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## Chapter 9

# COPPER, CHROMIUM AND ARSENIC IN SOIL AND PLANTS NEAR COATED AND UNCOATED CCA WOOD

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**Abstract:** For many years, Chromated Copper Arsenate (CCA) was widely applied as a wood preservative, and though its use for most residential uses has been phased out, concerns about CCA leaching into soil from existing structures remain. In this study, we determined the effects of coating CCA wood on reducing such leaching. Ten boxes were constructed, six of which were coated with opaque film forming (FF) or penetrating finishes (PF), filled with soil, and weathered for two years. The soil was periodically sampled up to two years, and then romaine lettuce, arugula, basil and chives were grown under greenhouse conditions in these boxes. After two years, average amounts of arsenic (As) in the soil 2 cm from the CCA wood was 29 mg/kg, dry weight, 27 from wood coated with PF finishes and six in those coated with FF finishes. Soil As in all samples 6 cm from the wood were near the background value of 3.4. The average amount of As in arugula grown 2 cm from the edge of the CCA wood was 60 mg/kg, dry weight, 61 in wood coated with PF finishes and 24 in those coated with FF finishes. Similarly, in chives the amounts were 75 in CCA, 75 in PF, 12 in FF, in lettuce they were 5 in CCA, 5 in PF, in 1.4 FF and in basil they were 6 CCA, 10 PF, 3 FF. The amounts of As in plants grown in the control boxes were all <1. Compared to uncoated CCA wood, there was no reduction in As in plants grown along the edge of CCA wood coated with penetrating finishes, while the reduction in plant As ranged from 50-84% in plants grown next to the opaque finished wood. The reduction in arsenic in samples grown 6 cm from the wood compared to 2 cm from the wood ranged from 55-84%. The amounts of arsenic in the arugula and chives exceed the British limit for plant As of 1 mg/kg (fresh weight). As a result, gardeners should avoid growing certain vegetables in soils near CCA wood.

**Key words:** Arsenic, CCA wood, plant uptake, soil, coating

## **1. INTRODUCTION**

For many years, chromated copper arsenate (CCA) was the predominant formulation used in the pressure-treatment process to preserve wood from decay and insect damage. A number of investigations, however, have shown that varying amounts of CCA can be dispersed from the wood by leaching, erosion, weathering, decay and physical dislodgement (Belluck *et al.* 2003; Lebow 1996; Stilwell and Gorny 1997; Stilwell *et al.* 2003; Lebow *et al.* 2000; Stilwell and Graetz 2001; Weis and Weis 2002; Townsend *et al.* 2003; Zagury *et al.* 2003). The potential environmental problems associated with this dispersal resulted in a phase out of its use in the US for most residential applications effective January 2004 (Fed Reg. 2002). However, CCA wood produced prior to the phase out is expected to remain in service for many years (Solo-Gabriele and Townsend 1999), and its use is still permitted for many applications outside the residential setting, such as, utility poles and docks.

One major application for this wood was in situations involving soil contact, including raised-bed gardens, fence posts, and utility poles. Rahman *et al.* (2004) has shown that Cu, Cr, and As from CCA wood used to construct raised garden beds diffuse into the soil. Elevated levels of these elements in soils near CCA wood stakes and utility poles have also been reported (Zagury *et al.* 2003; Lebow *et al.* 2004). Recent reports have also shown that As levels in plants increased when grown in soils near CCA wood (Shiralipour 2004; Rahman *et al.* 2004; Cao and Ma 2004).

A promising treatment for minimizing CCA dispersal is to coat the wood with a paint, stain, sealer or varnish, thus forming a barrier between the wood and the environment. These finishes may contain water repellents to prevent water penetration or may provide a physical barrier by way of film formation. Much of the information on coatings for CCA wood focuses on the durability of the finish to withstand weathering in a given application (Williams 1999). A limited number of studies have been conducted to assess the ability of finishes to reduce metal dispersal (Kizer 1987; Reidel 1991; Cooper *et al.* 1997; Stilwell 1998; Lebow *et al.* 2002; Lebow *et al.* 2003, US EPA 2005). These studies have shown that finishes can reduce the dispersal of the preservative by 50-90%. Film-forming finishes tend to be the most effective barriers but they are not preferred in situations where they can chip and flake. A recent study by the US EPA on finishes applied to CCA wood found that, after one year of weathering, certain penetrating finishes were nearly as effective as the film-forming finishes in reducing surface available As (US EPA 2005).

In this study, we tested the use of coatings to prevent preservative dispersal from CCA wood in a soil environment, by coating boxes made

from this wood with various finishes, both film-forming (FF) and penetrating finishes (PF), filling them with soil, and weathering them for two years. During this time, the soil was sampled periodically and after two years, plant uptake of arsenic was determined by growing romaine lettuce, arugula, basil and chives in these boxes under greenhouse conditions. Preliminary results on the first phase of this study have been reported earlier (Stilwell et al. 2005).

## 2. EXPERIMENTAL

A total of 10 boxes (27x28x14 cm) were constructed, eight using 3x15 cm CCA boards, one using an alternative preservative containing copper and quaternary ammonia (ACQ), and one control using untreated pine. The bottom of each box was constructed using 1 cm thick untreated plywood, with nine drainage holes (0.5 cm dia.). The CCA containing boxes were constructed using 2.5 m x 3 cm x 15 cm pine boards, purchased at a lumber yard, nominally treated with 6.4 kg/m<sup>3</sup> of CCA preservative by Universal Forest Products. The boards originated from three sets, and though differences in the treatment level in the wood between sets from the nominal amounts (mg/kg) of 1840 (Cu), 3120 (Cr) and 2800 (As) were observed, there was no correlation between bulk levels in the wood compared to the amounts leached into the soil (Stilwell et al., 2005). The ACQ wood contained 3073±58 (Cu), <20 (Cr), and <20 (As) (mg/kg), while the control wood and the plywood contained <20 mg/kg Cu, Cr, and As. All of the boards appeared new and were stored indoors until use.

