Final Report to Illinois-Indiana Sea Grant

Evaluation of Phosphorus Loading Following a Manure Spill and In-stream Sediment Amendment to Reduce Phosphorus Desorption.

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Abstract

Within the last 20 years in the U.S. manure spills have become a water quality issue and has been associated to the increase in the number of hogs coupled with a drastic decline in hog farms. Moreover, the current manure spill remediation method neglects phosphorus (P) and nitrogen (N) enriched stream sediments that act as a source of P and N in subsequent flow. Therefore, the objectives of this research were to (i) develop an understanding of P and N partitioning during a manure spill and the effectiveness of the current manure spill remediation method; (ii) determine the relationship between sediment particle size distribution and the depth of N and P sediment contamination following a spill; (iii) advance the current manure spill remediation practice via the establishment of sediment specific alum application rates to treat contaminated fluvial sediments following a manure spill; (iv) evaluate the sediment specific alum application rates to advance the remediation of P enriched fluvial sediments following a manure spill, under flow dynamics. Laboratory manure spill simulations were conducted using fluvarium techniques, swine manure, and sediments collected from drainage ditches located in the St Joseph River Watershed of Northeast, IN. All sediments acted as a significant sink for P released P to the water column at concentrations that significantly exceeded the Environmental Protection Agency total P (0.076 mg L^{-1}) nutrient criteria for the St Joseph River Watershed. Significantly greater sediment P concentrations were observed in the 0-1 cm depth relative to the 1-2 cm depth and the range of P concentration for the entire 0-2 sediment depth was 3 to 12 mg P kg^{-1}. A 54% greater rate of alum was required to remediate P in clay loam sediments collected from a small drainage area relative to sediments that contain at least 85% sand that were collected from a larger drainage area. Augmenting the current remediation practice with alum reduced the water column P concentration in subsequent flow below the EPA total P standard and significantly reduced the sediment labile P by 91, 70, and 70% for the sand, sandy loam, and

clay loam sediments, respectively. However, further investigation of the sediment alum treatments is needed to determine the time of effectiveness after application under flow conditions. The results from this research have the potential to advance the decision making of environmental protection agencies, manure spill first responders, and livestock producers when remediating manure spills that reach surface waters.

Introduction

Manure spills that reach surface water contribute large masses of contaminants such as nutrients, salts, organic compounds, heavy metals, microbial pathogens, antibiotics and natural and/or synthetic hormones (Peterson et al., 2000; Mallin and Cahoon, 2003; Raman et al., 2004). Conventionally, drainage ditches are used by emergency response teams, after the occurrence of a manure spill, to intercept contaminated water before it is transported downstream. The current recommended manure spill response plan for Midwestern states (IN, OH, WI, and IL etc.) and Canada is: (1) containment and isolation of the contaminated area within the ditch or stream by constructing temporary earthen dams; (2) dewatering of the contained area using standard pumping equipment; and (3) redistribution of the recovered waste and water mixture into an alternative storage system or land application of the mixture in compliance with state regulations (Illinois EPA, 1999; IDEM, 2002; Kewaunee County Univ. of Wisconsin-Ext., 2003). This current remediation method adequately removes the P and NH_4-N contamination from the water column, but neglects the nutrient enriched sediments that remain in the fluvial system. As a result, the water column continues to be impaired for weeks after the plume of the spill has passed (Burkholder et al. 1997).

Thus, there is a need to further characterize P contamination in a fluvial system following a manure spill under worst case scenarios (base flow conditions that would only slightly dilute swine manure in the event of a manure spill), based upon the evidence that the water column continues to be impaired by contaminated fluvial sediments. A greater understanding of the depth of P contamination in fluvial sediments will give a basis for the development of supplemental sediment treatments that could mitigate the impairment of the water column in subsequent flow.

The use of alum ($Al_2(SO_4)_3 \cdot 14H_20$) has been shown to decrease the P solubility in poultry litter (Sims and Luka-McCufferty, 2002; Delaune et al., 2004), swine manure (Smith et al., 2004), and dairy effluent (Kleinman et al., 2002). Moreover, studies also have demonstrated that alum can enhance sediment P retention and sorption properties and decrease the potential for P transport downstream. Haggard et al. (2004) amended sediments that were P enriched due to continued exposure to waste water treatment plant effluent with alum and $CaCO_3$. They observed a significant increase in sediment P buffering capacity (the ability for sediments to adsorb P) and found a significant decrease in the equilibrium P concentration (EPC_0) (the P concentration where net absorption and desorption of P is zero) and sediment labile P concentration. Smith et al. (2005), found a similar trend after amending sediments from tile fed agricultural drainage ditches, where the labile P concentration was decreased by 48-89% and the EPC_0 was significantly reduced from 0.051 to 0.004 mg L^{-1}.

Although the effectiveness of alum to increase the P sorption capacity of fluvial sediments has been established, data relating alum rates to differences in sediment physiochemical properties is lacking, and data supporting alum as a manure spill remediation technology is largely nonexistent. Considering the inability of current manure spill clean-up protocols to remediate P enriched fluvial sediments and the efficacy of alum to increase sediment P retention, there is a need to explore the benefits of incorporating alum treatment technology to remediate P loading in sediments following manure spills.

