Metal loss from treated wood products in contact with municipal solid waste landfill leachate

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ABSTRACT

The research presented in this paper evaluates the potential impact of municipal solid waste (MSW) landfill leachate quality on the loss of metals from discarded treated wood during disposal. The loss of arsenic (As), chromium (Cr), copper (Cu), and boron (B) from several types of pressure-treated wood (CCA: chromated copper arsenate, ACQ: alkaline copper quaternary, CBA: copper boron azole, and DOT: disodium octaborate tetrahydrate) using leachate collected from 26 MSW landfills in Florida was examined. The toxicity characteristic leaching procedure (TCLP), the synthetic precipitation leaching procedure (SPLP), and California's waste extraction test (WET) were also performed. The results suggested that loss of preservative components was influenced by leachate chemistry. Copper loss from CCA-, ACQ- and CBA-treated wood was similar in magnitude when in contact with landfill leachates compared to synthetic TCLP and SPLP solutions. Ammonia was found as one of the major parameters influencing the leaching of Cu from treated wood when leached with MSW landfill leachates. The results suggest that disposal of ACQ- and CBA-treated wood in substantial quantity in MSW landfills may elevate the Cu concentration in the leachate; this could be of potential concern, especially for a bioreactor MSW landfill in which relatively higher ammonia concentrations in leachate have been reported in recent literature. For the As, Cr and B the concentrations observed with the landfill leachate as the leaching solutions were over a range from some sample showing the concentrations below and some showing above the observed value from corresponding SPLP and TCLP tests. In general the WET test showed the highest concentrations.

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1. Introduction

The majority of waterborne wood preservatives contain one or more heavy metals. The most prevalent wood preservative in recent years, chromated copper arsenate (CCA), contains chromium (Cr), copper (Cu), and arsenic (As). Several studies have reported concerns about the human-health and environmental impacts of As and Cr from CCA-treated wood [1–9]. With the phase-down of CCA-treated wood from residential applications, newer formulations have been introduced. Copper is the primary component in many of these alternative preservatives; some preservatives also use boron (B). The alternative wood preservatives commercially available include alkaline copper quaternary (ACQ), copper boron azole (CBA), and disodium octaborate tetrahydrate (DOT).

Previous research has shown that pressure-treated wood products release metals to some extent when exposed to water [1,6,10–13]. One of the several scenarios where loss of metals from pressure-treated wood poses a concern is final disposal [14–17]. The large demand for pressure-treated wood products in the past few decades results in the disposal of significant amounts of these materials at the end of their service life [18–20]. Landfills represent one of the primary disposal methods for discarded pressure-treated wood products. Since treated wood does release metals in aqueous environments, a potential concern for a landfill operator accepting this material is elevated metal concentrations. Groundwater contamination is a concern at unlined landfills. For lined landfills, preservative chemicals can accumulate in the collected leachate. At most lined landfills, leachate is sent for treatment to an offsite wastewater treatment plant (WWTP), and because these facilities often impose pretreatment standards, elevated metal concentrations in leachate can result in extra fees to the landfill operator or possibly the denial of service. An additional concern with respect to elevated concentrations is the ultimate fate of these metals. In a WWTP, the metals in the leachate accumulate in the biosolids

and since biosolids are often land applied, this could result in the distribution of metals to the environment.

Laboratory leaching tests are widely used to assess the chemical releases of landfilled wastes. Several tests have been developed and used by regulatory agencies for this purpose. The toxicity characteristic leaching procedure (TCLP) was designed to simulate the leaching conditions that occur when a potentially hazardous waste is disposed of in a municipal solid waste (MSW) landfill [21,22]. MSW is the garbage produced by household and commercial activities. Wastes are deemed hazardous if the amount of chemical leached from the waste exceeds a set threshold, or toxicity characteristic (TC) limit. The TC limit for As and Cr is 5 mg/L for each; no TC limit is set for Cu. In prior studies, CCA-treated wood has been found to leach As using TCLP at concentrations greater than the TC limit [12,14,15]. While the Cu-based preserved wood products do not leach As or Cr, they do leach several times more Cu than CCA-treated wood, especially under TCLP [12]. Although the TCLP has been used to assess the leaching of metals from waste disposed of in MSW landfills, this procedure has potential limitations in simulating the leaching conditions that truly occur in a MSW landfill [23–26]. An alternative approach for assessing the ability of a pollutant to leach from waste co-disposed of in a MSW landfill is to perform a batch leaching test similar to the TCLP, but using actual landfill leachate as the leaching solution [25–28].

The objective of the research presented here was to assess the impact of landfill leachate quality (e.g., pH, ammonia concentrations of extraction fluid) on metal releases from discarded treated wood products. The release of As, Cr, Cu, and B was measured from several types of pressure-treated wood (CCA, ACQ, CBA, and DOT) using leachate collected from 26 MSW landfills. The TCLP, the synthetic precipitation leaching procedure (SPLP), and California's waste extraction test (WET) were also performed on these samples to compare the results between synthetic leachates and actual leachates. A goal of this study was to compare the impact on MSW landfill leachate quality on disposal of CCA-treated wood and As-free, treated wood products. The present study differs from previous treated wood batch leaching studies in that the leaching solutions used, in addition to regulatory-based solutions, included leachate collected from full-scale landfills.

2. Materials and methods

2.1. Sample collection and preparation

Sawdust samples of CCA-, ACQ-, CBA-, and DOT-treated wood were generated by cutting dimensional lumber into small blocks using a power saw. The dimensional lumber was purchased from different home improvement stores in Florida. Separate blades were used for each wood type to avoid cross contamination and the power saw was vacuum cleaned before samples of each wood type were prepared.

2.2. Determination of total extractable metal concentrations

Total extractable As, Cu, Cr, and B concentrations (arithmetic mean of three replicate ± corresponding standard deviation) in the treated wood sawdust samples were obtained through a hot acid digestion (Method 3050B [29]) and analyzing the digestates using inductively coupled plasma atomic emission spectroscopy (ICP-AES, Thermo Electron Corporation, Trace Analyzer) as per Method 6010B [29].

2.3. Landfill leachate collection

Leachate samples were collected from 26 MSW landfill sites in Florida. Eleven (Sites 2, 5, 7, 9, 10, 14, 16, 20, 24–26) of the 26 land-

fill sites were active landfills, which means they were accepting waste when samples were collected from the leachate collection system. The amount of waste accepted per day at these active sites ranged from 370 to 1200 metric tons per day. The remaining 15 sites were closed at the time of sample collection, some closed as recently as 2 years prior to sample collection, while several others had been closed for over 10 years. The lined cell areas from which leachates were collected for these 26 landfill sites ranged from 4.85 ha to 50.6 ha. The different sites provided the opportunity to collect wide varieties of leachate characterized by a wide range of water-quality parameters. Leachate samples were collected either from the sump of the leachate collection system (LCS) or from the leachate collection pipe outlet into a manhole of the LCS. The leachate samples were collected in 5-gallon HDPE containers while maintaining minimum headspace. Samples were collected using the facilities' existing pumping system when available ($n = 10$). For landfills which did not have a pumping station, leachate samples were bailed using a Teflon bailer ($n = 16$). Leachate parameters measured in the field immediately upon sample collection included pH and specific conductance. The leachate samples were transported to the laboratory and stored at 4 °C until the extraction test was performed. Separate aliquots of each leachate sample were collected in accordance with sampling procedures of different leachate parameter analyses.

