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Evaluating landfill disposal of chromated copper arsenate (CCA) treated wood and potential effects on groundwater: Evidence from Florida

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Abstract

Chromated copper arsenate (CCA) treated wood has been used for more than 50 years. Recent attention has been focused on appropriate disposal of CCA-treated wood when its service life ends. Groups in the US and Europe concerned with the possibility of arsenic migration to groundwater from disposed CCA-treated wood have proposed that consumers be required to dispose of the wood as a hazardous waste, in the most protective of landfills. We examined available data for evidence of arsenic migration from unlined construction and demolition (C&D) debris landfills in Florida, where CCA-treated wood is disposed. Florida was chosen because soil, groundwater, landfill design, weather, and levels of CCA-treated wood use make the state a uniquely sensitive indicator for observing arsenic migration from CCA-treated wood disposal sites, should it occur. We developed and quality-checked a CCA-treated wood disposal model to estimate the amount of wood and associated arsenic disposed. By 2000, an estimated 13 million kg of arsenic in CCA-treated wood was disposed in Florida; however, groundwater monitoring data do not indicate that arsenic is migrating from unlined C&D landfills. Our results provide evidence that highly stringent regulation of CCA-treated wood disposal, such as treatment as a hazardous waste, is unnecessary.

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1. Introduction

Lumber products are treated with chromated copper arsenate (CCA) to enhance their durability by preventing damage due to fungi, termites, and marine boring organisms. CCA-treated wood was developed in 1933, and has been used in industrial applications beginning before 1940 and in residential applications beginning around 1974 (DeVenzio, 1998). The CCA used to pressure treat wood is a water-based mixture containing 0.6–6.0% (by weight) of chromic acid, copper oxide, and arsenic acid (USDA, 1980). During the treatment process, CCA, at pH 1.6–2.5, is infused into wood at elevated pressure (AWPA, 2002). The resulting treated wood contains chromium(III) and arsenic(V) compounds, postulated to be chromium (III) arsenate and chromium(III) hydroxide (Bull, 2001) or chromium dimer-arsenic clusters that are stable over long periods of time (Nico et al., 2004). Researchers and regulators have been evaluating the potential for these materials to leach from CCA-treated wood in

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landfills, and for leachate to migrate into groundwater at levels of potential environmental concern (e.g., Tolaymat et al., 2000; Weber et al., 2002; Townsend et al., 2004; Jambeck et al., 2006).

Although researchers have shown that crushed particles of spent CCA-treated wood release chromium, copper, and arsenic using the toxicity characteristic leaching procedure (TCLP) (e.g., Townsend et al., 2005), spent CCA-treated wood discarded by end-users is exempt from federal regulation as a hazardous waste. This exemption has been challenged in a petition to the US EPA (BP/NCAMP, 2002). The European Commission recommended that CCA-treated wood be subject to separate collection as a household hazardous waste and disposed via incineration (Genedbien et al., 2002). The potential leaching and migration of arsenic from disposed CCA-treated wood has acquired greater significance in the US, partly due to the recent decrease from 50 μ g l⁻¹ to 10 μ g l⁻¹ in the US primary drinking water standard for arsenic (Federal Register, 2001).

CCA-treated wood disposal issues have been studied in Florida more extensively than in any other US state, and perhaps any other place in the world. A unique combination of factors in Florida make the state a sensitive indicator of the potential for arsenic in CCA-treated wood to migrate from landfills to groundwater. These factors include: (1) Due to the state's long coastline and temperate weather, Florida contains many exposed wood structures, for which CCA-treated wood was often used. Furthermore, homeowners in the Southeastern US are substantially more likely to use treated wood than those in the other regions of the United States (Vlosky and Shupe, 2002). (2) The natural background concentration of arsenic in Florida soil and groundwater is lower than in many other parts of the US, so incremental increases in arsenic levels are easily detectable (Ryker, 2001; Chen et al., 2002). (3) Florida allows construction and demolition (C&D) debris to be disposed in unlined landfills that contain no leachate collection systems. Wood comprises 24% of the mass of C&D waste in the United States (Tolaymat et al., 2000), and up to 60% of the wood mass in loads delivered to Florida disposal facilities may be CCA-treated wood (Solo-Gabriele et al., 2000). (4) Florida experiences more precipitation than most US states, leading to a greater potential for leachate production in landfills (NOAA, 2002). (5) Soils throughout many areas in Florida are sandy, lacking aluminum- and iron-oxide containing clays that are particularly effective at binding and immobilizing arsenic (Chen et al., 2002). (6) The water table is shallow in many areas of Florida (Fernald and Purdum, 1998), so the chemical constituents of any leachate that escapes from a landfill has little opportunity to be attenuated in soil through sorption or precipitation before reaching groundwater.

