Filtered Metals Control in Stormwater using Engineered Media

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ABSTRACT

To address water quality concerns and numeric effluent limits, the designers of bioinfiltration/bioretention systems will need to integrate water and soil chemistry into the selection of filtration media mixtures. For the "dissolved" metals, designers will need to consider the ratio of valence states of the metals as they consider the proportion of ion exchange resins versus organic-based media in the final media mixture. As the correlations between pollutant capacity and soil/media chemistry showed, metals' capacity is directly related to organic matter content and the effective cation exchange capacity of the soil. Available stormwater treatment organic media provides a wide range of treatment sites, but possibly smaller numbers of each site type, compared to ion exchange resins such as zeolites. An activated organic media, such as granular activated carbon (GAC), will have an increased number of surface active sites potentially available for treatment, but this media may not sustain plant growth and may not be desired as a component of bioretention media. Other trade-offs also have to be considered in a complete analysis of a potential media component, such as the trade-off between organic content for plant growth versus nutrient leaching. Finally, there is a lower limit to treatment, after which no further pollutant removal occurs, especially given the contact time requirements based on draindown times required for many bioretention devices. Slightly improved removals for many metals may be achieved with much longer contact times, but the substantial increase in surface area devoted to bioinfiltration may not be cost-effective (given the draindown time requirements). This paper uses "dissolved" copper as an example of how to use both soil and water chemistry to design an optimal bioretention media from a subset of potential mixture components.

KEYWORDS

Urban stormwater runoff treatment; filtered heavy metals; bioretention; biofiltration; soil chemistry; runoff water chemistry

INTRODUCTION

Many guidance documents, published studies, and online performance databases list percent removal rates for stormwater treatment practices. For filtration, infiltration, and biofiltration/bioretention treatment devices, these efficiencies (often extracted from the literature) may range from <30% to almost 100% for pollutants such as metals and phosphorus and the percent removals for these devices appear to vary in the different guidance documents for each state, but are rarely based on regional studies. With the development of Total Maximum Daily Load (TMDL) and other Clean Water Act program requirements to protect stream reaches and lakes around the U.S., it is imperative that reasonably accurate and defensible predictions of treatment device pollutant removals and effluent quality be made. This is not possible with the current guidance, which, in addition to being problematic, is often misapplied. At one site (Pitt et al. 2010), numeric effluent limits on metals, toxic organics, sediments, and other pollutants have been applied through the site's NPDES permit. While the numeric effluent limits for metals may be presented as total recoverable (total) metal concentrations in the effluent water (often based on wastewater total-to-dissolved fractions and not reflecting the greater fraction of particulatebound metals in untreated stormwater), such as on the permit described in Pitt et al. (2010), achieving those concentrations may require removal of both particulate-associated and filtered ("dissolved", or passing through a 0.45-µm filter) pollutants.

"Dissolved" pollutants are considered to be the most bioavailable to the aquatic biota and therefore the greatest toxicity/bioaccumulation concern when they are discharged to receiving waters. For metals, the fraction of a certain metal that passes through the 0.45-µm filter is dependent on the source water chemistry, so a consistent ratio of filtered to total metals is generally not observed (Figure 1). For metals such as copper and cadmium values shown in Figure 1, much of the total metals concentration can be particulate-associated (not filtered through a 0.45-µm membrane filter), while for others, such as thallium and antimony, most of the total concentration was associated with the filtered fraction, although the range of the filtered fraction can be highly variable, even for the same sampling location.



Figure 1. Total (solid fill) versus filtered (striped fill) metals concentration in example stormwater runoff.

Compared to the removal of "dissolved" pollutants, particulate-associated pollutant removal is relatively easy in (bio)(in)filtration systems. As the water passes through the pores of the media, the larger particulates are strained out and trapped in the pore spaces of the media. Assuming that pore-water chemistry does not affect this trapping mechanism such as through repulsive charges, this removal does not depend on the chemistry of the water and media, but on the pore size and the particle diameters. The removal of filtered pollutants, though, is dependent on the chemistry of the influent water and of the media. Laboratory testing and ranking of media has often been based on "synthetic" stormwater where the pollutants, particularly the metals, are in ionic form. However, researchers such as Morquecho *et al.* (2005) have shown that metals that pass through these filters are not just ionic, but also can be associated with colloidal particles (both organic and inorganic) (Table 1). These complexes have different charge states than the original metal ions and therefore will react differently than the metal ion by itself. A media's ability to remove "dissolved" pollutants thus should be expressed not in generic terms but instead as a function of both the soil and water chemistry.

