Retention of heavy metals by stormwater filtration systems: breakthrough analysis

Rétention par filtres des métaux lourds dans les eaux de ruissellement: analyse de courbes de rupture

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RÉSUMÉ

Les ouvrages de filtration, surtout ceux utilisant la biofiltration, sont largement utilisés pour protéger les milieux aquatiques des impacts des rejets urbains des temps pluvieux (RUTP), mais leur capacité à retenir les métaux lourds à long terme reste largement inconnue. Cet article présente une étude en laboratoire où des courbes de rupture sont créées pour trois types de biofiltres typiques à base de sol. Dans tous les cas, une rupture est observée pour le zinc (Zn), mais pas pour le cadmium (Cd), cuivre (Cu) ou plomb (Pb). Si un filtre est dimensionné de façon assez large par rapport à son bassin versant (au moins 2-3% de sa superficie), avec une profondeur d'au moins 0.5 m, il n'y aura pas de rupture de ces métaux lourds avant 10 ans (et probablement plus longtemps). Toutefois, après l'équivalent de 12 à 15 ans d'opération, les concentrations en Cd, Cu et Zn dans le sol ne respectent plus les lignes directrices pour la protection du milieu naturel et de la santé humaine. Il est donc possible qu'une fois usagé, le sol d'un tel filtre doive être classifié comme un sol contaminé, et donc soumis à un traitement spécifique avant élimination.

ABSTRACT

Fine media filtration systems, particularly biofiltration systems, are widely used to mitigate the impacts of stormwater on receiving waters, however their long-term capacity to retain heavy metals has not previously been assessed. Accelerated-dosing laboratory experiments were used to assess the likelihood of breakthrough occurring for three different types of soil-based filter media that are commonly used in stormwater biofilters. In all cases, breakthrough of zinc (Zn) was observed, but not of cadmium (Cd), copper (Cu) and lead (Pb). If filtration systems are sized so that they are large relative to their catchment (at least 2-3% of its area) or have a deep filter layer (at least 0.5 m deep), then breakthrough will not occur for at least ten years and probably longer. However, after the equivalent of 12 – 15 years of operation, Cd, Cu and Zn had accumulated in the filter media to levels that exceeded human health and/or ecological guidelines. Further, depending on the design, it is possible that spent filter media may be classified as contaminated soil and thus require special disposal.

KEYWORDS

Biofiltration, bioretention, heavy metals, soil, treatment capacity, urban runoff

1 INTRODUCTION

The negative impacts of hydraulically efficient urban drainage systems on receiving waters are widely recognised, and include problems such as increased peak flows, runoff volumes and pollutant concentrations (Novotny and Olem 1994; Paul and Meyer 2001; Walsh *et al.* 2005). Fine media filtration systems, particularly those that are vegetated (commonly known as biofiltration systems, bioretention systems, and rain gardens), are an increasingly popular technology for mitigating the deleterious effects of urbanization on the ecological health of receiving waters. These systems are engineered to reduce runoff peaks and volumes (Hatt *et al.* 2009), and improve water quality via mechanisms such as sedimentation, filtration, and sorption, as well as biological uptake if the filter is vegetated.

A number of studies at both the laboratory- and field-scale have demonstrated that fine media filters are highly effective at removing heavy metals from stormwater (e.g. Dietz and Clausen 2006; Davis 2007; Hatt *et al.* 2008). Sedimentation and filtration processes effectively remove particulates (Davis *et al.* 2009) and thus the particulate phase of heavy metals. It is expected that removal of particulate-bound heavy metals will continue until the end of a filter's useful life, which is generally assumed to occur when the surface becomes clogged. However, a considerable fraction of heavy metals in urban runoff can be present in dissolved form (Dean *et al.* 2005), for which chemical sorption is an important removal mechanism (Davis *et al.* 2009). It is possible that the adsorption capacity of the filter media may be exhausted before surface clogging occurs, resulting in breakthrough of heavy metals. If breakthrough occurs, the capacity of the system to retain dissolved heavy metals will be compromised and may result in unacceptably high concentrations of heavy metals being discharged to receiving waters. Dissolved heavy metals are more bioavailable than particulate-bounds metals and thus may be more detrimental to aquatic ecology (Kominkova and Nabelkova 2007), hence it is particularly important to ensure long-term retention of dissolved heavy metals.