2.4. Laboratory extraction procedures

Wood samples were subjected to 29 extraction solutions, 26 of which were the landfill leachates and 3 of which were those prescribed by the following regulatory leaching procedures: TCLP, SPLP [29], and WET [30]. The pH of the landfill leachates ranged from 5.75 to 8.10, a pH range typical of modern MSW landfills [31,32]. The TCLP solution was prepared by mixing 0.1 M acetic acid and 1N sodium hydroxide in a ratio of 1:11 and diluting the mix with DI water to achieve the pH of the solution as 4.93 ± 0.05. The SPLP solution was prepared by mixing sulfuric acid and nitric acid in a ratio of 3:2. The pH of the solution used was 4.20 ± 0.05. The WET, a supplementary test used in California, uses a buffered citric acid solution as the leaching fluid. The WET extraction solution was prepared by titrating a 0.2 M citric acid solution with 4.0N sodium hydroxide to a pH of 5.0 ± 0.05.

All the extractions were carried out at room temperature (approximately around 25 °C). The extraction procedure for the leaching with MSW leachates followed the TCLP procedure, with the exception of the extraction fluid (details presented in Table 1). For the WET test, nitrogen purging of the leaching fluid was conducted immediately before extraction. After extraction in a rotary extractor, samples were filtered using a pressurized filtration apparatus with a 0.7-μm borosilicate glass fiber filter (Environmental Express TCLP filter). The filtrate was preserved with a few drops of concentrated nitric acid (to reduce pH < 2.0). The extraction for each sample type was performed in triplicate for each of the 29 extraction fluids. The arithmetic mean of the concentrations from these three replicates and the corresponding standard deviation are presented in this paper. Results of analysis of blank samples were consistently below detection limits; matrix spike samples and calibration check samples showed recoveries between 87% and 110%.

2.5. Impact of solution pH on leaching

ACQ and CBA sawdust samples were leached with modified TCLP solutions prepared following the procedure presented in Section 2.4, but with additional pH adjustment using 1N NaOH to achieve desired pH values. The purpose was to evaluate the relative impact of pH on Cu leaching under conditions where the original

Table 1
Summary of leaching test procedures used in this study.

	Standardized leaching test			MSW leachate	Impact of solution pH
	TCLP	SPLP	WET		
pH of leaching solution	4.93 ± 0.05 (acetic acid and sodium hydroxide)	4.20 ± 0.05 (sulfuric and nitric acids)	5.00 ± 0.05 (citric acid and sodium hydroxide)	6.97[a]	Two set of extractions were conducted: 1. TCLP solution with pH adjusted to 5.97 and 7.65[b]; 2. DI water with pH adjusted to a range of values from 4.3 to 8.4[c]
Solid to liquid ratio (gram of waste to liter of solution)	100 g/2 L	100 g/2 L	200 g/2 L	100 g/2 L	100 g/2 L
Extraction period	18 ± 2 h	18 ± 2 h	48 h	18 ± 2 h	18 ± 2 h

[a] Average of 26 landfill leachates.
[b] The pH of the regular TCLP solution was adjusted to the desired pH value by adding 1N NaOH until the pH was achieved.
[c] The diluted HNO_3 and NaOH was added to the DI water to achieve five extraction solutions in the pH range of 4.3–8.4.

TCLP acetic acid content was the same, and pH values of 5.97 and 7.65 (within the typical range of MSW landfill leachate pH) were selected. Additionally, a pH impact experiment was also performed on ACQ and CBA sawdust samples by adjusting the pH of DI water (extraction fluid) with 1N HNO_3 and 1N NaOH as needed in the pH range of 4.3–8.4. The pH was monitored and adjusted throughout the leaching duration of 18 h. Other procedures for these leaching tests were similar to those for TCLP. Detailed procedures of the pH impact study are presented in Dubey [33].

2.6. Leachate analysis

Leachate collected from the landfills was characterized for typical leachate quality parameters, including metals, sulfides, alkalinity, total dissolved solids (TDS), biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), total organic carbon (TOC), and ammonia. US EPA methods [29] and other standard methods [34] were employed when applicable. The detection limit for As, Cu, Cr, and B were 12 μg/L, 4 μg/L, 4 μg/L, and 6 μg/L, respectively. Volatile fatty acids were measured by gas chromatography (Shimadzu GC 9-AM fitted with a column of 10% SP1000 and 1% H_3PO_4 in Chromosorb WAW 100/120) with a flame ionization detector (FID). Samples were centrifuged at 10,000 rpm for 10 min and the resultant supernatant was acidified (v/v) with 1 part sample to 9 parts of 20% H_3PO_4 containing 1000 mg/L of isobutryate.

3. Results and discussion

3.1. Total metal content in the sawdust samples

The CCA sawdust samples contained 2350 ± 50 mg-As/kg (arithmetic mean of three replicate ± corresponding standard deviation), 2890 ± 56 mg-Cr/kg and 1330 ± 10 mg-Cu/kg, which is equivalent to CCA Type-C treated wood with 5.4 kg/m^3 retention. The rated retention for CCA-treated wood as indicated by the manufacturer was 6.4 kg/m^3. The rated retention values reported by the manufacturer are based on the outer 1.5 cm (0.6 in.) of the wood. The total metal analysis results presented here were conducted on the sawdust collected through the entire cross section of the wood and thus the results were slightly lower and consistent with the manufacturer's rating. Copper and B concentrations were 2860 ± 40 mg-Cu/kg, 360 ± 20 mg-B/kg for ACQ-treated wood sample and 5420 ± 120 mg-Cu/kg, 810 ± 20 mg-B/kg, for CBA-treated wood sample. Calculated retention values based on the Cu concentrations for ACQ- and CBA-treated wood were 1.9 kg/m^3 as CuO and 3.6 kg/m^3 as CuO, respectively. Rated retention of ACQ- and CBA-treated wood was 4.0 kg/m^3 and 6.4 kg/m^3, respectively. The borate-treated wood contained 0.15% of B (1690 ± 15 mg-B/kg) by weight (retention value of 5.07 kg/m^3 as B_2O_3).

3.2. Landfill leachate characterization

Chemical characteristics of the leachates collected from the 26 landfill sites (Table 2) show that leachate parameters values covered a wide range as did data reported in literature. Conductivity readings correlated with the TDS values as expected. A number of landfill investigation studies [35] have suggested that the stabilization of waste proceeds in sequential and distinct phases. In general, any landfill site can be broadly classified as either being in an acidogenic phase or in a methanogenic phase based on the prevalent stage of waste decomposition at that particular site. Leachate pH and its VFA concentration are considered indicators of these phases. Although the transition from the acidogenic phase to the methanogenic phase is not distinct, landfill leachate with pH < 7.0 is generally considered to be in the acidogenic phase. The 25 landfill sites out of 26 from which the leachates were collected have been

Table 2
Composition of landfill leachate collected from 26 landfill sites.