	/				
		% Ionic	% Bound		
Zinc		15	85		
Copper		70	30		
Cadmium		10	90		
Lead		12	88		

Table 1. Fraction Ionic vs. Bound Metals in Filtered Fraction from Source Area Stormwater (data from Morquecho *et al.* 2005).

This paper describes the preliminary investigation of the impact of both water and soil chemistry on pollutant removal. This paper is based on several studies (referenced in the following discussions) of (bio)(in)filtration media. It focuses on generic measures of pollutant removal as a function of the water and soil chemistry. The preliminary conclusions in this paper will be further refined as data from ongoing research projects are produced.

INFLUENT WATER CHEMISTRY

Figure 1 and Table 1 emphasize the challenge of treating metals in stormwater runoff to low effluent concentrations. The fraction of the metals that are associated with particulate matter varies by metal, and as Pitt *et al.* (1999) also noted, by location on the site (Table 2).

	Sample Location							
	Roofs		Vehicle Service Areas		Storage Areas			
Constituent	Ave % Particulate	Ave % Filterable	Ave % Particulate	Ave % Filterable	Ave % Particulate	Ave % Filterable		
Zinc	12	88	40	60	99	1		
Copper	97	3	94	6	14	86		
Cadmium	88	12	97	3	64	36		
Lead	97	3	96	4	97	3		

 Table 2. "Dissolved" vs. Particulate-Associated Metals by Sample Location

In addition, for that fraction of the metal that passes through a 0.45-µm filter, much of that is not ionic, but instead is bound in forms that may be more difficult to treat. The first step in developing treatment trains for metals involves understanding the forms of the metals entering the treatment system. Modeling using Visual Minteq 3.0 and assuming no dissolved organic matter (DOM) in the system (Figure 2) shows that, for copper and zinc, the "dissolved" fraction can have a variety of valence states, ranging from +2 to 0 (for zinc) and through -2 for copper. This run was performed at pH = 7, based on the average pH of urban runoff reported in the literature; had the pH been different, the distribution of valence charges would change. It is anticipated that lowering the pH of the runoff will increase the percentage of the +2 valence charges, since metals tend to be more soluble at lower pH values. The addition of DOM increases the likelihood that the metals are bound into organic complexes.



Figure 2. Zinc and Copper Species Distribution by Valence State.

SOIL/MEDIA CHEMISTRY EFFECTS

In terms of metals removal, soil is composed of many different types of reaction sites, resulting from the natural processes that generate the soil – decomposition of organic matter at the surface and leaching into the lower soil horizons and degradation of the underlying rock through weathering processes. Many generalizations have been made in the literature about chemical behavior of soils based on textural designations; however, soil texture does not define the soil's pollutant removal ability. The soil's two physical properties of interest – porosity and intrinsic permeability – dictate the rate of water transport, and thus dictate the minimum contact time the pollutants have with the medium. If the pollutant can react with the soil in that time, it is reasonable to assume that pollutant removals will be good.

In general, the chemical makeup of soils is the following, with the different horizons having different percentages of each of these components. It is the percentage of these components that affects the number and types of pollutant removal sites available in the soil.

- ♦ Inorganic mineral matter (50 60% in total). This is soil material consisting mostly of oxygen, silicon, and aluminum, plus many other metals in small quantities.
- Organic mineral matter (< 10%). This is soil material derived from plant residues and consisting mostly of carbon, oxygen and hydrogen.

- Solutes. This component is the portion of the soil that includes water and mostly dissolved salts (plant nutrients).
- Air. Soil air is mostly composed of the same gases as the atmosphere, but in different ratios, plus byproducts of decomposing organic matter or from chemical reactions.