Previous studies of the treatment performance of fine media filters have been relatively short-term and thus have not been able to assess the likelihood of breakthrough occurring. The objective of this work was thus to examine if and when breakthrough is likely to occur, as well as to assess the level of soil contamination in biofilters after many years of operation. This laboratory study is the first step in a multi-stage project. In this experiment, the filter media was tested in isolation (separate from any effects of vegetation). The performance of these filter media in combination with vegetation will be tested in a subsequent experiment.

2 METHODS

A fully automated one-dimensional experimental rig was used to conduct accelerated dosing breakthrough tests (Figure 1, left). The rig consisted of three 50 L polyethylene tanks, from which stormwater was pumped at a controlled rate into six threaded glass columns (Chromaflex, Multi-Lambda Scientific, 2.5 cm diameter x 15 cm long) in upflow mode. The experimental approach was based on techniques commonly used in groundwater studies and these columns were chosen based on a review conducted by Smith and Dillon (1997). While the columns are small relative to the size of a field-scale filter, a small column size was necessary to achieve breakthrough in a reasonable timeframe. In general, problems with upscaling from laboratory to field conditions occur due to homogeneous laboratory conditions compared to heterogeneous field conditions. However, given that soil-based stormwater filters are engineered systems, they are relatively homogeneous at both the laboratory- and field-scales. Outflow from the columns passed through in-line electrical conductivity (EC) meters (HI 7635 with HI8636CL, Hanna Instruments) into tipping bucket rain gauges (RG 12, Environdata), which were used to measure flow.

To ensure even flow, each end of the columns contained a 20 µm porosity HDPE bed support, 2.5 cm of acid-washed medium quartz sand (Unimin, 16/30 FD, ~700 µm) and 2 cm of acid-washed fine filter quartz sand (Unimin, 50N, ~100 µm), which left a height of 6 cm for filter media (Figure 1, right). Prior to packing, the filter media were air-dried then sieved. The columns were filled with water during packing to avoid air bubbles. The sand and filter media were added in 2-3 mm lifts and compacted lightly in between. The pore volume of each column (i.e., the volume of open space) was calculated as the difference between the total column volume and the volume of filter media added.

Three different types of filter media that are commonly used in biofilters were tested in two separate experiments (Table 1). In the first experiment, three replicates of loamy sand (LS) and loamy sand mixed with 10% vermiculite and 10% perlite (by volume, LSVP) were tested. In the second experiment, duplicates of LS, LSVP and loamy sand mixed with 10% leaf compost and 10% mulch (by volume, LSC) were tested.

Semi-synthetic stormwater was used in the first experiment; the rationale for use and method of preparation has been described previously (Hatt *et al.* 2008), however in this case the stormwater was passed through a 75 µm filter to avoid blocking the inflow tubing and columns. Target heavy metal concentrations were Cd 4.5 µg/L, Cu 50 µg/L, Pb 140 µg/L and Zn 250 µg/L based on a stormwater quality review conducted by Duncan (1999). Stormwater was stored in the inflow tanks and continuously mixed using recirculation pumps. However, despite pre-filtering, there were problems with blocking in the first experiment, therefore in the second experiment stormwater was prepared using tap water and laboratory-grade chemicals only, and did not contain any sediment. Stormwater was continuously pumped into the columns at a rate of 330 mm/hr, which is representative of the target drainage rate for biofiltration systems (100-300 mm/hr, FAWB 2009). The total run time was 11 and 13 weeks for the first and second experiments, respectively.

