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The behavior and long-term fate of metals in simulated landfill bioreactors under aerobic and anaerobic conditions

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1. Introduction

An issue of debate in the solid waste community is the behavior and fate of metals disposed in municipal solid waste (MSW) landfills [1]. Metal concentrations in landfill leachate vary, for instance, by landfill age and the types of waste discarded, and concentrations are typically thought to become reduced as the waste is stabilized [2]. The fate and mobility of metal-bearing wastes is important on several fronts. Modern landfills in most countries utilize liner systems to prevent environmental contamination, and while the risk of chemical migration from well designed and operated containment systems is small, elevated pollutant concentrations in leachates raise concerns over long-term risks and policies related to land disposal of certain wastes. For example, in the US, concerns over the landfilling of discarded consumer products such as electrical devices [3,4] and treated wood [5,6] often center around the mobility of metals.

Multiple investigations have examined the potential for leachability of metal-bearing wastes under landfill conditions. Many of these studies utilized batch tests [3,7], but conditions occurring within a landfill are often much more complicated than can be measured in a short-term test. For example, during the initial stage of anaerobic waste decomposition, pH values are relatively low as a

ABSTRACT

The long-term behavior and fate of metals in leachate from four simulated bioreactor landfills were explored using lysimeters under both aerobic and anaerobic conditions for a maximum of 1650 days. Metal concentrations varied with time and stage of landfill activity. The behavior of selected metals (Al, As, Cr, Cu, Fe, Pb, and Zn) significantly differed between aerobic and anaerobic conditions. Leachate from the aerobic lysimeters contained greater concentrations of Al, Cu, and Pb compared to leachate derived from the anaerobic lysimeters (average concentrations of Al, Cu and Pb in the aerobic/anaerobic lysimeters were 8.47/0.78 mg/L, 1.61/0.04 mg/L and 0.10/0.03 mg/L, respectively). In the anaerobic lysimeters, As, Fe and Zn leached at greater concentrations (average concentrations of As, Fe and Zn in the aerobic/anaerobic lysimeters were 0.40/1.14 mg/L, 13.5/136 mg/L and 15.3/168 mg/L, respectively). Though no significant difference in overall Cr concentrations was observed in leachate samples from aerobic and anaerobic lysimeters, during the alkali and methane phases approximately 45% of Cr was presented as Cr(VI) under aerobic conditions, whereas no Cr(VI) was detected under anaerobic conditions.

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result of acid formation, and metals can more easily dissolve and migrate with leachate because of enhanced solubility [8]. Once the methanogenic phase is reached, leachate metal concentrations usually decrease as a result of increasing pH. However, other pathways for metal migration in landfills exist, including oxidation-reduction reactions [1], precipitation as sulfides [9], complexation with inorganic and organic ligands [10,11], sorption onto clays [12], and cation interactions [13]. To help account for these complex factors, simulated landfills can be utilized to more realistically replicate pollutant fate in landfilled waste and associated changes with time [4,14].

In this study, simulated landfill lysimeters were used to study the differences in metal leachate concentrations between landfill bioreactors (landfills operated to promote waste stabilization) under aerobic and anaerobic conditions. Since leaching behavior is dominated by pH, oxidation-reduction potential (ORP), and the presence of ligands [8,15], the behavior and fate of some metals in aerobic systems should differ from anaerobic systems. MSW landfills, including those operated as bioreactors and unless specifically operated using air addition, will be anaerobic. The motivation for including aerobic landfill conditions as part of this research was two-fold. First, the addition of air to landfills as a means of accelerating waste decomposition has been practiced on a limited scale [16]. Second, stabilized aerobic waste may provide an indication of the longer-term fate of metals in landfills after the predominant anaerobic decomposition has occurred [2]. In this study, aluminum (Al), arsenic (As), copper (Cu), chromium (Cr), iron (Fe), lead (Pb),

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- 1. peristaltic pump; 2. air purging; 3. leachate collection port; 4. wet-tip gas totalizer; 5. temperature controller; 6. gas-sampling bag; 7. hydraulic cylinder; 8. heating tape;
- 9. hydraulic jack; 10. plunger; 11. fabricated wastes

Fig. 1. Schematic of the aerobic and anaerobic lysimeters used for the research.

and zinc (Zn) in leachate from aerobic and anaerobic simulated landfill lysimeters were routinely analyzed and compared over an extended monitoring period (a maximum of 1650 days). Metal concentration trends are evaluated as a function of time, pH, and stabilization phase.

