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Attic dust and human blood samples collected near a former wood treatment facility ☆

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Received 6 October 2006; received in revised form 9 March 2007; accepted 23 March 2007

Abstract

A wood treatment facility operating in southern Alabama released dioxins and other hazardous substances into the surrounding community over a period of approximately 35 years. The facility used a variety of chemical insecticides including pentachlorophenol (PCP), chromated copper arsenate (CCA), and creosote (which contains polycyclic aromatic hydrocarbons (PAHs)) to treat wood. The health risks associated with the released contaminants are numerous and significant. To evaluate the historic exposure to the contaminants from the facility, blood samples and health surveys were collected from 21 current and past residents of the adjacent, isolated community and analyzed for concentrations of polychlorinated dibenzo-*p*-dioxins (dioxins) and polychlorinated dibenzofurans (furans). In addition, attic dust sampling was performed in 11 buildings located within a 1-mile radius of the former wood treatment facility. The average total dioxin toxic equivalent (TEQ) concentration in the residents' blood samples was 36.6 pg/g lipids. In the attic dust, the average total dioxin TEQ concentration, benzo[a]pyrene (PAH) TEQ concentration, and arsenic concentration were 145 ng/kg, 0.98 and 29.8 mg/kg, respectively. The concentrations of dioxins measured in the blood samples exceed the 90th percentile total dioxin levels found in the general US adult population. Concentrations of dioxin, arsenic, and PAHs found in the attic samples exceeded the US Environmental Protection Agency (USEPA) Region 4 soil exposure cancer risk preliminary remediation goal (PRG) values. These findings indicate a very significant potential for related health effects in these communities.

Keywords: Arsenic; Attic dust; Blood analysis; Dioxins; Polycyclic aromatic hydrocarbons

1. Introduction

A wood treatment facility operating in southern Alabama, from at least the 1960s to the mid 1990s, released dioxins and other hazardous substances into the surrounding community. The facility used a variety of chemicals including pentachlorophenol (PCP), chromated copper arsenate (CCA), and creosote as insecticides to treat wood. Wood waste and ash from incineration activities on site containing PCP, polychlorinated dibenzo-*p*-dioxins (dioxins), polychlorinated dibenzofurans (furans), creosote, chromium, and arsenic were dumped on the site and left exposed to the wind, creating a secondary airborne contaminant plume. Wastewater from the facility was also injected into boilers at the site, increasing the amounts of airborne contaminants in the community. The former wood treatment facility is located immediately adjacent to two isolated cities with a combined population of 2400 people (Fig. 1).

Dioxins and furans comprise a class of chlorinated congeners that are usually released as mixtures into the environment. The congeners activate a common toxicological mechanism by binding with a receptor protein, the Ah (aryl hydrocarbon) receptor, with varying degrees of potency (Webster and Commoner, 2003). The best-known congener, 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD), is a known carcinogen according to the International Agency for Research on Cancer (IARC) and the National Toxicology Program (NTP), having been the subject of many epidemiological and laboratory studies. Dioxins and furans are not produced commercially but occur as

☆ *Funding Sources*: Soil Water Air Protection Enterprise (SWAPE) and Environmental Litigation Group.

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doi:10.1016/j.envres.2007.03.011

