, P filtration/removal technologies). Using the Best Management Practice (BMP) Toolbox (Monaghan, 2009) and additional financial analyses, McDowell and Nash (2012) demonstrate the cost-effectiveness of these strategies in terms of cost, expressed as U.S. dollars (USD) per kilogram of P conserved. In general, management practices such as reducing soil test P and stream bank fencing were seen as the most cost-effective (0 to 200 USD kg^{-1}), followed by amendments (20 to >400 USD kg^{-1}) and edge-of-field mitigations (20 to >400 USD kg^{-1}). Notably, P removal technologies, either when implemented as amendments to soils or as filters for tile drain and stream water discharge, were the least cost-effective, ranging from 110 to >400 USD kg^{-1}. One of the review's key takeaway messages is that as scale and flow-path complexity increase, it becomes progressively more difficult and less cost-effective to treat P loss from a site. Therefore, P source management is often the best approach initially, and P removal technologies should be reserved for situations where they may provide additive P mitigation benefits when used in combination with other strategies. Overall, the review by McDowell and Nash (2012) illustrates the importance of nesting and targeting P management strategies to minimize P loss and maximize on-farm profitability.

Gaps in Our Knowledge and Future Research Direction

The papers in this special collection provide insight into the development, testing, and potential application of technologies to remove P from surface and groundwater. Batch and column experiments represent a critical first step in the design of P removal technologies, and these studies are the basis for screening prospective PSMs to determine their potential to remove P from agricultural drainage waters under a controlled set of conditions. There will be a continued need for laboratory studies to provide information on new PSMs as they emerge, as well as to study the effectiveness of physically and chemically modifying existing, well-characterized materials to match specific field conditions. These studies will also provide important empirical data to support the development and validation of theoretical P sorption models, which can then be used to predict the effects of variable influent chemistry on P removal efficiency in the field.

There is also a significant need for more long-term testing and monitoring of P removal technologies in field situations. Field trials highlight management requirements that may not be discovered or anticipated in the laboratory, such as the formation of preferential flow pathways in P removal structures and filters, clogging due to surface sealing and sedimentation, and the effects of animal activity and vegetation growth, to name a few. Developing protocols to address these management situations will be essential to ensure that P removal technologies maintain optimum system efficiency. In addition, field monitoring data will be vital for documenting the ability of P removal technologies to improve groundwater and surface water quality over time, as well as for supporting the development of predictive models to understand the impact of P removal technologies at field, farm, and watershed scales.

Finally, there is an important need for information on how best to target P removal technologies on farms to minimize costs and maximize water quality benefits. Although traditional conservation and nutrient management practices appear to be the most cost-effective strategies over the long term, P removal technologies may provide more immediate or additive water quality benefits, especially at badly overloaded legacy P sites where manure and fertilizer additions are already restricted to crop removal rates. Additional studies of this nature will be essential to understanding the costs and benefits of P removal technologies in the context of whole-farm- and watershed-scale P management.

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