Parameter	Present study (overall)	Range reported in literatures[a]	Leachates from landfills in acidogenic phase	Leachates from landfills in methanogenic phase
pH	6.97[b] (5.75–8.10)[c]	4.5–9.0[c]	6.60 (5.75–6.89)	7.30 (7.0–7.57)
Conductivity (ms/cm)	7.63 (1.54–31.2)	2.5–3.5	5.6 (1.54–11.82)	8.10 (2.90–13.3)
TDS (mg/L)	3830 (880–15,300)	NR[d]	3160 (880–8000)	3630 (1380–5540)
Alkalinity (mg/L as $CaCO_3$)	2500 (400–10,500)	NR	1660 (400–3750)	3890 (1050–4900)
TOC (mg/L)	380 (46–1890)	30–29,000	420 (46–1890)	280 (80–600)
BOD_5 (mg/L)	430 (7.5–2400)	20–57,000	500 (7.5–1980)	130 (25–285)
COD (mg/L)	2020 (220–10,930)	140–152,000	1860 (220–9660)	1400 (330–2840)
BOD_5/COD	0.18 (0.02–0.74)	–	0.2 (0.02–0.74)	0.11 (0.03–0.29)
Sulfides (μg/L)	2360 (10–32,000)	NR	206 (10–1450)	4670 (10–32,000)
Ammonia-N (mg/L)	325 (11.5–1620)	50–2200	220 (11–500)	350 (90–665)
Total VFA (mg/L)	215 (<1.0–3420)		498 (<1.0–3420)	7.7 (<1.0–53.1)
As (mg/L)	0.035 (0.012–0.165)	0.01–1.0	0.021 (0.012–0.037)	0.05 (0.012–0.165)
B (mg/L)	1.50 (0.02–5.50)	NR	0.8 (0.010–2.0)	2.15 (0.02–5.50)
Cr (mg/L)	0.043 (0.006–0.164)	0.02–1.50	0.03 (0.006–0.09)	0.05 (0.01–0.16)
Cu (mg/L)	0.031 (0.011–0.10)	0.005–10	0.05 (0.006–0.101)	0.02 (0.004–0.071)
Sites[e]	1–26	–	2–5, 7–10, 15–17, 19–20, 24	1, 6, 11–14, 21–23, 25, 26

[a] Reinhart and Grosh [32]; Kjeldsen et al [31].
[b] Average.
[c] Range (Min–Max).
[d] NR = not reported.
[e] Site 18 was not included in acidogenic and methanogenic classification.

classified into the two categories. Site 18, consisting of a vertical well at a bioreactor landfill, was not included in this classification as the leachate from this well was very different in terms of leachate parameters that were measured (pH = 8.10, TOC = 10,000 mg/L and COD = 10,900 mg/L).

Out of 25 landfill sites, 14 were found to be in the acidogenic phase and the remaining 11 in the methanogenic phase, based on the pH of the leachate samples from these sites. The average pH for the acidogenic and methanogenic leachate was 6.60 and 7.30, respectively. Another parameter which may indicate the phase of waste degradation in a MSW landfill is the volatile fatty acid (VFA) concentration in the leachate. Among the landfill sites identified as being in the acidogenic phase, the average VFA concentration was 498.2 mg/L with a range from <1.0 mg/L to 3420 mg/L. For the leachates identified as methanogenic leachate, only 4 out of 12 leachates had VFA concentrations above the detection limit with the average and range of concentrations being 7.6 mg/L and <1.0–53 mg/L, respectively. Concentrations of other leachate parameters evaluated were in same order of magnitude for both categories of leachate.

3.3. Average preservative loss from pressure-treated wood in contact with different leachates

The results of the average inorganic preservative components loss from the different pressure-treated wood samples evaluated in this study are presented in Fig. 1. The average concentration axis for CCA, ACQ, and CBA graphs are in logarithmic scale to present the data from different leaching tests together. MSW concentrations in the figure represent the average of various element concentrations obtained on leaching with 26 landfill leachates as extraction fluid.

3.3.1. Chromated copper arsenate-treated wood
The amount of As and Cr released from CCA-treated wood in TCLP, SPLP, WET, and MSW leaching (Fig. 1(A)) followed the same trend as observed previously in Townsend et al. [14]. The highest As loss was observed with WET (56 mg-As/L), followed by TCLP (11.3 mg-As/L), SPLP (7.7 mg-As/L), and MSW (4.4 mg-As/L), respectively. The average final pH values for WET, TCLP, SPLP and MSW leachates were 4.88, 4.84, 4.74, and 6.95, respectively (with initial pH as 5.00, 4.93, 4.20, and 6.97, respectively). The pH dropped for all the extractions except for SPLP where the final pH was higher than the initial pH. The pH of the SPLP solution was controlled by the

buffering capacity of the wood since the SPLP fluid has a very low buffering capacity. The average Cr concentration followed the same pattern as As with higher levels observed with the WET extraction (41 mg-Cr/L), followed by TCLP (3.7 mg-Cr/L), SPLP (1.95 mg-Cr/L), and average MSW concentrations (1.2 mg-Cr/L). The TCLP As concentration exceeded the TC limit of 5 mg-As/L. As (56 mg/L) and Cr (41 mg/L) concentration under the WET leaching test also exceeded the corresponding soluble threshold limit concentration (STLC) of 5 mg/L for both the elements.

Copper leached the most under WET (73 mg/L) followed by TCLP (11 mg/L), MSW (4.5 mg/L), and SPLP (2.2 mg/L). Similar observations were made by Townsend et al. [14] except that more Cu leached with MSW leachate than with TCLP. As can be observed from the pH values mentioned above, the pH of the SPLP, WET, and TCLP leachate was statistically similar (4.74–4.88), with the average pH of MSW (6.95) greater by more than 2 units when compared to the other three. Although the pH was higher for the MSW leachate than for SPLP, higher Cu leaching was observed with MSW leachate. This could be due to the presence of organic acids which tend to complex with Cu to produce soluble complexes. The average concentration of VFA in the leachate was 215 mg/L. Along similar lines, citrate in WET extraction fluid extracted more Cu than acetate in TCLP fluid because citrate makes more stable complexes than acetate due to its multi-dentate character [36]. One more point that needs to be noted here is that in the WET test twice the amount of sample is leached compared to other leaching tests reported here. WET uses a liquid-to-solid ratio of 10:1 whereas other leaching tests use a liquid-to-solid ratio of 20:1. A previous study [37] has shown that for most elements, including As, Cr, and Cu, the element concentrations in the extraction fluid increase as the liquid-to-solid ratio decreases. As stated earlier, Cu is not a TC element, but it is regulated in California. A Cu concentration of 73 mg/L in the WET leachate exceeded the STLC limit for Cu of 25 mg/L. Hence, the CCA-treated wood sample used in this study fails TCLP for As and fails WET for all three elements (As, Cr, and Cu).