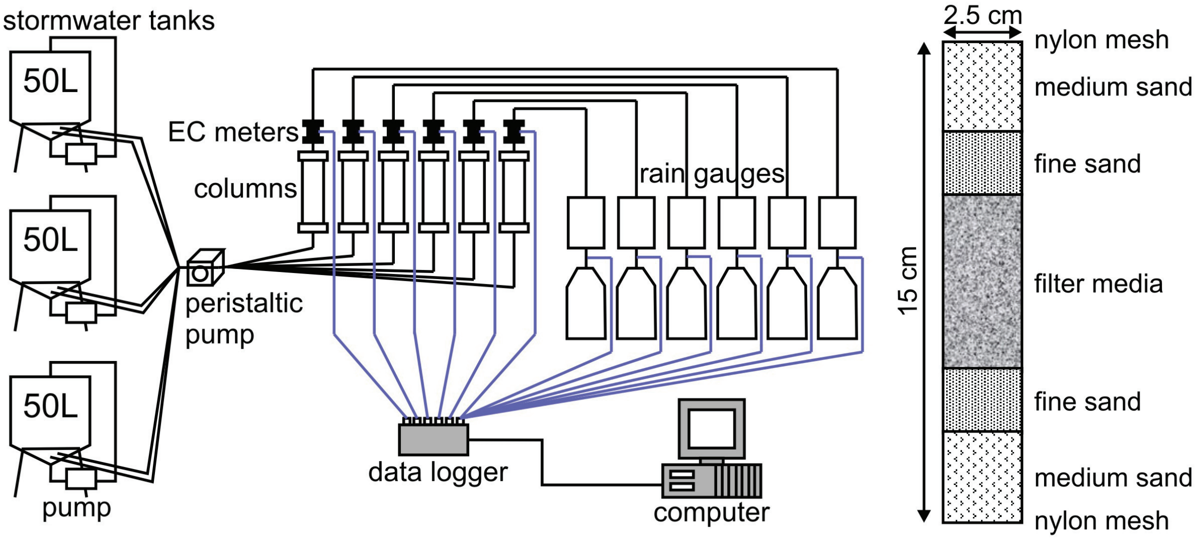

Figure 1. Schematic diagram of experimental rig (left) and one-dimensional test column (right).

Table 1. Filter media characteristics.

Filter media	TOC (mg/kg)	$CaCO_3$ (mg/L)	CEC (meq/100 g)	Cd (mg/kg)	Cu (mg/kg)	Pb (mg/kg)	Zn (mg/kg)
LS	41	138	17	< 0.5	< 5	< 5	13
LSVP	19	167	15	< 0.5	< 5	< 5	13
LSC	13	217	21	< 0.5	< 5	< 5	17

Inflow water quality samples were collected at 150 L intervals. Outflow water quality samples were collected after the equivalent of 0.5, 1, 2, 4, 7, 10, and 15 years of flow had passed through the columns. The annual flow volume was calculated based on a biofilter sized at 2% of its impervious catchment area and the mean annual rainfall for Melbourne, Australia (mean annual rainfall: 648 mm, BOM 2009), which equates to 13L inflow. Water quality samples were collected before the rain gauges to avoid contamination (and weighed to allow mass balance calculations) and analysed for cadmium (Cd), copper (Cu), lead (Pb), and zinc (Zn). All samples were acid-digested and analysed on an ICP-OES using standard methods and quality assurance procedures.

At the end of each experiment, where the end-point was approximately equivalent to 15 and 12 years of flow in the first and second experiments, respectively, the filter media was carefully removed from the column and divided into three equal parts from the top to the bottom, homogenised and sub-sampled. Sediment samples were acid-digested and analysed on an ICP-OES using standard methods and quality assurance procedures.

3 RESULTS AND DISCUSSION

3.1 Breakthrough

Inflow Cu concentrations were up to five times higher than typical stormwater concentrations in the second experiment, therefore only results from the first experiment were analysed for Cu breakthrough. Outflow concentrations of Cd and Pb were almost always below the instrument detection limit (3 µg/L, for Cd and Pb), thus results for these metals are not presented. The pH was relatively neutral for the duration of both experiments and did not change from inflow to outflow, where the mean ± one standard deviation was 6.3 ± 0.3 and 6.4 ± 0.4 for the inflow and outflow, respectively. In all cases, flow decreased with time due to blocking of the tubing and at the column inlets, even though the stormwater was pre-filtered. The rate at which flow decreased varied between filter media types, therefore outflow concentrations are presented as a function of pore volume (i.e., cumulative inflow volume (L) divided by the pore volume (L)) to enable direct comparison of results.

Zn breakthrough occurred in the LSVP filter media first, followed by LS and LSC (Figure 2). The number of pore volumes to breakthrough was estimated as 1500, 2000, and 2500 for LSVP, LS, and LSC, respectively; conservative estimates were made because the EC meters caused mixing of the outflow solution and likely resulted in retarded breakthrough. This order in which Zn breakthrough occurred followed the cation exchange capacity CEC of each filter type (Table 1). There was no Cu breakthrough for the entire duration of the experiment (Figure 3). Given that Zn has a higher mobility than the other metals considered here, it is unsurprising that Zn breakthrough occurred first.

