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Original Research

Lead Transport in Soils Amended with Municipal Solid Waste Ash

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Abstract

Municipal solid waste (MSW) is one of the major environmental problems in the Kingdom of Saudi Arabia (KSA). Many efforts have been made to reduce and recover MSW, but land disposal is still the most popular option. The current research aims to investigate the effects of municipal solid waste ash (MSWA) application rates on the mobility of Pb by 2 soils with their different physical and chemical properties. The soil was amended with MSWA at the rate of 0, 1, 2, 3, and 5% (w/w). Pb adsorption experiments were carried out by mixing 1.0 g of samples with initial Pb concentration varying from 0 to 300 mg L^{-1}. The results indicated that distribution coefficient (K_d) values of Pb^{2+} on sandy loam soil were higher than those on sandy loam soil. The application rate of 5% MSWA to loamy sand and sandy loam soils resulted in increases of K_d values by 36.6% and 29.0% more than the control soil (0%). MSWA amendment is most effective in reducing Pb mobility in the studied soils. The results suggest that MSWA could be used as a low-cost adsorbent for Pb^{2+} of contaminated soils.

Keywords: municipal solid waste ash, soils, distribution coefficient, Pb adsorption

Introduction

The volume of municipal solid waste (MSW) produced in the world is increasing annually, and disposing of such wastes is a growing problem. MSW generation has become an important issue in recent years due to the uncontrolled growth of the urban population and industrialization in Riyadh, Kingdom of Saudi Arabia (KSA). Total MSW production has increased to more than 12 million tons $year^{-1}$ and projections show that MSW production has exceeded 20 million tons per annum by 2015 and the per capita MSW production in KSA varied from 1.50 to 1.80 kg $person^{-1}$ day^{-1} [1]. The heavy metals Cr, Cu, Hg, Ni, Cd, Zn, and Pb are the most commonly found in municipal solid waste ash (MSWA), and Pb usually exists in the largest amounts [2]. These metals are harmful to the environment if there are no treatments. The environmental and technical problems have discouraged the reuse of MSWA. Even though pre-treatment increases the total cost, the treatment process enables the MSWA to be reused. Any one of the applications would be a great contribution to minimizing the waste and providing an alternative to landfill. Many of the applications of MSWA are still under investigation. The common method of MSWA disposal in KSA is dumping it into soils, because it is the cheapest way. The contaminated soils by MSWA application have the potential for groundwater pollution – especially with heavy metals [3]. Moreover, leachate from MSWA dumpsites affect the physical and chemical

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properties of the soils [4-5]. MSWA has received special attention for removal of heavy metals from soils and wastewater [6-7].

Contamination of soils by Pb has increased dramatically due to anthropogenic activity in recent decades. The presence of Pb in the environment is of a major concern because of its toxicity, bio-accumulating tendency, and threat to human health. The major sources of Pb are exhaust gases of petrol engines, pesticides, fertilizer impurities, emissions from mining and smelting operations, and the combustion of fossil fuels [8-10]. Clay content, soil pH, and Fe and Mn oxides are the most important factors affecting the mobility of Pb in soils. Although the leaching of Pb into groundwater often occurs at low pH, at higher pH Pb may precipitate as $Pb(OH)_2$ with low solubility [11]. The stabilization of Pb-contaminated soils using immobilizing amendments such as MWSA is a kind of remediation procedure practical for reducing Pb mobility in soils [12]. Stabilization of Pb in soils can be achieved by Pb adsorption, complexion, and precipitating of Pb. The transport and distribution of Pb through soils is important because of the possibility of contamination of both surface and ground water [13]. The dramatic growth in urbanization and population in Riyadh has brought some environmental challenges that need to be efficiently dealt with. One of these challenges is finding the proper management of hugely produced MSWA. Recycling of MSWA via landfill is a possible option of its management that has recently received more attention, although the impact of MSWA application on Pb transport into soils is not widely studied under arid soil conditions. The MSWA which has unique chemical characteristics may exert effects on Pb mobility and distribution in soils. However, there is limited information addressing the fate of Pb added by MSWA dumpsites into soils in arid and semi-arid regions. The objective of this study was to examine the effects of MSWA application rates on adsorption and mobility of Pb by two soils with their different physical and chemical properties.