Therefore, the objectives of this study were to (1) develop a comprehensive understanding of P and NH_4-N partitioning between fluvial sediments and the overlying water column during and following a manure spill, (2) determine the effectiveness of the current manure spill remediation plan to mitigate P desorption from manure exposed ditch sediments (3) investigate the influence of sediment particle size distribution on the depth of P contamination as a result of a manure spill (12:1 manure to water ratio) under base flow conditions using fluviarium techniques, (4) determine adequate application rates of alum and $CaCO_3$ needed to remediate fluvial sediments following a manure spill (5) evaluate the efficacy of the sediment alum treatments following a manure spill.

Narrative

Objective 1 and 2: To evaluate the partitioning of P during and after the occurrence of a manure spill in a fluvial system, a series of manure spill simulations were conducted within fluvaria packed with sediments collected from each sampling site (A full description of the material and methods, and results can be found in Armstrong et al., 2009). The fluvarium (stream simulator; 0.75 m long x 0.23 m wide) simulated a constant flow of water (0.06 L sec^{-1}), over a path of homogenous sediment for an estimated length of 1206 m. During the adsorption phase of the manure spill simulations, the relative water column soluble P concentration was significantly related (P <0.0001) with estimated distance travelled downstream for all three sediments (Figure1; Table1).

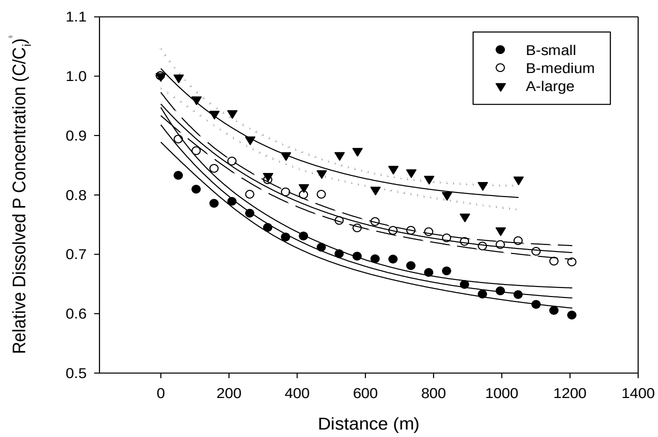

Figure1. Relative P concentrations in the water column during the adsorption phase of the manure spill simulation. The error bars represent 85% confidence intervals.

Table 1. Line equations from the adsorption and desorption manure spill phase regressions of water column P concentrations versus estimated distance travelled downstream.

Ditch Sediment	Line Equation	R^2	*P*
Adsorption			
B-small	y=0.61 + 0.30exp(-0.0025*distance)	0.98	<0.0001
B-medium	y=0.68 +0.26exp(-0.0024*distance)	0.97	<0.0001
A-large	y=0.78 + 0.23exp(-0.0027*distance)	0.88	<0.0001
Desorption			
B-small	y=0.08 + 0.104*(1-e^(-0.01*distance))	0.91	<0.0001
B-medium	y=0.03 +0.104*(1-e^(-0.006*distance))	0.94	<0.001
A-large	y=0.09 +0.11*(1-e^(-0.003*distance))	0.84	<0.001

Exclusive confidence intervals between sediment adsorption curves suggested that the rate of P adsorption during the manure spill simulation was not the same among sediments (Figure 1). B-small sediments (33.8 % clay) removed P from the water column at a significantly greater rate during the spill simulation, followed by the B-medium (6.1 % clay) and A-large (1.3 % clay) sediments. Sediments with greater clay and organic matter content removed P from the water column at a greater rate relative to sandy textured sediments during the adsorption phase of the manure spill simulation.

Immediately following the adsorption phase of the manure spill simulation, the current manure spill remediation method was simulated by removing the water from the stream simulator and allowing the sediment to remain in place. Subsequent flow was simulated via 23 hr of flow over the sediments with manure free simulated ditch water. Results from the desorption phase experiment suggested that the current remediation method is not effective at addressing the total fluvial system (water column and sediments) following a manure spill (Figure 2). After five hours of subsequent flow, the average water soluble P concentrations in the water column for the B-small, B-medium, and A-large sediments were 0.19, 0.15, and 0.21 mg L^{-1}, respectively.

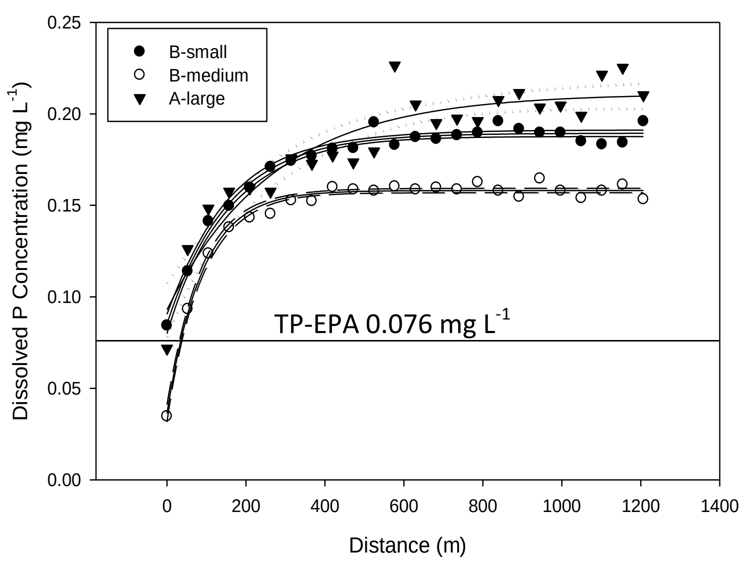

Figure 2. Phosphorus concentrations in the water column during the desorption phase of the manure spill simulation. The error bars represent 85% confidence intervals.