The coincidence of the preceding six factors in one geographic region makes Florida an area where effects on groundwater due to arsenic from CCA-treated wood wastes in landfills would most likely be observable. The objective of this analysis is to estimate the amount of CCA-treated wood being disposed in landfills in Florida and assess the impact, if any, on groundwater in the state.

Solo-Gabriele and Townsend (1999) modeled the amount of CCA-treated wood that will be disposed in Florida in coming years. Their estimates indicated that only a small fraction of CCA-treated wood sold in the state had been disposed to date, and as a result, the volume of treated wood discarded will increase dramatically within the next few years. Weber et al. (2002) have simulated the potential groundwater impacts expected due to the postulated future surge in CCA-treated wood disposal. They concluded that arsenic, which was present in leachate from field-scale C&D disposal test cells at concentrations greater than the drinking water standard, originated from CCAtreated wood. However, they did not report the amount of CCA-treated wood in their test cells. Jambeck et al. (2006) reported that the mass of arsenic leached was greatest in the first few months after disposal in their study of pilot-scale lysimeters in Florida, which contained a mixture of new (50%) and weathered (50%) CCA-treated wood cut into small blocks, but which contained no other C&D wastes. We found no published, full-scale field studies describing arsenic migration from CCA-treated wood to groundwater in unlined landfills.

In this report, we present a more refined model for CCA-treated wood disposal in Florida, using disposal data specific to the Southeastern US. These data, which characterize CCA-treated wood disposal in Florida, call into question the assumptions and results of the previously published disposal model (Solo-Gabriele and Townsend, 1999). Our model indicates that approximately 47% of the CCA-treated wood sold by 2006 in Florida has been disposed. Measured amounts of CCA-treated wood in Florida debris are consistent with our model results.

These results imply that arsenic measurements in groundwater at full-scale C&D disposal sites might already show signs of CCA-treated wood leaching and arsenic migration, if it occurs to a significant extent. The possibility that measurable arsenic migration to groundwater has occurred as a result of CCA-treated wood disposal in Florida's uniquely susceptible conditions is addressed here by evaluating arsenic concentrations in groundwater samples collected at unlined C&D disposal facilities in Florida and compiled in a regulatory database. The results of the evaluation are geared at determining appropriate disposal practices for obsolete CCA-treated wood.

2. Methods

2.1. Disposal model

The annual amount of CCA-treated wood disposed in Florida was forecast in a model using the production and expected service life of different CCA-treated wood products. Where measured data were unavailable, we made estimates and then performed a sensitivity analysis for those estimates to establish the impact of parameter uncertainty on our results and conclusions.