2. Materials and methods

2.1. Simulated bioreactor landfill lysimeters

A detailed schematic of the lysimeter is presented in Fig. 1. Four lysimeters were constructed, each consisting of a stainless steel column and carriage system. The 183-cm (6 ft) stainless steel body contained 5 front ports and 1 valve at the bottom for leachate collection. The front ports were used for air addition (in the case of the aerobic lysimeters). A small port located on the top flange was used for liquids addition. Perforations in the steel plate allowed added liquids to percolate into the waste. The amounts of gas produced from each lysimeter were monitored using a gas totalizer.

The carriage system was designed to support a hydraulic pressurizing unit installed at the top of each lysimeter for the application of an external load to the fabricated waste. The fabricated waste was composed, by dry weight, of typical MSW components including office paper (28%), cardboard (15%), food waste (15%), plastic (15%), and other materials based on MSW composition estimates reported for the US and Florida [17,18]. Additional metal sources were added: aluminum and galvanized steel sheet (Al, Fe, and Zn), crushed cathode ray tube (CRT) monitor glass (Pb), and chromated copper arsenate (CCA) treated wood (Cr, Cu, and As). A load of 98 kPa (2040 lb/ft²) was applied as the overburden pressure. Additional details of waste composition and processing methodology are provided in the online supplementary materials.

2.2. Experiment operations

Two lysimeters each were operated under aerobic and anaerobic conditions. Temperature and air addition were controlled to simulate aerobic or anaerobic bioreactor conditions. To main-

Table 1

Leachate characteristics of the aerobic and anaerobic lysimeters.

Parameter (mg/L)	Aerobic lysimeters		Anaerobic lysimeters	
	Acid phase	Alkali phase	Acid phase	Methane phase
рН	4.5-7.0	7.0-9.2	4.5-7.0	7.0-7.9
COD (mg/L)	6000-70000	200-9000	40000-80000	7000-45000
BOD (mg/L)	3000-50000	5-3000	18000-60000	700-18000
Alkalinity (mg/L as CaCO ₃)	500-14000	50-2400	13000-18000	9000-15000
Ammonia (mg/L)	30-300	10-450	100-1,600	700-1,500
Sulfide (µg/L)	0-200	100-1200	50-600	700-2800
Metals (mg/L)				
Al	0.40-10.9	3.50-16.6	BDL ^a – 2.10	BDL – 0.50
As	0.01-1.01	0.13-0.90	0.25-3.20	0.20-0.70
Cr	0.02-0.20	0.10-0.45	0.01-0.45	0.02-0.20
Cu	0.10-25.65	0.60-5.60	BDL ^a – 0.06	BDL – 0.10
Fe	1.50-190	0.90-8.10	12.10-600	5.60-330
Pb	0.01-1.70	0.01-0.10	BDL ^a – 0.12	BDL – 0.05
Zn	5.50-270	2.55-16.80	6.4-455.50	2.80-67.40
Percentage of leaching of selected metals (%) ^b)			
As	0.05%	0.38%	2.26%	0.10%
Cr	0.02%	0.16%	0.16%	0.06%
Cu	1.59%	1.69%	0.08%	0.06%

^a Detection limits: Al = 0.03 mg/L; Cu = 0.004 mg/L and Pb = 0.003 mg/L.

^b The initial concentrations of As, Cr and Cu in CCA-treated wood = 2350 mg/kg, 2890 mg/kg and 1330 mg/kg, respectively.

tain aerobic conditions, air was added and maintained at a rate between 70 and 120 mL/min; most of the operational period was at a rate of 70 mL/min. The temperature was maintained at 55 °C for both aerobic and anaerobic lysimeters using heating tape and a type T thermocouple wire (SRT201-160, Omega). Eight liters of deionized water was added to each lysimeter during waste compaction. After loading, 11L of additional water was added. The waste in the lysimeter was brought to field capacity (58% was targeted as this was the field capacity measured for this waste under the initial compaction conditions of the lysimeter). Approximately 200-300 mL of leachate was collected from the bottom and added back to the top of the lysimeter every week. Approximately 100 mL of collected leachate was used for analysis. In addition the volume used for analysis was supplemented by adding deionized water. Additional deionized water was also added to the aerobic lysimeters to replenish moisture lost through air stripping, maintaining the moisture content of the waste at field capacity (approximately 58%). Since the decomposed waste was exhumed for other research purposes at different times, the experimental periods for the aerobic and anaerobic lysimeters differed; the aerobic lysimeters were operated and monitored for 370 days (aerobic 1) and 1280 days (aerobic 2), and the anaerobic lysimeters for 760 days (anaerobic 1) and 1650 days (anaerobic 2).