Table 1. Selected physical and chemical properties of soils.

Parameters	Soils	
	Loamy Sand	Sandy Loam
Sand (%)	85.5	73.5
Silt (%)	8.00	19.0
Clay (%)	6.48	7.48
CEC (cmol kg^{-1})	4.85	6.46
$CaCO_3$ (%)	23.2	34.3
OM (%)	1.04	1.07
pH (soil paste)	7.80	8.07
EC (dS m^{-1})	1.0	1.6
Cations and anions (meq L^{-1})		
Ca^{2+}	4.31	8.51
Mg^{2+}	3.42	4.10
Na^{+}	1.13	2.05
K^{+}	0.69	0.72
HCO_3^{-}	4.25	7.56
Cl^{-}	1.91	3.64
SO_4^{2-}	3.82	4.50

Material and Methods

Soil Samples

Two representative surface soil samples (0-30 cm) were collected from Dierab farm (N 24°25′, E 46°34′) 40 km south west of Riyadh. The soil sample was air-dried and sieved through a 2-mm mesh. Soil texture, pH, electrical conductivity (EC), $CaCO_3$%, cation exchange capacity (CCE), and organic matter (OM) were determined using the standard methods described in [14]. The related chemical properties of the soils are presented in Table 1.

MSW Samples and Chemical Analysis

MSW samples were collected from four different dumping sites of Hail, KSA (N 27° 30′, E 41° 43′). The MSW was incinerated in a kiln that allowed for the generated gases to pass through a water bath and avoid air pollution. The MSWA was then screened through a 0.30 mm sieve to remove un-burnt MSW, ceramics, and broken glasses. The ash was further ground using a micro-milling machine for particle sizes less than 0.063 mm. The resultant ash was analysed for total $CaCO_3$ content using the method applied [15]. The CEC was determined using sodium acetate (pH 8.20) according to the method described in [16]. Total Cd, Pb, Cu, and Zn content were digested by a concentrated mixture of HNO_3-$HClO_4$ acids and measured using induction-coupled plasma-mass spectrometry (ICP-MS, Perkin Elmer, USA). The chemical properties of MSWA are presented in Table 2.

Particle Size Analysis of MSWA

The particle size distribution analysis of the MSW ash samples was determined using the laser diffraction spectrometry method using a laser diffracto-meter. The analysis was performed in triplicate of each sub-sample with water as a dispersing agent in the wet method and air as a dispersing agent in the dry method. The dried MSWA samples were placed in the laser diffraction machine and subjected to dispersion energy in the presence of a suspension medium. The ash particle sizes were determined from the intensity of the scattered light by the particles in suspension due to their different refractive indices.

Table 2. Chemical characterization of MSWA.

Parameter	Value
SiO_2	26.0%
Al_2O_3	27.3%
CaO	31.1%
Fe_2O_3	2.50%
MgO	3.50%
K_2O	4.20%
Na_2O	2.50%
P_2O_5	1.50%
TiO_2	1.40%
CEC	8.10 cmol kg^{-1}
OM	20.0%
$CaCO_3$	11.8%
pH	12.0
Particle size distribution	
2.0-1.0 mm	0.40%
1.0-0.50 mm	36.7%
0.50-0.25 mm	30.5%
0.25-0.125 mm	20.6%
0.125-0.063 mm	7.88%
Less than 0.063 mm	3.92%