Treese *et al.* (2009) highlighted the impacts of soil horizons on stormwater pollutant removal in two natural soils -a silt loam and a loamy sand.

The six primary chemical properties of soil that impact pollutant transport include the following (Evangelou 1998):

- 1. Permanent charge (permanent Cation Exchange Capacity [CEC]) not pH dependent, but are ions that will exchange at all pHs.
- 2. Variable charge (pH-dependent CEC)
- 3. Point of zero charge, ZPC (pH at which the net surface charge is zero or the pH where the CEC minus the Anion Exchange Capacity [AEC] is zero)
- 4. Inner-sphere/outer-sphere complexes (defined as strong surface complexes or innersphere complexes, as opposed to weak surface complexes or outer-sphere complexes)
- 5. Hydrophobic-hydrophilic potential (defined as the potential of soil to adsorb water)
- 6. pH buffering (ability of the soil to resist pH change).

Cation Exchange Capacity. Much of the groundwater protection offered by soils is associated with its ability to remove cationic (positively-charged) pollutants, measured as cation-exchange capacity (CEC) (Pitt, *et al.* 1999; Johnson *et al.* 2003). The CEC of a material is defined as the sum of the exchangeable cations that can be adsorbed at a given pH and is used to evaluate the ability of a soil to attract and retain phosphorus, heavy metals, and other targeted cations of concern. The CEC is pH-specific. As the organic content of the soil increases, so does its CEC content. Natural soils therefore vary widely in their CEC content. Organic soil amendments, such as compost, increase the CEC of a soil that is naturally low in organic material or clays.

It has been proposed that CEC should generally diminish over time in a soil used to treat stormwater runoff because the quantity of exchangeable cations decreases as cations, such as metals, are removed from the infiltrating water. However, as shown in Figure 3, this does not necessary happen for CECe, even when a soil layer has reached capacity (note the capacity of copper for the uppermost layer appears to be reached after 32 storm events [left], yet the CECe is not depleted in this layer [right], but has only undergone a sight decrease).

Anion Exchange Capacity. Similar to the cation exchange capacity, the anion exchange capacity (AEC) is the sum of the exchangeable anions that can be adsorbed at a given pH. It is a measure of the positive charges on the surface of the soil particles that can participate in pollutant removal. The AEC also is pH-specific. AEC values rarely are measured for stormwater filter/biofilter media. In general, soils where the predominant colloids are oxides of Fe and Al

may have a net positive charge and may contribute to ion exchange. According to Troeh and Thompson (2005), anion-exchange sites in soils result from: 1) amine groups in humus, 2) structures terminating in a cation at the edge of a silicate-clay material, 3) OH⁻ ionizing from materials such as Al(OH)₃ or Fe(OH)₃, and 4) isomorphic exchange of Ti⁴⁺ for Fe³⁺ in iron oxides. Anion exchange capacity can increase in a soil as the soil ages and weathering occurs.. AECs are typically low in natural soils.



Figure 3. Breakthrough of copper (left) in a natural soil column and corresponding CECe measurements in the 0 - 3" (0 - 7.5 cm layer)

Mineral Matter. Aluminosilicates, also often known as the clay minerals, make up a large fraction of soil particles < 0.2 mm ($< 200 \mu \text{m}$). Substituting aluminum for silica and substituting divalent cations for the aluminum is common. If the substitution replaces the given coordinating cation with a cation of lower valence, a permanent negative charge or CEC increase occurs (Evangelou, 1998). Although this is likely impractical in stormwater infiltration devices, this material may be regenerable through washing of the media with high concentrations of the readily exchangeable cation. The concern with road salt entering infiltration devices is that sodium may act as an exchangeable cation and wash out previously-captured heavy metals.