2.3. Analytical methods

The leachate parameters that were measured included pH, ORP (Accumet Co. Model 20), alkalinity (Standard method, 2320B), dissolved oxygen (DO) (YSI Inc. Model 55/12 FT), specific conductance (HANNA Instruments, Model H19033), biochemical oxygen demand (BOD) (Standard Method 5210B), ammonia (Standard Method 4500D), and chemical oxygen demand (COD) (Standard Method 5220D) [19]. Leachate samples were randomly selected during the experimental period and analyzed for hexavalent chromium (Cr(VI)) using the USGS Method I-1230-85 [20]. A total of seven leachate samples selected from each aerobic and anaerobic lysimeter were analyzed for Cr(VI).

Leachate samples were digested for metal analysis with nitric and hydrochloric acids following EPA method 3010A [21]. Digested samples were analyzed for seven metals (Al, As, Cr, Cu, Fe, Pb, and Zn) using Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES, Thermo Jarrell Ash Corp. Model 95970). Laboratory blanks, duplicates, and calibration samples were performed as appropriate. Analytes were not detected above the detection limits in any of the blanks. Metal concentrations in the aerobic and anaerobic lysimeters were statistically compared using an analysis of variance (ANOVA) test at the 0.05 level of significance.

3. Results and discussion

3.1. Leachate characterization

The results of general water quality parameters over time are shown in Table 1. Changes in pH and COD over the course of the experiment are presented in Fig. 2. These two parameters served as important indicators to describe the leachate condition and the extent of waste degradation and stabilization. The acid/alkali and acid/methane phases of the aerobic and anaerobic lysimeters in Table 1 were distinguished using leachate pH. Leachate from both the aerobic and anaerobic lysimeters remained acidic during the beginning of the experiment. Leachate pH in the aerobic lysimeters remained from 5.5–6.5 until day 170 and increased to 9 at day 210; it then remained relatively constant in the range of 8.5-9.3 (average of 8.8). An increase of pH values upon adding air has been reported in other studies, such as composting of organic matter [22,23]. The pH values in leachate from the anaerobic lysimeters were predominantly in the range of 5-6 in the first 500 days. After day 500, pH values began to increase and reached close to a pH of 8 at the end of the experiment. No considerable pH change was observed in the lysimeters after day 1000. The period of time required to stabilize the pH for the aerobic and anaerobic lysimeters was 200 and 600-800 days, respectively. Average pH measurements of approximately 8.6 (aerobic) and 7.8 (anaerobic) were observed at the end of the experiment. These results are similar to the ranges of pH observed in other landfill leachate studies [24-26].

The initial COD concentrations of the anaerobic lysimeters were near 80,000 mg/L, whereas the maximum COD concentrations of the aerobic lysimeters (aerobic 1 and 2) were 70,000 and 40,000 mg/L, respectively. The COD concentrations from both aerobic and anaerobic lysimeters decreased over time and the changes in COD corresponded to changes in pH; COD lowered as pH became stable. A common observation as the waste decomposition stabilizes is a decrease in degradable organic matter. The ranges of DO measurements in the aerobic and anaerobic lysimeters were 2–7 mg/L and 0.1–0.7 mg/L, respectively.



Fig. 2. Changes in pH and COD of the aerobic and anaerobic lysimeters over time.

3.2. Changes of metal concentrations in leachate from aerobic and anaerobic lysimeters

Changes in selected metal concentrations of the aerobic and anaerobic lysimeters with time are depicted in Fig. 3A–G. In addition, the distribution of metal concentrations in the aerobic and anaerobic lysimeters over the entire course of the experiment are compared using box-and-whisker plots. To show the entire range of metal concentrations, the *y*-axis was plotted with a logarithmic scale.

Relatively large concentrations of Al were observed from both aerobic (18 mg/L) and anaerobic (20 mg/L) lysimeters for the first 10–20 days (Fig. 3A). While the Al concentrations of the anaerobic lysimeters dramatically decreased below 0.5 mg/L within 100 days, no significant change in Al was observed from the aerobic lysimeters. Initially, As concentrations in the aerobic lysimeters were lower and then increased over time. However, As concentrations in leachate from the anaerobic lysimeters began high and continually decreased over time (Fig. 3B). The greatest As concentration was 3.2 mg/L on day 90. The As concentration then lowered below 1.0 mg/L after day 800 for the two remaining lysimeters (aerobic 2 and anaerobic 2).