Each paint or stain was applied in two coats. As shown in Table 1, the coatings consisted of oil-based, semi-transparent stains (two brands, one with and the other without alkyd resin ingredients), water-based coatings (two brands, one with a penetrating alkyd/acrylic formulation), an acrylic solid color deck stain, and a polyurethane enamel. Two of the boxes made from CCA wood were left uncoated, as were the control box and the box made using the ACQ preserved wood.

Table 1. Description of Coatings

Coating/Box #	Coating*	Base	Color	Cover
1	None			
2	Sealant	Water	Clear	Clear
3	Stain	Oil	Grey	Semi
4	Sealant	Oil	Clear	Clear
5	Stain	Oil	Gray	Semi
6	Solid Stain	Water	White	Opaque
7	Solid Enamel	Oil	Grey	Opaque

Coating/Box #	Coating*	Base	Color	Cover
8	None			
9	ACQ wood			
10	Untreated Pine			

\* Brand and Code: Coating 2, Behr, 300 with alkyd and acrylics; 3, Behr 1-765 deck and siding stain; 4, Thompson's; 5 Olympic, 53178 deck stain with alkyds; 6, Olympic, 53097 acrylic deck stain; 7, Sapolin, 40-9309 polyurethane floor and deck enamel.

The boxes were filled with a mixture of 90% soil (sandy loam) and 10% compost (by volume) and placed out to weather. The soil properties and sampling procedures are detailed in Stilwell et al. (2005). Briefly, after 107 days of weathering, the soil was sampled using a 2.2 cm diameter soil corer, at 0-3 cm from the wood to the box bottom, taking one sample from each of the four sides, 5 cm from the left corner. The procedure was repeated after 365, 547 and 731 days of weathering, except that the samples were taken 22 (day 365), 9 (day 547) and 14 cm (day 731) from the left corner of each side. Also on day 731, a soil sample was taken at the center of each of the four sides, 6 cm from the edge and one was taken at the center of each box. Inverted plastic test tubes were inserted to fill the void caused by the soil removal after sampling. Natural rainfall supplied most of the water, but in times of drought, the soil in the boxes was watered at a rate of about 2-3 cm per week (1 cm per application).

Elemental analysis of the soil and wood composite samples was determined, following nitric acid digestion, using a Thermo Jarrell Ash ICP-AES Atom Scan 16 atomic spectrometer (Stilwell and Graetz 2001). In samples containing low arsenic (<0.1 mg/l in solution) the more sensitive technique of graphite furnace atomic absorption (GFAA) was employed using a Perkin Elmer 5100 instrument.

After two years of weathering, arugula (*Eruca sativa*, rocket), romaine lettuce (*Lactuca sativa*), sweet basil (*Ocimum basilicum*), and chives (*Allium schoenoprasum*) were grown in these boxes in a greenhouse. The seeds were germinated in 1.2x1.2x2.6 plugs in a starter tray filled with growth media. After germination and sprouting (14 days lettuce and arugula, 21 days chives and basil) the seedlings were transplanted into the box soil. The arugula seedlings were planted, equally spaced, 2 cm from the box edge, four along one side and three along an adjacent side. Two seedlings were also planted, equally spaced, 8 cm from the edge of the two sides and one seedling was placed in the center of box. The lettuce seedlings were planted similarly along the remaining two sides, three seedlings 2 cm from the edge per side, and one seedling on each side, 8 cm from the edge. The chives and basil were planted in the corners of each box and with each type on opposite sides. Water was supplied as needed, typically 1 liter per box every other day. On seven occasions fertilizer was added to the water at a rate of 30 mg/l

N/P/K. Over the growing period approximately 300 mg of P was added to the approximately 10 kg of soil.

The entire plants were harvested after 21 days of growth for the lettuce and arugula, and after 28 days for the chives and basil, by cutting them off within 1 cm of the soil line. The arugula plants harvested along each box edge and 8 cm from each box edge were composited, forming four composites and one center plant sample per box. The lettuce plants along each edge were similarly composited, along with the two lettuce plants grown 8 cm from the edges, forming three composites per box. The basil and chives harvested from the box corners were combined, forming one composite of basil and one of chives per box. All the plants were rinsed with distilled water, dried at 80° C for 10 hours in paper bags, crushed and transferred to polypropylene containers. For percent moisture determination, the lettuce and arugula plants along one edge of each box were weighed prior to compositing, and three composites of the chives and basil were weighed right after harvest. The percent moisture in the plants were, arugula,  $91.4 \pm 0.3$  (n=10), lettuce,  $92.9 \pm 0.5$  (n=10), basil,  $90.1 \pm 0.3$  (n=3), and chives  $89.8 \pm 0.4$  (n=3). The percent moisture in these plants were in close agreement to those reported by the USDA (2005), arugula 91.7%, lettuce 94.6%, basil 91.0%, and chives 90.6%.

The plant tissue was analyzed by weighing 0.2-0.4 g of dried plant material into 50 ml plastic containers, adding 5 ml of conc. nitric acid, and digesting in a hot block (Digi-Prep Ms, SCP Science, Champlain NY) at 115° C for one hour. After adding distilled deionized water to the 50 ml mark, the plant digests were analyzed for copper, chromium, and arsenic, as described above.

Statistical analysis was carried out by using the analysis of variance (ANOVA) utility in Microsoft Excel 2003.

