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Metal Transport and Bioavailability in Soil Contaminated with CCA-Treated Wood Leachates

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A laboratory study was conducted to investigate metal transport and accumulation within soils contaminated with As, Cr, and Cu from CCA-treated wood leachates. New blocks of CCA-treated wood were leached using synthetic rainwater. Soil columns were constructed and filled with three different soils, including a sandy soil, an organic soil and a clay soil. The leachate was applied intermittently until 80 pore volumes were eluted through each column. Metal concentrations (Cu, Cr, and As) were measured in the leachate before passage through the columns as well as in each elutriate fraction collected. Chemical analysis was complemented with toxicity testing using Ceriodaphnia dubia, Selenastrum capricornutum, *and MetPLATE™. Following application of 80 pore volumes of leachate, the columns were dissected and the profile of the metal concentrations within each column was determined. A comparison of the arsenic, chromium and copper leaching patterns found arsenic to be the most mobile, with copper the most retained in the soil columns (As > Cr > Cu). Transport patterns of As differed in the three soil types, with observed mobility highest in the sandy soil and lowest in the clay soil. The three metals accumulated in the top layer of soil. Arsenic posed the greatest risk when soil concentrations were compared to risk-based target levels. Although metals were detected in soil elutriates, no toxicity was detected in any of the soil column elutriates using any of the three toxicity assays.*

Keywords CCA-treated wood, metals in soils, metal sorption to soils, metal transport MetPLATE, leachate toxicity, metal bioavailability

Introduction

Chromated Copper Arsenate (CCA) has been the predominant wood preservative used in the US in recent history. This chemical was used in approximately 75% of all treated wood produced in the United States and in approximately 97% of the wood treated with water-borne preservatives (AWPI, 1997). The type C formulation of CCA has been the one most predominantly used and contains the oxides of copper, chromium and arsenic in the ratio of 18.5% as CuO, 47.5% as CrO_3, and 34.0% as As_2O_5 (AWPA, 1999). Concerns have been raised over the leaching of metals (Cu, As, Cr) from CCA-treated wood and the subsequent potential impact of these leachates on soils and groundwater. Two of the metals (As and Cr) are potential human carcinogens (Bidstrup and Wagg, 1985; Chatterjee and Mukherjee, 1999; Nebert and Shertzer, 1997). Copper is a known aquatic toxicant (Flemming and Trevors, 1989).

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Soils can be contaminated from metals leached from CCA-treated wood in several manners. All three preservative elements in CCA-treated wood have been shown to leach out and accumulate in soils located under CCA-treated wood structures (Stilwell and Gorny, 1997; Townsend *et al*., 2003a). CCA-treated wood is sometimes inadvertently land applied as part of waste-derived wood mulch (Tolaymat *et al*., 2000); the smaller particle size of the wood mulch particles increases surface available for metal leaching from any CCA-treated wood present. In some locations (Florida, for example), CCA-treated wood is disposed in unlined construction and demolition (C&D) debris landfills (Townsend *et al*., 2003b). The CCA preservative elements, most notably arsenic, can become elevated in the leachate, and thus would result in contaminating underlying soil and groundwater.

Soils are widely used as receptacles for organic and inorganic contaminants such as metals. They are useful in mitigating the impact of contaminants on groundwater resources and surface waters. Metal contaminants tend to bind to soils, thus becoming non-bioavailable to the biota and to plant roots. The main mechanisms involved in metal binding to soils include adsorption, ion exchange, complexation with humic substances, and precipitation. Several studies have been conducted on metal binding to soils, namely the effect of soil texture, electrolyte concentration, pH, and the complexing ability of organic matter (Weng *et al*., 2002). Others have focused on metal speciation in soils (Balasoiu *et al*., 2001; Smith *et al*., 1999; Yong *et al*., 2001). In these studies, a sequential extraction procedure uses different reagents to obtain metal fractions, which include soluble, exchangeable, carbonate-bound, oxide/hydroxide-bound, organic matter-bound, and residual fractions, of which the first two fractions are generally considered to be biologically active or bioavailable and thus potentially toxic to the biota. These metal fractionation procedures need, however, to be complemented with toxicity testing to obtain information about the biological activity of metals in soils. Some investigators have used both chemical and toxicological approaches to assess the bioavailability of metals in solid matrices (Jang *et al*., 2002; Kong and Bitton, 2003; Schultz *et al*., 2004; De Vevey *et al*., 1993).