The disposal of CCA-treated wood was modeled using treated wood production data from 1970 through 1997 (Supplemental Table S1). Southern yellow pine (SYP) production data (Supplemental Table S2) were used to estimate the annual increase in CCA-treated wood use after 1997, since treated wood in Florida is primarily SYP. We used an American Wood-Preservers' Institute (AWPI) method to estimate the CCA-treated wood sales in Florida based on Southeast US production data (Parris, 2002). The Southeast Region accounted for 32.6% of US production in 1997 (Micklewright, 1998), which we extrapolated for all years. CCA-treated wood demand in Florida was estimated as a fraction of the production from the entire Southeast region, which includes Florida, Georgia, North Carolina, South Carolina, Virginia, and Puerto Rico. The method includes the following assumptions: use of residential products is proportional to the population in Florida relative to the region; use of industrial products (e.g. poles, ties) is proportional to the land area in Florida relative to the region; and use of marine products (e.g. piling) is proportional to the amount of coastline in Florida relative to the region. Florida contains 34% of the Southeast population (US Census Bureau, 2001), 23% of the Southeast land area, and 40% of the Southeast coastline (Millhouser et al., 1998). The Southeast CCA-treated wood production totals for residential, industrial, and marine products were multiplied by these factors, respectively, to obtain estimates for CCA-treated wood used in Florida annually.

Wood production data were available by product type (Micklewright, 1998). The use of each product differs, leading to different disposal patterns. We grouped products together according to similarity in use, leading to similarity in service life and time to disposal (Supplemental Table S3).

Researchers have shown that CCA-treated wood can remain sound outdoors for more than 50 years (e.g. Cooper, 1993). Cooper (1993) assumed a 25 year service life for CCA-treated wood products to estimate US and Canadian disposal patterns, made in the absence of measured data. More recently, published studies show that treated wood structures are replaced sooner than previously expected, for reasons including aesthetics, inadequate size, poor construction, and weather damage (McQueen and Stevens, 1998; Alderman et al., 2002). McQueen and Stevens (1998) polled Southeast US carpenters about the age of CCA-treated wood decks at the time of disposal, and reported the disposed deck age for 527 decks. The data were lognormally distributed, with an average age at disposal of nine years and standard deviation of five years. Alderman et al. (2002) summarized data from 580 contractors in Georgia, North Carolina, and South Carolina indicating that the average age of decks at removal was just less than 13 years. Other estimates of decking service life presented in peer-reviewed and popular literature (Truini,

1996; Rice et al., 2002) agree more closely with these studies (McQueen and Stevens, 1998; Alderman et al., 2002) than Cooper's assumption (Cooper, 1993). Based on these studies, we assumed an average disposal age of ten years, and a standard deviation of five years for outdoor residential CCA-treated wood products. A portion of the lumber included in the outdoor residential products category used in more permanent applications such as in near-soil portions of house frames would have a greater age before disposal; we estimated that this more permanent portion comprised 5% of the outdoor residential CCA-treated wood, and tested the model's sensitivity to this estimate.

We performed an analysis to gauge the potential influence of CCA-treated wood production trends on deck disposal data reported by McQueen and Stevens (1998). Assuming the number of decks disposed after a given service lifetime is proportional to the amount of CCA-treated wood produced in the year the deck was installed, we calculated the proportion of decks disposed per unit production for each service lifetime reported by McQueen and Stevens (1998). Based on this proportion, we calculated a "production-normalized" number of decks disposed for each service lifetime. The resulting alternative service lifetime distribution had an average of 16 years (versus nine years for the non-normalized data). This value was substituted into the model for sensitivity analyses.

The Florida CCA-treated wood disposal forecast model was programmed using MATLAB (The Mathworks, Natick, MA). The model simulates CCA-treated wood disposal using Eq. (1).

$$TW_{disposal(n)} = \sum_{i=1}^{7} \sum_{j=1970}^{n} [(TW_{used(i)(j)}) \times f_i(n-j,\mu_a,\sigma_a)] + \sum_{i=1}^{7} (1-\eta_i) \times TW_{used(i)(n)}$$
(1)

where TW (i.e. treated wood) is the CCA-treated wood volume used or disposed during a given year, *i* is one of the seven product types (Supplemental Table S3), *j* is the year, beginning in 1970 (the first year of CCA-treated wood demand data considered) through year n. An estimate of the efficiency of creating final products (e.g. decks, fences) from CCA-treated wood, η_i , is used to estimate cutoff waste, or wood that is disposed immediately as scrap. The discrete lognormal distribution function, f, with an arithmetic mean service life for product-type *i* of μ_a and arithmetic standard deviation σ_a was used to model disposal patterns for CCA-treated wood. We calculated the discrete distribution for each product, *i*, from a lognormal distribution with geometric mean, μ_i , and standard deviation, σ_i , chosen such that the derived discrete distribution would have the desired arithmetic mean and standard deviation (μ_a and σ_a). We calculated μ_i and σ_i according to Eqs. (2)-(4) using MATLAB's zero-finding function.