Objective 3: The depth of P contamination following a manure spill was also determined via manure spill simulations (12:1 ratio of swine manure to simulated ditch water was used to simulate a manure spill that reaches ditches under base flow conditions). Following the simulation of a 23 hr manure spill and the current manure spill simulation, sediment samples were collected from the 0-1 and 1-2 cm depths. After sediment collection samples were extracted for labile P (the complete description of the materials and methods are present in Armstrong et al., 2010, submitted).

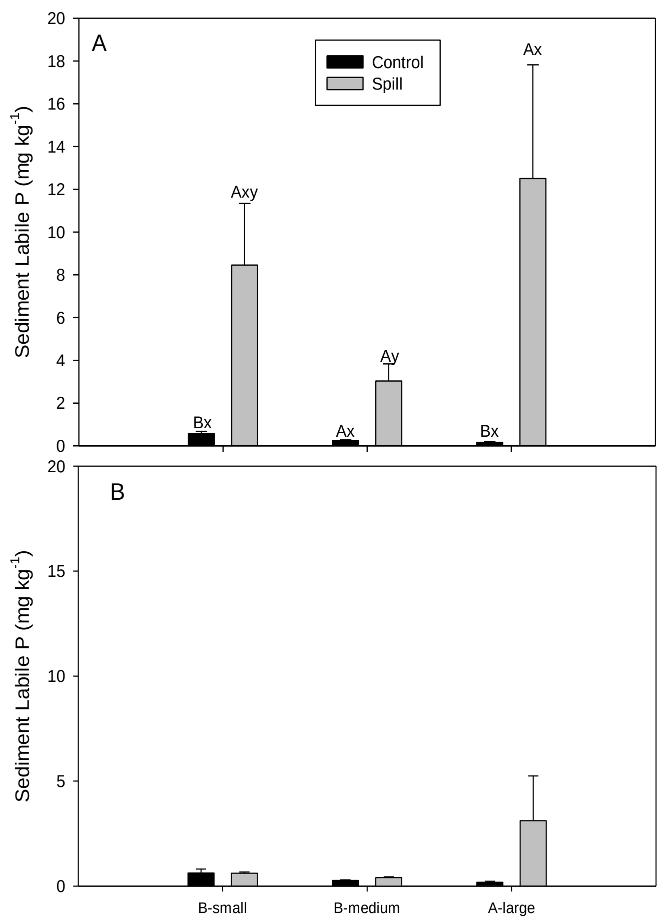

Figure 3. Labile P loading depth following the 23 hr manure spill simulation for each sediment type, at both depths (A) 0 - 1 cm and (B) 1 - 2 cm. Capital letters (A, B, C) indicate treatment mean differences within sediment type and depth. Lowercase letters (x, y, z) indicate sediment type mean differences within treatment and depth. (alpha level = 5%). Error bars represent the mean standard error.

At the 0 - 1 cm depth, sediments collected from the Large drainage area resulted in a slightly greater labile P concentration (12.4 mg kg^{-1}) than the Small drainage area sediments (8.45 mg kg^{-1}) and was significantly greater than the Medium P concentration (3.03 mg kg^{-1}; P = 0.001; Figure 3A). The sandier sediment, collected from the Large drainage area, appear to be more prone to serve as a significant P source to the water column relative to sediments with greater clay and silt contents. At the 1 - 2 cm depth, the Large sediment labile P concentration was 17 times greater compared to the control treatment sediment (Figure 3B). A possible explanation for this observation is the difference in pore size distribution among the sediments. The Large sediment contains the greatest sand content, and the least percentage of clay, and thus the largest pore sizes making the sediment more susceptible to translocation of P to the lower depths.

Table 2. Phosphorus sorption ratio (PSR) before and after the manure spill simulation. *Indicates significant difference of treatments within the medium sediments and 0-1 cm depth.

	Depth			
	0-1 cm		1-2 cm	
Ditch	Control	Spill	Control	Spill
	PSR			
Small	0.016	0.027	0.013	0.031
Medium	0.013	0.049*	0.015	0.013
Large	0.002	0.021	0.001	0.006

The differences of the average PSR (the ratio of extractable P in mmol to extractable Fe and Al mmol) for the control and spill treatments across all sediments confirmed the results observed in the sediment labile P concentration following a manure spill. In the 0 – 1 cm depth, the mean PSR significantly increased by 220% from 0.010 to 0.032, suggesting a considerable increase in extractable P (Table 2). At the lower depth (1 - 2 cm), there was no significant difference in labile P among the three sediments within the spill treatment, which was also reflected in the PSR values.

