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Removal of heavy metals from synthetic landfill leachate in lab-scale vertical flow constructed wetlands

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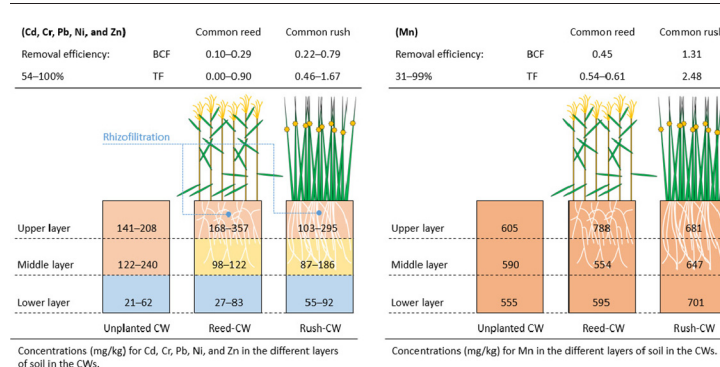
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HIGHLIGHTS

- CW showed high metal removal amounts for Zn, Cr, Ni, Cd, Fe, and Pb, but not Mn.
- Metal removal was low for high-strength leachate and short retention times.
- Cd, Cr, Pb, Ni, and Zn were highly accumulated in upper soil layers of planted CW.
- Rhizofiltration by plants is apparently helpful to decrease the dredging soil depth.
- Common rush had higher BCFs and TFs for the heavy metals than common reed.

GRAPHICAL ABSTRACT



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ABSTRACT

Synthetic landfill leachate was treated using lab-scale vertical flow constructed wetlands (CWs) in sequencing batch modes to assess heavy metal removal efficiencies. The CWs filled with loamy soil and pumice stone were unplanted or planted with common reed (*Phragmites australis*) (Reed-CW) or common rush (*Juncus effusus*) (Rush-CW). Synthetic leachate contained acetate, propionate, humate, ammonium, and heavy metals. Common reed grew almost vigorously but common rush partly withered during the 8-month experiment. The CWs reduced the leachate volume effectively by evapotranspiration and removed easily degradable organic matter, color, and ammonium. Furthermore, the CWs demonstrated high removal amounts for heavy metals such as Zn, Cr, Ni, Cd, Fe, and Pb, but not Mn from leachate. The metal removal amounts in the CWs were low for high-strength leachate (influent concentration increased from one time to three times) or under short retention time (batch cycle shortened from 3 days to 1 day). The Rush-CW showed slightly lower removal amounts for Cr, Ni, Mn, and Cd, although the Reed-CW showed lower Mn removal amounts than the unplanted CW did. However, Cd, Cr, Pb, Ni, and Zn were highly accumulated in the upper soil layer in the planted CW by rhizofiltration with adsorption compared with unplanted CW, indicating that the emergent plants would be helpful for decreasing the dredging soil depth for the final removal of heavy metals. Although the emergent plants were minor sinks in comparison with soil, common rush had higher bioconcentration factors and translocation factors for heavy metals than common reed had.

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1. Introduction

Leachate generated from waste landfills contains various toxic compounds at low concentrations, in addition to high concentrations of organic matter, color components, and ammonia-nitrogen (NH₄-N). The most toxic pollutants in landfill leachate are heavy metals. According to reviews (Baun and Christensen, 2004; Öman and Junestedt, 2008; Renou et al., 2008), typical metal concentrations in landfill leachate are the following: Cd (0.0001–0.13 mg/L), Cr (0.0005–1.6 mg/L), Fe (0.08–2100 mg/L), Mn (0.01–65 mg/L), Ni (0.03–3.2 mg/L), Pb (0.0005–1.5 mg/L), and Zn (0.00005–120 mg/L). The metal concentrations in young (acetogenic) leachate are usually higher than those in old (methanogenic) leachate (Kjeldsen et al., 2002; Öman and Junestedt, 2008).

A sustainable low-cost technology for leachate treatment is the constructed wetland (CW) especially in tropical/subtropical regions (Akinbile et al., 2012; Ogata et al., 2015; Sawaitayothin and Polprasert, 2006). Easily degradable organic carbon and NH₄-N can be removed effectively from landfill leachate by CWs (Stefanakis et al., 2014). Interest has been growing in removing heavy metals from leachate through CWs. Although various mechanisms are either still unknown or poorly understood, four mechanisms affect metal removal in CWs: (Fuji et al., 2017) adsorption to soils, sediments, particulates, and soluble organics; (Akinbile et al., 2012) precipitation as insoluble salts, principally sulfides and oxyhydroxides; and (Batty et al., 2002) uptake and induced changes in biochemical cycles by aquatic plants and microorganisms, (Baun and Christensen, 2004) deposition of suspended solids because of low flow rates (Cheng et al., 2002; Marchand et al., 2010). Several studies have demonstrated metal removal from landfill leachate by full-scale CWs (Mæhlum, 1995; Martin et al., 1999; Pevery et al., 1995), pilot-scale CWs (Akinbile et al., 2012; Bernad and Lauve, 1995; Bulc, 2006; Sawaitayothin and Polprasert, 2006; Yalcuk and Ugurlu, 2009), and lab-scale CWs (Lavrova and Koumanova, 2010; Mojiri and Ziyang, 2015). Although these studies using actual leachates were practical for assessment of the feasibility of metal removal in CWs, it is not easy to integrate the knowledge related to design and operation of CWs for effective metal removals because of complicated and variable leachate characteristics (Baun and Christensen, 2004). For example, studies comparing planted and unplanted systems often engender conflicting results related to metal removal in CWs (Marchand et al., 2010). Some researchers reported that the presence of plants did not enhance the metal removal of the CWs (Lee and

Schoilz, 2007; Zhang et al., 2007), although the others confirmed that the plants can certainly enhance it (Megtатели et al., 2009).

