



Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Calcium carbonate-based permeable reactive barriers for iron and manganese groundwater remediation at landfills

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ARTICLE INFO

Article history:

Received 19 June 2015

Revised 12 February 2016

Accepted 15 February 2016

Available online xxx

Keywords:

Landfill

Remediation

Iron

Manganese

Groundwater

Permeable reactive barrier

ABSTRACT

High concentrations of iron (Fe(II)) and manganese (Mn(II)) reductively dissolved from soil minerals have been detected in groundwater monitoring wells near many municipal solid waste landfills. Two *in situ* permeable reactive barriers (PRBs), comprised of limestone and crushed concrete, were installed down-gradient of a closed, unlined landfill in Florida, USA, to remediate groundwater containing high concentrations of these metals. Influent groundwater to the PRBs contained mean Fe and Mn concentrations of approximately 30 mg/L and 1.62 mg/L, respectively. PRBs were constructed in the shallow aquifer (maximum depth 4.6 m below land surface) and groundwater was sampled from a network of nearby monitoring wells to evaluate barrier performance in removing these metals. PRBs significantly ($p < 0.05$) removed dissolved Fe and Mn from influent groundwater; Fe was removed from influent water at average rates of 91% and 95% (by mass) for the limestone and crushed concrete PRBs, respectively, during the first year of the study. The performance of the PRBs declined after 3 years of operation, with Fe removal efficiency decreasing to 64% and 61% for limestone and concrete PRBs, respectively. A comparison of water quality in shallow and deep monitoring wells showed a more dramatic performance reduction in the deeper section of the concrete PRB, which was attributed to an influx of sediment into the barrier and settling of particulates from the upper portions of the PRBs. Although removal of Fe and Mn from redox impacts was achieved with the PRBs, the short time frame of effectiveness relative to the duration of a full-scale remediation effort may limit the applicability of these systems at some landfills because of the construction costs required.

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1. Introduction

The presence of a landfill can cause the formation of anoxic (i.e., reducing) conditions in the underlying aquifer due to both the cessation of natural atmospheric oxygen diffusion into the surficial aquifer and redox reactions consuming available oxygen. Microbial degradation pathways can oxidize landfill leachate organic matter with available electron donors. Once the more metabolically preferred electron acceptors are consumed (e.g., oxygen and nitrate), the oxidized Fe(III) and Mn(IV), naturally occurring as metal oxides, can be used by Fe and Mn reducing bacteria (Lepp, 1975; Nicholson et al., 1983; Lovley, 1991; Bjerg et al., 1995; Heron and Christensen, 1995; Rao, 2008; Wang et al., 2012; Nguyen et al., 2015). These dissolved metals are often detected during routine groundwater monitoring at landfill sites, potentially triggering regulatory action, including a requirement to remediate the issue.

Impacts are often noted in the form of decreased dissolved oxygen (due to Fe(II) oxidation to Fe(III)) and precipitation of iron oxide flocs and necrotic iron-reducing bacteria which produce a floating sheen on the water surface. In addition, the concomitant release of other trace contaminants, such as arsenic, that are bound onto iron minerals raises concerns particularly when the aquifer is used as a drinking water source (Keimowitz et al., 2005; de Lemos et al., 2006; Ghosh et al., 2006; Parisio et al., 2006; Di Palma and Mecozzi, 2010; Wang et al., 2012; Rachna et al., 2013).

Calcium carbonate-based materials (CCBMs) have been demonstrated to effectively remove di- and trivalent metal cations, copper, cadmium, chromium (Cr(III)), lead, nickel, and zinc from solution (Aziz et al., 2001, 2008). The use of CCBMs for ferrous iron remediation has been demonstrated in both batch (Mettler et al., 2009; Wang et al., 2013a, 2013b) and column (Wang et al., 2013c) laboratory-scale studies prior to the current field-scale pilot study. Soluble Fe and Mn, like other metal cations, can be removed from solution via sorption onto reactive media and precipitation of solid species (Aziz et al., 2001, 2008; Nano and Strathmann, 2006).

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Metal carbonate precipitates (FeCO_3 , MnCO_3 , $\text{CaFe}(\text{CO}_3)_2$) and metal hydroxides ($\text{Fe}(\text{OH})_2$, $\text{Mn}(\text{OH})_2$) are expected precipitates depending upon pH.

Permeable reactive barriers (PRBs), which utilize passive natural groundwater flow to expose groundwater contaminants (e.g., metals found in acid mine drainage, trichloroethylene) to *in situ* adsorptive/reactive materials (e.g., zero-valent iron; Richardson and Nicklow, 2002; Henderson and Demond, 2007), have shown to be effective at the field scale. PRBs can be more cost-effective compared to other conventional groundwater remediation options (e.g., pump and treat), and interest in utilizing passive treatment approaches such as PRBs has grown in recent years (US EPA, 2001a, 2001b). This study presents a field-scale test to evaluate CCBMs (limestone and crushed concrete) performance for *in-situ* remediation of Fe and Mn contaminated groundwater downgradient of an unlined municipal landfill.