3.2 Accumulation of heavy metals

As mentioned in the previous section, inflow Cu concentrations were up to five times higher than typical stormwater concentrations in the second experiment, therefore only results from the first experiment were assessed for pollutant accumulation. Inflow concentrations of Zn, Pb, Cd were comparable between experiments as well as consistent with typical stormwater concentrations, therefore combined results are presented for these metals.

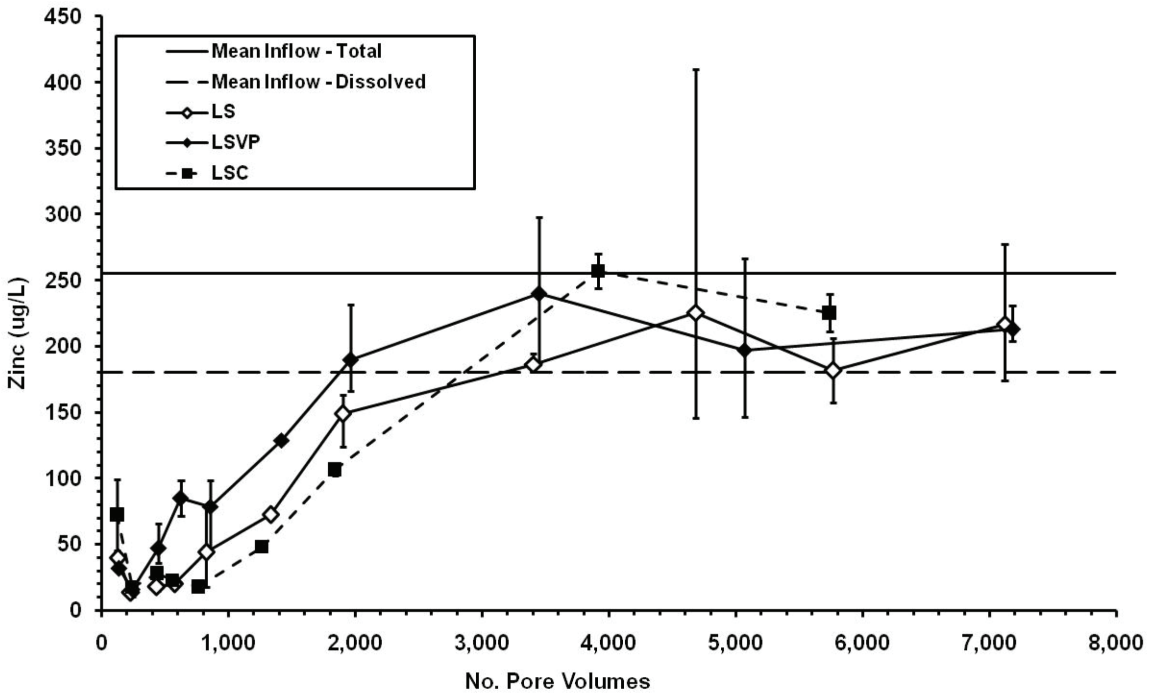

Figure 2. Experimental breakthrough curve for Zn and three filter media types. Mean outflow concentrations (n = 5: LS, LSVP, n= 2: LSC. Error bars indicate the range of observed values.