A model experimental system using lab-scale CWs with synthetic leachate is a useful tool to facilitate systematic understanding of the treatability of leachate by CWs (Soda et al., 2016; A et al., 2017). For example, Connolly et al. (2004) demonstrated effective NH₄-N removal from synthetic leachate in a lab-scale downflow reed bed system with effluent recirculation. To the best of our knowledge, no report of the relevant literature has described a study of heavy metal removal from synthetic leachate in a lab-scale CW. In this study, the removal efficiencies of heavy metals from synthetic young and old leachate in lab-scale CWs of three vegetation types (unplanted, planted with common reed, or with common rush) with different operations (influent loads/retention time) were investigated. Distributions of heavy metals accumulated in soil layer and plant tissues in CWs were also investigated. The knowledge obtained in this study will be helpful for evaluating potentials of CWs for heavy metal removal.

2. Materials and methods

2.1. Synthetic landfill leachate

Synthetic leachate developed in a previous study (Soda et al., 2016) was used with minor modifications by supplementing heavy metals. The synthetic leachate composition is presented in Table 1. The synthetic leachate contained organic matter, nutrients, and heavy metals (Cd, Cr, Fe, Mn, Ni, Pb, and Zn). The organic matter consisted of acetate and propionate as easily degradable components and humate (Sigma-Aldrich Co. Ltd.) as an only slightly degradable component. Young leachate and old leachate were designed respectively to simulate those in the acetogenic phase and the methanogenic phase in a landfill.

2.2. Lab-scale CWs

Lab-scale CWs were set up in a greenhouse in Osaka University in Suita City, Osaka Prefecture, Japan. A schematic diagram of the CW is depicted in Fig. 1. Each CW consisted of a plastic container (18 cm inner diameter × 63 cm height) with a recirculation pump. The CWs were filled from bottom to top with pumice stones (20–30 mm, 5 cm depth; Green Life Hyuga Corp., Miyazaki, Japan), medium loamy soil (10–15 mm, 15 cm depth; Green Plan Co. Ltd., Miyagi, Japan), and fine loamy soil (3–4 mm, 20 cm depth; Green Plan Co. Ltd.). The CWs were

Table 1
Compositions of synthetic landfill leachate and operational conditions of the CWs.

| | Run 1 (Old leachate) | Run 2 (Young leachate) | Run 3 ^a (High-strength young leachate) | Run 4 ^b (Young leachate with short retention time) |
|--|-------------------------|---------------------------|--|--|
| Period | 15 Jun.–3 Aug., 2012 | 10 Sep.–19 Nov., 2012 | 22 Nov.–30 Dec., 2012 | 11–25 Jan., 2013 |
| Batch cycle | 3 day | 3 day | 3 day | 3, 2, 1 day |
| Leachate composition | | | | |
| Sodium acetate (mg) | 0 | 100 | 200 | 100 |
| Sodium propionate (mg) | 0 | 5 | 10 | 5 |
| Sodium humate (mg) | 250 | 50 | 255 | 50 |
| NH ₄ Cl (mg) | 191 | 382 | 382 | 382 |
| CaCl ₂ ·2H ₂ O (mg) | 36.7 | 110 | 110 | 110 |
| MgCl ₂ ·2H ₂ O (mg) | 83.6 | 83.6 | 83.6 | 83.6 |
| CdCl ₂ ·2.5H ₂ O (mg) | 2.0 | 6.1 | 12.2 | 6.1 |
| NiCl ₂ ·6H ₂ O (mg) | 4.0 | 12.1 | 24.3 | 12.1 |
| ZnSO ₄ ·7H ₂ O (mg) | 4.4 | 13.2 | 26.4 | 13.2 |
| MnCl ₂ ·4H ₂ O (mg) | 3.6 | 10.8 | 21.6 | 10.8 |
| K ₂ Cr ₂ O ₇ (mg) | 5.7 | 17 | 34 | 17 |
| FeCl ₃ ·6H ₂ O (mg) | 4.8 | 14.5 | 29 | 14.5 |
| Pb(NO ₃) ₂ (mg) | 2.2 | 6.5 | 13 | 6.5 |
| Tap water (L) ^c | 1.0 | 1.0 | 1.0 | 1.0 |

^a The influent concentration of Run 3 increased twice after 9 days (3 cycles), three times after 18 days (6 cycles), and four times after 27 days (9 cycles).

^b Batch cycle of Run 4 shortened from 3 days to 2 days after 3 days, and to 1 day after 9 days.

^c Tap water contained nitrate-nitrogen at about 1.5 mg-N/L and phosphate-phosphorus at about 0.5 mg-P/L.

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