The objective of the research presented in this paper was to assess the behavior of metals leached from new CCA-treated wood when applied to three different soil types. CCA-treated wood leachate was simulated using a synthetic rainwater solution. The leachate was passed through columns containing the three soil types. Concentrations of the metals accumulated in the soils, as well as dissolved concentrations passing through the soils, were measured. An additional objective was to evaluate the ability of the soils to reduce the toxicity of the leachates by measuring the toxicity of the leachates passing through the soils using several toxicity assays.

Materials and Methods

Figure 1 outlines the approach used to study metal transport of As, Cu and Cr from CCA-treated wood leachates through soils and the resulting toxicity of the passing leaching. Following generation of CCA-treated wood leachate, As, Cu and Cr) concentrations and toxicity of the leachate and soil column elutriates were determined. Furthermore, metal profiles within the soil columns were determined.

SPLP Leaching of CCA-Treated Wood

The leaching solution outlined in the Synthetic Precipitation Leaching Procedure (SPLP) was used as the synthetic rainwater. The SPLP was developed by the EPA to simulate

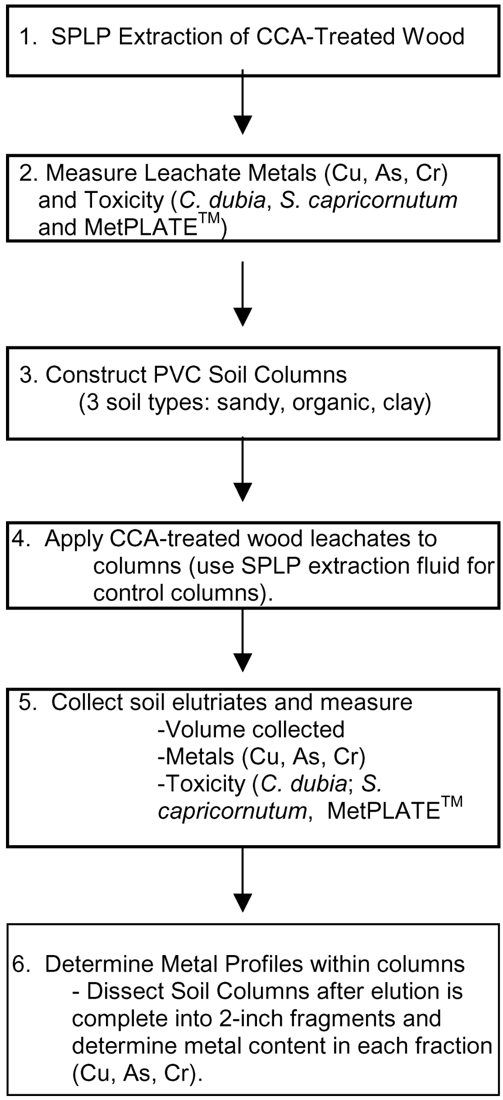

Figure 1. General approach used to study metal transport and toxicity in soil columns treated with leachates from CCA-treated wood.

waste or soil leaching that occurs during exposure to natural acidic precipitation (USEPA, 1996). CCA-treated wood was purchased from a home supply store in Gainesville, FL, in dimensional lumber form (2 × 4 × 8 in pieces). The wood was cut into 2 × 2 in blocks and extracted for 18 hrs with SPLP solution (pH = 4.2) using a large mechanical rotator. The resulting leachate was transferred to a pressure filtration apparatus and the solution was filtered through an acid-rinsed (20% HNO_3) filter (0.7 μm pore size). The solution was stored in washed and acid-rinsed 1 gal high density polyethylene (HDPE) containers at 4°C until further analysis. Small triplicate portions of the leachate were preserved with concentrated nitric acid for metal analysis. The generated leachate was subsequently applied to the soils described below.