Adsorption Experiment

Soil samples were mixed with MSWA @ 0, 1, 2, 3, and 5% (w/w) with particle sizes less than 0.063 mm. The adsorption experiment was carried out by mixing 2.0 g of soils-MSWA amended with 20 mL of $PbCl_2$ solution at different concentrations varying from 0 to 300 mg Pb L^{-1}. The mixture was shaken for 24 hours at room temperature and then centrifuged at 5,000 rpm for 20 minutes. The Pb concentration in the supernatant was measured using (ICP-MS, Perkin Elmer, USA). The amount of Pb adsorbed on soils was estimated according to the following equation:

$$X/m = (C_i - C_f)\ V/W \tag{1}$$

…where X/m is the amount of the Pb^{2+} adsorbed on soils (mg kg^{-1}), C_i is the initial Pb concentration (mg L^{-1}), C_f is the final Pb concentration (mg L^{-1}), V is the volume of solution used (mL), and W is the weight of soil sample (g).

The K_d values were estimated according to [17]:

$$K_d = S/C \tag{2}$$

…where K_d is the distribution coefficient (cm^{-3} g^{-1}), S is adsorbed Pb (mg g^{-1}) and C is equilibrium Pb concentration (mg L^{-1}).

Results and Discussion

Characterization of Soils and MSWA

The summary of the physico-chemical properties of soil (CEC, pH, EC, particle size distribution, and OM) are presented in Table 1. Soil pH, EC, OM, and clay content were consistently least variable, while sand, silt, CEC, and $CaCO_3$ were the highest. The texture of soils is dominated mainly by sand fraction, which ranged from 73.5% to 85.5%. The silt fraction ranged from 8% in loamy soil to 19% in sandy soil. The soil pH is alkaline with an average of 7.94. The EC value recorded for loamy sand soil and sandy soil belong to normal classes. The soils had a high $CaCO_3$ content ranging from 23.2% to 34.3%. The soils were low in OM content (1.0-1.6%) and CEC values (4.85-6.46 cmol kg^{-1}). The results of soil properties indicated that the soil pH is alkaline and EC values are non-saline. The soils have high $CaCO_3$ content and are low in OM and CEC values.

The mechanical properties of MSWA are expressed in terms of particle sizes. As shown in Table 2, the MSWA ranged from very fine to very coarse sand sizes. The coarse particles of size range from 1.0 to 2.0 mm and represent the largest component (36.76%) of the MSWA. The summary of the values that determined chemical properties of MSWA are presented in Table 2. The predominant chemical constituents of MSWA are aluminates, silicates, and lime. The oxides of Ca, Al, and Si content were higher, while oxides of Fe, K, Na, P, and Ti were moderately variable, and CaO was the most abundant compound (31.1%). MSWA had a lower CEC and $CaCO_3$ values and higher OM content (20.0%). The physico-chemical properties of MSW primarily depend on the nature of the parent MSW composition [18]. Alkalinity is an important MSW characteristic and results from the presence of Ca, Na, Mg, and OH, along with certain other trace metals [19].

Adsorption of Pb^{2+}

The adsorption data for Pb^{2+} on the two soils as well as mixtures of these soils with MSWA are presented in Tables 3 and 4. Amounts of adsorbed Pb^{2+} on soils increased with increasing initial Pb concentration from 25 to 300 mg L^{-1}. Loamy sand soil adsorbed much more Pb^{2+} than sandy loam soil. The application of MSWA has led to decreases in the ability of soils to absorb Pb^{2+}. With increasing application rates of MSWA, the amounts of Pb^{2+} decreased on both soils. Soils with high pH and CEC values adsorbed more Pb^{2+} than those with lower pH and CEC values [20-21]. The loam sandy soil was more likely to adsorb Pb^{2+} than the sandy loam soil. The results give a clear idea of the adsorption of Pb^{2+}

Table 3. Adsorption of Pb^{2+} on loamy sand soil mixed with different application rates of MSWA.