Objective 4: To determine a range of alum application rates to mitigate P desorption from fluvial sediments of different particle size distributions following a manure spill, an isotherm experiment was conducted using alum under oxic conditions. Sediments from the mixing depth experiment of objective 4 were treated with 5 rates of alum at 0, 1.6, 3.6, 5.6, and 7.6 mg g^{-1} that were dissolved in 10 mL of simulated ditch water. The alum rate required to mitigate P desorption from the sediments was considered to be the $Alum_{max}$ (the rate of alum needed to reduce the labile P fraction of sediments following the manure spill simulation under worst case scenario). Based upon the $Alum_{max}$ of each sediment type specific alum application rates were developed and evaluated. A full description of the material and method is present the draft manuscript: The Development of Alum Rates to enhance the Remediation of Phosphorus in Fluvial systems Following Manure Spills.

Sediments that were not amended with alum resulted in the greatest concentration of P to the water column that ranged from 2.4 to 12.3 mg P kg^{-1} (Figure 4.1). The sandy sediments desorbed significantly greater P (12.3 mg kg^{-1}), relative to the clay loam (6.0 mg kg^{-1}) and the clay loam desorbed P was significantly greater compared to the loamy sand (2.4 mg kg^{-1}) sediments. A significantly lower retention of P for the sandy sediments could be explained by lower Fe and Al oxide concentrations of the sand sediments relative to the clay loam and sand sediments, making the sandy sediments more prone to P release during the extraction procedure and a smaller P buffering capacity. Amending the contaminated sediments at a rate of 1.6 mg g^{-1} of alum mitigated the P desorption from the sandy sediments and reduced the desorbed P of the clay loam and loamy sand by 72 and 92 %, respectively. Treating the sediments at rates of 3.6, 5.6, and 7.6 mg g^{-1} of alum reduced the P desorption of the sandy and loamy sand sediments by 100 %, relative to sediments that received no treatment. However, amending the clay loam sediments at the same rates of alum resulted in a 92, 98, and 100 % reduction in desorbed P relative to the untreated clay loam sediments (Figure 4.).

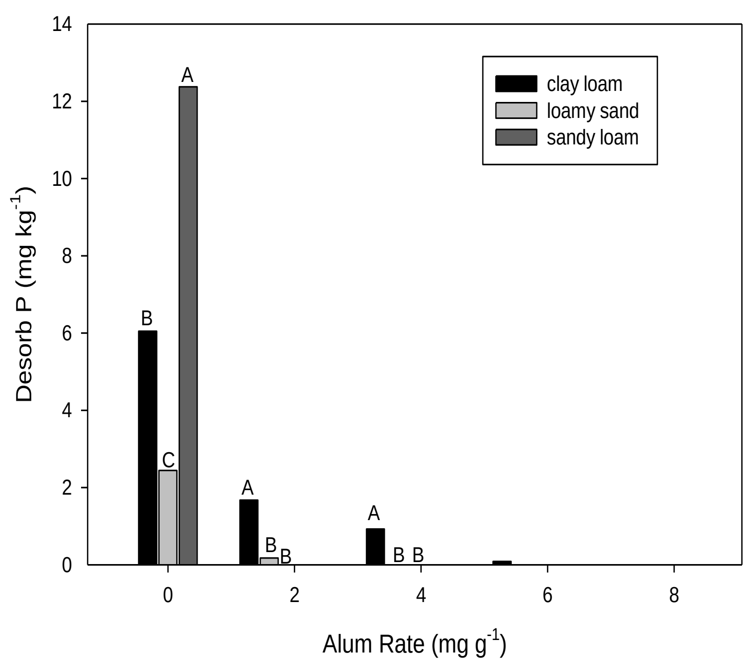

Figure 4. The impact of increasing alum rates on P desorption from manure contaminated sediments of different particle size distributions. Capital letters indicate significant difference (α=0.05) between sediment types within each alum rate.

These observations suggested that different $alum_{max}$ rates (the rate of alum needed to mitigate P desorption) were required to mitigate P desorption from manure exposed fluvial sediments of different particle size distributions and chemical properties. A greater rate of alum was needed to mitigate P desorption from the clay loam sediments (7.6 mg g^{-1}), relative to the loamy sand (3.6 mg g^{-1}) and sand sediments (1.6 mg g^{-1}). $Alum_{max}$ rates were also significantly correlated with chemical properties that influence the affinity of the sediments to sorb reactive P, such as Fe_2O_3 and Al_2O_3 percentages, P buffering capacity (the ability of the sediment to adsorb P from the water column), organic C, clay and silt, and CEC and AEC (Table 3.).

Table 3. Correlation R^2 values for the correlation analysis of sediment chemical and physical properties and $alum_{max}$ rates

	Alum max	Clay	Silt	Buff. Cap.	Org. C	Fe_2O_3	Al_2O_3	CEC	AEC
$Alum_{max}$	1								
Clay %	0.98	1							
Silt %	0.96	0.99	1						
Buff. Cap.	0.99	0.99	0.98	1					
Org. C	0.96	0.99	0.99	0.99	1				
Fe_2O_3	0.94	0.89	0.85	0.91	0.87	1			
Al_2O_3	0.97	0.93	0.90	0.95	0.92	0.99	1		
CEC	0.98	0.97	0.95	0.98	0.97	0.94	0.97	1	
AEC	0.75	0.84	0.87	0.81	0.87	0.6	0.69	0.8	1

Alum Rate Evaluation: Alum treated fluvial sediments resulted in less desorbed P relative to sediments that were untreated following a manure spill. Untreated contaminated sediments desorbed P at a significantly greater rate relative to sediments treated with alum or alum + $CaCO_3$ at all application rates (Figure 5).

