



Attenuation of metal contamination in landfill leachate by dewatered waterworks sludges



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ABSTRACT

Excess heavy metals in landfill leachate pose considerable environmental risk. The reduction of this risk using a by-product (dewatered waterworks sludges) is the main aim of this study. Bench-scale experiments were conducted towards understanding the key factors for the uptake of Pb, Cr and Cd by the sludges. Results indicate the maximum uptake for most of the sludges occurred at pH 4. Of the six models used for fitting the adsorption data for Pb, Cr and Cd; Freundlich, Temkin and Langmuir models gave the best fit in that order. Kinetic data was well fitted with the pseudo first order model for Pb and pseudo second order model for Cr; whilst the intraparticle diffusion model was found to adequately describe the kinetics for Cd. Principal component and canonical correlation analysis, FT-IR spectroscopy and sequential extraction revealed that: (i) organic carbon plays a key role in Cr adsorption and Pb through the carboxylate group, (ii) Cd adsorption is correlated to iron or aluminium (oxy) hydroxides, and (iii) the greatest amount of metals sorbed by the sludges were tightly bound and not extractable. These findings show that the sludges are potential adsorbents for Cr, Cd and Pb removal in practical applications.

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1. Introduction

Landfill leachate is one of major sources of heavy metals discharged into the environment, and this has continued to attract public concern due to potential deleterious impact of heavy metals on human health and environment. In the long term of landfill operation, the concentrations of leached heavy metal are continuously to decrease due to the process of precipitation and ligand exchange mechanisms. However, continuous discharge of heavy metals from landfill with low concentrations, can result in impairment of groundwater, surrounding soil and surface water quality. Therefore, heavy metals removal from landfill leachates is a growing area of research (Baun and Christensen, 2004; Mohan and Gandhimathi, 2009). Moreover, the growing awareness of the risks posed by heavy metals has been driving research into the development of sustainable technologies for their removal. In par-

ticular, the redistribution of heavy metals can adversely affect water resources and endanger the health of surrounding ecosystems and human populations; hence their removal is of significant importance.

A number of methods have been developed to remove heavy metals from landfill leachate. These include chemical precipitation wherein the metals precipitate from the solution in form of hydroxide as insoluble solid phase. However, the limitation of the chemical precipitation method is the high dose of the precipitant required, the sensitivity of the process employed to pH, and the need for disposal of the sludge generated. Ion exchange is another method which has been used, and it involves reversible interchange of ions between the solid and liquid phases, as there is no permanent change in the structure of the solid. However, this method is rarely used for heavy metals removal because the landfill leachate needs to be initially treated biologically and the suspended solids removed; and in addition, it has high operation cost (Kurniawan et al., 2006). Reverse osmosis can also be used; however, the drawbacks of this method are: small molecules can pass through the membrane, undesirable deposition of dissolved substances on the external surface of the membrane, and high energy consumption (Kurniawan et al., 2006).

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Table 1
Adsorption kinetic equations and their corresponding linear forms.

Kietic models	Equation	Linear plot
First order	$\frac{dq_t}{dt} = k_1(q_e - q_t)$	$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t$
Second orer	$\frac{dq_t}{dt} = k_2(q_e - q_t)^2$	$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$
Intraparticle diffusion	$q = k_d t^{0.5} + c$	

Adsorption is a major process responsible for attenuation of heavy metals contaminants in wastewater. Therefore, the study of adsorption processes is of utmost importance for the understanding of how heavy metals are transferred from a liquid mobile phase to the surface of a solid phase (Castaldi et al., 2015). Whilst various media have been assessed as adsorbents for heavy metals; the process could be made more attractive if the sorbent is inexpensive and does not require complicated pre-treatment or regeneration. In this regard, dewatered waterworks sludge can be a potential adsorbent for heavy metals removal from landfill leachate. Dewatered waterworks sludge are low-cost and easily available worldwide; and they are generated during the drinking-water treatment process. They are primarily composed of Fe/Al hydroxides which are often amorphous species; and they contain sediment and humic substances from the raw water.

Whilst dewatered waterworks sludges have been shown to have large surface areas and high affinity for heavy metals such as Cd, Cr, Cu, Hg, Zn, Pb and Se (Chiang et al., 2012); In this study, different dewatered waterworks sludges, collected from different areas in the UK, were used to investigate the combine effect of various physicochemical properties on the removal of Pb, Cr and Cd and the associated mechanisms. Therefore, attenuation capabilities of fourteen dewatered waterworks sludges towards selected individual heavy metals found in landfill leachate (Pb, Cr and Cd) were investigated. The specific objectives were: (1) to evaluate the physicochemical properties of different dewatered waterworks sludges that are relevant to heavy metals attenuation using principal component analyses (PCA) and canonical correlation analysis (CCA), (2) to investigate the characteristics of Pb, Cr and Cd removal by dewatered waterworks sludges through batch experiments investigating the equilibrium and kinetics of adsorption, (3) to study the nature of the interactions established between the metal ions and the surfaces of dewatered waterworks sludges, and (4) to ascertain the role of the inorganic and organic fractions of dewatered waterworks sludges in the metal attenuation processes.