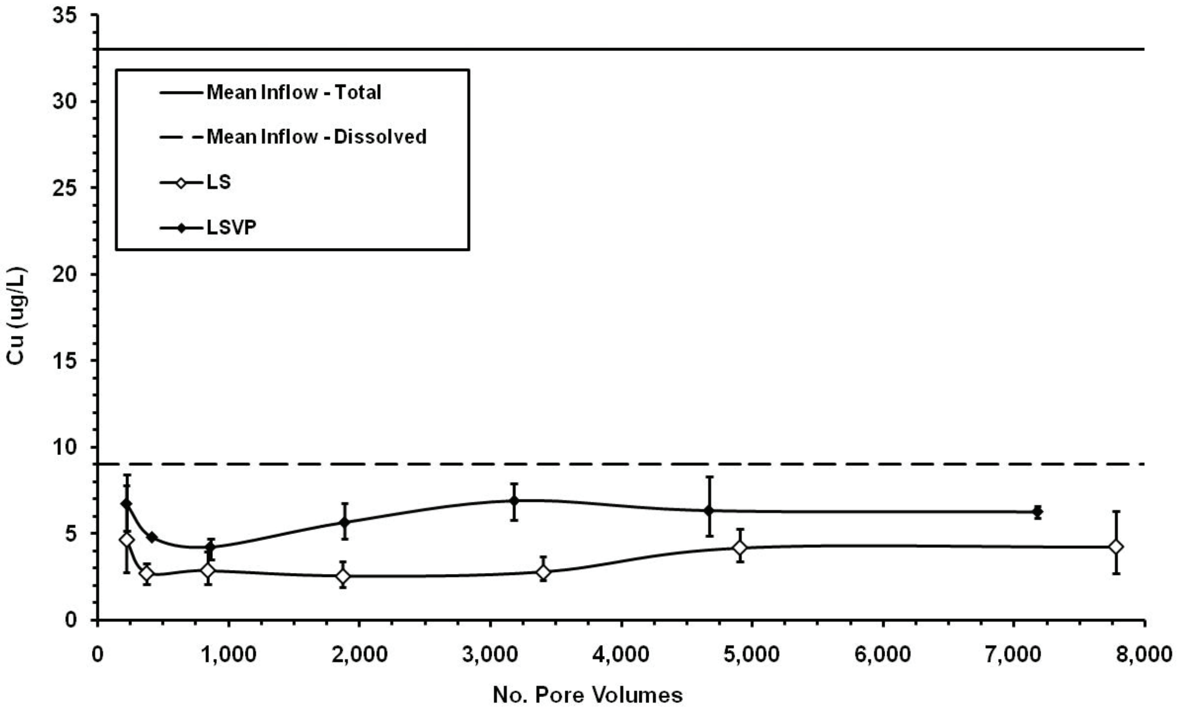

Figure 3. Experimental breakthrough curve for Cu and two filter media types. Mean outflow concentrations (n = 3). Error bars indicate the range of observed values.

Concentrations of Cu and Pb were generally highest in the first layers of the filter media profile and decreased through the profile, however there was a relatively even spatial distribution of Cd and Zn (Figure 4). In all cases, an increase in concentrations relative to raw filter media concentrations was observed throughout the profile (Table 1, Figure 4). However, given that these columns are only 6 cm, whereas the typical depth of fine media filters is at least 30 cm, these columns really only represent the surface layers. A previous study of pollutant accumulation in soil-based filters showed that heavy metals were largely trapped at the surface of the filters and rapidly decreased through the first 10 cm (Hatt *et al.* 2008), therefore it is not entirely unsurprising to observe this migration of metals through the profile here. Regardless, it can be seen that, after an equivalent of 12 – 15 years of operation, Cd, Cu and Zn levels exceeded ecological guidelines and Cd concentrations also exceeded human exposure guidelines (based on land use for parks, recreational open space and playing fields, NEPC 1999). Given that the majority of stormwater filtration systems are installed in areas that are accessible to the public, this finding suggests that management is required to prevent metals accumulating to levels of concern, as discussed in further detail below.

3.3 Practical implications

It is important to note that, although adding compost and mulch (i.e. organic matter) to the loamy sand increased the CEC and presumably delayed breakthrough of Zn, it has previously been shown that the use of filter media containing compost and mulch can result in significant leaching of nutrients (Hatt *et al.* 2008). Therefore, if removal of heavy metals and nutrients were both important, care would need to be taken to ensure a low-nutrient form of organic matter was used.