Organic Matter Content. Soil organic matter results from two major groups of compounds – the non-nitrogenous compounds (primarily carbohydrates) and the nitrogenous compounds (compounds derived from proteins). Seventy to eighty percent of the organic matter by weight in soils is humic substances (condensed polymers of organic compounds). They have a wide range of molecular weights and a large number of functional groups which participate in reactions in the soil that remove large quantities of pollutants. Heavy metal and soil colloid (including soil clays, humic substances or a combination) interactions result from ion exchange, surface adsorption, or chelation reactions. Humic substances may form complexes with heavy metals because of their oxygen-containing functional groups on their surface. The retention of heavy metals in native soils by the clays and humics varies with ionic strength, pH, clay mineral type, functional group type, and types of competing cations (Evangelou 1998). Commonly, the amount of metal ions adsorbed increases with increasing pH for humics and clays. The reactions that may occur in humic substances include the following:

- Protons (H^+) compete with cations for organic binding sites
- Hydroxyl ions (OH) competes with humic substances for the cationic-metal ion
- Soft metals (Ca^{2+} , Mg^{2+} , etc.) compete with the hard/heavy metals for organic functional groups.

Complexation of metals also is a viable reaction in humic substances. These complexation reactions may be weak (outer-sphere of the particle) and reversible, or relatively strong (inner-sphere). Inner-sphere cations can be displaced only by other cations that can form inner sphere complexes (Evangelou 1998).

Soil pH. As noted above, soil pH plays a strong role in the resultant reactions. Soil pH is a controlling factor for CEC and AEC, which in terms describes the ability of a soil to remove charged pollutants. Soil and water pH also affect precipitation reactions. Many metals form sparingly soluble hydroxide precipitates at certain pHs. These precipitates would be expected to remain in the soil profiles once created, unless the soil and water pH change sufficiently to cause the precipitate to dissolve and the metal to be transported further.

Oxidation-Reduction Potential. Many metals in soil-water systems, such as iron and manganese, have the potential to change oxidation states. In oxidized soil-water systems, the system is electron deficient, and the higher oxidation states are more stable. The reverse is true for reduced environments. The implication for pollutant removal and retention is that the lower valence states of these metals generally are more soluble. This solubility may be mitigated by the presence of ions that will react with these lower valence ions to form insoluble compounds (Evangelou, 1998).

The impact of redox chemistry on pollutant retention is especially important as infiltration devices are generally quiescent between storms. Water remains in the pores and microbial activity may consume the oxygen, changing the water from aerobic to anaerobic conditions. This may result in the release of previously-captured pollutants. Also, adsorption under anaerobic conditions has been shown to lead to weaker bonding between phenolic compounds and activated carbon (Abuzald and Nakhla, 1994). As shown in Johnson *et al.* (2003), Clark (2000), and Clark and Pitt (2009), heavy metal retention, except for iron, was good under anaerobic conditions, but previously-trapped nutrients were released from the organic media.

Impact of Soil Chemistry on Pollutant Capacity as Measured in Column Tests. Table 3 summarizes the results of a Pearson correlation used to investigate potential relationships between soil chemistry parameters, as reported by the Pennsylvania State Agricultural Services Lab, and the batch-tested filtered-metal capacities of six stormwater treatment media. The analytical methods for this work are described in Pitt *et al.* (2010). The significance level for this correlation matrix was set at 0.10, since the number of columns, and thus data points, is much smaller than for the soil results reported above.

Compared to the batch testing, the CECe is not correlated with the estimated media capacities for the listed metals. Organic matter is correlated only with Sb. Soil C, though, is correlated with the capacities for Cd, Cu and Ni. In the batch testing (data not shown), C and OM almost tracked each other in terms of their relationships with a specific metal's capacity. In the column study, though, this was not the case. One possible reason for this is the subset of media chosen for this

experiment: two sands, two zeolites, a coconut based granular activated carbon (GAC), and peat moss. The batch testing added several other organic media to the testing, although the percentage of organic media did not change between the column testing and batch testing. GAC typically is heat and/or acid activated and much of the organic matter is removed, leaving inorganic carbon. In the smaller data set, the use of only one organic media may have affected whether OM was critical in predicting capacity. In this series of column tests, soil Cu content was negatively correlated with zinc capacity. In the batch testing, soil Cu content had no impact on zinc removal. Soil Mg also demonstrated less of an influence on capacity.