Table 1
Some characteristics of the soils under study[a]

Characteristic	Soil type		
	Sandy soil (SS)	Clayey soil (CS)	Organic soil (OS)
PH	6.2	5.0	6.5
Eh (mV)	328.1	435.3	302.6
Moisture Content (%)	1.7	5.5	13.2
Bulk Density (kg/L)	1.5	1.4	1.2
% Organic Carbon	0.5	0.4	1.9
% Organic Matter	1.3	1.2	5.6
% Sand	93.0	53.8	87.2
% Silt	4.9	11.2	9.6
% Clay	2.1	35.0	3.2

[a]From Song *et al.*, 2005.

Soils Used

Three soil types were used. A sandy soil was chosen because it is representative of the soils prevailing in north-central Florida. Its metal retention capacity was compared to a clay soil and to an organic soil. The sandy and clay soils were collected by means of a hand auger from the top 4 feet below the surface from a site identified by soil scientists in Gainesville, FL. The organic soil was collected from the first few top inches of a creek bed in Gainesville, FL. (Table 1) shows some characteristics of the soils under study (Song *et al.*, 2005).

Soil Columns

Twelve PVC columns of 1 ½ inches internal diameter and 18 inches in length were constructed. Each column was equipped with a stainless steel mesh, which was covered with 1½ inches of gravel to aid in drainage. Four columns each were filled with 15 inches of the sandy soil (columns SS-1, SS-2, SS-3, and SS-C), clay soil (columns CS-1, CS-2, CS-3, and CS-C), and organic soil (columns OS-1, OS-2, OS-3, and OS-C). After the soil was packed inside the columns, a small glass fiber filter (0.7 μm pore size) cut to fit the internal diameter of the columns was placed on top of the columns. The filter was used to distribute the leachate more evenly on top of the soil columns. Finally, a cap with a small hole drilled in the center was placed on top of each column.

Except for the control columns (SS-C, CS-C, and OS-C), all columns were intermittently (roughly weekly) treated with SPLP extract of CCA-treated wood at 4 pore volume intervals until 40 pore volumes had been eluted. The pore volumes were obtained by adding water to the soil column and measuring the difference in mass before and after saturation. After 40 pore volumes were passed, elutriates were collected at 54, 60, 64, 76, and 80 pore volumes. The control columns were treated in a similar manner, except that the SPLP leaching solution was used in lieu of the CCA-treated wood leachate. The leachate was delivered to the columns by means of an 8-channel peristaltic pump (Manostat Carter™ model cassette pump) set at a flow rate of 0.8 mL/sec.

The volumes of the column elutriates fractions were recorded and the elutriates were filtered (0.7 μm pore size) and stored for chemical analyses and toxicity testing. The metal

(Cu, As, and Cr) profiles within the columns were determined at the end of the leaching experiments. The contents of each column were extruded onto a clean tray and extra care was used to ensure that the soil remained intact. The soil was cut into two-inch fragments, which were dried overnight at 105°C.

Metal Analysis

All glassware employed in the metal analysis was washed with Liquinox™, rinsed with 20% nitric and hydrochloric acids, and triple rinsed with deionized water. Leachate samples were first digested using USEPA Method 3010A (USEPA, 1996) and metals were analyzed according to USEPA Method 6010B using Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES; Thermo Jarrel Ash Enviro 36). Dried soil samples were digested using USEPA Method 3050B and similarly analyzed using ICP-AES.