C_0 (mg L^{-1})	Application rate of MSWA									
	0%		1%		2%		3%		5%	
	C_e (mg L^{-1})	S (mg g^{-1})	C_e (mg L^{-1})	S (mg g^{-1})	C_e (mg L^{-1})	S (mg g^{-1})	C_e (mg L^{-1})	S (mg g^{-1})	C_e (mg L^{-1})	S (mg g^{-1})
25	0.0047	4.70	0.005	4.50	0.004	4.100	0.004	3.850	0.004	3.50
50	0.0097	9.70	0.009	9.10	0.008	8.300	0.008	8.050	0.007	7.10
100	0.0195	19.5	0.019	18.5	0.017	17.0	0.016	16.1	0.015	14.8
150	0.0292	29.2	0.027	27.3	0.025	25.2	0.024	24.2	0.022	22.3
200	0.0385	38.5	0.036	36.2	0.034	33.7	0.032	32.4	0.029	29.1
250	0.0481	48.1	0.046	45.5	0.042	42.3	0.040	40.4	0.037	36.9
300	0.0575	57.5	0.055	54.7	0.051	51.0	0.049	48.7	0.045	45.2
K_d	**41.95**		**44.92**		**49.08**		**51.76**		**57.33**	
R^2	0.999		0.999		0.999		0.999		0.999	

C_0: initial Pb concentration; C_e: equilibrium Pb concentration; S: adsorbed Pb; K_d: Distribution coefficient (cm^3 g^{-1}).

on loamy sand soil and sandy loam amended with different rates of MSWA ranging from 1.0 to 5.0%. Soils amended with MSWA may change the soil's ability for Pb^{2+} adsorption because of increases in the pH of soils. Soil pH plays a major role in the adsorption of Pb because it directly controls the solubility of $Pb(OH)_2$. In addition, soil pH affects net surface charges of soils [22-23]. Although the mobility of Pb in soils is controlled by adsorption and desorption characteristics of soil, the mobility of Pb depends not only on its distribution among several species, but also on diffusion coefficients [24]. [25] studied the effect of residence time on Pb adsorption on hydrous Fe oxides and they reported that residence time had significant effect on the amount of Pb, which could be ascribed to the strong Pb into soil complexes. Reduction of Pb mobility by MSWA application may be two mechanisms: increasing pH and higher specific surface area, which causes the precipitation of insoluble phases and promotes Pb adsorption via surface complexation [26-27].

Distribution Coefficient

The calculated K_d values are presented in Tables 3 and 4. The results indicate that Pb^{2+} was distributed in soil solution and the adsorbed phase, in which a great portion accounted for the adsorbed phase. Lower K_d values indicate that most of the Pb^{2+} present in the soil system remain in the solution and will be available for transporting into soils, whereas higher values as reported

Table 4. Adsorption of Pb^{2+} on sandy loam soil mixed with different application rates of MSWA.

C_0 (mg L^{-1})	Application rate of MSWA									
	0%		1%		2%		3%		5%	
	C_e (mg L^{-1})	S (mg g^{-1})	C_e (mg L^{-1})	S (mg g^{-1})	C_e (mg L^{-1})	S (mg g^{-1})	C_e (mg L^{-1})	S (mg g^{-1})	C_e (mg L^{-1})	S (mg g^{-1})
25	0.0040	4.00	0.004	3.80	0.003	3.45	0.003	3.30	0.003	3.10
50	0.0079	7.90	0.008	7.70	0.007	7.10	0.007	6.70	0.006	6.30
100	0.0161	16.0	0.015	15.4	0.014	14.1	0.014	13.9	0.013	12.7
150	0.0241	24.1	0.023	23.3	0.022	21.9	0.020	20.1	0.020	19.6
200	0.0324	32.4	0.031	31.1	0.029	28.6	0.028	27.6	0.026	26.1
250	0.0406	40.6	0.039	39.0	0.036	36.1	0.034	34.1	0.033	32.6
300	0.0481	48.1	0.047	46.7	0.043	43.2	0.041	41.2	0.039	38.5
K_d	**52.04**		**54.26**		**59.41**		**62.95**		**67.15**	
R^2	0.999		0.999		0.999		0.999		0.999	

C_0: initial Pb concentration; C_e: equilibrium Pb concentration; S: adsorbed Pb; K_d: Distribution coefficient (cm^3 g^{-1}).