2. Material and methods

2.1. Site description

The Klondike Landfill, located in Escambia County, Florida, is a closed unlined municipal solid waste (MSW) landfill that operated from 1976 until 1982. In 1987, regulation-driven semi-annual groundwater monitoring at the site detected iron (Fe) and manganese (Mn) at concentrations that substantially exceeded regulatory thresholds (0.3 and 0.05 mg/L, respectively) and health-based guidelines (4.2 and 0.33 mg/L) in monitoring wells downgradient of the landfill. Fig. 1 presents a site map showing compliance well

groundwater Fe data measured during regulatory monitoring five months prior to PRB installation.

Following successful demonstration of CCBM for Fe removal in laboratory columns (Wang et al., 2013c), PRBs were installed in the north-west portion of the site, between the western landfill boundary (i.e., downgradient of the landfill) and a regulatory monitoring well (DW-4S). PRBs and monitoring wells were installed in the “shallow” hydrogeologic unit; a clayey sand layer is present to approximately 5 m below land surface (bls) in this area. Historical semiannual groundwater monitoring data for monitoring well DW-4S show total iron (Fe_{TOT}) levels ranging from 20 to 50 mg/L and Mn levels in the range of 0.5–2.0 mg/L prior to PRB installation. Groundwater velocity at the site (calculated based on shallow aquifer hydraulic conductivity of approximately 7.93 m/day, a hydraulic gradient of 0.006 m/m, and an estimate effective porosity of 0.375) was reported to be approximately 0.13 m/day (Geosyntec, 2005). Groundwater levels during the first year of the study were approximately 0.44 and 0.48 m bls at the limestone and crushed concrete PRBs, respectively; a period of drought during the second year after PRBs were installed and caused the groundwater level to drop to mean depths of 0.73 and 0.88 m bls during the third year of monitoring.

2.2. PRB and monitoring well network installation details

Two permeable reactive barriers (PRBs) were installed approximately 20 m downgradient of the landfill boundary. Limestone (7–10 mm particle diameter) and crushed concrete (70–150 mm particle diameter) were selected as reactive materials based on

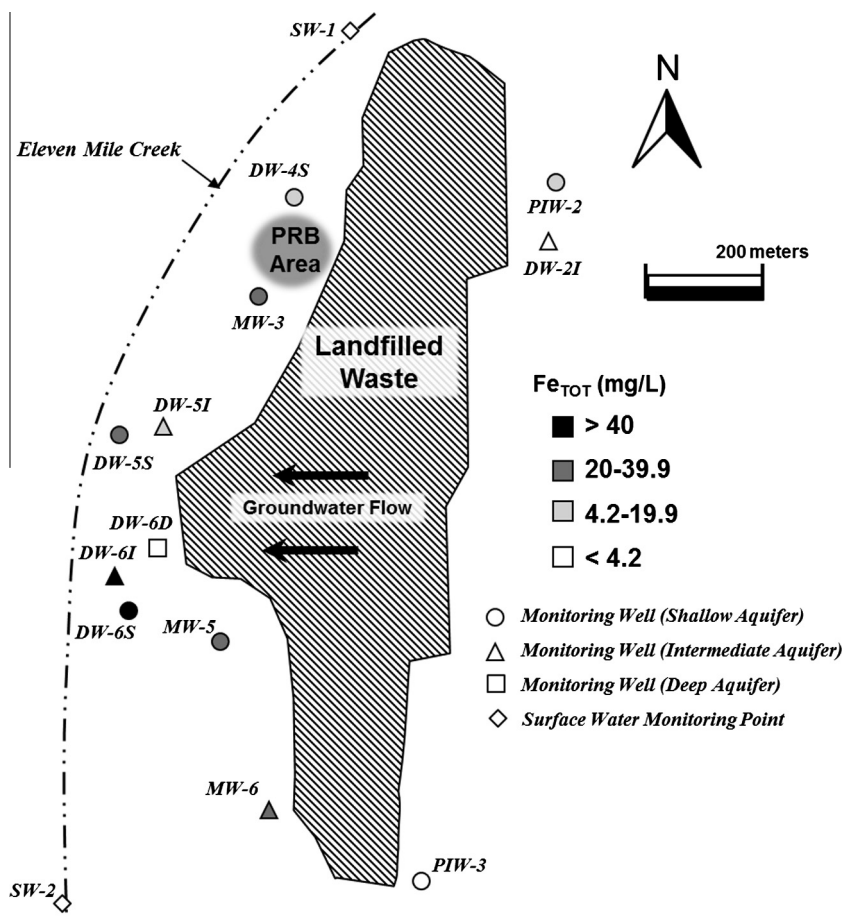


Fig. 1. PRB area location and regulatory sampling points at Klondike Landfill (Fe_{TOT} data reflective of sampling 5 months prior to PRB installation).

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