Toxicity Testing of Soil Elutriates

Three aquatic toxicity assays were employed: the *Ceriodaphnia dubia* 48-h acute toxicity assay, the *Selenatastrum capricornutum* 96-h chronic algal growth inhibition assay, and the MetPLATE™ acute heavy metal toxicity assay. The *C. dubia a*cute test consists of exposing for 48 hrs neonates (<24 h of age) to the soil elutriates and dilutions thereof (USEPA, 1985; 1994). Lethality was assessed by lack of mobility upon examination of each organism. The green alga *Selenastrum capricornutum* (renamed *Pseudokirchneriella subcapitata*) was utilized in the standard 96-hr chronic algal assay (USEPA, 1994). The algal cells were exposed for 96 h under controlled light and temperature to five dilutions of toxic sample in triplicate as well as to negative controls. Following exposure, the total cell density was assessed by cell counting with a hematocytometer under 200× magnification. MetPLATE™ is a short-term bioassay that was used to test specifically for the presence of heavy metal toxicity in the soil elutriate samples. MetPLATE™ kits were obtained from the Department of Environmental Engineering Sciences at the University of Florida (see http://www.ees.ufl.edu/homepp/bitton/ for more information). The assay was performed in triplicate with six serial dilutions in a 96-well microplate to determine the EC_{50} (% extract that inhibits 50% of enzyme activity) of the aqueous samples. The MetPLATE test was performed according to Bitton *et al.* (1994).

Results and Discussion

Chemical and Toxicological Characteristics of the CCA-Treated Wood Leachate Used to Treat Soil Columns

Results of the ICP-AES analysis of the contaminants in the SPLP extract of CCA-treated wood are shown in Table 2. Arsenic leached the most of the three metals at a concentration of 1052 μg/L. The copper concentration in the leachate was 533 μg/L, while chromium leached less, resulting in a concentration of 307 μg/L. The leachate concentrations were generally within the range of concentrations reported for leaching of CCA-treated wood blocks of similar size (Townsend *et al.*, 2004; Stook *et al.*, 2004). The concentrations were lower than those reported by Stook *et al.* (2005), who also measured toxicity of CCA-treated wood leachates. Stook *et al.*, however, leached samples that were much smaller in particle size compared to the blocks leached here. All three toxicity tests showed that the leachates were highly toxic, with *S. capricornutum* being the most sensitive assay. EC_{50}s

Table 2
Metal content and toxicity of SPLP[a] leachate from CCA[b]-treated wood

	μg/L
Metal contaminant	
Cr	307
Cu	533
As	1052
	EC_{50} %v/v
Toxicity	
C. dubia	1.50
S. capricornutum	0.32
MetPLATE™	6.10

[a]SPLP = Synthetic Precipitation Leaching Procedure.
[b]CCA = Chromated Copper Arsenate.

of the toxicity assays expressed in percent of the leachate were 1.5, 0.3, and 6.1% for *C. dubia, S. capricornutum*, and MetPLATE™, respectively.

Transport of Leachate Metals Through Soil Columns

Figure 2 displays the metal concentration profiles in elutriates from the soil columns following application of the CCA-treated wood leachates. The maximum Cr concentration in the elutriates (based on an average of the three replicate columns) from the sandy soil columns (SS-1, SS-2, SS-3) was 121 μg/L. In comparison, the maximum Cu concentration was 45 μg/L and the maximum As concentration was 479 μg/L after 80 pore volumes of leachate were applied to the columns (Figure 2A). The As concentrations were nearly 50 times the impending As safe drinking water standard (10 μg/L; USEPA, 2002).

The results of the metal analyses of the elutriates from the organic soil columns (OS-1, OS-2, OS-3) are shown in Figure 2B. The mean metal concentrations in elutriates collected from these columns were as high as 136, 87.0, and 383 μg/L for Cr, Cu and As, respectively. As found for the sandy soil, As concentrations were greater than Cu and Cr. Results for the clay soil columns are presented in Figure 2C. The mean chromium concentrations in elutriates collected from the clay soil were as high as 94 μg/L after 80 pore volumes. The mean copper concentration in the soil elutriates was as high as 40 μg/L, thus displaying very little mobility in the clay soil. The mean arsenic concentrations peaked at 112 μg/L (USEPA, 2002). Metal concentrations in elutriates from the control columns were below detection limits.