	Water-Phase Metals Removal Capacity									
	Cd	Cu	Fe	Рb	Ni	Мn	Zn	cr	ΤI	Sb
Soil CEC _e										
Soil OM										+
Soil C	+	+			+					
Soil N	+				+					
Soil P										
Soil pH										
Soil K										
Soil Mg										+
Soil Cu							-			
Soil Zn										+
Soil Ca				-						

Table 3. Correlations between Chemistry Parameters of Stormwater Treatment Media and

 Pollutant Removal Capacity.

Treese *et al.* (2009, 2010) (Figure 3) showed that for the upper organic layer of the soil, breakthrough occurred when the soil Cu content was 15 mg/kg. However, using the batch tests, the calculated capacity was approximately 40 mg/kg for the same soil. The functional capacity, or the capacity at which substantial downward migration of the saturation front could be expected, should be taken as 15 mg/kg, and not the 40 mg/kg. This also highlights the challenges of translating simple batch laboratory tests to field applications. Batch testing typically encourages equilibrium conditions and provides time for the pollutants to migrate into the media, plus attach to the surface sites. Column tests in stormwater field operations, where the media are

free-draining, rarely provide sufficient contact time for pollutants to migrate to the interior pores of the media. Only the surface sites are readily used for pollutant removal.

Trade-Offs. Pollutant removals in (bio)(in)filtration media generally occur through ionexchange reactions. Therefore, it is important to know what ions are being exchanged during pollutant removal and to determine whether these exchanged ions are problematic in the effluent water and discharge location. An example of the trade-offs in pollutant capture versus ion export is shown in Figure 4 for a GAC-sand column of varying depths. In this case, the capture of cadmium was excellent and was not a function of column depth. However, deeper columns results in greater export of potassium from the media, likely as a result of increased ion-exchange reactions occurring in the media due to increasing media contact as the residence time increases. These ion exchange reactions likely involve the capture of the metals as well as the capture of the hydrogen (hydronium) ion, since the pH of the effluent water was at least 1 pH unit higher than the influent water for these columns.

Based on the results in Figure 4, it appears that a treatment media depth of 14 inches would be ideal since cadmium capture is not a function of depth in the ranges investigated during the supporting study, and it would minimize export of potassium from the media. However, this is not true for all pollutants. This GAC provided excellent capture of nitrate until its removal capacity was saturated. Increasing the GAC depth increased nitrate capture substantially, but it also increased phosphate leaching.



Figure 4. Ion exchanges for a coconut-hull GAC-sand mixture as a function of column depth for cadmium (left) and potassium (right).

These trade-offs greatly affect the design of (bio)(in)filtration devices. While metals may be captured well, as noted above, these trade-offs affect the capture or release of other pollutants. Figure 5 highlights these trade-offs for nitrate and phosphate with various percentages of the

coconut-based GAC and peat moss in the column. Another trade-off that has been addressed in the literature has been the fraction of organic matter that is incorporated into the design. Treese *et al.* (2010) describe how the disturbance of native soil profiles results in initial high releases of nutrients. Hunt *et al.* (2006) state that the use of compost in bioretention should be limited to low-phosphorus compost. Compost whose phosphorus content exceeds the needs of the plants grown in it will leach phosphorus in the passing stormwater runoff and will export both nitrogen and phosphorus. Pitt, *et al.* (1999) showed that compost-amended soils exported substantialy greater amounts of phosphorus compared to unamended soils, but only for about three years.



Figure 5. Trade-offs during design of (bio)(in)filtration systems – nitrate capture versus phosphate release.

DESIGNING A (BIO)(IN)FILTRATION SYSTEM TO TREAT "DISSOLVED" HEAVY METALS

Filtered or "dissolved" copper is used as an example of how to integrate the water chemistry and soil media information to design a (bio)(in)filtration system. Assuming a runoff water pH of approximately 7, filtered copper can exist in multiple forms. If no DOM is in the runoff, it can be assumed that approximately 25% will have a +2 valence charge, 10% will have a +1 valence charge, 65% will have a 0 valence charge and less than 1% will have a -2 valence charge. With DOM in the system, it could be assumed that approximately one third of the copper will be associated with organic complexes (and the percentages described above would be that portion that was not associated with organic complexes).