3.3.2. Alkaline copper quaternary-treated wood
Average inorganic preservative component loss from ACQ-treated wood under WET, SPLP, and TCLP and with MSW landfill leachate is presented in Fig. 1(B). The average final pH of the extracts under different leaching tests was 4.89 for WET, 5.70 for SPLP, 4.86 for TCLP, and 7.03 for MSW leachates. No significant change in pH of TCLP, WET and landfill leachate was observed when com-
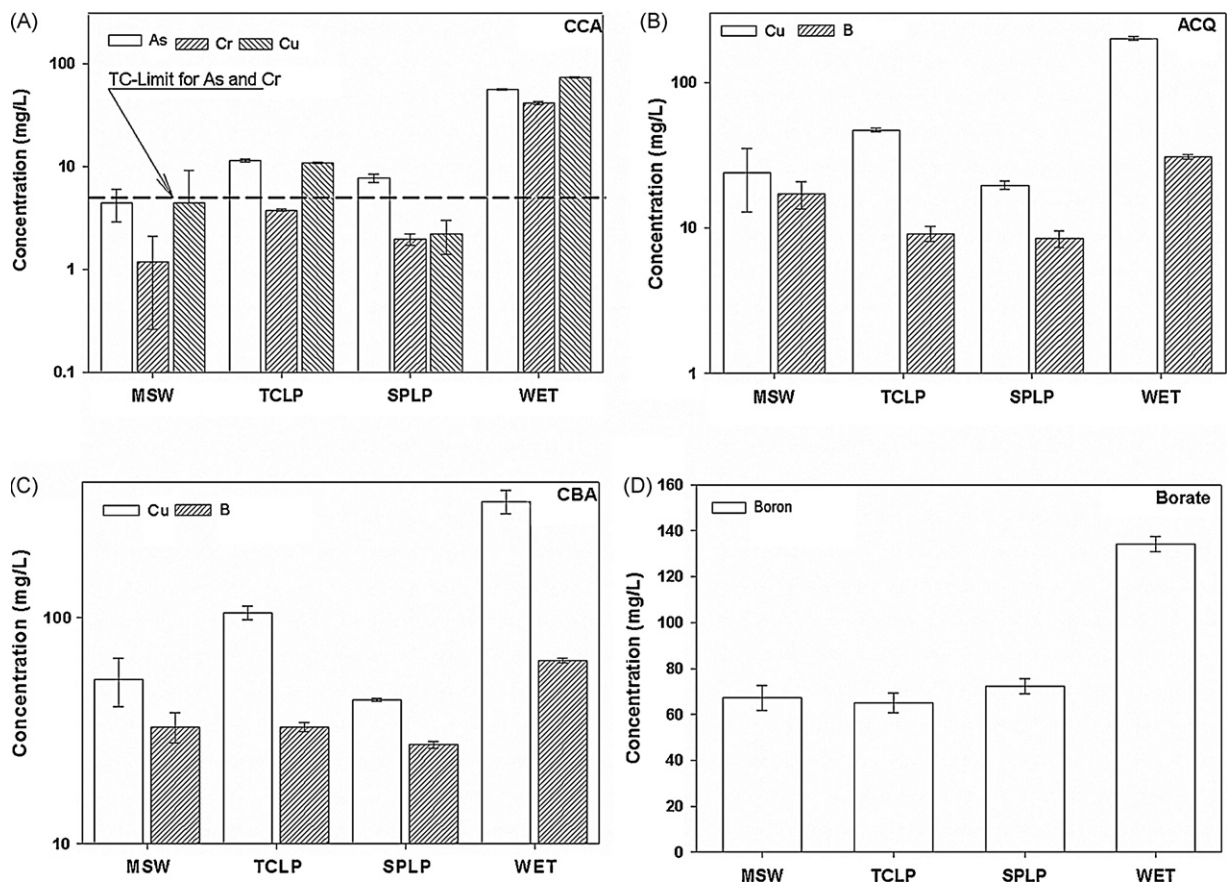
Fig. 1. Average preservative loss from wood products in contact with various leaching fluids (A) CCA, (B) ACQ, (C) CBA, (D) DOT (error bar represents standard deviation of three replicates for TCLP, SPLP and WET; for MSW leaching the error bar represents standard deviation of 78 samples (three replicate per leachate from 26 sites).

pared to initial values (Table 1), but the final pH of the SPLP extract increased by 1.5 units. Copper leaching for ACQ followed the same trend as for the CCA sample presented in the previous section. Higher Cu loss was observed under WET (200 mg/L) followed by TCLP (47 mg/L), MSW (24 mg/L), and SPLP (20 mg/L). A similar trend was observed for SPLP and TCLP Cu leaching for ACQ-treated wood by Stook et al. [12]. A comparatively lower pH of TCLP and WET, along with presence of citrate in WET and acetate in TCLP, resulted in more Cu leaching out in these extractions. The average Cu concentrations for SPLP and MSW leaching were similar, although pH was higher for MSW. This again could be due to organic acids in MSW leachates [36]. Copper concentrations for ACQ samples also exceeded the STLC limit (for WET) of 25 mg/L for Cu. Therefore, the ACQ-treated wood sample used in the present study would qualify as a hazardous waste in California. Boron loss from ACQ sawdust was highest for WET leachate (31 mg/L), followed by the concentrations in MSW (17.2 mg/L), TCLP (9.1 mg/L), and SPLP (8.4 mg/L).

3.3.3. Copper boron azole-treated wood

For CBA-treated wood, copper loss was highest under WET (average final pH = 4.90) followed by TCLP (average final pH = 5.05), MSW (average final pH = 6.83), and SPLP (average final pH = 5.54) (Fig. 1(C)). Average Cu concentrations in the leachates samples were 327 mg/L for WET, 104 mg/L for the TCLP, 53.7 mg/L for the MSW leachates, and 31.0 mg/L for SPLP. Stook et al. [12] also observed more Cu loss from CBA-treated wood under TCLP extraction test as compared to observed SPLP concentration.

When CBA sawdust samples were extracted with modified TCLP solutions at pH of 5.97 and 7.65, similar concentrations of Cu were measured compared to the non-adjusted TCLP. A pH impact experiment revealed that for the pH range of 4.3–8.4, Cu concentrations

were reduced marginally as the pH of the extraction solution was increased (results are presented in detail in Dubey [33]). These two observations indicated that pH was not a major factor influencing Cu loss from CBA-treated wood in the pH range encountered in this experiment. The presence of multiple inorganic (ammonia, Cl⁻, SO_4^{2-}, CO_3^{2-} etc.) and organic ligands (i.e. CH_3COO^-) in a MSW leachate would lead to the formation of soluble Cu complexes, resulting in the higher Cu concentration in the extracted fluid when the treated wood is in contact with landfill leachate. Boron leaching of CBA samples followed the same trend as for the ACQ samples, with highest loss reported for WET leachate (65 mg/L) followed by MSW (33 mg/L), TCLP (32 mg/L), and SPLP (17 mg/L).

3.3.4. Borate-treated wood

The average final pH values of the extracts were 4.95 (WET), 4.86 (TCLP), 6.85 (SPLP), and 7.10 (average for leaching with MSW leachates). As observed for the other treated wood products, a substantial increase of pH was observed in the SPLP extraction fluid (from 4.20 to 6.85). Highest B loss was observed under the WET extraction (134 mg/L). TCLP, SPLP, and MSW leachates (average of 26 extractions in triplicates) concentrations were in the statistically similar range of 65 mg/L, 72 mg/L, and 62 mg/L, respectively. Higher concentrations were observed with the lower final pH of the extraction fluid.