As concentrations clearly presented the greatest elevation with respect to risk-based drinking water standards. Chromium did exceed its respective drinking water standard in some elutriates, but it should be noted that this standard was derived Cr in the hexavalent form (Cr^{6+}). No analysis of chromium speciation was conducted; however, it is expected that all of the chromium in the soil elutriates was in the form of Cr^{3+}. Thus, there would be little risk associated with chromium transport through soils.

A comparison of the leaching patterns of the three metals shows that arsenic was the most mobile while copper was the most retained in the soil columns (As > Cr > Cu). Using

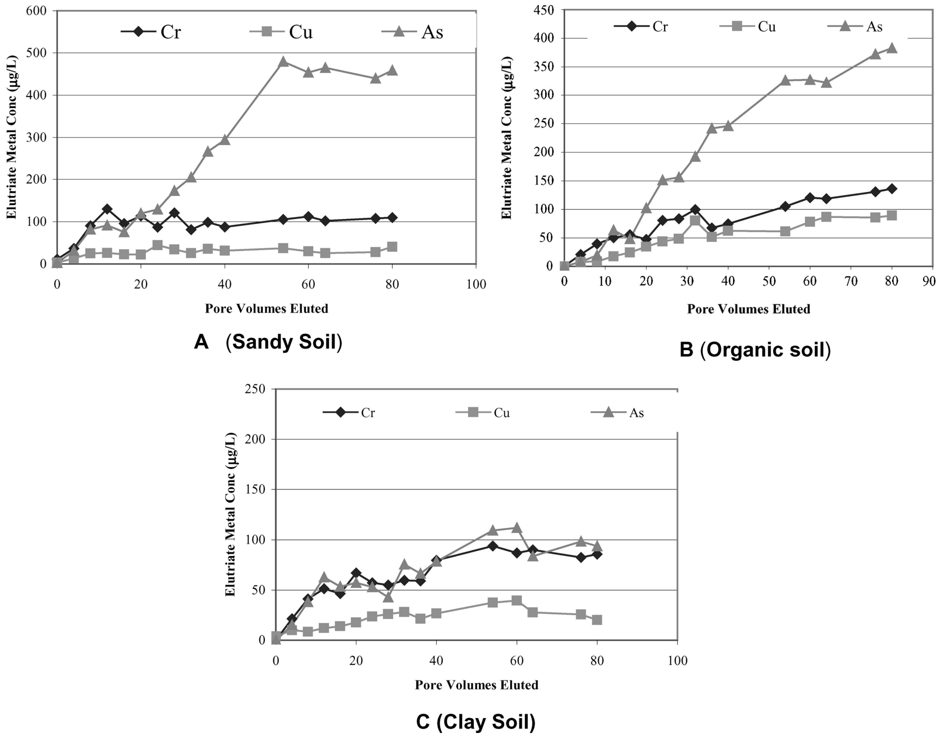

Figure 2. Transport of metals in CCA-treated wood leachates through soils. (*note*: different scales of metal concentration on the Y axis).

batch leaching of a CCA-contaminated sandy soil, Jang *et al.* (2002) showed a similar leaching pattern for As, Cr, and Cu. Batch studies of water extraction of soils from wood impregnation sites in Finland also showed that As was the most easily extracted from soils (Schultz *et al.*, 2004). Although these studies utilized batch tests, they nonetheless support our finding of higher mobility of As in soil columns.

The elevated mobility of As and Cr compared to Cu results from the fact that, in soil solutions, Cu is the only cationic species of the three metals tested. In addition to adsorption to soils, copper forms strong complexes with soil organic matter (Balasoiu *et al.*, 2001; Tyler and McBride, 1982). On the other hand, arsenic and chromium display anionic sorption behavior and require the formation of coordination bonds with iron and aluminum oxides, which form bridges between oxyanions and soil organic matter (Chirneje *et al.*, 2003; Khaodhiar *et al.*, 2000; Sarkar *et al.*, 2004). Cai *et al.* (2002) correlated soil arsenic retention with iron and aluminum content.

Figure 2 also shows differences in the transport patterns of As among the three soil types under study. Arsenic mobility was highest in the sandy soil and lowest in the clay soil. Similar results were reported by Schultz *et al.* (2004), who showed that levels of extractable As were significantly lower in clay soil relative to a sandy soil.

