

Enhancement of Phosphorus Removal in Bioretention Cells by Soil Amendment

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Abstract. *Phosphorus (P) removal in bioretention cells has been highly variable. The objective of this study was to find inexpensive filter media with high P sorption and adequate hydraulic conductivity. Batch sorption experiments were conducted to screen filter media. Incorporation of fly ash increased P sorption of two Oklahoma soils, Teller loam and Dougherty sand significantly. Fly ash addition decreased the hydraulic conductivity of the sand exponentially. Maximum sorption capacity predicted by Langmuir isotherms was only 23.8 mg/kg for Dougherty sand, but 385 mg/kg for Dougherty sand with 5% by weight fly ash, and 82.0 mg/kg for expanded shale. Dougherty sand released most sorbed P while P released by the sand/fly ash mixture was negligible. A linear equilibrium convection-dispersion transport model was applied to estimate retardation factors by fitting observed breakthrough curves (BTCs) obtained from column flow-through experiments. Phosphorus BTC in Dougherty sand suggested its retardation factor to be close to one, while retardation factor of Dougherty sand with 2.5% and 5% fly ash and expanded shale was 199, 470, and 15.7, respectively. Incorporation of a sand/fly ash layer in bioretention cells could improve P removal dramatically.*

Keywords. Bioretention, Stormwater, Runoff, Phosphorus, Sorption, Hydraulic conductivity, Fly ash, Transport, Retardation

Introduction

Urban stormwater runoff, one of the main sources of non-point source pollution, is the second largest pollution source for estuaries and the third largest for lakes (USEPA, 2002). Phosphorus (P) has been long recognized as a limiting nutrient in some surface waters (Correll, 1998). Overenrichment of P in lakes, reservoirs, and stream backwater may lead to eutrophication. Stormwater runoff, especially the runoff from streets, fertilized lawns, and golf courses, can transport a significant P load (Garn, 2002; Moss et al., 2006; Waschbusch et al., 1999). In Wisconsin, the runoff from fertilized lawns had total P of 4 mg/L and dissolved P of 0.93 mg/L (Garn, 2002). In Oklahoma, the runoff from golf course fairways had dissolved P as high as 8 mg/L (Moss et al., 2006). Thus, P reduction from stormwater runoff may be of significance to protect the quality of recipient water bodies.

Bioretention cells have gained popularity as stormwater best management practices (BMPs) because of their design flexibility, appealing landscape aesthetics, and perceived effectiveness for reduction of pollutants. Field and laboratory studies on pollutant removal in bioretention cells were undertaken by several researchers (Davis et al., 2001, 2003; Hsieh and Davis, 2003, 2005a, 2005b; Hunt and Jarrett, 2003). Bioretention cells can remove heavy metals, oil and grease, and total suspended solids efficiently (Davis et al., 2003; Hsieh and Davis, 2003, 2005b). However, P removal has been highly variable, and in some cases the P production was noted (Hunt and Jarrett, 2003; Hsieh and Davis, 2003, 2005a). Hunt and Jarrett (2003) argued that high P-index soils placed in the cells might contribute to the P production. So far little effort has been made to improve P removal in bioretention cells.

The primary mechanisms of P removal in bioretention cells are filtration, biological uptake, and storage in planting soils and filter media (PGDER, 2002). Cost-effective filter media is particularly needed by bioretention cells. Fly ash, expanded shale, slag, red mud, gas concrete, and cement can remove P effectively (Agyei et al., 2002; Akay et al., 1998; Cheung and Venkitachalam, 2000; Forbes et al., 2004; Johansson and Gustafsson, 2000; Oguz et al., 2003; Ugurlu and Salman, 1998). Fly ash, the waste product of burning coal, is abundant. Expanded shale, a porous material manufactured by firing shale at 2000 °F, is also readily available but at greater cost. However, little research has been undertaken for using fly ash and expanded shale in bioretention cells.

