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Evaluation of applicability of filling materials in permeable reactive barrier (PRB) system to remediate groundwater contaminated with Cd and Pb at open solid waste dump sites

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ABSTRACT

This study aimed to identify appropriate filling materials for a permeable reactive barrier (PRB) system to treat groundwater contaminated with trace metals in the vicinity of solid waste landfills in Sri Lanka. Mixtures of alluvial loamy soil, coconut shell biochar, and laterite clay brick in different proportions were tested as easily-available PRB adsorbents. A series of adsorption and desorption experiments were carried out to investigate the effects of initial concentration, pH, ionic strength, and multiple competitive trace elements on Cd and Pb adsorption onto the tested adsorbents. In addition, hydraulic conductivities (K_s) of the tested adsorbents under different compaction levels were measured to examine a suitable packing condition for the PRB system. Results showed that the Langmuir model performed well for fitting Cd and Pb adsorption isotherms and maximum adsorption capacities (Q_m) for Pb (2.1–15.3 mg/g) became higher than those for Cd (0.8–6.8 mg/g). All tested adsorbents showed low leaching of adsorbed metals with high hysteresis indices in desorption studies. In the multiple trace element solution, the existence of other trace metals (Cu, Zn, Ni) had a significant effect on Cd adsorption but less on Pb adsorption. The three mixed adsorbents had no dependency on the initial pH and ionic strength of the solution, while the single material showed a low dependency in both Cd and Pb adsorption. The inclusion of brick was effective to improve the hydraulic property and measured K_s values for the 75% brick mixed materials resulted of $>10^{-4}$ cm/s at high compaction levels ($D_r = 90\%$ and 100%). Three mixed materials can be strongly recommended as a PRB filling material to treat landfill leachate based on their reactivity and hydraulic properties.

1. Introduction

Groundwater contamination caused by the intrusion of untreated wastewater plumes containing organic and inorganic contaminants, radionuclides, and heavy metals are widespread throughout the world (Alloway, 2013; Mulligan et al., 2001). Spilling, leaching and improper underground storage of fuel, chemicals, septic systems, hazardous wastes, road salts, agrochemicals, (Grimm et al., 2008) and uncontrolled open waste dumping (Chaney et al., 2004; Sewwandi et al., 2013; Udayagee et al., 2017) are known to be typical causes of groundwater contamination. Especially, the open dumping of non-sorted wastes causes a serious water pollution mainly with heavy metals, such as Cd, Pb, Hg, Ni, Mn, Cu (Chen, 1996 ; Udayagee et al., 2017; Wijesekara et al., 2014). As reported in previous studies, heavy metals such as Pb, and Cd exceed their concentrations than the effluent water quality standards in Sri Lanka (Sewwandi et al., 2013). Thus, effective remediation techniques are highly demanded to reduce the risk of spreading the contaminated water plume throughout the aquifers and minimize the negative effects on the ecosystem.

In order to treat contaminated groundwater, conventional pump-and-treat methods combining groundwater extraction and ex-situ treatment are used widely (Naftz and Davis, 1999). This option is costly and often ineffective in meeting long-term protection standards (USEPA, 1999), and sometimes not applicable in developing countries due to economic and technical limitations. In recent years, permeable reactive barrier (PRB) systems have received much attention as practical and cost-effective techniques for in-situ treatment of contaminated groundwater (Carey et al., 2002; Obiri-Nyarko et al., 2015; Regmi, 2009). Thus the PRB system is expected to treat the groundwater contaminated with landfill

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leachate originated from existing waste dumping sites with no lining system.

The concept of PRB systems has been developed to take advantage of a natural groundwater gradient in order to flow contaminated water through a highly permeable reactive material (Carey et al., 2002; Regmi, 2009), instead of pumping up the water as done in ex-situ treatment techniques (Naftz and Davis, 1999). As the groundwater flow passes through the PRB, target contaminants are trapped (adsorbed, immobilized, transformed to a non-toxic form, etc.) due to various physical, chemical, and biological reactions occurring in/on the reactive barrier materials (Di Natale et al., 2008; Obiri-Nyarko et al., 2015). Therefore, system components of PRB and its hydraulics have to be critically evaluated.

A thorough understanding of the adsorptive and hydraulic capacities of the reactive media ensures the efficient immobilization of contaminants and maintain the passive flow (Carey et al., 2002; USEPA, 1999). Many reactive media combinations can be used in PRB, and numerous media and mixtures of media are being investigated to treat a variety of contaminants. Zero-valent iron (ZVI) = Fe°, has been used widely as a PRB material (Statham et al., 2015; Suzuki et al., 2012), and as reported in ITRC (2005), more than 60% of PRBs were made of ZVI by 2004. The greater reduction potential (−440 mV) and precipitation capacity of ZVI augment the transfer of electrons to contaminants by activating various chemical reactions. In addition to ZVI, zeolite (Vignola et al., 2011), calcite (Turner et al., 2008), and FeS-coated sand (Han et al., 2011) have been used as PRB filling materials in various field scale and laboratory scale studies. Several biomaterials such as coconut husk powder (Sewwandi et al., 2014), crab shells (Vijayaraghavan et al., 2011), peanut shells (Han et al., 2011), sawdust (Sewwandi et al., 2012), activated neem leaves (Babu and Gupta, 2008), and coir pith (Parab et al., 2006) have been recognized as potential low-cost adsorbents in PRB systems due to their high performances in heavy metal removal from wastewater. Even though numerous materials have been tested for their reactivity in heavy metal removal, the hydraulic properties have not been evaluated with the consideration of PRB applications in Sri Lanka. The hydraulic conductivity of the compacted reactive material in PRB is essential to ensure long-term treatment efficiency. In particular, the hydraulic conductivity of PRB should be greater than that in the surrounding soil to ensure the groundwater flow through PRB without any external energy input (Carey et al., 2002; Obiri-Nyarko et al., 2015; Regmi, 2009; Smith et al., 2003). Thus, it is important to introduce a single material or a mixture of materials, which fulfill these key requirements of PRB system.