A valence charge of +2 indicates that ion exchange may be viable for that portion of the copper. Therefore, an ion exchange resin such as a zeolite could be considered. However, a valence charge of +1, particularly for a copper hydroxide complex, may not be readily removed by ion exchange. The complex is likely to be larger than most of the lattice openings on traditional ion exchange resins such as zeolites. If the percentage of -2 copper was greater, then anion exchange may need to be considered. The valence charge of 0 certainly will not be removable by ion exchange and therefore, other removal mechanisms need to be considered. The valence charge of 0 and +1 may need to be removed through the use of a media that participates in what appear to be non-charge reactions. The molecules themselves may not have a valence charge but may have a dipole and participate in weak ionic bonding to the media or they may be bound by other mechanisms that are not well characterized.

Applying this to the selection of media, the batch capacity testing indicates that copper capacity is related to CEC_e , OM content, soil C content, soil N content, and soil Mg content. Table 3 from the column capacity testing indicates that capacity is related to soil C. CEC_e and soil Mg content relate to the ability of the media to participate in ion exchange reactions. Therefore, at least one component in a media mixture should provide excellent ion exchange, such as would be found with a good zeolite. This media should be able to participate in reactions with the +2 copper and a portion of the +1 copper, although the +1 copper may not be as strongly bound and may be displaced if a more preferable exchangeable ion approaches the media's removal site. Soil OM, soil C, and soil N all relate to the organic matter content and indicate that these are sites that may participate in a variety of reactions and may be able to remove pollutants that do not carry a valence charge. Therefore, an organic component should also be incorporated. Sand may be needed for structure (to minimize compaction) and for controlling the flow rate to a level that allows for sufficient contact time.

These results do not address the effects of the biological component of biofiltration systems. They focus on the media results only. It is anticipated that the addition of plants to these systems may improve their ability to capture and retain "dissolved" pollutants, but that much of the effects of the plants will be on nutrients. Metals capture likely will continue to be in the media itself and until the surface media capacity is exhausted, it likely will occur in the top 3 cm of the bioinfiltration system.

CONCLUSIONS

To address water quality concerns and numeric effluent limits, the designers of bioinfiltration/bioretention systems will need to integrate water and soil chemistry into the selection of filtration media mixtures. For the "dissolved" metals, designers will need to consider the ratio of valence states as they consider the proportion of ion exchange resins versus organic-based media in the final media mixture. As more of the total metal concentration has either a 0 or +1 valence charge or as more is associated with organic complexes, the smaller the fraction of an ion exchange resin, such as a zeolite, are needed. For metals such as thallium, where few inorganic and organic complexes are formed and where the predominant valence state is +2, increasing the amount of zeolite in the final media mixture is important for improving removal. Therefore, the final media mixture will be based on the pollutants of interest and their water chemistry. As the soil chemistry correlations showed, capacity is directly related to OM and CECe content for many metals. Since organic matter will contribute to CECe, this is not

surprising. The organic media, though, provides a wider range of treatment sites. Activating an organic media, such as granular activated carbon, will increase the number of surface active sites for treatment, but this media will not sustain plant growth.

Other trade-offs also have to be considered in a complete analysis of these media. As described briefly in this paper, for the media evaluated in Pitt *et al.* (2010), increasing the amount of the media to improve nitrate removal and retention, but also increases the amount of phosphate released. The tradeoff may be based on an optimization analysis between the damage caused by reduced nitrate treatment and the additional release of phosphate. Finally, there is a lower limit to treatment, given the draindown times required for many bioretention devices.

Slightly improved removals may be achieved with longer contact times for many metals, but the substantial increase in surface area devoted to (bio)(in)filtration may not be cost-effective (given the draindown time requirements). This paper used "dissolved" copper as an example to show how to use both soil and water chemistry to design an optimal bioretention media from a subset of potential mixture components.

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