The objective of this study was to find cost-effective filter media with both high P sorption capacity and adequate hydraulic conductivity to improve P removal in bioretention cells. Batch sorption experiments were conducted to screen filter media. Hydraulic conductivity of filter media was examined by a falling head permeameter to meet infiltration capacity requirement for bioretention media. Sorption isotherms and desorption experiments were conducted to further characterize P sorption and desorption of filter media. Phosphorus retardation factors of filter media were estimated by fitting P breakthrough curves (BTCs) obtained from column flow-through experiments using a linear equilibrium convection-dispersion transport model. Results were combined to identify the best filter media for P removal in bioretention cells.

Materials and Methods

Soils and Other Materials

Teller loam (Thermic Udic Argiustoll) and Dougherty sand (Thermic Arenic Haplustalf) were collected from field locations in Payne County, Oklahoma. Soil samples were air dried and passed through a 2 mm sieve. Teller loam contains 52% sand, 31% silt, and 17% clay.

Dougherty sand has 98% sand and 2% silt and clay. Fly ash was obtained from the Sooner Power Plant at Red Rock, Oklahoma. Fly ashes from power plants burning different types of fuels could have different chemical compositions. The fuel source of the Sooner Power Plant is subbituminous coal from the Power River Basin, Wyoming. Table 1 lists the major chemical compositions for fly ash used in this study as determined by X-ray fluorescence (XRF) analyzer (Philips PW-2400) at Activation Laboratories Ltd., Ontario, Canada. Limestone was taken from a local aggregate supplier and crushed to pass a 2 mm sieve. Peat moss was commercially obtained from Premier Horticulture Inc., PA and ground to pass a 2 mm sieve. Two expanded shales were obtained from two plants of Buildex Inc., KS, located at Marquette, KS and New Market, MO, and labeled as M-shale and N-shale, respectively. Materials were tested by the Soil, Water and Forage Analytical Laboratory (SWFAL) of the Department of Plant and Soil Sciences, Oklahoma State University in accordance with the procedures of ASA and SSSA (ASA and SSSA, 1982; SSSA and ASA, 1996). Other relevant properties of materials are also presented in Table 1.

Table 1. Relevant properties of soils and other materials.

Materials	pH ^A	CEC ^B , meq/100g	Exchangeable Ca ^C , mg/kg	Extractable P ^D , mg/kg	K _d , mL/g
Teller loam	6.2	~ 9	1140	71	0.41
Dougherty sand	6.3	~ 1	148	14	2.08
Fly ash	11.5	~ 78	14300	13	2180
Limestone	9.0	~ 4	457	6.7	12.1
Peat moss	2.9	~ 7	821	23	-5.79 ^E
M-shale	6.4	~ 10	1180	32	280
N-shale	8.6	~ 1	192	18	1.21
Major chemical composition, %					
Fly ash	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	CaO
	38.1	18.4	5.93	5.43	22.9

^A pH was measured in batch sorption experiments

^B CEC was determined by summing the amount of exchangeable Ca, Mg, K, Na, and Al displaced by ammonium acetate (SSSA and ASA, 1996)

^C Exchangeable Ca was displaced by ammonium acetate and determined by ICP-AES (Spectro CIROS^{CCB})

^D Extractable Mehlich3 P was determined by the ascorbic acid method (LACHAT, Quickchem 8000)

^E Negative K_d indicated the P production

Distribution Coefficients

Two grams of each sorbent were placed in 50 mL polypropylene centrifuge tubes with 40 mL solution containing 1 mg/L P as sodium phosphate (Na₃PO₄·12H₂O) (0.05 solid/water ratio). Ionic strength was fixed with 0.01 mol/L potassium chloride (KCl). The tubes were shaken on a rotary agitator at 30 RPM and 23±2 °C for 24 hrs. Then the suspensions were centrifuged and the pH measured. The supernatants were filtered through a 0.45 µm Glass/Nylon filter, acidified

and analyzed for P by ICP-AES. Phosphorus distribution coefficients (K_d) (ASTM, 2004) was calculated as:

$$K_d = \frac{(C_0 - C)V}{M_s C} \quad (1)$$

where K_d (mL/g) is distribution coefficient, C_0 (mg/L) is the initial P concentration in the blank, C (mg/L) is the equilibrium P concentration in the solution, V (mL) is the volume of solution, and M_s (g) is the mass of sample.