here indicate lower mobility and stabilization of Pb^{2+} and sub sub-sequentially higher retention in the soils [28]. The application of 5% MSWA to soils increased K_d values to 36.6% and 29.0% more than the control (0%). The results indicated that K_d values were significantly correlated with MSWA application rates, with R^2 value ranging from 0.974 to 0.993 (Fig. 1). In a previous study [29], Pb transport was studied in soil amended with different rates of MSWI by applying Pb solution of 150 mg L^- at the rate of 0.09 cm min^{-1} for loamy sand soil and 0.035 cm min^{-1} for sandy loam soil. The results indicated that extremely low Pb concentrations in the effluent solution were observed in leaching solution with time passing. In general, Pb^{2+} transport has been primarily found in the top 5 cm of the soil surface, which shows low mobility in the studied soils. This is usually attributed to differences in soil texture, sand, clay, and $CaCO_3$ content.

Pb is strongly bound to OM and is rather immobile in soils except at extremely high concentrations [30]. The highest K_d values for Pb^{2+} were found at 5% application rate of MSWA, followed by 3%, 2%, 1%, and control. MSWA shows higher adsorption capacity for Pb^{2+} because of its higher pH values [31]. The higher obtained K_d values with lower Pb^{2+} concentrations were probably related to different soil properties. Otherwise, Pb^{2+} adsorption becomes unspecific at higher metal concentrations, resulting in lower K_d values [32]. The transport and distribution of Pb^{2+} in soils is directly related to amounts of Pb^{2+} adsorbed [33-34]. Pb is mainly adsorbed onto specific soil adsorption sites while with higher concentration; soils lose some of their ability to bind Pb^{2+} ions. Loamy sand soil has higher K_d values than sandy loam soil at all rates of MSWA application (Tables 3 and 4). The results indicated that the K_d values were influenced mainly by soil pH, $CaCO_3$%, texture, and CEC values of soils. MSWA has the potential for Pb^{2+} removal from soils due to its chemical characterization (Table 2). The results suggest that K_d values can be used for predicting Pb adsorption on soils. The K_d constants of Pb^{2+} in soils can be related to its mobility and toxicity. The difference in Pb adsorption on soils has been reported [35-36]. Soils have the ability to immobilize Pb^{2+} mainly due to adsorption properties that may be affected by physical and chemical characterization of the soils, such as CEC, pH, textures, and the properties of Pb [37-38].

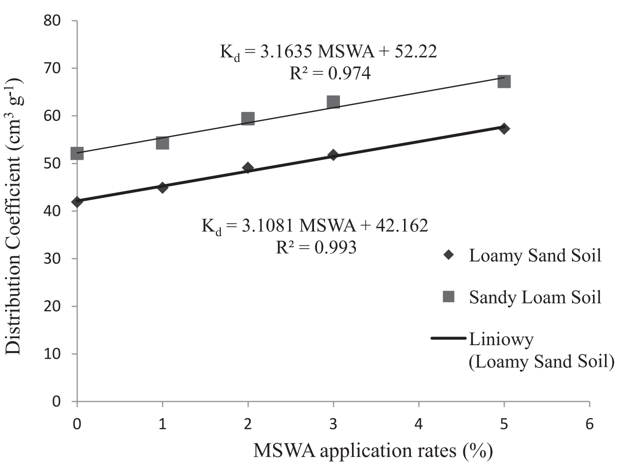

Fig. 1. Relationship between distribution coefficients of Pb and application rates of MSWA.

Conclusion

The different properties of soils, such as CEC, pH, and contents of silt and clay lead to the obvious differences in their ability for adsorption and mobility of Pb. The addition of MSWA, which contains high content of OM, has increased the soil's ability for Pb^{2+} adsorption due to the increased K_d values. The results indicate that MSWA has high power on Pb^{2+} retention in soils surface and thus may be used to remove Pb^{2+} from contaminated soils.

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