The initial Cr concentrations of the anaerobic lysimeters were higher than those of the aerobic lysimeters, as shown in Fig. 3C. However, Cr concentrations in the anaerobic lysimeters gradually decreased below 0.05 mg/L by day 450. After day 500, increases in Cr concentrations were observed in the anaerobic lysimeter. The Cr concentration in the aerobic lysimeter gradually decreased after day 700 to just above 0.01 mg/L at the end of the experiment. Overall, Cu concentrations from the aerobic lysimeters were one to three orders of magnitude higher than those of the anaerobic lysimeters, as shown in Fig. 3D. Copper concentrations of the aerobic lysimeters decreased after day 600 and reached similar levels to that of the anaerobic lysimeters, at the end of the experiment. For the anaerobic lysimeters, Cu concentrations gradually decreased for the first 450 days and slightly increased afterward. Copper levels in the anaerobic lysimeters were relatively constant after day 1000.

Iron concentrations of the aerobic lysimeters (110 mg/L)were initially higher than those of the anaerobic lysimeters (20-22 mg/L), as shown in Fig. 3E. Iron concentrations of the aerobic lysimeters increased to 250 mg/L on day 30, and then dramatically decreased to less than 10 mg/L throughout the experiment. In the anaerobic lysimeters, Fe concentrations increased from 20 to 600 mg/L for the first 450 days and then dropped to less than 1 mg/L. Lead concentrations in the aerobic lysimeters dramatically increased to 1.7–2 mg/L within 30 days, then decreased to levels less than 0.1 mg/L (Fig. 3F). In the anaerobic lysimeters, no dramatic changes in Pb concentrations were observed throughout the experiment. As shown in Fig. 3G, leaching patterns of Zn were similar to those of Fe for both aerobic and anaerobic lysimeters (see Fig. 3E). Relatively high concentrations of Zn were observed from both aerobic and anaerobic lysimeters for the first 150 days. The highest Zn concentrations from the aerobic and anaerobic lysimeters were 270 and 450 mg/L, respectively. Zinc concentrations substantially decreased for aerobic and anaerobic lysimeters to below 10 mg/L after day 600 and day 1000, respectively.

3.3. Factors affecting leachate metal concentrations

Multiple factors influence the metal concentrations in landfill leachate. The benefit that makes simulated landfills valuable – the ability of these systems to account for multiple different influences – also limits their ability to conclusively determine the causes of leachate concentration changes. For example, metal concentrations are understood to be pH dependent. However, in addition to pH, concurrent factors such as the amount of water that has contacted the waste over time and the oxidizing or reducing conditions present, also affect resulting concentrations. To further examine the different factors, the data are presented in several additional manners.

Table 1 summarizes the range of leachate concentrations in each phase for both aerobic and anaerobic lysimeters. For the aerobic lysimeters, the average metal concentrations for Cu, Fe, and Zn were greater in the acid phase than in the alkali phase, while Al, As, and Cr



Fig. 3. Changes in metal concentrations of the aerobic and anaerobic lysimeters over time.

showed higher average concentrations in the alkali phase. For the anaerobic lysimeters, metals were predominantly leached during the acid phase, with the exception of Cr.

The results demonstrated that aerobic conditions have the potential to increase the solubility of certain metals (Al, Cr, Cu, and Pb). Flyhammar and Hakansson [27] concluded that the mobility of heavy metals would increase as the landfill becomes oxidized and this was dictated by complexation reactions with dissolved organic substances. Martensson et al. [15] also observed almost two times greater concentrations of selected heavy metals in aerobic landfill leachate compared to those in anaerobic landfills as a result of a

decrease of buffering capacity and metal binding capacity under aerobic conditions. The other plausible explanations include the solubility changes of metals, such as Al and Cr, as pH increases. Fig. 4 presents the metal concentrations as a function of pH instead of time. Many of these plots display the classic amphoteric shape that many metals exhibit, with the greatest leachability occurring at the extreme ends of the pH range. Also influencing these plots, however, are different redox states and liquid-to-solid ratios.