Phosphorus K_d was also determined for the mixtures of Teller loam or Dougherty sand with various levels of fly ash to examine the effect of fly ash addition on P sorption of soils.

Hydraulic Conductivity

A falling head permeameter (McWhorter and Sunada, 1977) was used to determine the saturated hydraulic conductivity of Teller loam, Dougherty sand, expanded shale, and the mixtures of Dougherty sand with various levels of fly ash. A 4 cm inner diameter and 15 cm long acrylic column was packed with each material. The column was connected to a glass tubing reservoir containing 0.01 mol/L calcium sulfate (CaSO_4) solution. Water flowed upward through the column, and the hydraulic gradient ranged from 2.03 m/m to 3.79 m/m.

Sorption Isotherm and Desorption

Two grams of Dougherty sand, Dougherty sand with 5% fly ash (D+5%), and M-shale were placed into 50 mL polypropylene centrifuge tubes. Forty mL 0.01 mol/L KCl solution containing 1, 3, 6, or 11 mg/L P was added to each Dougherty sand and M-shale sample, while 3, 6, 11, or 29 mg/L P was added to each D+5%F sample. The sorption procedure was the same to the aforementioned procedure. Data were fitted to the linear form of Langmuir equation by a linear regression and Freundlich equation by a nonlinear regression. The isotherm equations are

$$\frac{C}{S} = \frac{C}{S_m} + \frac{1}{S_m b} \quad (2)$$

$$S = K_f C^n \quad (3)$$

where S (mg/kg) is the amount of P sorbed per unit mass, and S_m , b , K_f , and n are adjustable parameters. Langmuir S_m represents the maximum sorption capacity of materials. Freundlich distribution coefficient (K_f) provides a measure of sorption capacity.

Desorption describes the tendency of materials releasing sorbed P under diluted concentrations. Phosphorus desorption experiments were conducted on these three materials following the sorption experiments. Initial P concentration was extended to 114 mg/L for D+5%F. After removing the previous solution from the tubes 20 mL 0.01 mol/L KCl solution was added. The tubes were shaken thoroughly to disperse the sorbent and placed on the rotary agitator for another 24 hr at 23 ± 2 °C. Then the same procedures of separation and analysis used in the sorption experiment were conducted.

Column Flow-through Experiments and Transport Modeling

Dougherty sand, Dougherty sand with 2.5% fly ash and 5% fly ash (D+2.5%F and D+5%F), and M-shale were packed into acrylic columns with 14.4 cm inner diameter and 14.3 cm long. Influent containing 1 mg/L P passed upward through the columns with a loading rate of 3 cm/hr

(8 mL/min). Influent and effluent samples were collected periodically and their pH measured. Effluent samples were turbid in the early period of experiments for Dougherty sand and M-shale. Those samples were centrifuged to remove suspended particles. The test on Dougherty sand lasted 13 days and produced about 150 L of effluent. For other materials, the experiments lasted three weeks, and approximately 240 L of effluent was produced. All samples were acidified and analyzed for P by ICP-AES. Breakthrough curves (BTCs) were constructed by plotting normalized concentration versus pore volumes.

Phosphorus transport modeling was conducted by fitting BTCs using a one-dimensional linear equilibrium convection-dispersion transport model in CXTFIT 2.1 in the STANMOD software package (Simunek et al., Riverside, California), which was developed for evaluating solute transport in porous media using analytical solutions of the convection-dispersion equation by the U.S. Salinity Laboratory. It was assumed that there was no P production or decay. Retardation factors (R) and hydrodynamic dispersion coefficients (D) were estimated under the third-type inlet boundary and step input conditions. Column characteristics and modeling parameters are presented in Table 3.