$$\mu_a = \sum_{(n-j)=0}^{\infty} (n-j)f_i(n-j)$$
(2)

$$\sigma_i^2 = \ln\left(\frac{\sigma_a^2}{\mu_a^2} + 1\right) \tag{3}$$

$$\mu_i = \ln(\mu_a) - \frac{\sigma_i^2}{2} \tag{4}$$

For example, in 1980, disposal was forecast by determining a discrete annual binning of the 1980 outdoor residential CCA-treated wood total that is approximately lognormal, and has an arithmetic average and standard deviation of the model user's choice (in this case ten years, so that the average year of disposal is 1990 and the standard deviation is five years). The simulation indicates that wood for outdoor residential use that was purchased in 1980 exhibited peak disposal levels during 1987 (Supplemental Figure S2).

The CCA-treated wood used in Florida was translated into disposal distributions for each year and product type as described for the example year, 1980. The volume of CCA-treated wood disposed during each year is the sum of disposal among all CCA-treated wood product types from all previous years, as described by Eq. (1).

We also forecast the amount of arsenic disposed in the wood. The product categories differ 10 fold in the amount of CCA initially infused into wood, or the CCA retention levels, from 4.0 to 40 kg of CCA (as CrO_3 , CuO and As_2O_5 , delivered in aqueous solution at a ratio of 2.6:1.0:1.8 by weight for the most common CCA formulation, i.e. CCA "Type C") per cubic meter of wood. We assigned an arsenic content to each product category, and in some cases, subdivided products into several categories having different CCA retention levels (Supplemental Tables S4, S5, and S6).

The sapwood portion of SYP, which dominates the treated wood market in Florida, is the only section of wood into which CCA can be infused; heartwood remains largely untreated. In the base-case model, we assumed that 100%of all products were comprised of sapwood, so we multiplied the disposed wood mass from Eq. (1), by the CCA retention level (Supplemental Table S6), the volume fraction of treated sapwood required by industry standards (Supplemental Table S6), and the fraction of arsenic in CCA Type C (0.22) to estimate the amount of arsenic in disposed wood. In the sensitivity analysis, we alternatively used industry data on the sapwood content of products (Supplemental Table S7).

2.2. Analysis of groundwater monitoring data

We evaluated the likelihood that arsenic from CCAtreated wood in landfills is impacting groundwater near unlined C&D landfills. This was done using chemical concentration data from a database of groundwater monitoring at C&D landfills that is required by law in Florida (described in FDEP, 2000). The database includes sampling events from March, 1997 through May, 2001.

Three types of sampling wells are associated with C&D landfills in Florida: compliance, detection, and back-

ground wells. Compliance wells are located downgradient from landfills and are subject to compliance with groundwater quality standards. Detection wells are located within the leachate-groundwater mixing zone and Florida groundwater standards are not applicable at these wells. Background wells are located upgradient from landfills. Data from wells that were not designated as one of these types were not included in the analysis. We compared groundwater data with the arsenic drinking water standard $(10 \ \mu g \ l^{-1})$, which is typically the most stringent groundwater data.

Arsenic was undetectable in 651 of the 776 samples (84%) in which arsenic measurements were made. The Florida database does not include information about detection limits. We surveyed the available laboratory reports that were used to generate the database and determined representative detection limits. The most frequent detection limit for arsenic was $5 \ \mu g \ l^{-1}$. We calculated the average arsenic concentrations in groundwater in two ways: by assigning a concentration of $5 \ \mu g \ l^{-1}$ to all nondetects, which is likely to provide a conservative estimate of the average, and by assigning a concentration of 0 μ g l⁻¹ to all non-detects, which provides the lowest limit of the average. We tested the statistical significance of apparent differences between the average arsenic concentrations in background and compliance wells using two different non-parametric methods: the Wilcoxon Rank-Sum Test and the Kolmogorov-Smirnov test. Lacking detection limit data, we conducted both significance tests in three ways: by setting all non-detects equal to zero, $2.5 \ \mu g \ l^{-1}$, or 5.0 μ g l⁻¹.