3.4. Variation of preservative leaching with different landfill sites

3.4.1. Arsenic and Cr leaching as a function of landfill leachate source

Arsenic leaching from CCA-treated wood samples extracted with various landfill leachates as the leaching fluid has been presented in Fig. 2(A). The arsenic concentration in the leachate from
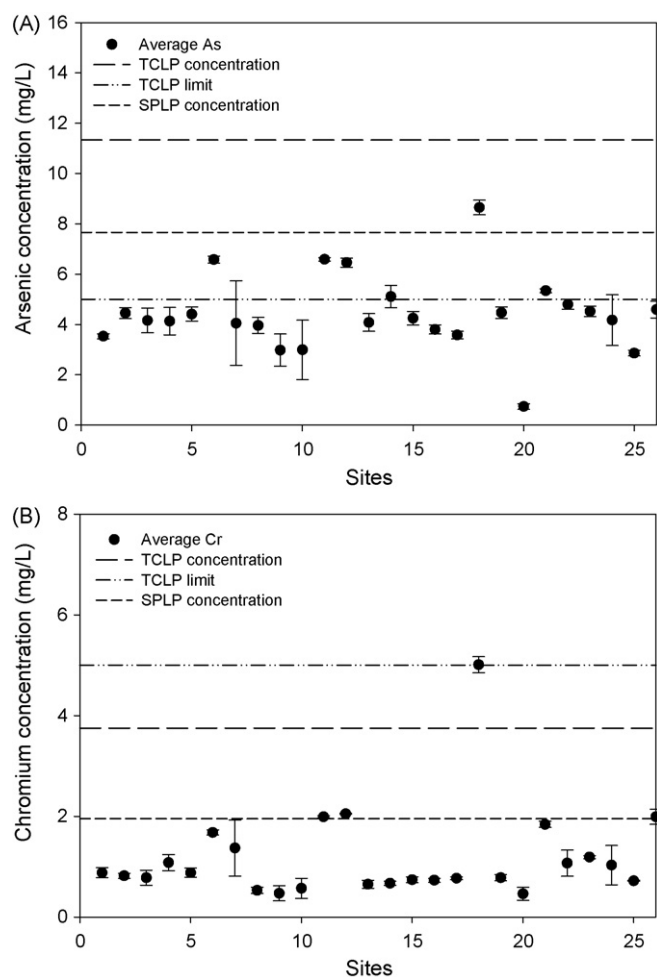
Fig. 2. Arsenic and chromium loss from CCA-treated wood samples in contact with various landfill leachates (error bars represent standard deviation among replicates), (A) Arsenic, (B) Chromium.

Site 18 was the highest among the MSW leachates, was higher than the corresponding SPLP concentration, but was lower than the TCLP concentration. The As concentrations in 6 out 26 CCA samples extracted with MSW leachate exceeded the TC limit for As. For the remaining 20 CCA leachates, As concentration varied between 0.74 mg/L and 4.80 mg/L. No specific trend with leachate pH was observed for As leaching. The pH of the leachate samples (used for extraction) varied from 5.80 to 8.10. In a previous study investigating the pH impact on preservative leaching from CCA-treated wood, As concentration was found to be similar to the samples in this pH range [14].

The average TCLP concentration was greater than that measured using actual landfill leachates. Thus, while some recent studies suggest that As may leach more in actual landfill leachate than does under TCLP [25,28], the opposite was observed for CCA-treated wood samples in the current study. A previous study [26] also found that As leaching from TCLP and MSW leachates vary with the pH of the waste material being evaluated. For the waste materials evaluated in that study [26], it was found that waste with a pH greater than 7 leached more arsenic with landfill leachates compared with TCLP, while the waste with pH = 2.9 through 6.3 leached more arsenic with TCLP compared with nine landfill leachates used in the study. The pH of the CCA sawdust used in the present study was 4.83 (similar to the second category of the waste evaluated in study by Dubey et al. [26]). On comparison of the As concentrations in extracts produced on leaching of samples with different

MSW landfill leachates with the corresponding leachate parameters (i.e., of extraction solution), it was observed that there was a general trend of higher As concentration in the extract produced with MSW leachate that had a higher alkalinity concentration ($R^2 = 0.38$).

Chromium loss from CCA-treated wood samples leached with various landfill leachates as the leaching fluid is presented in Fig. 2(B). The Cr concentration (5.1 mg/L) in the CCA leachate produced from MSW landfill leachate from Site 18 exceeded the TC limit for Cr (5 mg/L). The TCLP concentration of 3.74 mg-Cr/L was greater than that measured using most of the actual landfill leachates. Here again, although some recent studies suggest that chromium may leach more in actual landfill leachate compared to TCLP [25,28]; the same was not observed for Cr waste evaluated in this study. This again could be the result of the pH of the CCA sawdust as explained for As in the previous paragraph. Four out of 26 CCA leachate samples with MSW leachates exceeded the corresponding SPLP concentrations. For the remaining 22 CCA leachates, Cr concentration varied between 0.46 mg/L and 1.99 mg/L. When comparing the Cr leaching from different landfill leachates with leachate parameters, no trend with leachate pH was observed but there was a general trend of higher Cr concentration in the extract produced from the landfill leachates that had a higher alkalinity concentration ($R^2 = 0.55$).

3.4.2. Boron leaching as a function of landfill leachate source

Boron loss from pressure-treated wood samples leached with several landfill leachates as the leaching fluid has been presented in Fig. 3. For the DOT sawdust sample, the B concentration varied from 61 mg-B/L to 76 mg-B/L. If 100% B leached out from DOT-treated wood the leachate concentration would be 84.5 mg/L. Out of 26 landfill sites, three samples had concentrations higher than the corresponding SPLP concentration. Ten of the DOT leachates were characterized by concentrations between the TCLP and SPLP concentrations. The remaining samples had concentrations below the TCLP concentration. In general, B leached at higher concentrations from DOT-treated wood for all of the leaching fluids relative to that for ACQ- and CBA-treated wood; this is likely due to the presence of higher boron concentrations in DOT-treated wood. For the ACQ- and CBA-treated wood, in general TCLP and SPLP, B concentrations were lower than B concentrations in the leachate produced on leaching with MSW landfill leachates. Variation of the B concentrations with various landfill sites followed similar patterns for both ACQ and CBA samples with relatively higher concentrations recorded for the CBA samples.

3.4.3. Copper leaching as a function of landfill leachate source

Copper leaching from pressure-treated wood samples leached with several landfill leachates as the leaching fluid has been presented (Fig. 4). For the CCA sample, Cu concentrations varied from 0.8 mg-Cu/L to 23 mg-Cu/L. Out of 26 landfill leachates, two samples had concentrations higher than the corresponding TCLP concentration (Fig. 4(A)). Both of these had highest concentrations of ammonia (1620 mg/L and 625 mg/L) in the leaching fluid. Copper concentration was found to be higher than the SPLP concentration in 13 samples.