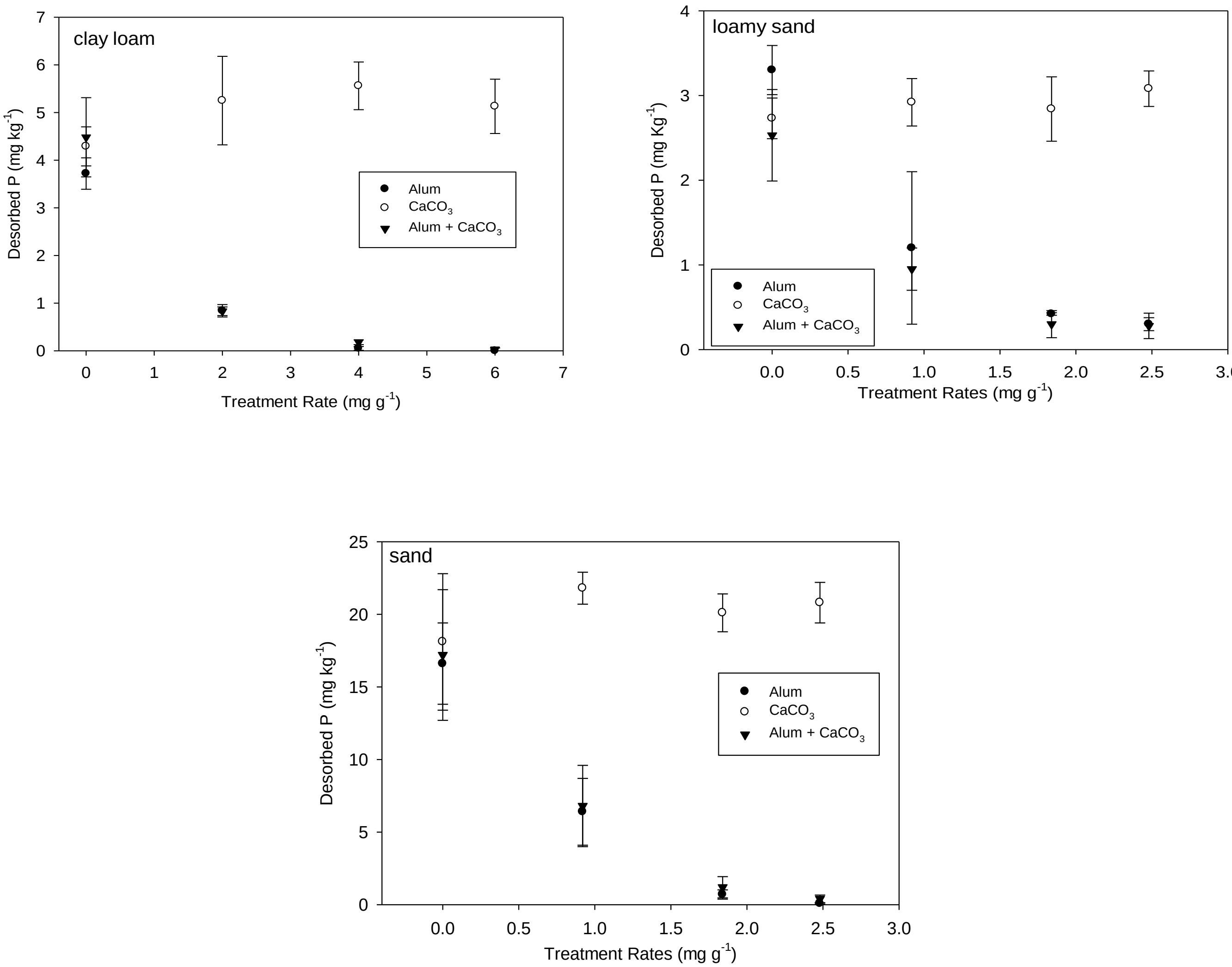

Figure 5. Relationship of sediment desorbed P and added alum, $CaCO_3$, and alum + $CaCO_3$ in the sand sediment.

Objective 5: Evaluate the efficacy of the sediment alum treatments following a manure spill.

Alum sediment treatment rates were evaluated under flow conditions using fluvariums and manure spill simulations. Three scenarios were simulated in this phase of the study:

(1) Uncontaminated stream flow remediated using the current remediation practice for manure spills and the simulation of subsequent flow for 23hr, following remediation. (Hereafter referred to as the Control treatment).

(2) Stream flow that was contaminated with a 12:1 ratio of swine manure to simulated ditch water (simulated ditch water = water spiked to a $CaCl_2$ concentration of 2.5 m*M* to simulated natural salt content of the ditch water), remediated using the current remediation practice for manure spills, and reentry of subsequent flow for 23hr, following remediation (Hereafter referred to as the CRP treatment).

