



# Geochemical modelling of heavy metals in urban stormwater biofilters



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## ABSTRACT

Complex metal compounds from urban wet deposition can be retained in stormwater biofilters and degraded to simple inorganic metal compounds. A mechanistic model that describes the bulk breakdown rate, accumulation, and leaching of copper (Cu), lead (Pb), and zinc (Zn) is presented. The model accounts for aqueous complexation, mineral adsorption, and kinetic methylation of these three key metals found in urban stormwater. The model was tested against experimental pollutographs from inflow and outflow of a stormwater biofiltration system over a period of 100 days. Parameter calibration resulted in  $R^2 \geq 98\%$  and residuals lower than 12% against cumulative effluent water and metal mass. The leaching concentration of Cu and Pb was linearly correlated to the hydraulic conductivity as well as equilibrium and kinetic rate constants; Zn leaching concentration was correlated by a power law to these parameters. It was found that  $\pm 20\%$  change in these parameters returned changes in Cu, Pb and Zn concentrations within about  $\pm 52\%$ ,  $\pm 45\%$  and  $\pm 96\%$ , respectively. Scenarios including inflow events with different frequencies and intensities resulting in the same annual total inflow were simulated. They showed that the maximum annual metal load in the biofilter outflow was obtained for the inflow combination with the lowest frequency and highest intensity. This model can be effectively used to assist in designing biofilters and assessing their long-term performance.

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## 1. Introduction

Currently, half of the world population lives in urban areas (WB, 2012), and the urban population is expected to increase in the future (UNPD, 2002). The increasing urbanization alters the urban hydrologic cycle, with high peaks and volumes of surface runoff and greater atmospheric pollution. Urban dusts are found to be polluted with metals, mainly stemming from combustion of fossil fuels in vehicles, and to have high ecological risk (Kabir et al., 2014). These polluted dusts are conveyed by stormwater runoff into receiving water bodies (Gnecco et al., 2005), thus resulting in degraded surface water quality (Ahiablame et al., 2012).

Green infrastructures that mimic natural ecosystems are increasingly embedded in urban environments to collect and treat stormwater. Among these, stormwater biofilters represent an effective and sustainable way to combine water flow management

with urban pollution mitigation (Hatt et al., 2009a). Biofilters typically consist of a trench filled with engineered soils supporting selected plants; stormwater is allowed to flow vertically through the soils, where various physicochemical processes occur (Davis et al., 2009). Biofilters have been shown to reduce peak flow and stormwater runoff volumes as well as retain more than 80% of the influent stormwater pollutants including suspended solids, nitrogen, phosphorus (Davis et al., 2001; Kim et al., 2003; Passeport et al., 2009) and metals (Feng et al., 2012; Blecken et al., 2009a, 2009b).

Although many laboratory and field experiments have been performed to provide guidelines to biofilter design and ascertain the ability of biofilters in removing a series of pollutants from urban stormwater (Hatt et al., 2009b; Bratieres et al., 2008; Hunt et al., 2006), modelling of biofilters has received less attention. The long-term hydrologic balance of biofilters have been described using both deterministic (Brown et al., 2013) and stochastic models (Daly et al., 2012). In addition to the water balance, chemical retention and degradation mechanisms in soils have been used in reactive transport models (Vezzaro et al., 2011; Randelovic et al., 2016). Some reactive transport models emphasise the chemical retention efficiency, but neglect the retention mechanisms and kinetics, and use empirical reduction factors to estimate the quality improvement of effluent water (Rossman, 2004). Other models

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describe the pollutant retention efficiency by first-order kinetic reactions, but generally lump multiple processes into a single reaction, thereby simplifying the processes taking place in the biofilter without accounting for specific time scales characterizing each process (Wong et al., 2006). Some other models (mainly related to wetlands) have a higher level of complexity because they include flows and reaction of pollutants in reaction networks (Lee et al., 2002). However, the use of these complex models for biofilter design is limited by the uncertainty in model structure and parameters (Bertrand-Krajewski, 2007), leading modellers to commonly prefer lumped and conceptual models. This uncertainty is magnified when dealing with metals, such as copper (Cu), lead (Pb), and zinc (Zn), because there is a general lack of information regarding their sources, and it is difficult to measure all possible chemical forms that may be present in an urban environment. Additionally, a comprehensive understanding of metal geochemical cycle and interaction with the soil matrix and the organic fractions are not currently available or limited to only some pathways (Obropta and Kardos, 2007; Vezzaro et al., 2012).

An attempt to quantify the uncertainty in Cu and Zn retention using a lumped model, where few redox reactions have been used to describe the soil geochemistry, is provided in (Vezzaro et al., 2012). The metal retention mechanism linked to physical and chemical protection to the mineral and organic soil matrix, the degradation mechanism of metal oxides (specifically for Cu, Pb and Zn), and the kinetics behind the formation of labile organic metal compounds have been neglected in models thus far.

In this study, we present a model that couples the water dynamics of biofilters with the geochemistry of Cu, Pb and Zn. The hydraulic and geochemical parameters of the model are calibrated against field experimental data of metal pollutant loads. A sensitivity analysis of the main parameters characterizing the geochemistry of Cu, Pb and Zn is shown, and a scenario analysis of the dependence of the metal retention efficiency on hydrometeorologic conditions is presented.