For the ACQ-treated wood, in general, the TCLP Cu concentration was higher than Cu concentrations in the leachate produced on leaching of ACQ sawdust samples with landfill leachate with the exception of Site 13 (47.8 mg/L). In 15 out of 26 landfill leachate extractions, Cu concentrations in the resulting leachates were higher than the corresponding concentrations in the SPLP extracts (Fig. 4(B)). The final pH of ACQ landfill leachate extracts varied from 5.91 to 7.47. In the pH impact experiment on this pH range for ACQ-treated wood samples, Cu concentration reduced to 27 mg/L at pH = 8 compare to 41 mg/L at pH = 4.8 (see Dubey [33]) for details).
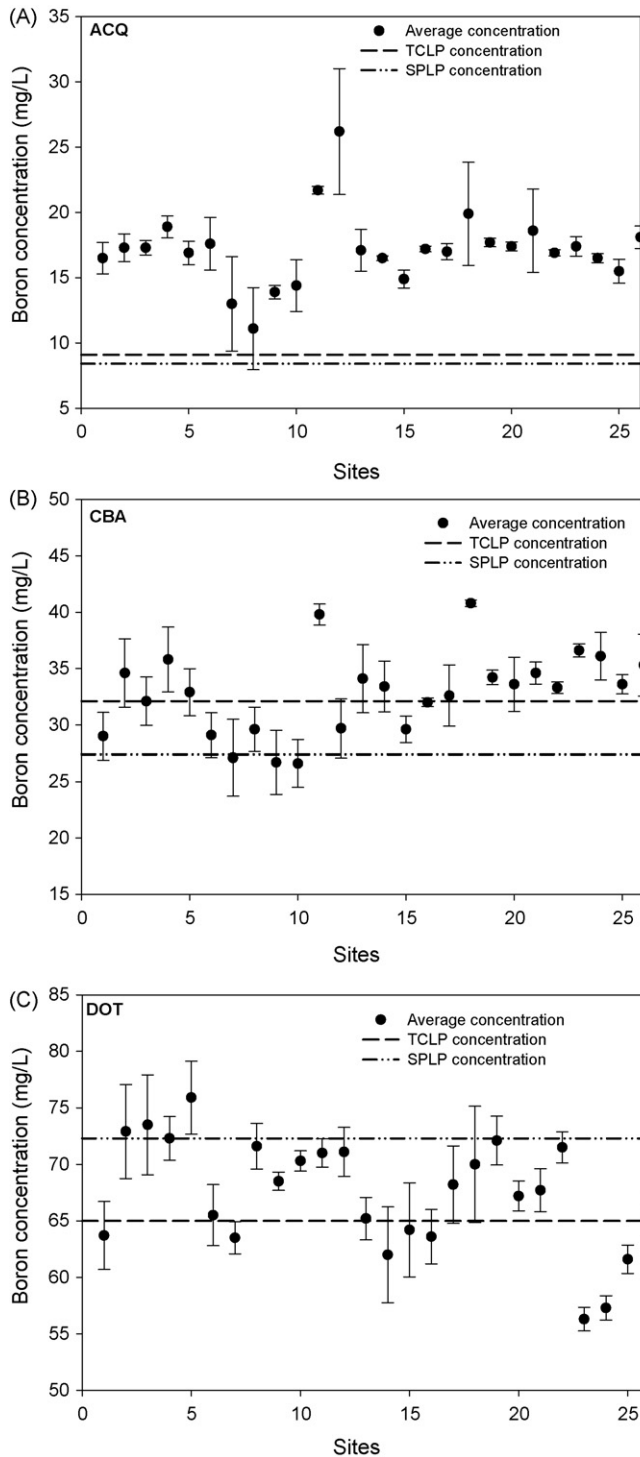
Fig. 3. Boron leaching from ACQ-, CBA- and DOT-treated wood samples leached with various landfill leachates as the leaching fluid (error bars represent standard deviation among replicates).
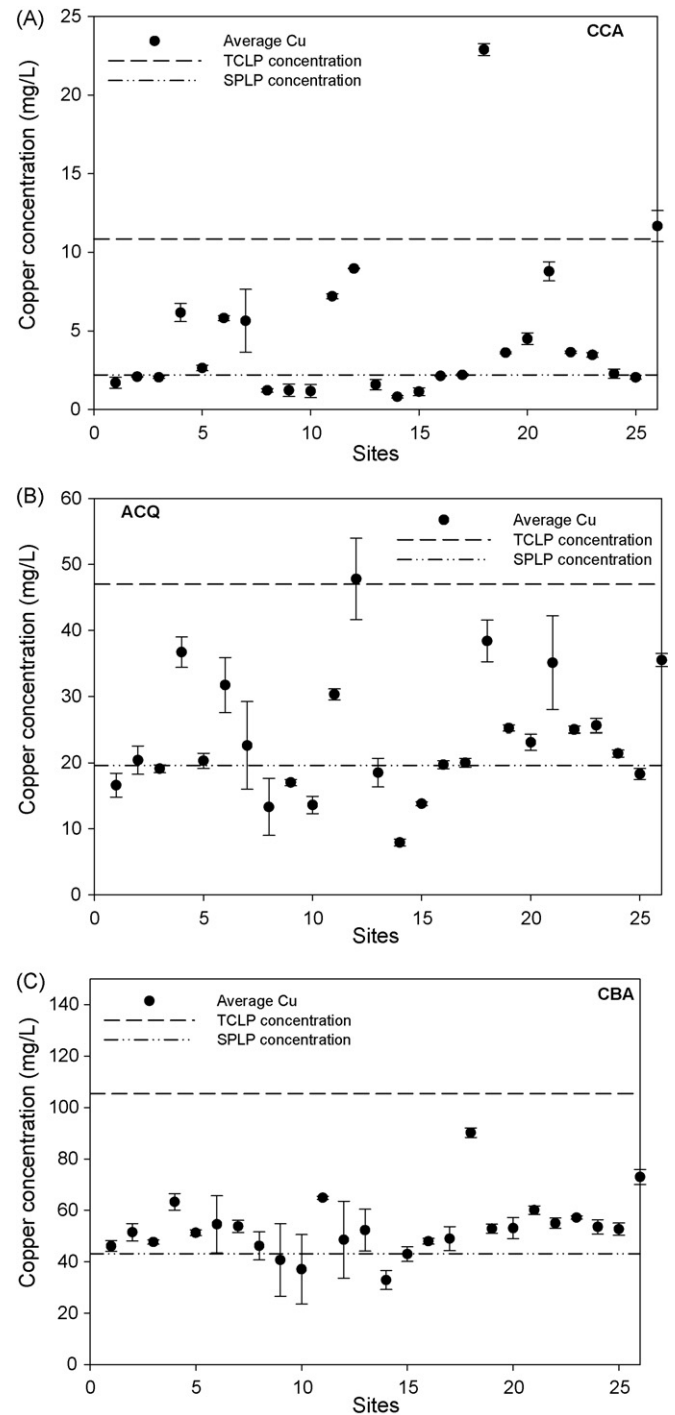

Fig. 4. Cu leaching from pressure-treated wood samples leached with several landfill leachates as the leaching fluid (error bars represent standard deviation among replicates) (A) CCA, (B) ACQ, and (C) CBA.

Lower pH in the TCLP extract compared to MSW leachate extracts could have impacted Cu leaching.

For the CBA samples the average Cu concentrations in the leachate produced from MSW leachates was lower than the TCLP Cu concentration. Comparing the impact of pH on Cu leaching from CBA, it was found that reduction in the concentration of Cu leaching from CBA-treated wood as a function with the increase in pH of the leaching fluid was not as pronounced as for ACQ samples. The total Cu concentration in the CBA sawdust samples used in the

leaching tests was 5420 mg/kg, which is nearly four times the Cu concentration in CCA (1330 mg/kg) and nearly two times the total Cu concentration in ACQ (2860 mg/kg). In the Cu-rich CBA system, the hydrated Cu-ion makes a temporary complex with amine (or ammonia) solution, which helps to minimize metal corrosion in the treatment plant and improve Cu penetration and distribution in the treated lumber [38]. The high amine and Cu concentration in CBA-treated wood compared to CCA- and ACQ-treated wood may have resulted in higher Cu leaching as Cu has been shown to make stable soluble complexes with ammonia in the pH range observed in
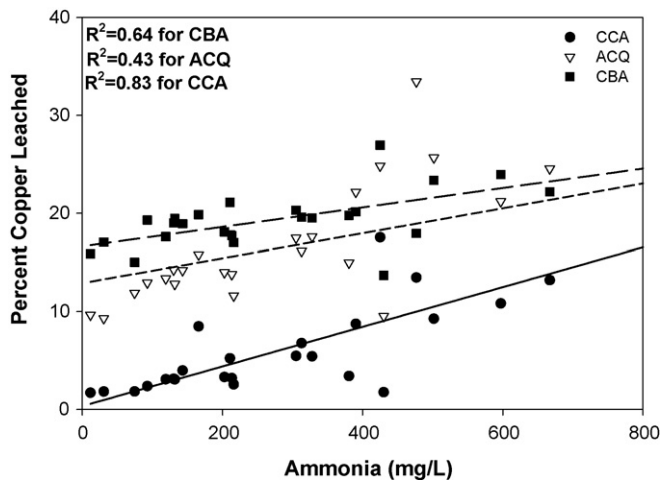
Fig. 5. Cu concentrations in the extracts as a function of landfill leachate–ammonia.

the experiment (details in next paragraph). The overall trend of Cu concentrations on leaching with different leachates follows similar pattern for the three wood types. On the graphs for copper concentration vs. sites, the three plots with CCA, ACQ and CBA show nearly identical patterns.