Metal Profiles in the Sandy Soil Columns

After elution of 80 pore volumes of CCA-treated wood leachate, the columns were dissected into two-inch sections in order to shed further insight on the mobility of metal contaminants

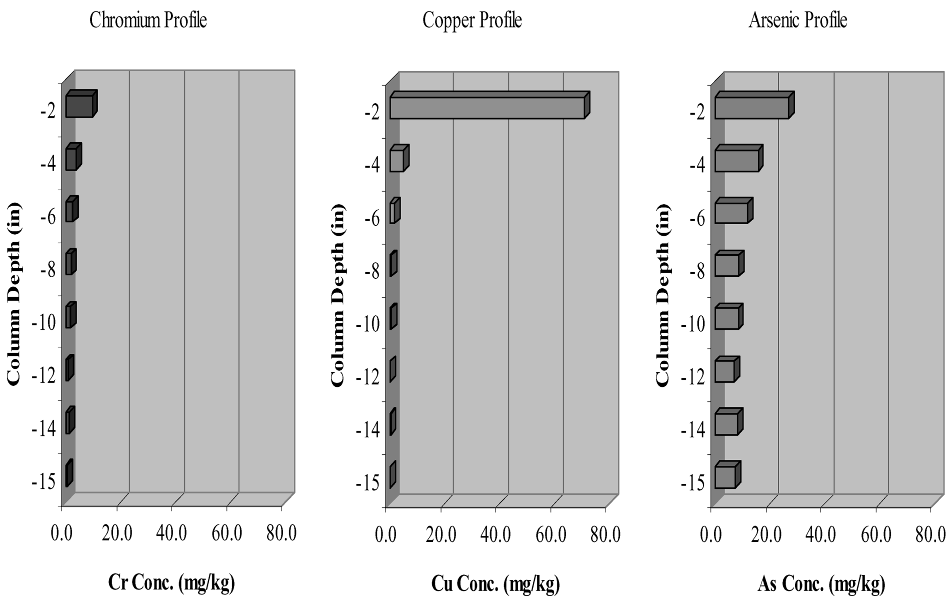

Figure 3. Metal profiles in the sandy soil columns.

within the depth of the columns. As an example, the metal profiles in the sandy soil columns are shown in Figure 3. As was found throughout the depth of the column ranging from 26.7 mg/kg top 2 inches of the column down to 7.5 mg/kg at 14–16 inches in depth. The mean Cr concentration ranged from 9.7 mg/kg in the first two inches of the column and dropped to 0.7 mg/kg at 16 inches below the surface. As expected from the soil elutriate results above, the Cu profile in the sandy soil columns indicates that this element was the most attenuated among the three metal contaminants found in CCA-treated wood. The Cu concentration in the first two inches below the surface of the sandy soil columns was 71 mg/kg, dropping to 5 mg/kg at 4 inches, 1.8 mg/kg at 6 inches, and less than 0.8 mg/kg at all lower depths.

The results again support the findings of the soil elutriate analysis that As was the most mobile of the constituents. Townsend *et al*. (2003a) evaluated soil concentration profiles collected from underneath CCA-treated structures and compared the relative amount of As, Cu and Cr retained in the soil to what would be expected based on typical CCA-treated wood leaching. They observed that ratio of As to Cr and Cu was not as great as might be expected based on leaching studies, and that greater mobility of As through the soil (relative to the other metals) might be a cause. The results of the soil column study presented here corroborate this observation.

Toxicity of Soil Elutriates

Soil elutriates were tested for toxicity using the *Ceriodaphnia dubia* acute test, *Selenastrum capricornutum* chronic test, and MetPLATE^{TM}, an acute test that is specific for metal toxicity. Despite the presence of copper, chromium and arsenic in elutriates collected from all three soil types, no toxicity was observed in any of the fractions collected from these columns. Based on elutriates metal contents, the most sensitive toxicity test used in this study, *Selenastrum capricornutum* chronic test, should have shown some toxicity for arsenic and chromium, when considering the documented EC_{50}s for these two metals (Call *et al*., 1981). This may have been due to a reduction of metal bioavailability resulting from alkalinity, hardness, suspended solids, and organic or inorganic ligands in the soil elutriates (Huang *et al*., 1999).