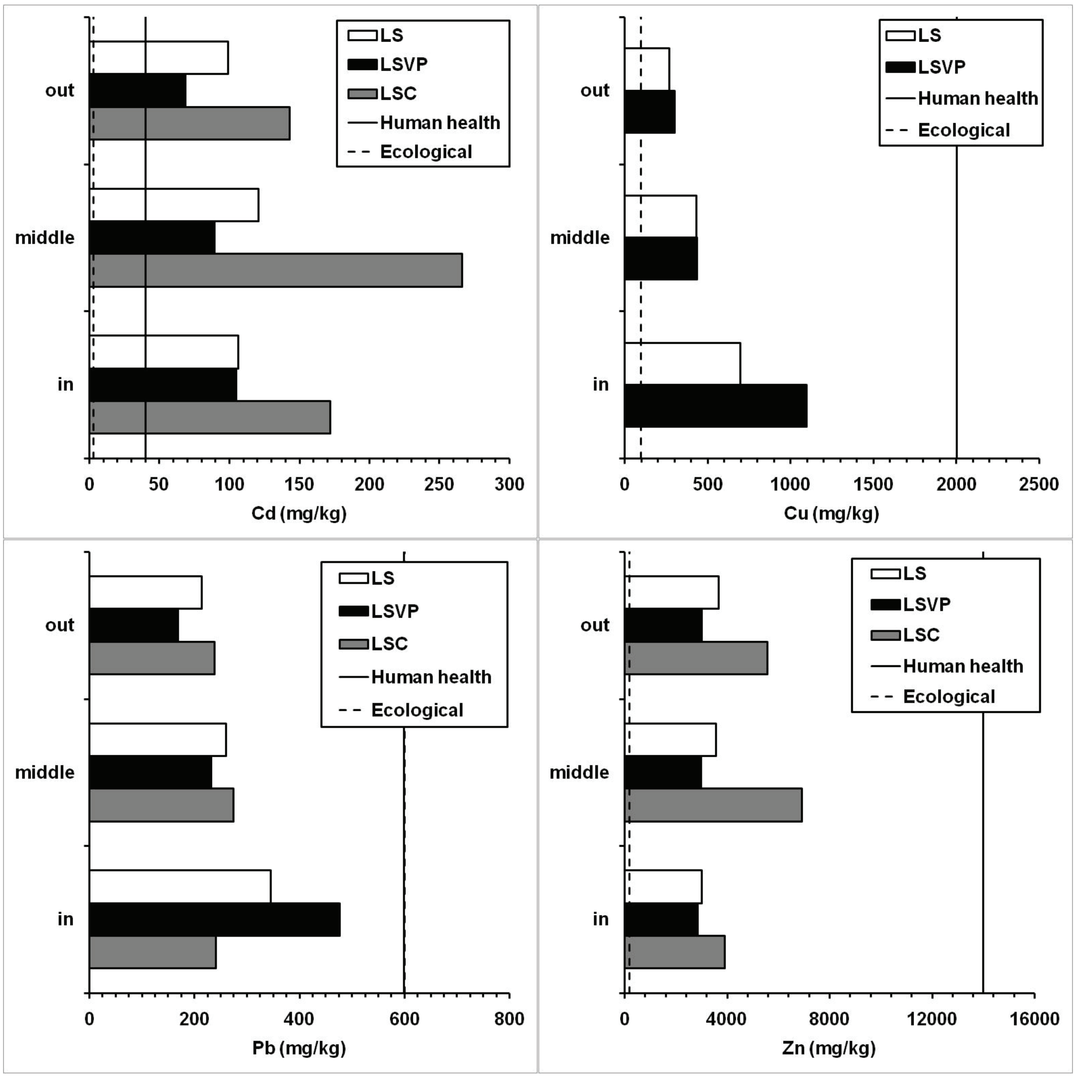

Figure 4. Spatial distribution of four heavy metals through filter media profile. Mean concentration: n = 5 (LS, LSVP for Cd, Pb, Zn), 3 (LS, LSVP for Cu), and 2 (LSC for all metals). Concentrations include native filter media concentrations. The vertical solid line indicates the guidelines for human health and the vertical dotted line indicates the guideline for ecological health (NEPC 1999).

3.3.1 Predicted years of operation

The predicted time until Zn breakthrough occurs is presented in Table 2 for two Australian cities in different climates, and a range of filter media depths and sizes relative to the impervious catchment. These calculations are based on the assumption that dissolved metals will be primarily adsorbed in the surface layers until the exchange sites in this zone are fully occupied, at which point dissolved metals will migrate into the next layers and be trapped, and so. It can be seen that breakthrough will occur quickly if a filter is small relative to its catchment or has a shallow filter media depth. Such a filter would have high associated maintenance requirements as it would be necessary to regularly remove the spent filter media, and then reset the system. In the case of biofilters, revegetation would also be required, which would compromise the treatment capacity while the system re-established, particularly with respect to nutrient removal, which typically requires mature vegetation to be effective (Bratieres *et al.* 2008). On the other hand, if the filter is large relative to its catchment or the filter media is deep, then it may be many years until breakthrough occurs (Table 2). Given that it is has been identified that designing large systems is also important for maintaining an adequate long-term infiltration capacity (Le Coustumer *et al.* 2009), it is recommended that the filter area be increased in preference to using a deeper filter media layer wherever practicable. Consideration of breakthrough in the design process is particularly important for high rainfall areas, such as Brisbane, where breakthrough has the potential to occur very quickly. It is important to note that these results are considered to be indicative of the worst-case scenario because the columns were operated in upflow mode, hence there was no ponding of water prior to percolation through the media. Under normal operating conditions, stormwater temporarily ponds on the surface of a filter, which facilitates sedimentation of particulate-bound metals. Finally, water was pumped through the filter columns at a rate of 330 mm/hr, which is at the upper end of the range of recommended drainage rates for filter media (FAWB 2009) and thus the contact time between the water and filter media was likely to be lower than normal. Nevertheless, it would be pertinent to use these results as conservative estimates of breakthrough.