3. Results and discussion

3.1. Disposal forecast model

Fig. 1 presents the results of the disposal forecast model for the volume of CCA-treated wood and associated arsenic mass in Florida. New residential use of CCA-treated wood ceased at the end of 2003 as part of a change in the CCA product label specifying permitted uses. Thus the total amount of CCA-treated wood used and ultimately disposed will decrease in coming years, as is evident in Fig. 1, after 2006. Our model indicates that annual CCA-treated wood disposal is currently at the peak annual disposal rate. Residential treated wood is generally treated to a lower CCA retention level than wood for industrial or marine uses, so post-2006 changes in the forecast for arsenic disposal are not as substantial as changes in the wood volume disposed after 2006.

Due to a paucity of published, measured data for certain model parameters, we estimated these parameters. The estimated parameters include the service life for several CCA-treated wood product types, the standard deviation around the service lives (assigned as 5 years for all product categories), the proportion of products assigned to subcategories containing different CCA retention levels

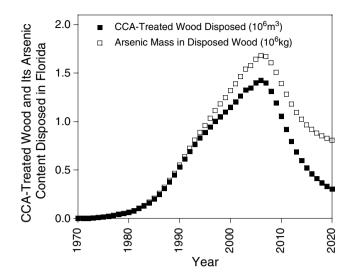


Fig. 1. Results of MATLAB data-based model for CCA-treated wood disposal in Florida. The maximum annual disposal rate of CCA-treated wood by volume and the maximum annual disposal rate of arsenic in CCA-treated wood occur in the year 2006.

(Supplemental Table S4), the amount of CCA-treated wood discarded immediately after purchase as cut-off scrap waste, and the amount of residential outdoor lumber used in house framing. In order to test the robustness of our model, we performed a sensitivity analysis for these parameters, our use estimate, which was based on population, coastline, and land area in Florida, and the percent of treatable sapwood in CCA-treated wood products. We chose a range of values for each estimated parameter, and reproduced the model with the newly substituted values.

In sensitivity analyses, the year of peak CCA-treated wood disposal and cumulative mass of arsenic disposed by 2000 were most strongly influenced by the service life of the largest product category - the outdoor residential product category. The annual rate of disposal is important because each individual landfill cell only accepts waste during a several-year segment of time. Cells that are open during years of higher CCA-treated wood disposal rates (e.g., 1990s and beyond) will contain a waste mixture composed of a relatively high proportion of CCA-treated wood. The influence of changing the service life for that product category between 5 and 25 years dramatically impacts the estimate of disposed wood (Supplemental Figure S3). Even when the service life is extended to 20 years, a value twice that supported by other studies (McQueen and Stevens, 1998; Alderman et al., 2002), 2006 disposal rates are 69% of the maximum - significantly greater than the value of approximately 47% in the previously published CCA-treated wood disposal model for Florida (Solo-Gabriele and Townsend, 1999). Only when we adopted their assumed 25 year service life for outdoor residential products, our results (i.e., 2006 disposal is 47% of the maximum, which would occur in 2020) were similar to those of Solo-Gabriele and Townsend (1999). This confirms the

importance of service life versus modeling approach or other parameters.

Table 1 presents the results of sensitivity analyses for parameters in the disposal model. The first two results columns in Table 1 describe the volume of CCA-treated wood disposed over time, and show that under nearly all scenarios tested, current disposal is near expected peak levels. The last column of results in Table 1 describes the mass of arsenic in disposed CCA-treated wood, for use in evaluating groundwater data, later. This analysis demonstrates that the only parameter to which the model's key predictions are sensitive is the service life of residential CCA-treated wood. This is the most certain among all values used in the model; because it is based on published data from two separate studies (McQueen and Stevens, 1998; Alderman et al., 2002). The model's insensitivity to the other parameters implies that uncertainty in these variables will not affect the outcome of the model and subsequent conclusions. Thus the sensitivity analysis provides confidence in the base-case model results.