(3) Stream flow that was contaminated with a 12:1 ratio of swine manure to simulated ditch water for 23hr, remediated using the current remediation practice plus sediment specific alum amendment rates developed in chapter 4, and the reentry of subsequent flow for 23 hr, following remediation (Hereafter referred to as the Alum treatment).

Differences in sediment P adsorption during the manure spill was similar to results of objective 1, where sediment with the greatest clay content removed the greatest mass of P from the water column and sediments of greater sand content removed significantly less P from the water column (a full description of the methods and results can be found in draft manuscript: Evaluation of Sediment Specific Alum Rates Under Flow Conditions to Reduce Phosphorus Desorption following a Manure Spill).

Following the simulation of the current manure spill remediation methods and 23 hr of subsequent flow, the estimated mean equilibrium dissolved P concentrations in the water column were 9.60, 6.79, 5.87 mg L^{-1} for the clay loam, loamy sand, sand sediments, respectively (Figure 6). The equilibrium concentrations observed were compared to the EPA total P nutrient criteria for ecoregion VI (0.076mg $L^{-1;}$ USEPA, 2000) to give some water quality perspective. The comparison indicated that after the manure spill clean-up the resultant equilibrium P concentrations were 126, 89, and 77 times greater than the EPA standard and were equivalent to 32, 27, and 19% of the equilibrium P concentration in the water column immediately before the remediation process began.

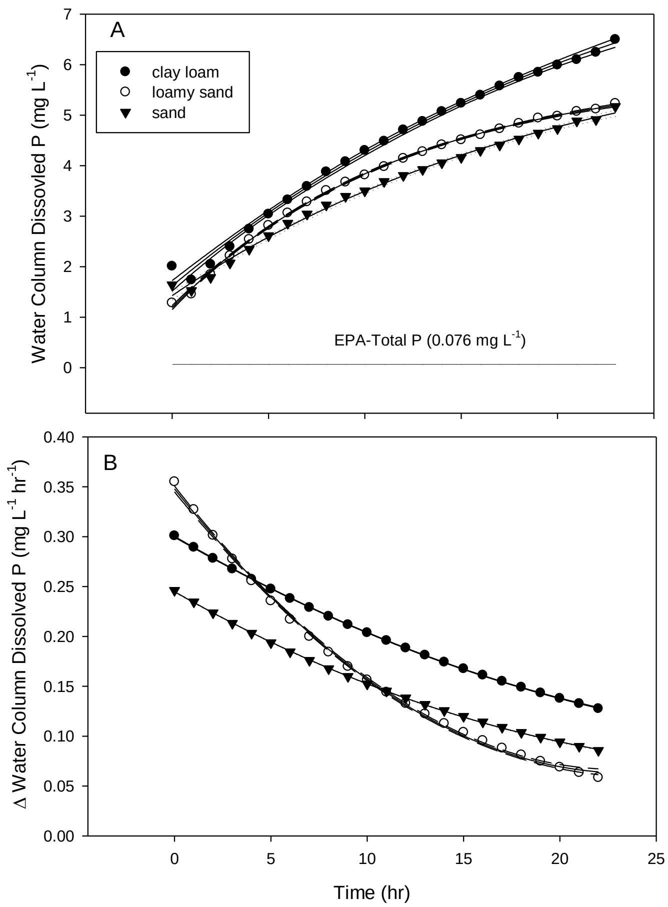

Figure 6. Desorbed P concentrations from sediments of the CRP treatment regressed against time (A). Instantaneous change of dissolved P regressed against time (B). The error bars represent 85% confidence limits.

Augmenting the current remediation practice with sediment-specific rates of alum resulted in equilibrium concentrations that were below the EPA total P nutrient criteria for all sediments (Figure 7A). The loamy sand sediments resulted in the greatest P concentration (0.067

mg L^{-1}) after the alum treatment, followed by the clay loam sediments (0.01 mg L^{-1}) and the (0.00 mg L^{-1}) sand sediments. The resultant water column P concentrations after 23 hr of circulated flow for alum treated sediments were 98, 99, and 100% lower for the loamy sand, clay loam, and sand sediments, respectively, relative to the P concentration in the water column of the same sediments remediated without alum (Current Recommended Practice).