Implications

The results of these experiments show that the soil column elutriates created by passing CCA-treated wood leachates through soil did contain concentrations of As (and sometimes Cr) that exceeded the standards for groundwater quality (USEPA, 2002). It is important to consider the concentration of the source leachate in relation to what might be expected to be discharged to the environment from a treated wood structure or from disposed wood. The source leachate used in this study was created by leaching blocks of CCA-treated wood; the same leachate was used throughout. The As, Cr and Cu concentrations in this leachate are expected to be higher than would be expected from structures because of the size reduction in the batch tests. The concentrations from actual structures will decrease over time, whereas in this study the concentration remained the same. Nonetheless, the fact that As is the most mobile of the preservative elements and also has the lowest water quality standard suggests that the risk from arsenic leaching should be evaluated further.

Arsenic mobility in Florida soils has been previously documented (Weigand, 1999; Cai *et al*., 2002). In a study to evaluate the impact of arsenic-based herbicides used on golf courses, the Dade County Department of Environmental Resource Management (DERM) and Florida Department of Agriculture and Consumer Services (FDACS) showed groundwater arsenic levels at potentially deleterious concentrations with a maximum level of 815 μg/L (Weigand, 1999). In a two-step sequential extraction of soils from golf courses in South Florida contaminated with herbicidal arsenic, Cai *et al*. (2002) concluded that arsenic was relatively mobile in soils. As regards the impact of CCA-treated structures (utility poles, fences, and decks) on surrounding soils in Florida, Chirenje *et al*. (2003) showed that the degree of impact of treated utility poles on surrounding soils varied with the age of the poles, with newer poles (0–2 and 2–5 years old) resulting in far greater soil contamination than older poles (5–10 years old).

Khan (2004) found that As leached from CCA-treated wood did reach concentrations above the drinking water standard after passing through one ft of sandy soil after 12 months. Some initial As concentrations captured from runoff from the deck were on the same order of magnitude as the batch leachate in the study, but did decrease over time, however. The impact on groundwater from underneath a CCA-treated wood structure will be dictated by the type of soil, the amount of runoff, the depth to the water tables and the degree of dilution in the underlying groundwater (among other things). Perhaps a more serious issue with respect to groundwater contamination is from areas where concentrated leachates may be disposed continually over time. This is possibly the case with unlined C&D debris landfills that accept C&D debris. Jambeck (2004) found leachate As concentrations above those in the leachate used for this study. The mobility of As from CCA-treated wood leachates as indicated in this study thus raises questions about possible groundwater contamination at C&D debris landfills.

The profiles of the metal contaminants within the soil columns revealed that although arsenic is more mobile it, along with Cr and Cu, becomes concentrated in the surface soil layers. This raises the concern over possible human health impacts because the majority of the metals were found in the first two inches of the column, the soil layer humans, particularly children, are most likely come in contact with. Arsenic concentrations in the top two inches of the columns were 32 to 82 times greater than the Florida Department of Environmental Protection's risk-based target levels for direct human exposure in residential settings (0.8 mg/kg) (FDEP, 1999). Concerns similar to these led the industry to phase out the production of CCA-treated wood for most residential uses. The wood preservatives that have replaced CCA do not contain arsenic or chromium but do contain larger amounts of Cu. The results reported in this study found that copper displayed the poorest mobility, and thus Cu leaching from the new preservative treated wood will likely become bound up in the top layer of soil. While Cu does not pose the same magnitude of human health risk as As or Cr, it can result in gastric distress if consumed at sufficiently high levels; Florida's risk-based direct exposure soil threshold for Cu is 110 mg/kg. The impact of elevated Cu concentrations in areas with high likelihood of human exposure merits additional evaluation.

Acknowledgements

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