Results and Discussion

Distribution Coefficients

Measured P distribution coefficients (K_d) are summarized in Table 1. Distribution coefficient is an index of materials' P affinity. Fly ash had K_d an order of magnitude greater than expanded shale from Marquette, KS (M-shale) and three orders of magnitude greater than Dougherty sand. Therefore, fly ash was identified as a better additive to amend soils. The predominant mechanism for high P sorption of fly ash is calcium phosphate precipitation. Fly ash used in this study has 14,300 mg/kg exchangeable Ca (Table 1), which is at least one order of magnitude higher than that of the two soils. Calcium can dissolve from fly ash and form calcium phosphate precipitates (Agyei et al., 2002; Cheung and Venkitachalam, 2000; Ugurlu and Salman, 1998). Using X-ray diffraction Ugurlu and Salman (1998) directly observed the formation of calcium phosphate precipitate in a fly ash P sorption experiment. It is interesting that expanded shales from two locations exhibited significantly different P sorption. The difference may be a result of the variation of physicochemical properties of shales. Expanded shale from New Market, MO (N-shale) was excluded from the further experiments. M-shale, with the second highest K_d , was also examined in the sorption isotherm, desorption, and column flow-through experiments. However, shale was not investigated as a soil additive.

The addition of fly ash increased P sorption of soils dramatically (Figure 1). With the addition of 5% fly ash, the K_d of Teller loam and Dougherty sand were elevated from 0.41 mL/g and 2.08 mL/g to 49.3 mL/g and 398 mL/g. It was noted that the change of P sorption of Dougherty sand was more significant than Teller loam. The reason for this phenomenon was not understood clearly. It might be attributed to the higher extractable P in Teller loam (Table 1), which is 71 mg/kg and four folds higher than Dougherty sand and fly ash. If this characteristic is similar to other sands, it would favor sand/fly ash mixtures in bioretention cells.

Hydraulic Conductivity

The hydraulic conductivity of Teller loam, Dougherty sand, and M-shale was 0.29 cm/hr, 40.3 cm/hr, and 39.2 cm/hr, respectively. Fly ash had an extremely low hydraulic conductivity, which could not be measured by the procedure used. The infiltration rate of soils in bioretention cells is recommended to be at least 2.54 cm/hr (1.0 inch/hr) (PGDER, 2002). Due to its low hydraulic conductivity, Teller loam is not appropriate to be used in bioretention cells or amended with fly

ash. The hydraulic conductivity of Dougherty sand and its mixtures with various levels of fly ash are presented in Figure 2. The hydraulic conductivity dropped exponentially with increasing fly ash content. To keep the hydraulic conductivity of amended soils higher than 2.54 cm/hr, the incorporation rate of fly ash should be less than 5.8% calculated from the exponential relationship (Figure 2). Therefore, D+2.5%F and D+5%F were selected for further experiments, but only D+5%F was investigated in sorption isotherm and desorption experiments along with Dougherty sand and M-shale.

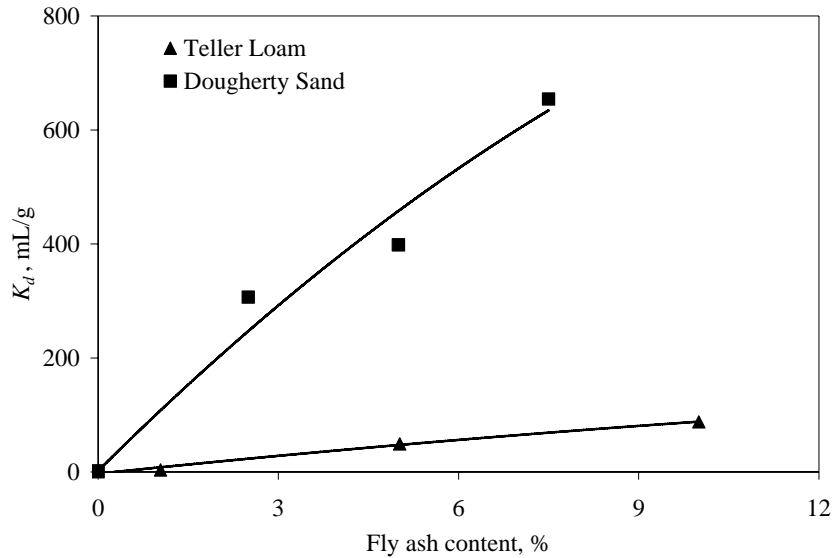


Figure 1. Effect of the fly ash addition on P sorption of soils.