Once metals are dissolved in water, they can be exposed to metal precipitating ions, such as sulfide, carbonate, and organic matter, and can subsequently be precipitated by creating a water insoluble



Fig. 4. Lysimeter leachate metal concentrations as a function of pH.

metal–ligand complex [28]. A common sink for some metals in anaerobic landfill systems (e.g., Cu, Pb) is the formation of sulfide precipitates. The solubility of some metals may be greatly influenced by oxidation state and reduced in the presence of oxygen. For example, the water solubility of Fe(II) is substantially greater than that of Fe(III) and relatively small amounts of Fe were measured in the aerobic lysimeters. Fe(II) can oxidize to Fe(III) mineral, such as Fe(OH)₃, at high redox conditions, and thus be entrained in the waste matrix; aeration is one of the most widely used remediation techniques for iron removal from iron polluted groundwater [29,30].

High concentrations of As, Cr, Fe, and Zn were observed in the acid phase of the anaerobic lysimeters. The low pH that developed before the onset of a reducing environment in the anaerobic lysimeters may have resulted in increasing metal solubility. The metal concentrations, however, were decreased to a large extent upon entering the methane phase as a result of increases in pH and sulfide concentrations. Erses and Onay [31] reported that approximately 90% of Cu, Ni, Fe, and Zn were attenuated by sulfide precipitation in the methane phase. A great reduction of some metals in land-

fill leachate during the methanogenic phase was also observed by Bilgili et al. [24].

To better evaluate the factors impacting leachate metal concentrations other than pH, concentrations were plotted as a function of time after the leachate reached relatively stable conditions (see Fig. 5). Using pH and organic content of the leachate as an indicator, day 600 and 1000 were designated as times when leachate became stabile in the aerobic and anaerobic lysimeters, respectively. During this phase, metal concentrations were observed to dramatically drop in the aerobic lysimeter. According to research conducted to predict the long-term fate of heavy metals in landfills, heavy metals would be released as air intrudes, resulting in oxidation of humic substances and sulfides [2,8]. The loss of the buffering capacity of landfill leachate in the presence of oxygen may also result in a pH drop due to waste degradation, and thus may enhance the metal mobility [8]. However, the research results show that after the organic fraction of the waste completely decomposed, the heavy metal concentrations in leachate dropped to a large extent. Mott et al. [32] explained that sorption of a variety of heavy metals may take place on the surface of amorphous oxides of iron. Heavy metal



Fig. 5. Lysimeter leachate metal concentrations with time once leachate stabilization is reached (day 0 = day 600 for aerobic lysimeters and day 1000 for anaerobic lysimeters).

removal from wastewater and landfill leachate by aeration would be a good example of metal sorption on iron oxide [33].

Although the concentrations of some metals changed to a degree in the anaerobic lysimeter, no dramatic changes in metal concentrations were observed after leachate became relatively stable (Fig. 5). This indicates that metal concentrations could be lowered as the leachate becomes stable, but a trace level of heavy metals may remain for an extended time under anaerobic conditions. With regard to the long-term fate of heavy metals in landfill leachate, these results suggest that metal concentrations could temporarily increase when air is intruded into a landfill, but would ultimately decrease with time.

3.4. Metal leaching differences illustrated using CCA-treated wood

In this research, CCA-treated wood was used as a source of As, Cr, and Cu. In addition to the implication of these results from a landfill operating perspective, the three metals represented here (As, Cr, and Cu) provide good examples of how individual metals behave differently in aerobic and anaerobic landfill environments. The relative amount of the three metals leached in each landfill environment can be more clearly understood by comparing the leaching percentages of As, Cr, and Cu (Table 1). Overall, As leaching under anaerobic conditions was greater compared to aerobic conditions. Overall Cr leaching in the aerobic and anaerobic lysimeters was not significantly different. Copper leaching in the aerobic lysimeter, however, was substantially greater than that in the anaerobic lysimeter.

Arsenic leaching is strongly dependent on pH and ORP condition. The greatest As leaching occurred in the anaerobic columns at low pH values. Similar pH values under aerobic conditions, however, leached substantially less As. A possible cause is that under reducing conditions, the predominant form of arsenic will be As(III), which is more soluble and mobile than As(V), the predominant phase under oxidizing conditions [6].