2. Materials and methods

2.1. General physicochemical characterization

Dewatered waterworks sludges were collected from fourteen drinking water treatment works located in the United Kingdom. The treatment plant locations are kept anonymous on request; and samples obtained were simply labelled using a sequential alphabetic code generated from the location names. The dewatered waterworks sludge samples were air-dried and ground to pass a 2-mm mesh sieve, and then used for the characterization and adsorption tests. The chemical and physical characteristics of the sludges were determined as follows: The pH sludges were determined according to the standards BS ISO 10390:2005; while the point of zero charge (PZC) was determined using the solid addition method (Mohan and Gandhimathi, 2009). Surface area was measured using Ethylene Glycol Monoethyl Ether (EGME) method (Cerato and Lutenegeger, 2002). The physicochemical characteristics of the dewatered waterworks sludges are presented in Table 1. The sludges were also subjected to X-ray diffraction analysis using a Phillips PW3830 x-ray diffractometer in order to

determine their mineralogy. Elemental metal composition of the sludges were determined using inductively coupled plasma optical emission spectroscopy (ICP-OES) after digestion in a microwave with 15.8 M of HNO₃ and 11.65 M of HCl (1:1 HCl: HNO₃). Al and Fe oxalate of the sludges were determined following the method of McKeague and Day (1966).

2.2. Adsorption study – kinetic and equilibrium tests

To examine the kinetics of heavy metals adsorption by the sludges, batch experiments were used to investigate the kinetics of the adsorption process. Initial metal concentration for individual solutions was taken as 5 mg L⁻¹ for Cd, 1 mg L⁻¹ for Cr and 0.5 mg L⁻¹ for Pb. The concentration of the heavy metals used were chosen based on their typical concentration in young landfill leachate in the UK (Baun and Christensen, 2004; Thornton et al., 2000). The solutions for each heavy metal was prepared in the laboratory using CdSO₄ · $\frac{8}{3}$ H₂O, Cr(SO₄)₂ · 12H₂O and PbCl₂ salts, respectively for Cd, Cr and Pb. To investigate the effects of adsorbent dosage and equilibration time, different masses of the sludge samples (0.1, 0.5 and 1.0 g) were equilibrated with 100 ml each of heavy metal solutions (5 mg L⁻¹ for Cd, 1 mg L⁻¹ for Cr, and 0.5 mg L⁻¹ for Pb); contained in 250 ml bottles for 1–96 h using a rotary shaker. The mixture at each specified time was withdrawn, filtered and analysed for each heavy metals uptake using Eq. (1). The kinetics of the adsorption process for each metal ions by the sludges was analysed by fitting the kinetic data to the pseudo first-order equation, pseudo second-order equation, and the intraparticle diffusion models (Table 1).

$$q_e = \frac{(c_o - c_e)}{m} v \quad (1)$$

where C_o and C_e (both in mg L⁻¹) are the initial ($t=0$) and final heavy metals concentrations at equilibrium (q_e), respectively, v is the volume of the solution (L) and m is the mass of dewatered waterworks sludges used (g). q_t is the amount of adsorbate adsorbed at time t (mg g⁻¹), k_1 is the pseudo-first-order rate constant (min⁻¹) and t the contact time (min), k_2 is the pseudo-second-order rate constant (g mg⁻¹ min⁻¹), k_d and C is the rate constant for intraparticle diffusion in (mg g⁻¹ min^{-1/2}) and the intercept, respectively.

The initial rates of intraparticle diffusion can be further supported by calculating the pore diffusion coefficient \bar{D} (cm² s⁻¹) using Eq. (2); where $t_{1/2}$ (min) is the time for half of the adsorption and r is the average particle radius of the adsorbent particles.

$$\bar{D} = \frac{0.03r^2}{t_{1/2}} \quad (2)$$

For the equilibrium experiments, a contact time of 48 h and an optimal dosage of 10 g L⁻¹ were used as predetermined from batch kinetic experiments. To obtain equilibrium data for determining the heavy metals adsorption capacity and to investigate the effect of solution pH, 1 g of the sludges were equilibrated with 100 ml of the heavy metals solution (with initial pH ranging from 2 to 9). The range of concentrations of the heavy metals used were 0.05–1 mg Pb L⁻¹, 0.05–1 mg Cr L⁻¹ and 0.05–5 mg Cd L⁻¹. This wide range of heavy metals concentration was chosen to study the variability in heavy metals adsorption capacity and its dependence on the initial concentration of the heavy metals; and to investigate the effect of low initial metal concentration on the adsorption, giving the low concentration of heavy metals typically found in UK landfill leachates (Baun and Christensen, 2004; Thornton et al., 2000). A possible reason for the low concentration is that the heavy metals in landfill leachate have been found to be bound in or onto particulate matter, which settles in the settling tank and this reduces the concentration of the heavy metals in the landfill leachate (Øygard et al., 2007). After the set equilibrium time (48 h), the mixtures were

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