### **3. RESULTS AND DISCUSSION**

#### **3.1 Arsenic Leached**

The average soil arsenic levels next to the wood over time for different treatments are given in Figures 1 and 2. The results from the uncoated CCA boxes (Box 1 and 8) were combined (n=8) in computing the averages for each weathering time period. All other averages were an individual box (n=4). Arsenic levels in the soil samples from the uncoated CCA boxes increased with time of weathering (Figure 1). Furthermore, the average arsenic level in soil samples taken from the uncoated boxes, after 365 days of weathering, exceeded the State of Connecticut limit of 10 mg/kg (State of

CT 1996). The results in Figure 2 show that after 731 days of weathering, As in soil from the uncoated CCA boxes was not significantly different ( $p=0.43$ ) from the As levels in soils from boxes coated with the penetrating finishes (coatings 2-5). The lowest soil As levels were from boxes coated with opaque finishes. These levels,  $6.8\pm 0.6$  (coating 6) and  $4.6\pm 1.5$  (coating 7) mg/kg As, though elevated with respect to the As in soils from the control box ( $3.0 \pm 0.2$  mg/kg) maintained a level below the 10 mg/kg State of Connecticut limit throughout the two-year period. The amounts of As in the soil next to the wood after two years of weathering,  $29\pm 7$  mg/kg, was within the range of 12-56 mg/kg As found in soil next to CCA wood in a survey of six raised garden beds by Rahman et al. (2004).

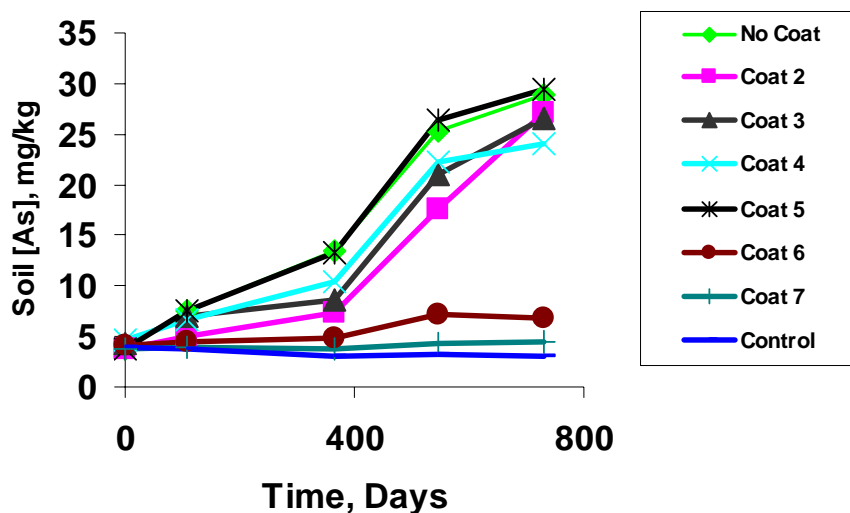


Figure 1. Comparison of soil arsenic versus time for different wood coatings (see Table 1)

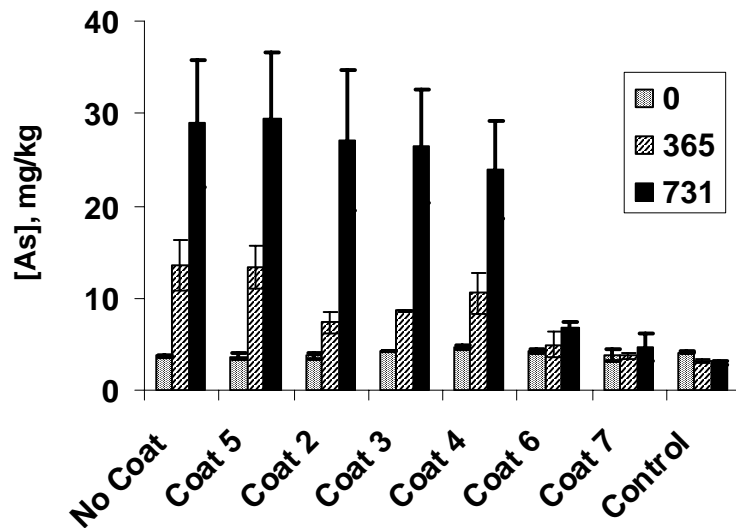


Figure 2. Average soil arsenic after 0, 365, and 731 days of weathering, ranked by coating effectiveness (107 and 547 day data omitted for clarity)

The percent reduction in soil As with different finishes and weathering time is given in Figure 3. The percent reduction was calculated by subtracting the amount of arsenic in soil from the control box from that in soil from each coated box, and dividing this by the difference between the arsenic in soils from uncoated boxes and the control boxes, i.e.  $100 * (\text{Coat Value} - \text{Control Value}) / (\text{No Coat Value} - \text{Control Value})$ . The opaque acrylic finish (#6) reduced the arsenic level by about 80% while the polyurethane based finish (#7) was around 95% effective over the entire two-year time. Opaque finishes were also found to be the most effective coating to reduce arsenic dislodged from surfaces (Kizer 1987; Stilwell 1998). The oil-based, deck and siding stain (#3), the sealant with alkyd and acrylics (#2) and the oil-based sealant (#4) were less effective and reduced the arsenic level by only 30-60%. In addition, the barrier appears to be breaking down after 1.5 years of weathering for these finishes (#2-4) since the percent reduction in soil arsenic was noticeably diminished compared to the one-year values (figure 3). The oil-based stain (#5) which had no apparent effect on arsenic leaching in this soil environment was found earlier to reduce arsenic dislodged from surfaces (Stilwell 1998).