Although overall Cr concentrations were low, the Cr concentrations increased slightly upon entering the methane phase in both anaerobic lysimeters. No Cr(VI) was detected during this phase. Thermodynamically, the solubility of Cr(III) would be lower at pH 6 through 10 [34,35]. Other similar research showed that Cr concentrations in leachate from an anaerobic landfill (simulated) were lower when pH was stabilized around neutral [24,36]. However, relatively high concentrations of Cr at neutral pH and reducing conditions have been observed when CCA-treated wood was present as the major Cr source in landfills [37,38]. A plausible explanation is that some Cr is present in CCA-treated wood as Cr-As complex [39] and, thus, Cr release may be subject to As solubility at low ORP conditions. Another explanation is that Cr solubility increased as a result of lower Fe concentrations; the Cr-Fe complex reduces Cr solubility lower than the Cr-OH complex. In the presence of Fe. Cr may have precipitated with Fe rather than OH due to rapid kinetics [40]

At lower pH values, Cr in an aerobic landfill can be present as an ionic form, such as Cr(VI), under oxidizing conditions, and the cumulative mass may increase over a period of time. Although total concentrations of Cr leached were not considerably high, thermodynamically Cr(VI) becomes a major Cr species in the environment formed upon air intrusion. It was found that the average percentage of Cr(VI) contained in total Cr in the aerobic lysimeters was approximately 45%, whereas no Cr(VI) was detected in the leachate samples from the anaerobic lysimeters.

High Cu leaching under aerobic conditions was also noticeable. Copper solubility is controlled by several Cu-containing minerals with Fe and sulfide. In addition, Cu-sulfides may coexist with the sulfides of other metals such as Zn, Pb, and As [41]. Copper is easily precipitated as a Cu–sulfide complex in anaerobic conditions, thus Cu concentrations in the anaerobic lysimeters were substantially low. Thermodynamically, ionic Cu is predominantly present at a pH less than neutral and under highly oxidizing conditions. However, it may not be sufficient to account for relatively high Cu concentrations in the aerobic lysimeter under alkaline conditions. The other important factor to control Cu solubility in aerobic conditions may be microorganism-mediated leaching. Edwards et al. [30] reported high Cu concentrations in drinking water at alkali conditions. The water color changed to blue; Critchley et al. [29] explained that the Cu-rich blue water was caused by microbial activity. In this research, blue colored water was also observed from the condensate created between the aerobic lysimeters and a gas totalizer.

4. Summary and conclusions

This research explored the long-term behavior and fate of metals leached from simulated landfill lysimeters loaded with fabricated MSW and a variety of metal sources. The experimental results presented here add to previous research exploring the issue of heavy metals in landfills and leachate [1-3,24,26,31] by providing a controlled examination of the difference encountered between aerobic and anaerobic environments, both during acid phases of waste decomposition and stabile phases when the majority of biological activity has been completed. The behavior of selected metals (Al, As, Cr, Cu, Fe, Pb, and Zn) significantly differed by aerobic/anaerobic conditions and leachate pH. Leachate from the aerobic lysimeters contained higher concentrations of Al, Cu, and Pb compared to leachate derived from the anaerobic lysimeters. In the anaerobic lysimeters, As, Fe, and Zn leached at higher concentrations. Although the median of Cr concentrations in the aerobic lysimeters was greater than that of the anaerobic lysimeters, Cr concentrations highly varied over phases of the aerobic and anaerobic lysimeters.

Two metal-containing wastes of current concern, electronic waste and CCA-treated wood, were specifically examined, and provided a good contrast of metal leaching behavior among elements and under differing conditions. Concentrations of Pb in leachate (a result of CRT glass), usually low in most landfill environments, was greatest during the acid phase in the aerobic lysimeters. While landfills are for the most part reducing environments, Cr(VI) accounted for nearly 45% of the total Cr found in leachate during the alkali phase of the aerobic lysimeters. Large amounts of As were leached during the acid phase of the anaerobic lysimeters and gradually decreased, while relatively low As concentrations were maintained in the aerobic lysimeters for the entire period.

Metal concentrations in leachate of the aerobic lysimeter dramatically reduced as waste stabilized, while no significant changes in most of the metal concentrations were observed in the anaerobic lysimeter. If it is assumed that an anaerobic landfill at the end of waste stabilization will transition to aerobic conditions as a result of air intrusion, metal concentrations in landfill leachate may thus be expected to become substantially lower with time. As more engineered landfills reach their final capacity and are closed, future studies should examine metal concentrations in leachate from operating systems to confirm whether these observations hold true under expected full-scale conditions.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jhazmat.2011.07.119.

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