To further assess the quality of the model, we surveyed the literature for field measurements of disposed CCA-treated wood to compare with our model's predicted volumes. The 1998 field data were the most comprehensive, and provide support for our model results. The total C&D waste disposed in Florida in 1998 was 5.4×10^6 metric tons (FDEP, 2001). Approximately 25% of the mass of C&D waste is wood (ERL and C.T. Donovan, 1992; McKeever, 1998), or about 1.35 million metric tons. Assuming that approximately 38% of the mass of waste wood is CCAtreated wood, based on the results of studies discussed below, the 1998 CCA-treated wood disposed was 513000 metric tons. The density of SYP is 0.51 metric tons m^{-3} (AWPA, 2002), so the volume of CCA-treated wood disposed in Florida in 1998 was approximately 1.01×10^6 m^3 . This compares favorably with our model estimate for CCA-treated wood disposal in 1998 of 1.05×10^6 m³. The previously published model yielded approximately 115000 m³ for 1998 (Solo-Gabriele and Townsend, 1999), or only 4% of the waste wood stream – a percentage much lower than in the authors' own studies of waste wood at recycling facilities, where CCA-treated wood was purposefully excluded (Solo-Gabriele et al., 2000).

The estimate that 38% of the 1998 waste wood stream was CCA-treated wood originates from the following information. Most disposed CCA-treated wood field measurements are taken in the recycling waste stream. However, treated wood is purposefully excluded from recycling activities because the presence of CCA constituents in waste wood may limit some of the potential end-uses of recycled wood products (e.g. Smith and Shiau, 1998). Tolaymat et al. (2000) examined wood chips that were sorted and pre-processed for recycling and found that 6% of the wood mass was CCA treated wood in 1996. Solo-Gabriele et al. (2000) examined seven waste wood loads that had been sorted to exclude CCA-treated wood, found that still, 16% of the mass was CCA-treated

Table 1

Key results from sensitivity analysis of estimated disposal model parameters compared to base-case model result (first row)

Parameter varied from base case	Parameter value used in base case	Alternative parameter value	Affect of using alternative parameter values			
			Max. annual CCA-treated wood disposal (10 ⁶ m ³)	Year of max. wood disposal	Cumulative arsenic disposed by 2000 ^b (10 ⁶ kg)	
Base-case results	_a	_	1.4	2006	13	
Service life mean for outdoor residential category	10 yrs	13 yrs 16 yrs	1.4 1.4	2009 2012	10 7	
Service life standard deviation for outdoor residential category	5 yrs	2.5 yrs 10 yrs	1.6 1.3	2008 2003	12 14	
Percent disposed as cutoff scrap $(1-\eta_i)$	2.5%	0% 10%	1.5 1.4	2006 2003	12 14	
Fraction of lumber products used above ground	66.5%	0 96.5%	1.4 1.4	2006 2006	13 13	
Percent outdoor residential lumber used in house framing	5%	0% 10%	1.5 1.4	2006 2006	13 12	
Mean service life for all products except outdoor residential	100%	50% of base case 200% of base case	1.4 1.4	2006 2006	13 13	
Service life std. dev. for all products except outdoor residential	5 yrs	2.5 yrs 10 yrs	1.4 1.4	2006 2006	13 13	
Florida's share of Southeast US utility pole use Florida's share of Southeast	23% 34%	8% 38% 19%	1.4 1.4 0.8	2006 2006 2006	13 13 8	
US lumber/timber use	5770	49%	2.0	2000	18	
Florida's share of Southeast US marine-use wood Products' sapwood content	40% 100%	25% 55% From Table S7	1.4 1.4 1.4	2006 2006 2006	12 13 12	

^a Not applicable.

