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Leaching of cadmium, chromium, copper, lead, and zinc from two slag dumps with different environmental exposure periods under dynamic acidic condition

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ABSTRACT

Over the past few decades, zinc smelting activities in Guizhou, China have produced numerous slag dumps, which are often dispersed on roadsides and hill slopes throughout the region. During periods of acid rain, these exposed slags release heavy metals into surface water bodies. A column leaching study was designed to test the potential release of the heavy metals cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), and zinc (Zn) under simulated acid rain events. Two slags with varying environmental exposure periods were packed in columns and subjected to leaching solutions of pH 3.5, 5.5, or DI H_2O at intervals of 1, 7, 14, 28, 56 d. Pulse concentrations of Cd in leachate were found above 5 μg/L, Cr, Pb, and Zn $>$10 μg/L, whereas, Cu reached 10 μg/L. After five leaching events, the leachability (percentage of cumulative heavy metal leached after five leaching events as in its respective total concentration in slags) of Cd was 0.05 percent and 0.035 percent from the old and young slag, respectively. Cr (0.035 percent and 0.05 percent) was greater than Cu (0.002 percent and 0.005 percent) and Zn (0.006 percent and 0.003 percent), while the lowest leachability was observed for Pb (0.0005 percent and 0.0002 percent) from the old and young slags, respectively. Reaction rates (release amount of heavy metals in certain period of leaching) of heavy metals in the leachates demonstrated the sequence of Zn > Cr > Cd, Cu > Pb. Leaching release of heavy metals was jointly affected by the pH of leaching solution and mineral composition of slags (including chemical forms of Cd, Cr, Cu, Pb, and Zn). Environmental exposure period of slags, resulting in the alteration of minerals, could affect the release process of heavy metals in leaching as well.

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1. Introduction

Metal smelting activities have been identified as a significant source of toxic metal pollution to the atmosphere, water bodies, soils, and crops (Little and Martin, 1972; Rieuwerts and Farago, 1996; Basta and McGowen, 2004; Bi et al., 2009; Yang et al. 2011). Smelting slag dumps have received a large amount of attention as potential pollutant sources (Kierczak et al., 2013). These slag dumps are normally rich in highly toxic metals such as Pb, Cd, and Zn (Yang et al., 2006; Navarro et al. 2008; Ettler et al., 2009), which can re-enter the surrounding environment through rainwater (Ash et al., 2013), leaching (Navarro et al., 2008; Houben et al., 2013), or dispersal by surface weathering under acidic soil conditions (Kierczak et al., 2013). In addition, these slag dumps are frequently deposited on roadside near towns and rivers, increasing the likelihood of human contact via contaminated irrigation water, consumption of garden vegetables and field crops, and inhalation of dusts (Yang et al., 2011; Douay et al., 2013). Thus, local residents, especially children, receive potential exposure risk to toxic metals in metal smelting regions (Pelfrene et al., 2013).

Release of toxic metals from smelting slags is dependent on pH, metal concentration, metal bioavailability, texture, and geochemical composition of slags, due to their highly heterogeneous in structural materials and chemical and mineral compositions (Ettler et al., 2009; Ash et al., 2013; Kierczak et al., 2013). Under surface weathering conditions, porous slags with increased reactive surface areas release more metals, and phase assemblages of slags significantly affect the availability of metals (Kierczak et al., 2013). Ash et al. (2013) stated that rainwater simulation solution can leach more Cd, Mn and Zn from smelting slags with time than DI water, and slag heterogeneity and contact time are the prominent factors for the toxic metal release from smelting slags. A pH-static leaching experiment indicated that the

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largest amount of Pb leached from primary Pb slags was at 1 yr's contact time and the lowest Pb in leachate was at pH ~7.5 (Vitkova et al., 2011). However, a 24 h slag-leaching experiment revealed that stream water and distilled water merely leached metal (metalloid) less than 0.1 percent of total content in the solid (Kierczak et al., 2013). Some researchers observed metal containing silicates and oxides (such as Zn containing phyllosilicate and magnetite, and franklinite, willemite, and hemimorphite) in smelting slag-affected soils (Manceau et al., 2000; Vespa et al., 2010), suggesting a natural attenuation mechanism (Vespa et al., 2010).

Zinc smelting activities using traditional techniques (nicknamed as manger furnaces) were widespread in northwestern Guizhou, China in the 1980s and 1990s. It was reported that coarse zinc (ingot) production surged from 4540 t in 1985 to 61,800 t in 2001. Such large scale activities of zinc smelting resulted in a large quantity of slag being deposited near the towns of Magu and Hezhang, while no barriers were constructed between slag dumps and rivers or irrigation fields. High concentrations of Pb, Zn, and Cd were detected in these smelting slags (Yang et al. 2006; Bi et al. 2009). Under the local condition of dolomite/limestone geological settings, carbonate and bicarbonate are supposed to buffer release capacity of toxic metals from slags that are subjected to weathering. However, this region is frequently affected by acid deposition resulting from the combustion of coal with high sulfur content (Zhao et al., 1988; Tanner et al., 1997). Under long term surface weathering conditions, release of toxic metals from acidic leaching of slags remains uncertain. This study aimed at dynamic leaching of toxic metals from slags with varying environmental exposure periods. It was hypothesized that the leachability of toxic metals from slags is related to slag exposure time. The objectives of this study were to survey the leaching behavior of Cd, Cr, Cu, Pb, and Zn from slags under simulated acidic conditions, and to discuss the limitation of pH and slag properties on the leachability of these metals.