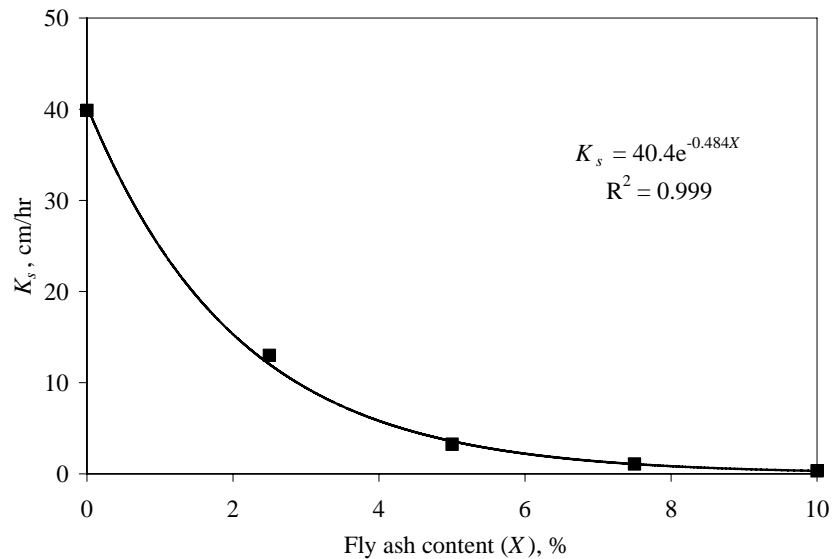


Figure 2. Effect of the fly ash addition on hydraulic conductivity of Dougherty sand.

Sorption Isotherm and Desorption

Sorption data for Dougherty sand, D+5%F, and M-shale fitted both Langmuir and Freundlich equations well ($r^2 > 0.914$). The fitted parameters are listed in Table 2. Dougherty sand with 5% fly ash had the highest Langmuir maximum sorption capacity (S_m) and Freundlich distribution

coefficient (K_f), which were 385 mg/kg, and 203 L/kg, respectively (Table 2). Expanded shale also showed a degree of P sorption capacity with S_m of 82.0 mg/kg, and K_f of 52.9 L/kg.

Table 2. Estimated isotherm parameters for Dougherty sand, D+5%F, and expanded shale.

	Langmuir			Freundlich		
	S_m , mg/kg	b , L/mg	r^2	K_f , L/kg	n	r^2
Dougherty sand	23.8	0.278	0.948	4.93	0.622	0.914
D+5%F	385	2.89	0.998	203	0.295	0.985
M-shale	82.0	3.30	0.997	52.9	0.254	0.986

The results of desorption are shown in Figure 3. Shale desorbed a small amount of sorbed P averaging 6.7% of the initially sorbed P. Dougherty sand with 5% fly ash released the negligible P. However, Dougherty sand released a large amount of P averaging 42% of the initially sorbed P, which means that Dougherty sand cannot provide long-term P storage, and the sorbed P will be desorbed to water flow with low P concentration. Forbes et al. (2004) also reported that Mason sand has little P storage capacity comparing with expanded shale. It was noted that even when the P initial concentration was up to 114 mg/L, D+5%F still released a negligible amount of the sorbed P. Thus, P sorption in D+5%F may be irreversible, and D+5%F can provide long-term P retention. M-shale can also exert a smaller, but significant long-term P retention.