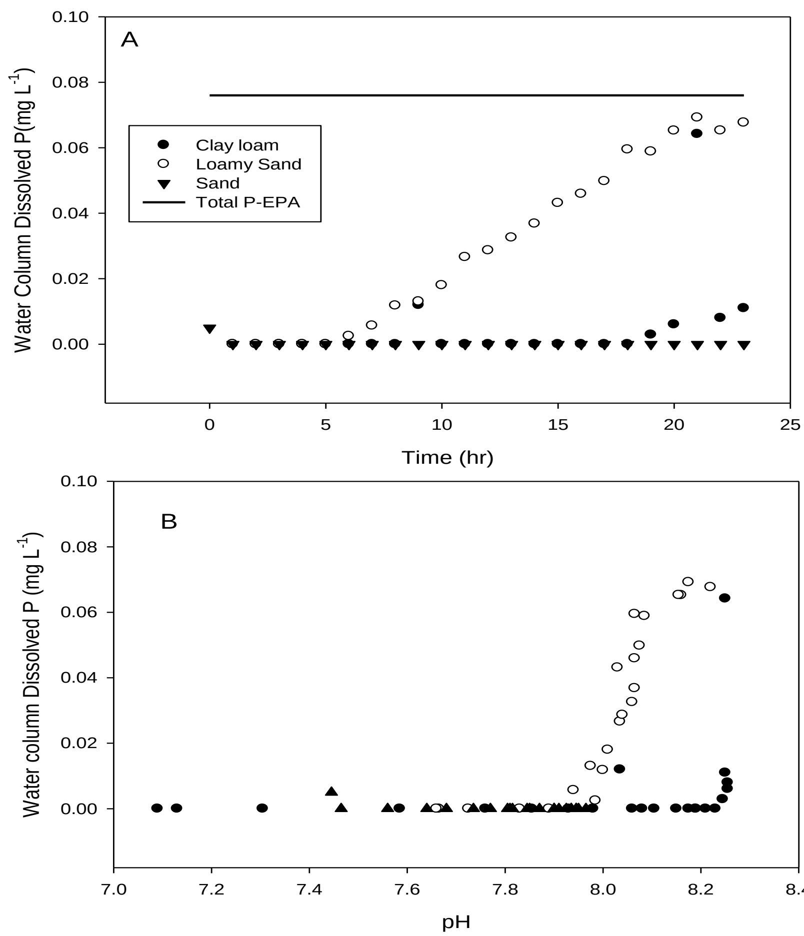

Figure 7. Release of dissolved P following remediation using the current recommended practice and alum treatment (A) and the regression of the water column dissolved P against water column pH (B).

After six hours of flow, the P concentration in the water column of the loamy sand alum treated sediment began to increase linearly and the same was observed for the clay loam alum treated sediment at the18th hour. However, dissolved P desorption from the sand sediment was mitigated and no P desorption occurred throughout the entire subsequent flow experiment. One possible explanation for the increase in P concentrations for both loamy sand and clay loam sediments is an underestimation of the alum application rate needed to maintain a pH <8 in the water column.

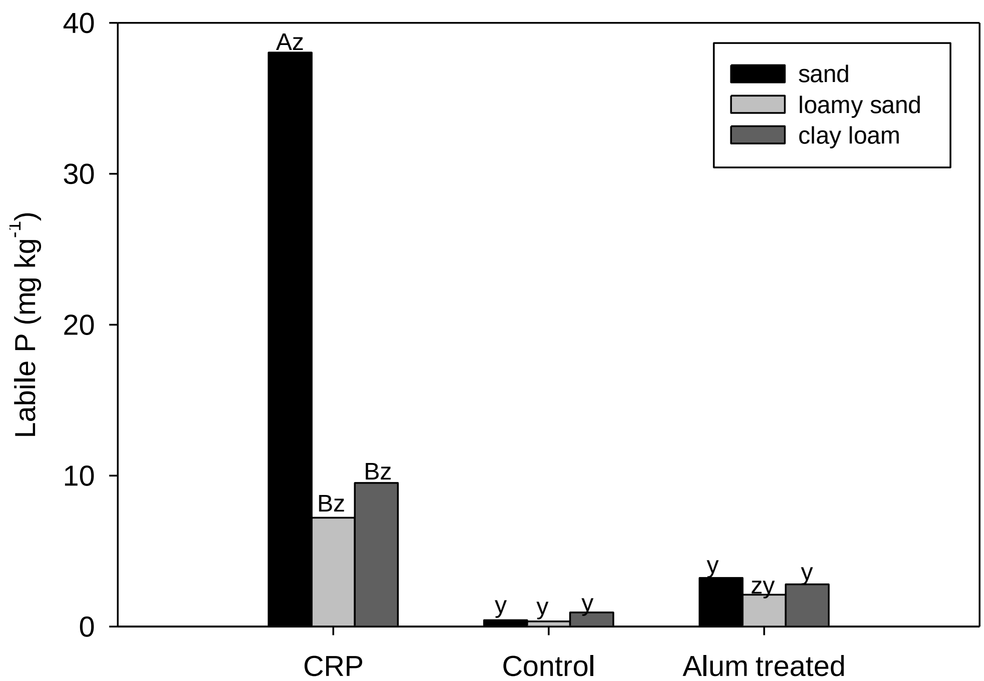

Figure 8. Sediment labile P concentrations following the subsequent flow experiment. Capital letters (A, B, C) represent statistical differences in sediment type within treatments and lower case letters (y, z) represent defenses between treatments within sediment types.

Data from sediment samples collected immediately after the 23 hr subsequent flow study confirms trends observed from the water samples and indicated significant differences among sediment types within treatments and among treatments within sediment types (Figure 8.). The comparison of sediment types within the CRP treatment suggest that the sand sediments resulted in significantly greater labile P concentration (38.0 mg kg^{-1}) relative to the clay loam (9.52 mg kg^{-1}) and loamy sand (7.21 mg kg^{-1}) sediments. There were no differences found within the control and alum treatments among sediment types. Ultimately these findings suggested that remediating the manure spill without the augmentation of alum resulted in significant greater labile P in the sediments relative to the control even after 23 hr of subsequent flow. However, remediating the fluvial system with the CRP and the application of alum reduced the labile P in the contaminated sediments by 91, 70, and 70% for the sand, loamy sand, and clay loam, respectively.