2. Materials and method

2.1. Slags

Two zinc mining slags were sampled from the smelting region in northwestern Guizhou, China. The first had an environmental exposure of ten years while another one was exposed for only two years. Slags were air dried, cracked, and passed through a 1 mm sieve before analysis. The pH of slags was determined in slurry using a pH meter (Multi 3420, WTW, Germany) at a DI water to slag ratio of 1:1 (V/W) after the mixture was shaken for 30 min on a reciprocating shaker. The two year old slag, had higher pH (8.1) and heavy metal concentrations of Cd 25 mg/kg, Cr 87 mg/kg, Cu 101 mg/kg, Pb 1.0 percent, and Zn 1.27 percent, as compared with the ten year old slag with a pH of 3.9 and metal concentrations of Cd 10 mg/kg, Cr 40 mg/kg, Cu 65 mg/kg, Pb 616 mg/kg, and Zn 0.10 percent (Table 1). X-ray diffraction results showed that the dominant minerals in the old slag were quartz, calcite, gypsum, and magnesite, and in the young slag were quartz, calcite, gypsum, montmorillonite, illite, and dolomite (Fig. 1).

2.2. Column leaching

Considering the main chemical composition of regional acid deposition is sulfate (Tanner et al., 1997), acid leaching solution were constructed using 0.02 N sulfuric acid by adjusting DI water to pH 3.5 and 5.5. During each leaching event, 285 mL of solution was applied, totaling 1425 mL of acid solution, which corresponds to half the annual rainfall in the slag sampling region. For comparison, deionized water from a Mini power machine (with pH 5.9) was used for a leaching control.

About 1.0 kg of slag material was packed into a Plexiglas column (30.5-cm long, 6.6-cm inner diameter), with several 5-mm diameter holes at the bottom. The design of the column is described by Yang et al. (2008). Prior to the leaching experiment, the slag columns were saturated with deionized water for three days from the bottom of the column, to remove air pockets and keep the equilibrium of chemical and biological reactions. Leaching was conducted with five leaching events at days 1, 7, 14, 28, and 56. The leaching solution was supplied using a peristaltic pump at a speed of ~2.5 mL per minute. The leachate was collected from each leaching event and analyzed for pH, conductivity, and heavy metal concentrations. All experiments were run in triplicate.

2.3. Chemical analysis

The pH and EC of leachate were determined using a pH/conductivity portable meter (Multi 3420, WTW, Germany) according to US EPA method 150.1. Concentrations of heavy metals (Pb, Zn, Cd, Cu, and Cr) in the leachate after filtering through a 0.45 μm membrane were determined using an Inductively Coupled Plasma Mass Spectrometry (ICP-MS Platform ICP, GV instruments, UK) following US EPA method 200.8.

2.4. Quality assurance and quality control (QA/QC)

QA/QC for ICP-MS was accomplished using initial calibration (standard curve) verification (ICV), QC sample, matrix spike sample and laboratory blank. The recoveries of ICV were in 90–110 percent; Recoveries of QC sample were in the ranges of 94–10, 91–114, 94–107, 94–114, and 91–119, respectively for Cd, Cr, Cu, Pb, and Zn (EPA 200.8). Matrix spike recoveries were 80–120 percent. Laboratory blanks (DI H_2O) were less than method detection limits.

2.5. Leachability

Leachability of metals from slags was calculated as

$$\text{Leachability (percent)} = \Sigma\,(C_{i,j} \times V_i)/M_j \quad (1)$$

where i represents leaching events, j is heavy metals Cd, Cr, Cu, Pb, and Zn, C is the concentration of metals in leachate, V_i is the leachate volume, and M is the mass quantity of element j in 1 kg slag.

2.6. Reaction rate

Release amount of metals ($A_{i,j}$, nmol/kg) from slags at each leaching event was calculated as

$$A_{i,j} = C_{i,j} V_0 t_i / W \quad (2)$$

where $C_{i,j}$ is the concentration of heavy metal J in leachate at leaching event i, V_0 is the velocity of the peristaltic pump, t_i is leaching time of each leaching event i, and W is the mass quantity of slags in the column (1 kg).

Table 1
Chemical compositions of slags used in this study.

Samples		pH	EC (μS cm^{-1})	Total C(g kg^{-1})	Total Al (mg kg^{-1})	Total Fe (mg kg^{-1})	Total Ca (mg kg^{-1})	Total Mg (mg kg^{-1})
Old slag	Mean	3.9	2779	48.1	14,037	77,208	3849	1088
	Stdev	0.02	14.6	0.42	700	3103	189	50.1
Young slag	Mean	8.1	2331	67.8	17,505	61,195	21,513	1984
	Stdev	0.02	14.9	2.65	469	904	359	46.8

Samples		Total P (mg kg^{-1})	Total Cd (mg kg^{-1})	Total Cr (mg kg^{-1})	Total Cu (mg kg^{-1})	Total Pb (mg kg^{-1})	Total Mg (mg kg^{-1})
Old slag	Mean	212	9.98	39.3	64.9	616	1028
	Stdev	7.2	0.464	0.952	2.58	73.8	20.1
Young slag	Mean	310	25.0	87.1	101	9728	12,721
	Stdev	8.4	1.17	3.19	3.41	1214	148

Stdev: standard deviation.

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