Table 2. Predicted time to breakthrough of Zn for filters in two different climates and with varying filter media properties, depth, and size relative to the catchment. Melbourne: temperate climate, annual rainfall = 648 mm. Brisbane: sub-tropical climate, annual rainfall = 1149 mm (BOM 2009).

		Years to breakthrough							
		Melbourne				**Brisbane**			
		% of impervious catchment				**% of impervious catchment**			
Media	**Depth (m)**	**1**	**2**	**3**	**5**	**1**	**2**	**3**	**5**
LS	0.3	4	9	14	23	2	4	6	10
LS	0.5	7	15	23	39	3	7	10	18
LS	0.7	10	21	32	54	5	10	15	25
LSVP	0.3	3	7	10	17	1	3	4	8
LSVP	0.5	5	11	17	29	2	5	8	13
LSVP	0.7	8	16	24	41	3	7	11	19
LSC	0.3	5	11	17	29	2	5	8	13
LSC	0.5	9	19	29	48	4	9	13	22
LSC	0.7	13	27	41	68	6	12	19	31

3.3.2 Disposal of spent filter media

All three filter media types were classified as Category B contaminated soil at the end of their useful lifespan (Table 3). If filter media is classified as fill material, no license is required to dispose of this material, although reuse must not give rise to environmental or health impacts. If material is classified as Category B or C, the material must be disposed of to a licensed landfill using a certified transport system. If it were a priority to avoid special disposal of spent filter media, then the biofilter should be sized at 3 – 5% of the impervious catchment. Further, if the top 2 – 5 cm of the filter media were to be scraped off and replaced at two year intervals (as recommended for avoiding surface clogging, e.g. Urbonas 1999; Hatt *et al.* 2008), then it is entirely likely that special disposal would not be required, particularly given that these results are considered worst-case scenario, as discussed above. This would also prevent metals accumulating in the system to the point that they were of concern to human and ecological health. We stress that the possibility that filter media may be classed as contaminated soil at the time of disposal should not be viewed as a failure, however it is a management issue that asset owners need to be aware of.

Table 3. Maximum heavy metal concentrations in three filter media types and classification of soil according to level of contamination. [a](EPA Victoria 2007).

		Cd	**Cu**	**Pb**	**Zn**
max concentration in filter media after 12 – 15 years of runoff application (mg/kg)	LS	166	13577	533	4496
	LSVP	118	12641	794	3596
	LSC	266	19111	274	6920
max concentration allowed in soil to be classified as (mg/kg)[a]	fill material	3	100	300	200
	Category C	100	5000	1500	3500
	Category B	400	20000	6000	140000

4 CONCLUSIONS

If designed well, fine media filtration systems will effectively remove both particulate-bound and dissolved heavy metals from stormwater in the long term. Breakthrough of Zn, a relatively mobile heavy metal, can occur, however it is not expected that breakthrough of Cd, Cu, Pb will occur during the operating life of a filter (i.e., breakthrough of Zn or physical clogging will occur first). It is important to size filters so that they are large relative to their impervious catchment or, if this is not feasible, specify a deep filter layer to avoid breakthrough. Use of a filter media with a high cation exchange capacity will also delay breakthrough. If these design considerations are taken into account, it is likely that physical clogging will occur before breakthrough. However, filter media will almost definitely exceed guidelines for human and ecological health at the end of a filter's operating life, and may also be classified as a contaminated soil, thus requiring special disposal.

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