^b In 2000, there were 98 C&D landfills operating in Florida (FDEP, 2002).

wood. They estimated that visual sorting reduces the percent of CCA-treated wood in wood waste by an increment of 15% to 20% (Solo-Gabriele et al., 2000). These data imply that CCA-treated wood may have comprised between 31% and 37% of waste wood in Florida in 1998. Solo-Gabriele et al. (2000) measured the mass of CCA-treated wood in three individual loads of unsorted C&D wood waste. Mixed, unsorted wood waste loads contained 16% and 60% CCAtreated wood, or an average of 38% CCA-treated wood by mass. This value is supported by Southeastern US data showing that treated wood production, which is predominantly CCA-treated wood, has been 30% or more of the total wood production since 1982 (Supplemental Table S8).

The model's predicted CCA-treated wood disposal patterns apply on average statewide. However, each individual landfill operates in a unique locality during a finite time range, so it is acknowledged that the statewide disposal model would not replicate any individual landfill's waste composition. For that reason, the most important conclusion that can be drawn from the results of the disposal model is not the particular year of peak disposal, but that under any of the modeling scenarios in Table 1, most C&D landfills that accept CCA-treated wood in Florida would likely contain a substantial amount of that material well before the period when the groundwater monitoring data were collected (e.g., from Table 1, $7-18 \times 10^6$ kg of arsenic was disposed in CCA-treated wood by 2000, at which time there were 98 C&D landfills operating in Florida. Simplistically dividing these numbers suggests that a hypothetical "average" C&D landfill would contain on the order of 70000 to 180000 kg of arsenic). That result, combined with evidence that most arsenic leaching would occur in the initial months after disposal (Jambeck et al., 2006), suggests that there was ample opportunity for substantial arsenic quantities to leach prior to the groundwater monitoring described next.

3.2. Groundwater monitoring data analysis

We evaluated arsenic concentration data for groundwater collected from wells at unlined C&D landfill sites compiled in a database by the FDEP (2000). Typically, each landfill had 5–6 monitoring wells, from each of which 2–3 samples were collected spanning the course of one year. Fig. 2 shows that the average arsenic concentration in samples from each of the well types tested at unlined C&D landfills is less than the drinking water standard, 10 µg l⁻¹. The data do not support the idea that landfill leachate is contributing arsenic to groundwater. Detection wells, by statute, must be located hydraulically downgradient and less than 15 m from the disposal area (unless

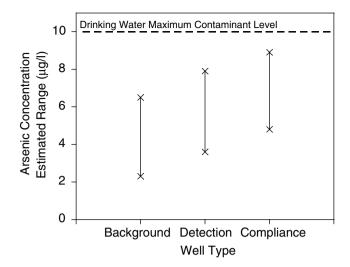


Fig. 2. Average arsenic concentration measured in groundwater at unlined C&D landfills in Florida between 1997 and 2001 estimated using two different assignments (0 μ g l⁻¹ and 5 μ g l⁻¹) for the 651 samples (of 776 total) in which arsenic was measured but not detected. There is no significant difference in concentrations among averages for the three well types.

impracticable), and thus contain leachate before it has been fully attenuated into the aquifer. They are designed to provide an "early warning" if contaminants are migrating from a landfill. Thus, constituents migrating from the landfill into groundwater will be present at higher concentrations in detection wells than in compliance wells, which are located at the edge of each landfill's state-designated "zone of discharge" - typically no further than 30 m from the landfill. Because the average arsenic concentration in detection wells is not greater than in compliance wells, it appears that arsenic is not migrating from unlined Florida C&D landfills to groundwater. There is no significant difference ($\alpha = 0.05$) among the average arsenic concentrations from background, detection, and compliance wells. Many sites had no detection wells; data from those sites were excluded for significance tests involving the average concentrations measured in detection wells.

Arsenic was undetectable in all samples at more than half of the landfills: arsenic was not detected in any samples at 34 out of 62 sites. Arsenic is detectable in largely unattenuated C&D landfill leachate (detection wells) at the same frequency as in background wells. Arsenic was detected in 14% of the detection well samples and in 15% of the background well samples.