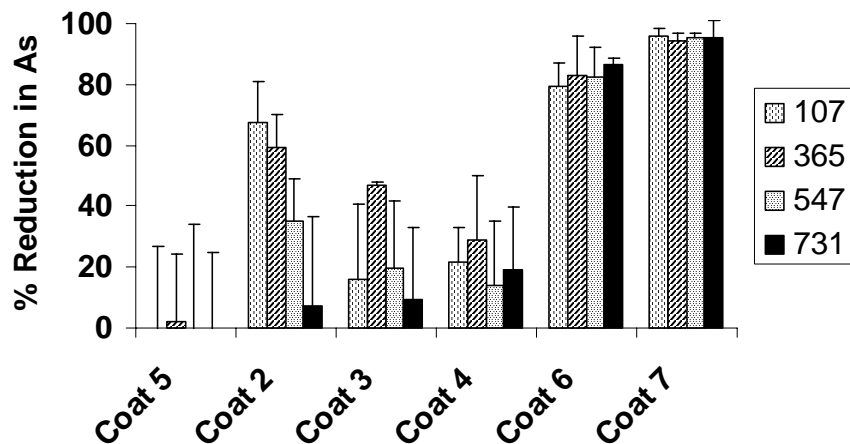


Figure 3. Percent reduction in soil arsenic levels with coating after 107, 365, 547, and 731 days of weathering

Shown in Figure 4 is the relationship between soil As and distance from the box edge. The concentrations of As in soil 6 cm away from the edge of the boxes is significantly less than the levels in soil next to wood. The average arsenic in all samples 6 cm from the box edge were at most 0.7 mg/kg higher than the average of  $3.1 \pm 0.2$  mg/kg in the control soils samples taken at this time. At the box center (13 cm), As levels in all treatments were within 0.3 mg/kg of the control except for Box 4 sample which was 0.6 mg/kg higher. Thus, beyond 6 cm from the edge of the wood the soil arsenic levels is reduced to well within 1 mg/kg of background levels of 3-4 mg/kg in this type of soil. This immobilization of As by the soil is likely due to the presence of Fe and clay which are known to fix As (Lebow 1996). Lateral decreases in soil As, reaching background levels within 15-130 cm from the CCA wood, has also been observed next to raised beds (Rahman et al. 2004), fences (Shiralipour, 2004), and traffic sound barriers (Stilwell and Graetz, 2001).

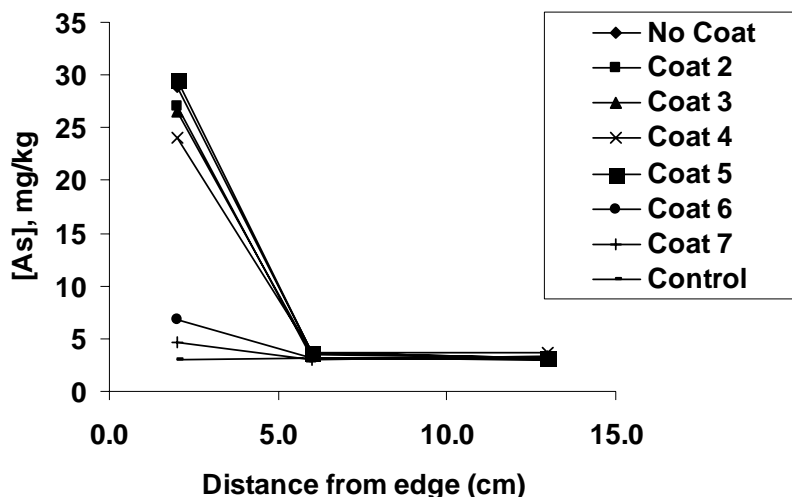


Figure 4. Soil arsenic with distance from box edge after 731 days of weathering

### 3.2 Copper and Chromium Leached

The average copper levels in soils from various treatments are given in Figure 5. The copper in the soil samples next to uncoated CCA wood increased modestly, from 23 mg/kg initially, to 36 mg/kg after two years of weathering, a 55% increase over the two-year period. The increase in copper in soils next to wood coated with the penetrating finishes increased to a lesser extent, from 16% (coat 4) to 35% (coat 2), while the copper in soils next to wood coated with the film-forming finishes (coat 6&7) increased the least, 10-15%, over the two-year period. In contrast, the copper in soil samples next to the ACQ treated wood increased from 24±0.5 mg/kg, to 80±25 after two years (Figure 5), a greater than three-fold increase. The greater copper content in the soils next to the ACQ wood is due in part to the fact that the ACQ wood contained about 2.3 times more copper than the CCA wood, 3073±58 mg/kg in the ACQ wood compared to 1360±370 mg/kg in the CCA wood. However, the copper content in the soil samples next to the ACQ wood increased over the weathering time to 56 mg/kg, about a factor of four, over the average 13 mg/kg increase in soil copper next to CCA wood. This increase is higher than the 2.3 expected from the difference in concentrations between the two materials, suggesting a faster leaching rate in the ACQ wood than in the CCA wood, consistent with the findings of Stook et al. 2005. The copper in soil samples away from the

wood (6 and 13 cm) were all within 2 mg/kg of the background value of 23 mg/kg.

The average chromium levels in the soil for different treatments exhibited only limited increases compared to the pre-weathering values. For example, the increase in the soil Cr in the uncoated wood treatment increased from the initial value of 10±2 mg/kg, to 12±2 (day=107), 13±1 (day 365), and 13±2 (day 547) and 13.6 (day 731). Due to these small increases in soil Cr, coupled with the variation in baseline Cr (range 9-11 mg/kg) we could not evaluate the effects of coatings on Cr leaching.