Potential Applications or Benefits

This study assessed the current remediation of manure spills that reach surface waters and quantified the impact of manure exposed sediment following a manure spill. The current methods used to clean-up fluvial systems after the occurrence of manure spills should be improved to adequately remediate the entire fluvial system: water column and sediments following a manure spill that reaches surface waters. The study confirmed that during a manure spill, all sediments are capable of serving as P sink during the manure spill. In addition, it was determined that sediments adsorbed P from the water column rapidly for the first 5 hr and slower adsorption occurred as the pH of the system increased and the system approached P equilibrium. The data also demonstrated that following the current remediation practice, all sediments released P to the water column at concentrations that exceed the EPA nutrient criteria for total P in ecoregion VI, where the sediments were collected.

Furthermore, findings from this study demonstrated that alternative manure spill remediation methods could be employed to increase the effectiveness of remediation and decrease the recovery time of manure contaminated surface water. Augmenting the current remediation practice with sediment specific rates of alum reduced the water column P concentration in subsequent flow below the EPA total P standard and significantly reduced the sediment labile P by 91, 70, and 70% for the sand, sandy loam, and clay loam sediments, respectively. Sediment alum rates, based the physiochemical properties and depth of P contamination during the spill, were developed in this study for sediments collected in the Midwestern Corn Belt region. Clay loam sediments that contained the greatest clay, Fe, Al, and organic carbon contents required an alum rate equivalent to 14lb/1000ft^2, and the Loamy sand sediments required 9.6 lb/1000ft^2. The sand sediments required a rate of 9.61b/1000ft^2 based upon sediment properties for the first centimeter of sediments; however to account for a deeper P contamination depth (1-2 cm), a rate of 14lb/1000ft^2 was applied, which is equivalent to a rate 50% greater than the rate required for the 0-1 cm depth.

With the establishment of sediment specific alum amendment rates, it is possible to supplement the current method of treating manure spills with alum to adequately remediate the fluvial system. For example, after damming the contaminated stream with earthen dams and removing the contaminated water column, the surface area of the contaminated stream can be acquired. Immediately following, the sediment texture can be determined and the most feasible rate of alum can be surface applied to reduce further impairment of the fluvial system via P desorption from the contaminated sediments.

Ultimately, I believe that the findings of this study have exposed environmental managers to the fate of P during and after a manure spill and could advance the remediation of manure spills that reach surface waters

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Keywords: Manure Spills, Phosphorus, Remediation, Nutrient Transport, Nutrient Fate, Adsorption, Desorption

Lay Summary

The impact of P loading and fate of P following a manure spill has become a research priority, due to contributions of P to surface waters by manure spills. Therefore, the objectives of this study were to (1) develop an understanding of P partitioning between fluvial sediments and the overlying water column during and following a manure spill, (2) determine the effectiveness of the current manure spill remediation plan to mitigate P desorption from manure exposed ditch sediments (3) investigate the influence of sediment particle size distribution on the depth of P contamination as a result of a manure spill (4) determine adequate application rates of alum and $CaCO_3$ needed to remediate fluvial sediments following a manure spill (5) evaluate the efficacy of the sediment alum treatments following a manure spill.

Using laboratory manure spill simulations we determined that, during a manure spill, bed sediments within the stream act as a sink for phosphorus, and after the spill the sediments act as a phosphorus source resulting in the release of P after remediation. Furthermore, we determined that during a manure spill the top 2 cm of sediments served as temporary storage for P and were

responsible for the release of P back to the water. During an isotherm study, we determine aluminum sulfate application treatment rates for sediments of different textures. In addition, we observed that treating the contaminated sediments following a manure spill with aluminum sulfate reduced P release back to the water by at least 90% compared to sediments that receive no treatment following a manure spill under flow conditions.

Publications:

Armstrong, S. D. 2010**.** Phosphorus and Nitrogen Partitioning In A Fluvial System During and After A Manure Spill: The Development of A Supplemental Sediment Chemical Treatment. Ph.D. dissertation. Purdue University, West Lafayette IN.

Armstrong, S. D., D. R. Smith, P. R. Owens, A. B. Leytem, B. C. Joern, C. H. Huang, and L. Adeola. 2010. Phosphorus and nitrogen loading depth in fluvial sediments following manure spill simulations. Canadian Journal of Soil Science (Accepted).

Armstrong, S. D., D. R. Smith, B. C. Joern, P. R. Owens, A. B. Leytem, C. H. Huang, and L. Adeola. 2009. Transport and fate of phosphorus during and after manure spill simulations. J. Environ. Qual. 39:345-352.

Armstrong, S. D., D. R. Smith, B. C. Joern, P. R. Owens, A. B. Leytem, C. H. Huang, and L. Adeola. 2010. The development of alum rates to enhance the remediation of phosphorus in fluvial systems following manure spills (Draft to be submitted to Water Environment Research by Nov. 2010).

Armstrong, S. D., D. R. Smith, B. C. Joern, P. R. Owens, A. B. Leytem, C. H. Huang, and L. Adeola. 2010. Evaluation of sediment specific alum rates under flow conditions to reduce phosphorus desorption following a manure spill.(Draft to be submitted to the Journal of Soil and Water Conservation by Dec. 2010).

Awards:

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