Six (of 62) unlined C&D landfill sites exhibit average arsenic concentrations in compliance wells greater than the drinking water standard for arsenic (assuming nondetects are equal to a $5 \ \mu g \ l^{-1}$ detection limit) (Table 2). However, at three of those six sites, the average arsenic concentrations in background wells is also greater than the drinking water standard, and similar to the concentrations in compliance wells, indicating that the arsenic is from a source other than the landfill (sites B, D, and F from Table 2). At one of the three remaining sites, the average arsenic concentration in compliance wells is driven by a single occurrence where arsenic was detected (at 29 μ g l⁻¹); furthermore, arsenic was not detected in the subsequent sampling event, which is an indication that the single detection may have been the result of sample contamination, or the presence of soil particles in the sample (site E from Table 2). Finally, there was no significant difference ($\alpha =$ 0.05) between the average arsenic concentration in background well samples and in compliance well samples at the two remaining sites (sites A and C).

Subsequent to initial preparation of this analysis, FDEP provided us with additional groundwater data, including samples taken through 2003. Summary statistics and time trends were calculated for the new data (not shown) to determine if the results were comparable. The data were consistent with the results described above: no arsenic migration was apparent though 2003.

In conclusion, aggregate groundwater data from the 62 unlined C&D landfill sites and data from individual sites indicate no appreciable arsenic migration to groundwater, despite an estimated 12.8 million kilograms of arsenic disposal in CCA-treated wood in Florida by 2000. Notwithstanding Florida's unique conditions leading to a likelihood of observing arsenic impacts from disposed CCA-treated wood, we found a lack of associated arsenic migration to groundwater at unlined disposal sites. This may be attributable to limited solubility from wood surfaces or to limited migration because of landfill and soil chemistry. Arsenic solubility from wood surfaces may be limited by the relatively insoluble form of arsenic identified in CCA-treated wood, chromium arsenate (Bull, 2001), and

Table 2

Groundwater data for the six unlined C&D landfills in the FDEP database in which the average arsenic concentration (assuming non-detects, "ND," are equal to a 5 μ g l⁻¹ detection limit) in compliance wells exceeds the drinking water standard (10 μ g l⁻¹)

2	No. of	No. of arsenic	Arsenic concentration in groundwater samples from compliance wells ($\mu g \ l^{-1}$)				
	samples	detections	Average $(ND = 0)$	Average $(ND = 5)$	Minimum	Maximum	
A	9	5	67.3	69.5	ND	384	
В	19	6	17.1	20.5	ND	110	
С	5	3	10.3	12.3	ND	50	
D	4	2	8.8	11.3	ND	24	
E	4	1	7.3	11.0	ND	29	
F	6	1	6.5	10.7	ND	39	

the fact that wood is commonly disposed as large (i.e., lumber-sized) pieces such that a significant portion of the wood is not surface-exposed, and thus is not subject to rapid leaching, but slow, diffusion-controlled leaching (e.g., Jambeck et al., 2006). We are not aware of studies that examined arsenic precipitation and dissolution in C&D landfill leachate or adsorption onto solid constituents typically present in C&D landfills (e.g. concrete, asphalt), but clearly the chemical characteristics of both the solids in the landfill and the soil are also important for arsenic mobility. The refined estimate for CCA-treated wood disposal, showing a significant amount of CCA-treated wood disposed in Florida, and field-scale groundwater data at disposal sites, showing no evidence of arsenic impacts from unlined disposal sites, indicates that requiring disposal in hazardous waste landfills, would be unnecessary.

4. Note added in proof

While this manuscript was in press, we became aware of a recent two-part publication (Khan et al., 2006a,b) by the Florida research group whose earlier work we challenge in this document. In it, groundwater data near Florida C&D landfills were evaluated and older CCA-treated wood disposal estimates were revised and now match our data much more closely, although their conclusions still differ substantially from ours.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere. 2006.05.063.

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