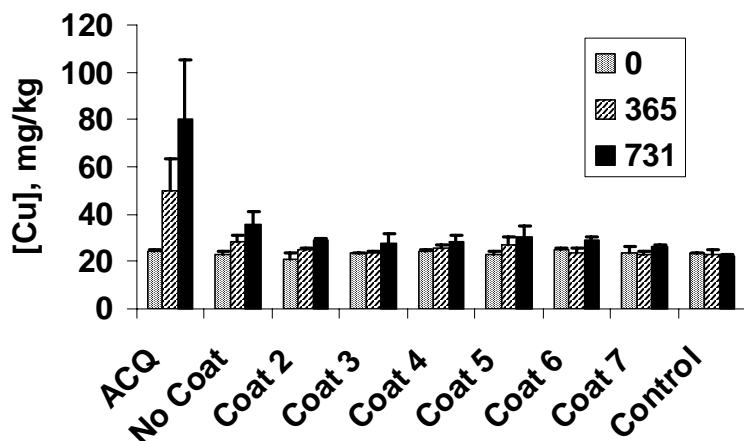


Figure 5. Average soil copper after 0, 365, and 731 days of weathering (107 and 547 day data omitted for clarity)

### 3.3 Plant Uptake

The amounts of As in the plants and soil at the box edges, with finish type, are shown in Table 2. Average amount of As in the soil next to the box edges was 29±7 mg/kg next to uncoated CCA wood, 27±6 next to wood with PF coatings and only 6±2 next to wood with FF finishes. Average amounts of As (mg/kg, dry weight) in arugula grown 2 cm from the CCA wood was 60±0.1 (4 composites), 61±13 (8 composites) from wood coated with PF finishes and 24±7 (4 composites) in those coated with FF finishes. Similarly, the amounts in chives were, 75 (CCA), 75 (PF), 12 (FF); lettuce 5 (CCA), 5 (PF), 1.4 (FF); basil 6 (CCA), 10 (PF), 3 (FF). The amounts of As in plants grown in the control boxes were all <1 mg/kg. Clearly, there was no reduction in plant As when plants were grown next to the non-opaque

finished wood, while the reduction in plant As ranged from 50-84% in plants grown next to the opaque finished wood. The amounts of arsenic in the arugula and chives grown in the CCA boxes exceeded the British limit for plant As (Thornton 1994) of 1 mg/kg on a fresh weight basis (10-14 mg/kg dry weight basis).

Table 2. Arsenic (mg/kg, dry weight basis) in soil and plants next to CCA and control wood boxes coated with penetrating (PF) or film forming (FF) finishes.

Finish	Soil	Arugula	Chives	Basil	Lettuce
None	29 ± 7	60 ± 0.1	75 ± 19	6 ± 2	4.9 ± 0.6
PF	27 ± 6	61 ± 13	75 ± 24	10 ± 3	4.8 ± 0.5
FF	5.7 ± 1.6	24 ± 7	12 ± 3	3 ± 0.3	1.4 ± .25
Control	3.0 ± 0.2	0.5 ± 0.2	<0.2	0.9 ± 0.7	0.2 ± 0.01

The amounts of As in the soil and in plants grown with distance from the edge of the box and type of finish are shown in Figure 6. Although the plant As followed the trends in soil As, and the amounts of As in plants grown 6 cm from the box edge compared to 2 cm from the edge were lowered by 55 to 84%, these amounts were well above the background levels in plants grown in the control soil. Furthermore, the As levels in arugula plants grown in the box center (13 cm from the edges) did not decrease significantly from the levels in plants grown 6 cm from the edge. Also, the As in the arugula plants grown 13 cm from the edge in the CCA boxes, ranging from 7 to 18 mg/kg, was significantly above the 0.5 mg/kg As levels in arugula plants grown in the control boxes, even though the soil As in the center of the box was at or near background. This increase in uptake of As in the plants probably results from root growth into areas of As contamination (Miliss et al., 2004).

Evidence suggesting that the As originating from the CCA wood was generally more available to plants is shown by a comparison of the uptake factors, given in Figure 7. The uptake factor is the dry-weight concentration of plant As divided by the soil As. The uptake factor in arugula ( $2.4 \pm 0.7$ ) and chives ( $2.6 \pm 0.7$ ) is greater than one, showing that these plants actually concentrate the As from the CCA soil, but not in the control soil, where the uptake factor is much less ( $0.14 \pm .04$  arugula;  $<0.06$ , chive). In lettuce this effect is less pronounced ( $0.21 \pm 0.08$ , CCA soils,  $<0.06$ , control soils), and in basil there is no difference ( $0.4 \pm 0.1$ , CCA;  $0.3 \pm 0.2$  control). Increased plant availability of As in CCA soil was also noted by Cao and Ma (2004). They determined that the percentage of water soluble As in the soil, the fraction available for plant uptake, was much higher in CCA contaminated soils (3-14%) than in uncontaminated soils ( $<1\%$ ). The continuous leaching of a fresh supply of As from the wood may also account for the increased phytoavailability. Jacobs et al. (1970), found that the extraction of As in

NH<sub>4</sub>Cl, which is related to the plant As, decreased substantially over a six month aging period compared to As freshly spiked into soils.