## 2. Model description

The model describes a biofilter consisting of a ponding zone, a soil layer, and a drainage layer (Fig. 1). The drainage layer hosts a perforated pipe to collect leaching water; no submerged zone is included in this model (Blecken et al., 2009a). Each of these three zones is considered as a bucket with a given capacity, and water and metal compounds can flow from one zone to the other. The model combines the water balance (Lintern et al., 2012a) and the metal geochemistry of each of these three zones of the biofilter.

### 2.1. Water balance

The model describing the water flows was presented in (Randelovic et al., 2016; Lintern et al., 2012a; CRCE, 2009) and is briefly summarized here. The inflow to the biofilter,  $Q_I$ , is calculated as

$$Q_I = R \cdot A + Q_R, \quad (1)$$

where  $R$ ,  $A$ , and  $Q_R$  are the rainfall rate, biofilter surface area, and the stormwater runoff diverted to the biofilter, respectively (See Appendix A-1 for all notations used in this paper).

The overflow,  $Q_O$ , is assumed to be regulated by a rectangular weir and is calculated as

$$Q_O = \begin{cases} C_w \cdot L_w \cdot (h_p - h_w)^{3/2} & \text{for } h_p > h_w \\ 0 & \text{otherwise} \end{cases}, \quad (2)$$

where  $C_w$ ,  $L_w$ ,  $h_w$ , and  $h_p$  are the weir coefficient constant, weir length, weir height, and the pond water depth, respectively.

Water from the ponding zone infiltrates the soil layer at a rate  $Q_F$ , which is calculated as

$$Q_F = \min \begin{cases} A \cdot \frac{h_p}{\Delta t} + Q_I \\ A \cdot K_s \\ A \cdot h_c \cdot \varphi \cdot \frac{(1-s)}{\Delta t} \end{cases}, \quad (3)$$

where  $K_s$  is the saturated soil hydraulic conductivity,  $\varphi$  is soil porosity,  $s$  is soil saturation,  $h_c$  is the soil media depth, and  $\Delta t$  is the time step of the numerical simulation.

The evapotranspiration rate,  $ET$ , is assumed to be driven by  $s$  (Daly et al., 2009). In a well-watered vegetated soil,  $ET$  remains approximately constant at  $ET_{max}$ . When  $s$  falls below a threshold,  $s_t$ , the vegetation starts experiencing water stress and  $ET$  decreases linearly. As soil moisture reaches the wilting point,  $s_w$ , transpiration stops and only evaporation contributes to  $ET$ . Evaporation decreases linearly from a maximum value  $E_w$  and ceases when soil moisture reaches the hygroscopic point,  $s_h$ . Accordingly, the  $ET$  flux,  $Q_{ET}$ , can be calculated as (Daly et al., 2009)

$$Q_{ET} = \begin{cases} A \cdot E_w \cdot \frac{s - s_h}{s_w - s_h} & \text{for } s_h < s \leq s_w \\ A \cdot \left[ E_w + (ET_{max} - E_w) \cdot \left( \frac{s - s_w}{s_t - s_w} \right) \right] & \text{for } s_w < s \leq s_t \\ A \cdot ET_{max} & \text{for } s_t < s \leq 1 \\ 0 & \text{otherwise} \end{cases} \quad (4)$$

According to the Darcy's law, leaching from the soil media to the drain,  $Q_L$ , can be calculated as

$$Q_L = A \cdot K_s \cdot s^{(2\beta+3)} \cdot \left( \frac{h_p + h_c}{h_c} \right), \quad (5)$$

where  $\beta$  is the soil pore volume index that varies with the soil textural properties. The rate of change of  $h_p$  and  $s$  depends on the net inflow and outflow as

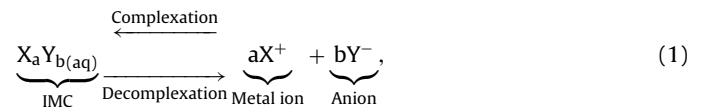
$$\frac{dh_p}{dt} = \frac{Q_I - Q_O - Q_F}{A}, \quad (6)$$

$$\frac{ds}{dt} = \frac{Q_F - Q_{ET} - Q_L}{A \cdot h_c \cdot \varphi}, \quad (7)$$

which describe the moisture dynamics in the biofilter as a function of time, and sinks and sources of water.

### 2.2. Metal biogeochemistry

A geochemical cycle of various organic and inorganic metals was presented in (Kabir et al., 2014), where a generic set of aqueous chemical species was described by equilibrium aqueous complexation reactions, equilibrium adsorption reactions, and kinetic methylation reactions. In the soil water, metal ions and reactive anions can form various species of inorganic metal compounds (IMCs), which are the simplest form of metal compounds, through complexation reactions. The generic expression for complexation-decomplexation reactions is of the form



where  $a$  and  $b$  are the stoichiometric coefficients of aqueous metal ion,  $X^+$ , and anion,  $Y^-$ , respectively, and  $X_a Y_{b(aq)}$  is the aqueous IMC. The equilibrium reaction constant of Reaction (1),  $K$ , is determined by the mass-action law as

$$K = \frac{[X^+]^a [Y^-]^b}{[X_a Y_{b(aq)}]}, \quad (8)$$

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