Efforts were made to identify what factor in a leachate influenced Cu leaching. Variation of the amount of Cu leached with ammonia concentration in the leachate has been presented (Fig. 5). As observed in the figure, there is a relationship between ammonia concentration in the landfill leachate and Cu concentration in the extract produced on leaching with MSW leachates ($R^2 = 0.83$ for CCA, 0.43 for ACQ and 0.64 for CBA) leaching. Adding ligands such as NH_3 to Cu (II) in aqueous solution leads to the stepwise formation of soluble stable Cu complexes in the pH range of 6–9 [39]. Humar et al. [40] have also observed that higher amounts of Cu leached out from chromated copper borate (CCB)-treated wood when a mixture of ammonia and oxalic acid was used as an extraction solution compared to when only oxalic acid was used as the leaching solution. This hypothesis was further tested by spiking the leachate collected from Site 20 with different ammonia concentrations. The ammonia spiked leachate was used as a leaching fluid and the ACQ sample was leached with this solution. Fig. 6 presents the Cu concentration as a function of ammonia concentration in
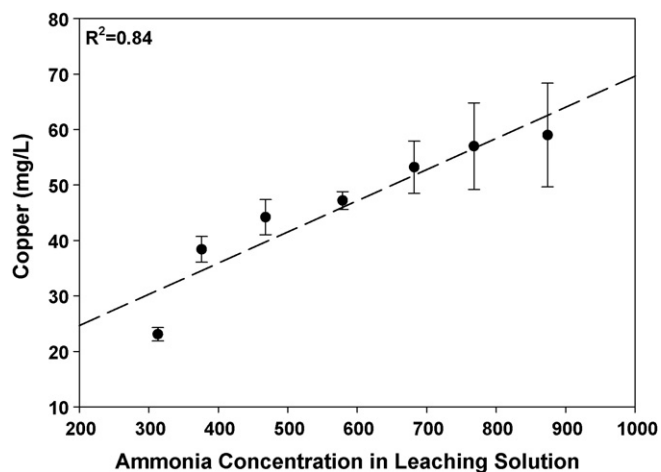
the leaching solution. As the figure shows, there was a co-relation between ammonia concentrations in the leaching solution and the amount of Cu leached. The pH of the extract was in the range of pH 6.0–7.6.

More and more landfill sites are adopting bioreactor technology for faster stabilization and other benefits associated with this. A higher ammonia concentration has been observed compared to traditional landfills since in bioreactor landfills the rate of ammonification increases with moisture addition and/or recirculating leachate [41,42]. Higher ammonia in the bioreactor landfill leachate may be a concern for leading to elevated Cu concentration in the leachate on disposal of Cu-rich waste such as ACQ, CBA, and even CCA-treated wood.

As mentioned in the previous section, the leachate quality of the Site 18 leachate was much different as compared to other leachates. The pH of the leachate was 8.10. The BOD_5, COD, and Ammonia concentrations were 2400 mg/L, 10,900 mg/L, and 1620 mg/L, respectively. The Cu leaching was highest in the leachate produced on leaching with this leachate and statistically different as compared to leaching with other leachates for CCA and CBA samples. For ACQ sample, Cu leaching with leachate from this site was the second highest among the Cu concentrations on leaching with the 26 landfill leachates.

3.5. Comparison of leaching by acidogenic and methanogenic leachates

The Cu, Cr, B, and As concentrations in the leachates produced by leaching with landfill leachates have been grouped separately for acidogenic leachates and methanogenic leachates (Site 18 was not included in this classification as mentioned previously). The average concentrations and corresponding standard deviations for different elements is presented in Table 3. The statistical 't-test' was performed on the concentrations from the two leachate types and the P-value corresponding to the 't-test' is also presented in Table 3. Comparing different elements in Table 3, the average preservative concentrations following leaching with these two groups of leachates were significantly different except for boron concentration from CBA samples. Metal concentrations observed in samples created using the Site 18 leachate (results were not included in the acidogenic and methanogenic group as compared above) were in general different compared to the other leachates; the relatively higher pH and organic content of this leachate may have influenced the leaching pattern of metals.

4. Regulatory and disposal implications

The CCA-treated wood has been reported to exceed the TC limit for As in many instances [14,15]. A similar observation was made in the present study. Limitations to TCLP have been identified and many recent studies have suggested that TCLP does not always predict the leaching concentration in MSW landfills [25–28]. In some of these studies, TCLP was found to over-predict the MSW leaching concentrations and in some other cases the reverse was observed. For example, Jang and Townsend [27] found lead from electronic devices to leach more with the TCLP compared to MSW leachates. Both, Hooper et al. [25] and Halim et al. [28] measured greater arsenic releases with actual landfill leachates compared to TCLP. Hooper et al. [25] hypothesized that negatively charged oxy-anions such as arsenic are unlikely to complex with negatively charged acetate ions in the TCLP solution, and thus other factors in the landfill leachate caused arsenic to leach more. Halim et al. [28] postulated that organics in the municipal landfill leachate may lead to a reducing condition resulting in the conversion of arsenic (V) to arsenic (III), which is more soluble. Additionally, they suggested

Fig. 6. Cu concentrations from ACQ sample as a function of spiked ammonia concentrations in the leaching solution (Leachate sample from Site #20 was spiked with several ammonia concentrations and each ammonia-spiked solution was used as a leaching fluid).

Table 3
Comparison of preservative leaching using acidogenic and methanogenic leachates for leaching.

Preservation type	Elements	Concentration in acidogenic leachate	Concentration in methanogenic leachate	P-value ($\alpha = 0.01$)
CCA	Arsenic	3.72 ± 1.10	5.00 ± 1.7	0.0004
	Chromium	0.79 ± 0.3	1.32 ± 0.6	0.00001
	Copper	2.72 ± 1.7	4.95 ± 3.5	0.0015
ACQ	Copper	20.6 ± 6.1	26.6 ± 11.7	0.011
	Boron	16.0 ± 2.5	18.4 ± 4.0	0.005
CBA	Copper	49.5 ± 9.2	54.3 ± 11.3	0.06
	Boron	31.7 ± 5.1	33.6 ± 4.6	0.104
DOT	Boron	68.8 ± 5.0	65.1 ± 5.5	0.004

that under TCLP conditions the arsenate would precipitate with calcium, but under landfill leachate conditions carbonate would preferentially precipitate with calcium, thus resulting in greater arsenic releases.

In the present study, the average As concentration produced by extracting with MSW leachate was below the TCLP concentration. Furthermore, the concentrations from different extraction tests (except WET) were in similar orders of magnitude. Dubey et al. [26] have found a trend with arsenic waste samples with pH < 7 leaching higher concentration of in TCLP than MSW leachates and for the As-waste samples with pH > 7 (of waste), higher concentration leached with MSW leachate compared to TCLP.