The significant accumulation of As in plants reported here is consistent with recent reports showing increased As in plants when grown in soils near CCA wood (Cao and Ma 2004; Rahman 2004; Shiralipour 2004). Cao and Ma (2004) determined the As levels in carrots and lettuce grown in pots containing CCA contaminated soil (27 and 43 mg/kg As). The amounts of As in the lettuce, which ranged from 4-32 mg/kg dry weight, and in carrots which ranged from 9-44 mg/kg, increased by a factor of 2-10 with the addition of phosphorus, and decreased by 80% or more with the addition of biosolid amendments. They concluded that growing vegetables in soils near CCA-treated wood may pose a risk of As exposure. Rahman et al. (2004) conducted a similar study using CCA contaminated soil (40-50 mg/kg As) from raised beds that were at least 10 years old. The As content in carrots, spinach, buckwheat and beans grown in pots containing the CCA soil ranged from an average of 0.32 mg/kg dry weight (bean pods) to 3 mg/kg in unpeeled carrots. In plants grown in control soil, taken 1.5 m from the beds, the As was <0.1 mg/kg except in unpeeled carrots where it was 0.2-0.3. Shiralipour (2004) grew lettuce and turnip in pots containing soils taken 0 to 135 cm from a fence constructed using CCA wood. The As decreased from 31 mg/kg in soils taken directly under the fence to 1.3 mg/kg in soils 135 cm from the fence. The As in the plants grown in soil taken under the fence ranged from 3 mg/kg dry weight in carrot leaf to 6 mg/kg in lettuce leaf and turnip root. There was around a 50-70% reduction in plant As in plants grown in soils taken 15 cm from the fence and when grown in soils 30-45 cm from the fence the plant As was indistinguishable from background levels. The large variability in the plant As in these studies could be partly due to differences in plant species and soil properties.

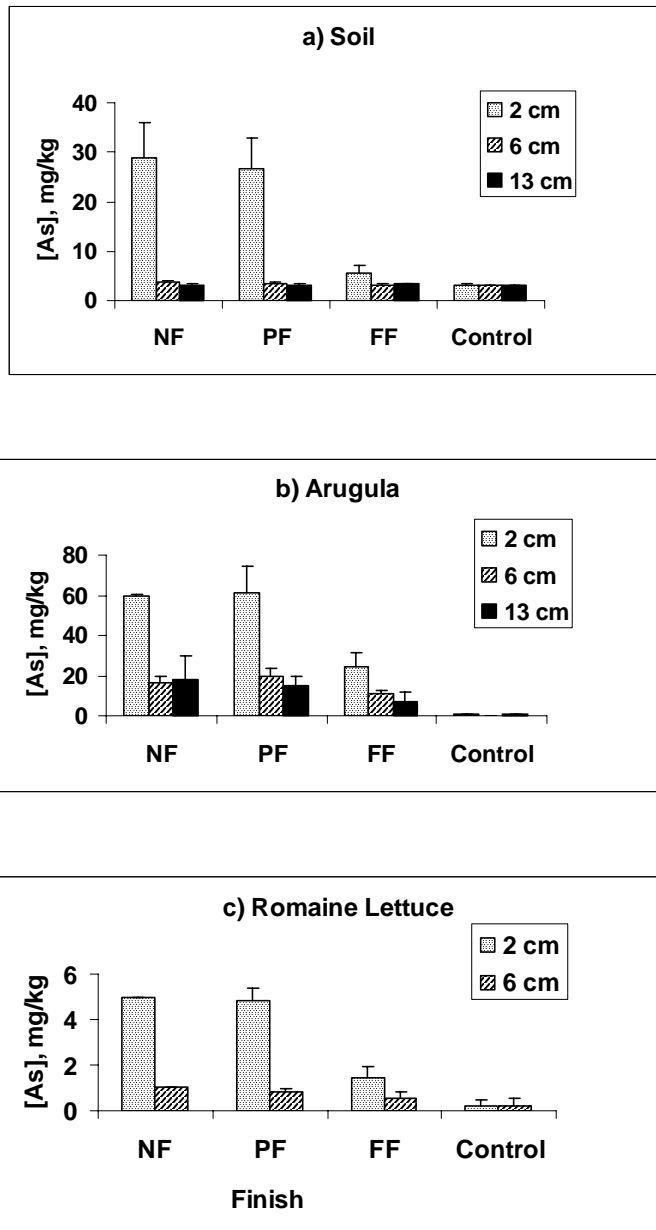


Figure 6. Soil and plant As with distance from edge of box and type of finish. a) Soil, b) Arugula, c) Romaine lettuce (NF= No Finish, PF= Penetrating Finish, FF= Film Forming finish).

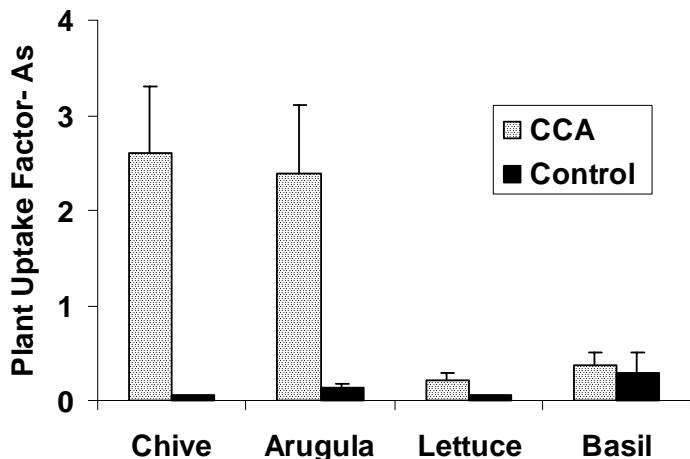


Figure 7. Plant uptake factor for As ( $[As]_{plant}/[As]_{soil}$ ) in plants grown in soil 2 cm from the wood.