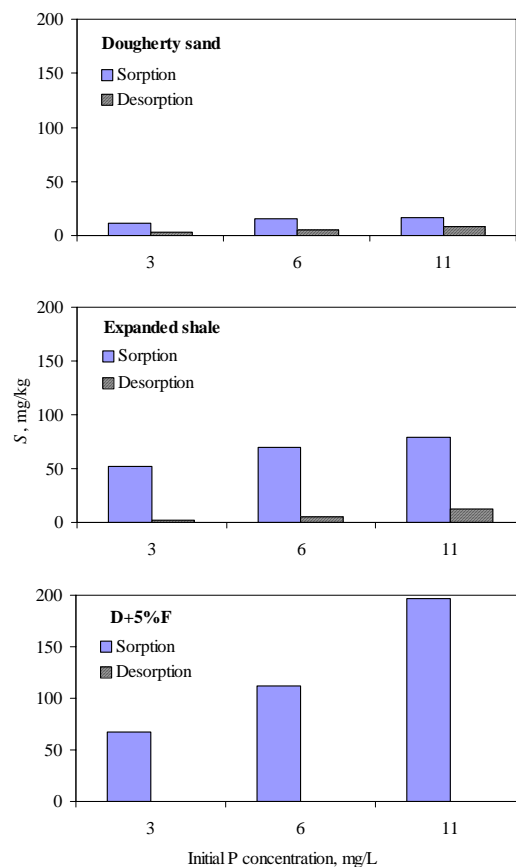


Figure 3. Initial sorption with P concentration ranging from 0.5 mg/L to 10 mg/L, followed by desorption.

Column Flow-through Experiments and Transport Modeling

Total mass of input and output P are summarized in Table 3. Dougherty sand had little P retention with only 2% mass removal. Adding 2.5% and 5% fly ash to Dougherty sand increased the mass removal to 66% and 85%, respectively. Expanded shale exerted 40% mass removal. Overall, a Dougherty sand/fly ash mixture retained a large fraction of input P under continuous loading for more than 300 pore volumes. Phosphorus breakthrough curves are presented in Figure 4. A P flush was observed in the Dougherty sand column, but no P flush in the D+2.5%F, D+5%F, or shale columns. In the BTC of the Dougherty sand column, the P flush only occurred during the approximately first 10 pore volumes, and then the P effluent concentration dropped back close to the influent concentration. Therefore, the amount of this additional P was negligible. The initial P flush may be an artifact of the test procedure. When packing the column, the material was wetted with 9% by weight deionized water to achieve a better packing. Because Dougherty sand contains 14 mg/kg extractable P (Table 1), that P could be released into the pore water and eluted to form the initial peak. The P sorption capacity of D+2.5%F, D+5%F, and M-shale are high, thus P was bound tightly with sorbing sites, and not released into the pore water. It was also noted that the actual BTCs became flat for D+2.5%, D+5%, and shale near the end of the experiments. These leveled-off BTCs may imply a long-term P sorption. Cheung and Venkitachalam (2001) found that fly ash can exert a steady P removal over a longer period. Forbes et al. (2004) reported that shale has a sustained ability of P retention. Thus, the long-term P sorption may prevent Dougherty sand/fly ash mixtures and expanded shale from exhibiting a normal breakthrough.

Table 3. Phosphorus column experiments and transport modeling.

	Dougherty sand	D+2.5%F	D+5%F	M-shale
P input from the influent, mg	141	240	236	249
P output in the effluent, mg	138	82.4	35.2	149
Mass removal, %	2	66	85	40
Bulk density, g/cm ³	1.55	1.76	1.82	0.92
Porosity (θ)	0.415	0.336	0.312	0.653
Pore-water velocity (v), cm/hr	7.14	8.70	9.41	4.53
Residence time ^A , hr	2.0	1.6	1.5	3.2
Solid/water ratio ^B	3.7	5.2	5.8	1.4
Hydraulic dispersion coefficient (D), cm ² /hr	--	43.4	249	404
Retardation factor (R)	1 ^C	199	470	15.7
MSE	--	0.001273	0.005592	0.001356
K_d ^D from column experiments	0	37.8	80.4	10.4
K_d from batch sorption	2.08	307	398	280

^A Calculated by dividing the length of column by pore-water velocity

^B Estimated from the relationship of $2.65(1-\theta)/\theta$

^C Estimated from the actual BCT assuming negligible retardation indicates a retardation factor of 1

^D Calculated based on the relationship of $R = 1 + K_d\rho/\theta$ for a linear equilibrium transport model