There are numerous advantages of using combinations of materials rather using a single material as PRB components, though the single materials have been used frequently in past applications. A mixture of materials with different geochemical and geophysical properties (e.g., Adsorption capacity, graded particle size distribution) may improve the hydraulic conductivity, expand the contaminant removal mechanisms, and accelerate the removal efficiency. Further, the long-term performance of the barrier improve the sustainability (Hamidpour et al., 2010).

The objectives of this study were to investigate adsorption, desorption behaviors and hydraulic conductivity of selected reactive materials and their mixtures in order to evaluate their applicability to PRB systems in Sri Lanka.

2. Materials and methods

2.1. Materials used

The basic physical and chemical properties of the three materials tested in this study are shown in Table 1. A loamy alluvial soil (hereafter "soil") taken from Bangadeniya, North Western Province, Sri Lanka, is a sandy clay loam soil and was categorized as an Entisol (Mapa et al., 2010). According to Paranavithana et al. (2013), the soil is rich in micro pores in the range of 0.5–50 μm, and its surface charge is a predominantly permanent negative charge. The soil was used for laboratory tests after air-drying and sieving with a 2-mm mesh.

Table 1
Basic physical and chemical properties of the tested materials.

Location	Soil Bangadeniya, Sri Lanka	Biochar Matale, Sri Lanka	Brick Kandy, Sri Lanka
Particle size (mm)	< 2.00	< 0.075	2.00–4.75
Moisture content in air dry (%)	3.4	3.2	0.4
pH	4.7	8.8	5.4
EC (μS/cm)	59	139	47
BET surface area (m^2/g)	28.5	212	14.4
Specific gravity	2.66	1.51	2.77
Loss on ignition (%)	12.3	60	0.9

Coconut shell biochar (hereafter "biochar") is an inexpensive and easily available material in tropical countries including Sri Lanka (Babel and Kumiawa, 2004). According to (Liu and Zhang, 2009; Paranavithana et al., 2016) the coconut shell biochar is moderately alkaline and predominant in basic functional groups. The large surface area of biochar was also noted in the same studies which is positively affect in heavy metal adsorption. The biochar was collected from a biochar producing industry in Sri Lanka, air-dried, ground by a mill, and fine granules (<75 μm) were used for laboratory tests, in order to facilitate better mixing with other tested materials.

Burnt laterite brick used in construction (hereafter "brick"), commonly available in Sri Lanka, was used to improve hydraulic conductivities of the tested adsorbents in this study. The brick was crushed and sieved, and coarse granular particles, (<2.00–<4.75 mm) were prepared. Although the brick has been identified as a good heavy metal adsorbent at lower contaminant concentrations, (Bibi et al., 2015; Djeribi and Hamdaoui, 2008), the brick was expected mainly to provide a good matrix with other material for PRB adsorbents to ensure a high permeability.

In this study, three materials were mixed in different proportions on a dry mass basis, with a total of nine combinations were prepared to analyze their adsorption and desorption properties and hydraulic conductivities. The nine mixed PRB testing materials were Soil 100%, Biochar 100%, Brick 100%, Soil 50% + Biochar 50%, Soil 75% + Biochar 25%, Soil 25% + Biochar 75%, Soil 25% + Biochar 25% + Brick 50%, Soil 37.5% + Biochar 37.5% + Brick 25%, and Soil 12.5% + Biochar 12.5% + Brick 75%.

2.2. Batch experiments for adsorption and desorption studies

The test conditions for batch adsorption and desorption experiments are summarized in Table 2. Batch adsorption and desorption experiments were conducted as per to the standard method recommended by the Organization of Economic Cooperation and Development (OECD, 2000). All experiments were conducted using a 1:10 solid: liquid ratio. Heavy metal solutions were prepared by using $PbCl_2$, $CdCl_2$, $CuCl_2$, $NiCl_2$, and $ZnCl_2$ salts (<99.5%, Wako Pure Chemical Industries, Ltd., Japan). Clean 50 mL Violamo centrifuge tubes (AO1350002, Sigma-Aldrich, USA) were used to shake the testing materials and solutions in a reciprocating shaker for 24 h at 100 rpm at 20 °C. After shaking, the samples were centrifuged at 8000 g for 15 min (TOMY LC-200, Japan) and supernatants were filtered through a membrane filter (Millipore 0.23 μm). Then, the supernatants were diluted to predetermined dilutions, and contaminant concentrations were measured by atomic adsorption