Differences in uptake of As by plant species was demonstrated by Thornton (1994). Thornton determined the As content in vegetables grown in garden soils contaminated with As from mining activity in south-west England. The As uptake was highest in lettuce and lowest in beans. Plant uptake was found to increase with increasing phosphorus in the soil and decreased with increasing iron content, presumably due to competitive sorption reactions between phosphorus and arsenic in the soil and with precipitation reactions with iron to form insoluble iron arsenates. Other studies on plant uptake of arsenic under laboratory conditions (Burlo et al., 1999; Carbonell-Barrachina et al., 1999; Cox et al., 1996; Onken and Hossner, 1995) have confirmed that arsenic levels in plant tissue are dependent on the type of plant, the part of the plant (root vs. shoot), the concentration and form of arsenic in the soil and in the soil solution, and the amounts of phosphorus and iron in the soil.

The copper and chromium contents were also determined in the plants. The Cr content in all of the plant tissue samples were below the detection limit of 3 mg/kg. The Cu content in plants grown in soils next to the CCA wood were not any different than the amounts in plants grown in the control box. There was, however, a slight increase in the Cu content of arugula plants grown next to the ACQ wood. In these samples the Cu content in plants grown 2 cm and 6 cm from the ACQ was  $16 \pm 2$  (n=2) and  $14.2 \pm 1.3$  (n=2) mg/kg, respectively, compared  $10.2 \pm 1.0$  (n=5) in the plants grown in the control soil. The average Cu content (mg/kg) in all of plant tissue

samples (excluding the arugula grown in the ACQ box) was  $5.8 \pm 1.1$  (n=30), lettuce;  $10.4 \pm 1.2$  (n=45), arugula;  $6.2 \pm 1.2$  (n=10) chives, and  $11.9 \pm 1.4$  (n=10) basil.

#### 4. CONCLUSIONS

Over the two-year weathering period the As levels in soils within 2 cm of the uncoated CCA wood increased from  $3.7 \pm 0.1$  to  $29 \pm 7$  mg/kg. Moreover, within one year of weathering, the arsenic next to uncoated CCA wood increased to levels that not only exceeded the State of Connecticut limit of 10 mg/kg, but which were also on the upper bounds of As limits (2-26 mg/kg) set by other local, state and federal government agencies (Belluck et al. 2003). This contamination, however, appears to be localized to soil within a few cm of the CCA wood. Soil samples, taken 6 and 13 cm from the box edge after two years of weathering, were at, or near background levels for As.

Only minor increases in the copper and chromium content occurred in the soil next to CCA wood over this two-year period. The relatively minor increases in Cu, and Cr, reflects one, the relatively low amount of Cu in the wood, and two, the lower leaching rate of Cr (Lebow 1996; Stilwell and Gorny 1997). All of the copper levels in the soil samples from all treatments were much less than the State of CT limit of 2500 mg/kg (State of CT 1996). In no case did the Cr level approach the State of CT limit of 100 mg/kg (hexavalent Cr) or 3900 for trivalent Cr (State of CT 1996).

Opaque coatings formulated using acrylics or polyurethane when applied to CCA wood reduced the migration of arsenic from the wood into the surrounding soil by 80% to 95%, which kept the As levels in the soil below the regulatory limit over the entire two-year weathering period. Other coatings, either oil- or water-based, but with clear or semi-transparent coverage, while initially reducing the arsenic migration up to 60%, did not appear to exhibit any protective properties after two years of weathering. Clearly, the film-forming opaque finishes are effective in reducing leaching and dislodgeable arsenic from CCA treated wood. The penetrating semi-transparent and transparent finishes, though useful in above ground situations, proved to be very limited when used in contact with soil.

The plant uptake of As followed the order Chives > Arugula > Basil > Lettuce. Compared to plants grown next to uncoated CCA wood, there was no reduction in plant As when grown along the edge of CCA wood coated with penetrating finishes, while in plants grown next to opaque finished wood the reduction in plant As ranged from 50-84%. The As reduction in plants grown 6 cm from the wood compared to 2 cm from the wood ranged



from 55-84%. The amounts of arsenic in the arugula and chives were significant and exceeded the British limit for As in edible plants of 1 mg/kg, fresh weight basis (10-14 mg/kg, dry weight basis). The As in the basil was near the limit and the lettuce plants were all below the limit.