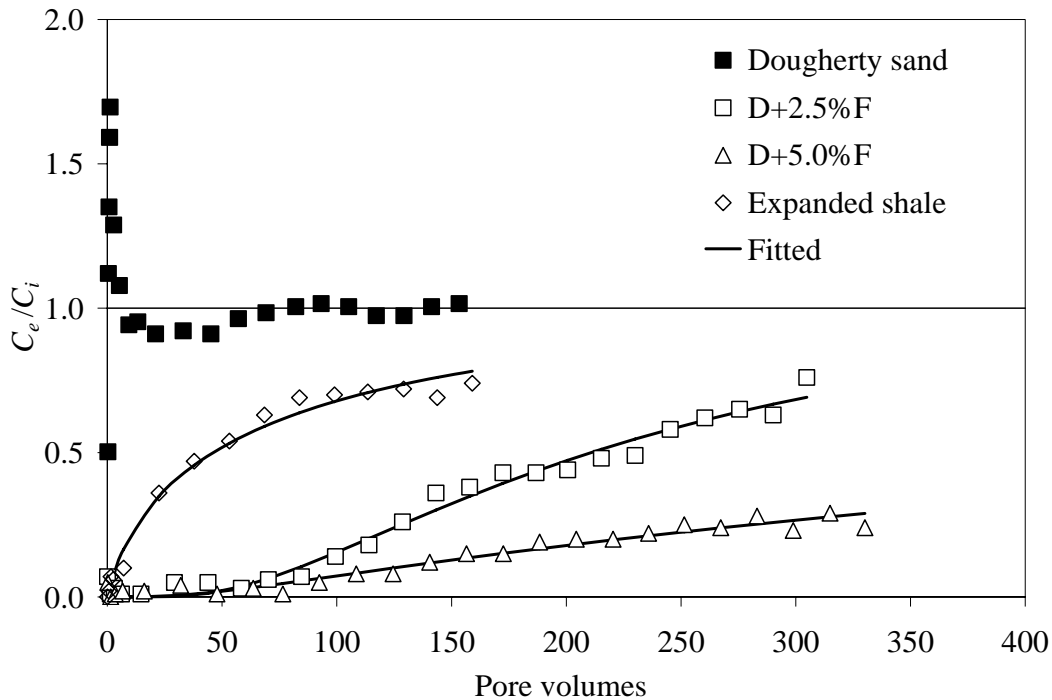


Figure 4. Observed and fitted phosphorus BTCs.

The transport model CXTFIT 2.1 could not fit the BTC in the Dougherty sand column. Judging from the actual BTC, there was no P retardation in Dougherty sand. Thus, the P retardation factor of Dougherty sand was assumed to be 1. The transport model fitted the BTCs of D+2.5%, D+5% and shale columns very well with a highest mean square error (*MSE*) of 0.005592 (Table 4). With the addition of 2.5% and 5% fly ash in Dougherty sand, retardation factors (*R*) were increased to 199 and 470, respectively. The retardation factor of shale was 15.7. In short, it is apparent that D+5% removed P more efficiently than any other filter media.

Comparison was made between distribution coefficients from column experiments and those from batch sorption experiments (Table 3). The batch sorption resulted in much higher K_d . The discrepancy can be explained by the difference in hydrological condition (batch reactor versus continuous flow), and solid/water contact time. Sorption is mainly dependent on solid/water ratio and solid/water contact time. Although the solid/water ratio was increased in the column experiments, the solid/water contact time was decreased greatly. The residence time of water in the columns ranged from 1.5 hours to 3.2 hours (Table 3), far below 24 hours. The lower K_d in the column experiments indicated that the adverse effect of reducing contact time overweighed the positive effect of increasing solid/water ratio. The difference for expanded shale appeared more significant than others, which may be the result of dual porosity factors.

Conclusion

The P removal of bioretention cells has been highly variable due to the diverse properties of soils. Soil amendment with fly ash improved P sorption. However, the addition of fly ash decreased the hydraulic conductivity of soil exponentially. Column flow-through experiments and P transport modeling indicated that the mixture of Dougherty sand and fly ash was more efficient to remove P than Dougherty sand and expanded shale. The incorporation of sand/fly ash infiltration layer in bioretention cells is expected to enhance P removal. However, pilot and field studies are still needed to evaluate the performance of this kind of bioretention cells.

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