The As concentrations produced with MSW leachates varied from 0.3 mg/L to 8.4 mg/L, with the majority of samples having a concentration between 3 mg/L and 5 mg/L. The results of the present study shows that CCA-treated wood, if disposed of in a MSW landfill, could potentially increase the As and Cr concentrations in the leachates, a concern for leachate management. Several cases are being cited in the landfill industry where leachates had to be hauled over a long distance from the local WWTP for treatment due to high concentrations of pollutants, such as As, in the leachates. As stated earlier, most of the WWTP end up sending off their end-product "biosolids" for land application. The WWTP is generally not designed to treat heavy metals and, therefore, high concentration of heavy metals in the leachate will be transferred to the biosolids, which may limit its reuse.

The Cu-based alternatives studied were found to leach Cu greater (a degree of magnitude higher) than CCA-treated wood. The Cu concentrations released using the landfill leachate varied somewhat among the sites. If the Cu-based alternatives are disposed of in MSW landfill, the present study indicates that it may lead to elevated concentrations of Cu in the MSW landfill leachates; Cu is toxic to aquatic organisms at very low concentrations [43] and has also been listed as one of the heavy metal toxicants to methanogens [44].

High boron leaching was also observed when MSW leachates are used as the extraction fluid. The boron concentrations for all the samples exceed the groundwater cleanup target level of 630 μg/L. The elevated concentrations of boron in the landfill leachate will also pose a concern for biosolid reuse as soils with boron concentration greater than 100 mg/kg are toxic to certain plants [45].

A variation of preservative leaching from treated wood in different extractions solutions was observed. The preservative leaching was found to vary with leachate chemistry when landfill leachate was used as an extraction fluid. Trends of higher element leaching with higher concentrations of certain leachate parameters has been identified and presented in the previous sections. Although using landfill leachates instead of TCLP fluid does give an advantage of having organic and inorganic constituents (the factors which influence metal leaching) similar to what will be encountered in a real landfill; there are many factors that cannot be simulated in a laboratory leaching test that occur in a landfill (e.g., sorption, precipitation, reducing conditions). Heavy metals can be removed from solution by these mechanisms.

Other factors that might differ in laboratory tests relative to actual landfill conditions include temperature and decomposition. As previously stated, the batch leaching experiments in this study were carried out at ambient temperatures (25 °C). Landfill temperatures frequently reach as high as 55 °C as a result of biological activity, and temperature has previously been observed to impact metal leachability from treated wood. In laboratory batch leaching tests conducted at several temperatures, the fraction of As leached was nearly twice as much at 35 °C compared to leaching at 15 °C, nearly 2.5 times as much for Cr, with no change observed for Cu [46]. If wood were to significantly decompose in the landfill, this would likely impact leachability. Wood has been found, however, to decompose relatively slowly in anaerobic landfills; in one study no significant loss of mass was observed in wood samples excavated after 19 years and 29 years of disposal in a landfill, with samples as old as 46 years showing only an 18% carbon loss [47]. The impact of both of these conditions merits additional exploration.

Jambeck [48] has simulated disposal of CCA-treated wood in a MSW landfill environment. Two percent CCA-treated wood by weight was disposed with other components of MSW. The maximum As, Cr and Cu concentration observed in the leachate was 3.8 mg/L, 4.0 mg/L and 0.6 mg/L, respectively. Extrapolating (assuming linear relationship) to a scenario of 5% treated wood disposal (TCLP assumes 5% co-disposal) in a MSW landfill, the maximum concentration would be 9.5 mg/L, 10 mg/L and 1.5 mg/L for As, Cr and Cu, respectively. This linear extrapolation is not proposed to be an accurate forecast for leachate concentrations, as other factors will influence the leachability of metals. But the comparison is useful as a preliminary assessment of how the results from the batch leaching tests compare with predicted concentrations from the simulated landfill results. In the leaching tests with CCA-treated wood in this study, As, Cr and Cu concentrations measured with TCLP were 11.3 mg/L, 3.75 mg/L and 10.9 mg/L, respectively. Comparing the two set of concentrations, it was found that TCLP concentration was similar for As in two experiments. Chromium was under predicted and Cu was over predicted with TCLP. Copper is less soluble in anaerobic conditions and has been found to precipitate out in the presence of sulfides generally present in reducing landfill conditions [49,50]. Comparing the concentrations measured with landfill leachates, it was found that As and Cr concentrations measured in the leachate were lower than the predicted lysimeter concentrations. Copper concentrations in batch test with leachates were higher than the predicted lysimeter value for most of the CCA leachates. It should also be noted that although the preservative concentrations were lower in the simulated landfills compared to the concentrations observed in batch leaching tests of the present study, the concentrations in experimental lysimeter were significantly elevated for As and Cr even with 2% co-disposal of treated wood in simulated MSW landfill, a concern for leachate management and treatment.

A recent review study by Solid Waste Association of North America (SWANA) applied research foundation's disposal group concluded that "MSW landfills can provide for the safe, efficient,

and long-term management of disposed products containing RCRA heavy metals without exceeding limits that have been established to protect public health and the environment" [51]. The study has focused on RCRA heavy metals and has collected leachate data from USEPA Leach 2000 database on these elements and compared them to the corresponding TC limit. The RCRA heavy metal concentrations were below the TC limit for all the leachate concentrations reported. When the leachate concentrations were compared with the four local government pretreatment standards at four counties at different parts of the country, it was found that average concentrations of As and Hg in the landfill leachate exceeded one out of four pretreatment standards. For the treated wood products As and Cr are on RCRA TC list (Cu is not included in this list). With the new As drinking water limit of 10 μg/L, the pretreatment standard for As may also be subsequently lowered at many of the WWTP facilities. Therefore, even though disposal of treated wood products may not result in leachates exceeding TC limit, but it may result in increase in leachate treatment cost and management problem for a landfill operator and potentially could also limit the reuse of biosolid produced at the WWTP receiving the leachates.

5. Conclusions

In this study CCA-, ACQ-, CBA- and DOT-treated wood products were leached with 26 different landfill leachates and also with TCLP, SPLP and WET procedure. The research presented evaluates the potential impact of municipal solid waste (MSW) landfill leachate quality on the loss of metals from discarded treated wood during disposal. Leaching of preservative components was influenced by leachate chemistry. Copper leaching from CCA-, ACQ- and CBA-treated wood was similar in magnitude when leached with landfill leachates compared to TCLP and SPLP concentrations. It is hypothesized that during anaerobic phase of landfill Cu is less soluble and will be precipitated out of solution. Over longer period of time, if landfills are slightly aerobic or when with operating a landfill as aerobic bioreactor the concentration of Cu in leachate may become elevated. Arsenic and Cr concentrations in TCLP solutions in contact with CCA-treated wood were found to be in the same range as in MSW leachate solutions with the majority of the concentrations below the TCLP concentration. Ammonia was found as one of the major parameter influencing the leaching of Cu from treated wood when leached with MSW landfill leachates. On spiking a leachate sample with different concentration of ammonia prior to extraction of ACQ sawdust sample, a correlation of Cu concentration in the leachate with ammonia concentration in spiked extraction solution was observed. The results suggest that disposal of ACQ and CBA-treated wood in substantial quantity in MSW landfill may elevate the Cu concentration in the leachate; this could be of potential concern especially for a bioreactor MSW landfill in which relatively higher ammonia concentrations in leachate have been reported in recent literature.

Acknowledgements

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