## REFERENCES

- Belluck, D.A., Benjamin, S.L., Sampson, J., and Johnson B. 2003. Widespread arsenic contamination of soils in residential areas and public spaces: an emerging regulatory or medical crisis? *International J. Toxicol.* 22, 109-128.
- Burlo, F., Guijarro, I., Carbonell-Barrachina, A.A., Valero, D., and Martinez-Sanchez, F. 1999. Arsenic species: effects on and accumulation by tomato plants. *J. Agric. Food Chem.* 47, 1247-1253.
- Cao, X. and Ma, L.Q. 2004. Effects of compost and phosphate on plant arsenic accumulation from soils near pressure treated wood. *Environ. Poll.* 132, 435-442.
- Carbonell-Barrachina, A.A., Burlo, F., Valero, D., Lopez, E., Martinez-Romero, D., and Martinez-Sanchez, F. 1999. Arsenic toxicity and accumulation in turnip as affected by arsenic chemical speciation. *J. Agric. Food Chem.* 47, 2288-2294.
- Cooper, P.A, Ung, Y.T., and MacVicar, R. 1997. Effect of water repellents on leaching of CCA from treated fence and deck units, an update. IRG/WP 97-50086. International Research Group: Stockholm, Sweden.
- Cox, M.S., Bell, P.F., and Kovar, J.L. 1996. Differential tolerance of canola to arsenic when grown hydroponically or in soil. *J. Plant Nutrit.* 19, 1599-1610.
- Federal Register. 2002. Notice of receipt of requests to cancel certain CCA wood preservative products and amend to terminate certain uses of CCA products. *Fed Regis.* 67, 8244-8246.
- Jacobs, L.W., Syers, J.K., and Keeney, D.R. 1970. Arsenic sorption by soils. *Soil Sci. Soc. Amer. Proc.* 34, 750-754.
- Kizer, K. 1987. Report to the legislature. Evaluation of hazards posed by the use of wood preservatives on playground equipment. California Office of Environmental Health Hazard Assessment, Department of Health Services, Health and Welfare Agency (1987).
- Lebow, S.T. 1996. Leaching of wood preservative components and their mobility in the environment. Summary of pertinent literature. Gen. Tech. Report FPL-RP-93. US Department of Agriculture, Forest Service, Forest Products Laboratory, Madison, WI.
- Lebow, S.T., Brooks, K.M., Simonsen, J. 2002. Environmental impact of treated wood in service. Symposium proceedings on enhancing the durability of lumber and engineered wood products, February 11-13, 2002, Kissimmee, FL, pp 205-216, Forest Products Laboratory, Madison, WI.
- Lebow, S.T, Williams, S.R., Lebow, P. 2003. Effect of simulated rainfall and weathering on release of preservative elements from CCA treated wood. *Environ. Sci. Technol.* 37, 4077-4082.
- Lebow, S., Foster, D., and Evans, J. 2004. Long-term soil accumulation of chromium, copper, and arsenic adjacent to preservative-treated wood. *Bull. Environmental Contamination and Toxicology* 72, 225-232.
- Millis, P.R., Ramsey, M.H., John, E.A. 2004. Heterogeneity of cadmium concentration in soil as a source of uncertainty in plant uptake and its implications for human health risk assessment. *Sci. Total Environ.* 326, 49-53.

- Onken, B.M. and Hossner, L.R. 1995. Plant uptake and determination of arsenic species in soil solution under flooded conditions. *J. Environ. Qual.* 24, 373-381
- Rahman, F.A., Allan, D.L., Rosen, C.J. and Sadowsky, M.J., 2004. Arsenic availability from chromated copper arsenate (CCA)-treated wood. *J. Environ. Qual.* 33, 173-180.
- Reidel, D., Galameau, D., Harrison, J., Gregoire, D.C., and Bertrand, N., 1991. Residues of arsenic, chromium, and copper on and near playground structures built of wood pressure-treated with CCA type preservatives. Health and Welfare Canada (unpublished).
- Shiralipour, A. 2004. Arsenic uptake released from CCA treated lumber by Florida vegetable crops, Report HW155-04. Florida Center for Solid and Hazardous Waste Management, Gainesville, FL.
- Solo-Gabriele, H. and Townsend, T. 1999. Disposal practices and management alternatives for CCA-treated wood waste. *Waste Manage Res.* 17, 378-389.
- State of Connecticut 1996. Remediation standard regulations. RCSA 22a-133k.
- Stilwell, D.E. and Gorny, K.D. 1997. Contamination of soil with copper, chromium, and arsenic under decks built from pressure treated wood. *Bull. Environ. Contam. Toxicol.* 58, 22-29.
- Stilwell, D.E. 1998. Arsenic from CCA-Treated Wood Can Be Reduced By Coating. *Front. Plant Sci.* 51(1), 6-8.
- Stilwell, D.E. and Graetz T.J. 2001. Copper, chromium and arsenic levels in soil near traffic sound barriers built using CCA pressure-treated wood. *Bull. Environ. Contam. Toxicol.* 67, 303-308.
- Stilwell, D., Toner, M., and Sawhney, B. 2003. Dislodgeable copper, chromium, and arsenic from CCA-treated wood surfaces. *Sci. Total Environ.* 312, 123-131.
- Stilwell, D.E., Musante, C.L. and Sawhney, B.L. 2005. Effect of coatings on CCA leaching from wood in a soil environment. In: *Environmental Impacts of Preservative Treated Wood*. Univ. of Florida, Gainesville FL (in press).
- Stook, K., Tolaymat, T., Ward, M., Townsend, T., Solo-Gabriele, H., Bitton, G. 2005. Relative Leaching and Aquatic Toxicity of Pressure-Treated Wood Products Using Batch Leaching Tests. *Environ. Sci. Technol.* 39, 155-163.
- Thornton, I. 1994. Sources and pathways of arsenic in South-West England: Health implication. In: *Arsenic Exposure and Health*, chapter 6, pp 61-70. W. Chappell (Ed.), Science and Technology Letters, Northwood, England.
- Townsend, T., Solo-Gabriele, H., Tolaymat, T., Stook, K. and Hosein, N. 2003. Chromium, copper, and arsenic concentrations in soil underneath CCA-treated wood structures. *J. Soil and Sediment Contam.* 12, 779-798.
- USDA (United States Department of Agriculture). 2005. Food and Nutrition Information Center. (<http://www.nal.usda.gov/fnic/etext/000020.html>)
- US EPA (United States Environmental Protection Agency). 2005. Evaluation of the effectiveness of coatings in reducing dislodgeable arsenic, chromium, and copper from CCA treated wood. Interim data report. EPA Report: EPA/600/R-05/050.
- Weis, J.S. and Weis, P. 2002. Contamination of saltmarsh sediments and biota by CCA treated wood walkways. *Marine Pollution Bull.* 44, 504-510 (2202).
- Williams, R.S. 1999. Finishing of Wood. *Wood handbook-Wood as an engineering material*. Gen Tech Rep. FPL-GTR-113, Forest Products Laboratory, Madison WI. Ch 15
- Zagury, G.J., Samson, R. and Deschenes, L. 2003. Occurrence of metals in soil and ground water near chromated copper arsenate-treated utility poles. *J. Environ. Qual.* 32, 507-514.