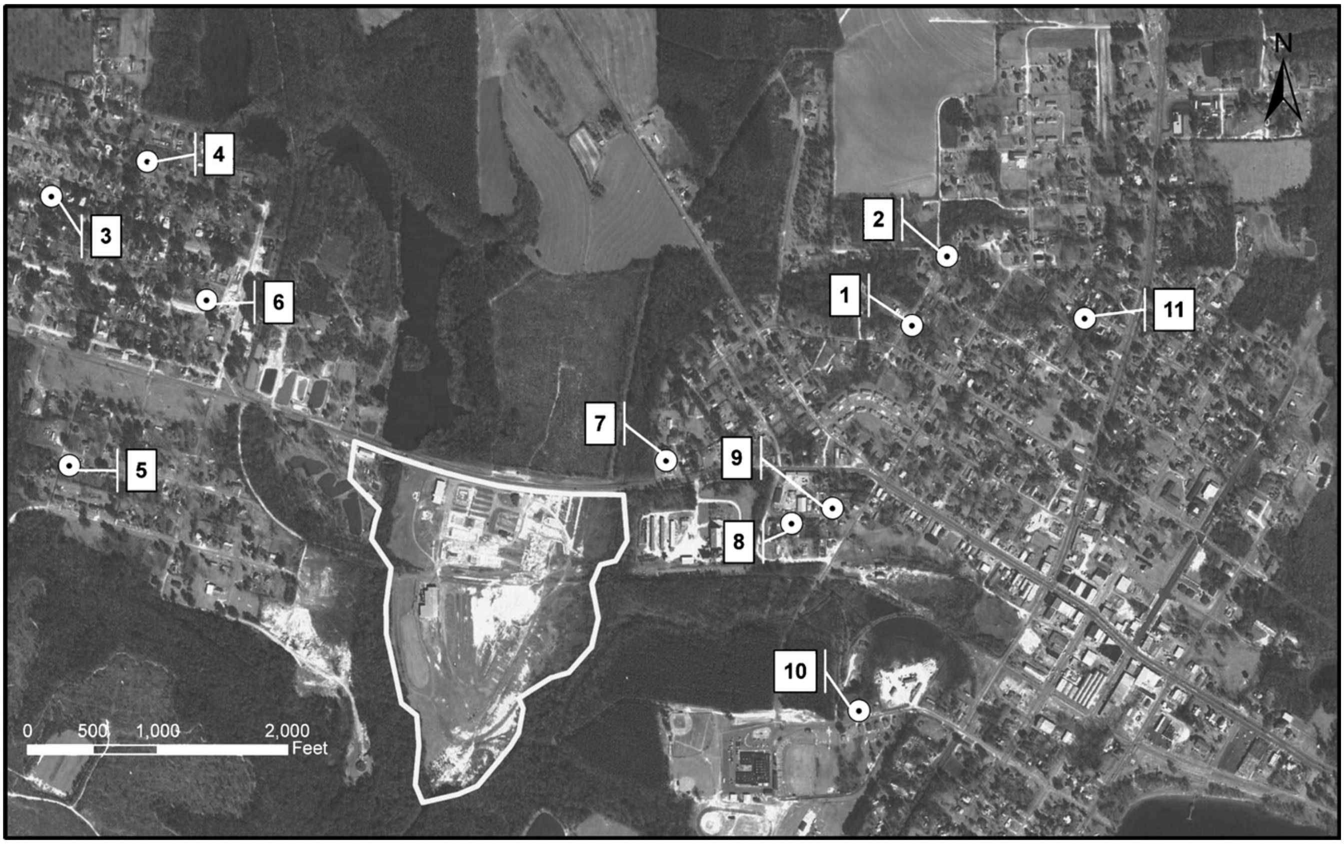

Fig. 1. Aerial photograph of the former wood treatment plant, surrounding communities, and approximate attic dust sample locations.

impurities in the manufacture of certain chlorinated compounds and as byproducts of certain incineration and combustion processes. PCP, a wood preservative used at the former wood treatment facility, contains dioxins and furans as impurities (Webster and Commoner, 2003). The more highly chlorinated dioxin and furan congeners are more likely to be created by burning of PCP-treated wood, particularly octacloro-dibenzo-p-dioxin (Paepke et al., 1989).

Sampling attic dust is a useful means of evaluating exposure, since dust settling within attics is often preserved from weathering, providing a ‘‘time capsule’’ of contaminants associated with dust (O’Connor and Sabrsula, 2005; Dahlgren et al., 2003). Over the years, dust from the outside environment infiltrates the attic and settles. The levels of contaminants within the attic dust provide a means to evaluate the resident’s potential historical exposure to those contaminants via airborne dust. The importance of sampling attic dust is not to evaluate the risks associated with exposure to the attic dust, but to use the attic dust to assess what levels of contaminants were present in the outdoor dust to which the residents were likely exposed. There are no other published papers that simultaneously measure attic dust and human blood for dioxins and furans.

Sampling human blood for dioxins and furans is indicative of past exposure because of their relatively long half-life in blood. The serum half-life in human was estimated at 7.1 years (range of 2.9–26.9 years) for TCDD in a group of 36 Vietnam veterans (Pirkle et al., 1989). A subsequent study of 343 Vietnam veterans, which included the 36 veterans in the prior study, calculated a half-life estimate of 8.7 years (95% CI of 8.0–9.5 years) (Michalek, 1996). In a separate study of occupational exposure, the estimated half-lives for dioxin congeners other than TCDD ranged from 3.5 to 15.7 years and half-lives for furan congeners ranged from 3.0 to 19.6 years (Flesch-Janys et al., 1996).

The National Health and Nutrition Examination Survey (NHANES) is a biomonitoring, cross-sectional, representative survey designed to collect information about the health and diet of the civilian, non-institutionalized population of the United States (CDC, 2005). The data from the NHANES is useful because it can be compared to blood samples collected from the community surrounding the former wood treatment. The NHANES is performed every few years by the Centers for Disease Control and Prevention (CDC) National Center for Health Statistics. For the NHANES, approximately 3162 people were sampled for dioxins/furans in blood in 1999–2000 and 2001–2002. The data are presented as average concentrations of each particular chemical or congener for particular sexes, age groups, racial groups, or as a whole. The 50th, 75th, 90th, and 95th percentile concentrations are reported. No further analysis of the data is performed by the CDC, but all data are available on their website.

Please cite this article as: Hensley, A.R., et al., Attic dust and human blood samples collected near a former wood treatment facility, Environm. Res. (2007), doi:10.1016/j.envres.2007.03.011

Table 1
USEPA Region 4 PRG values for relevant compounds (USEPA, 2006)

Compound	PRG for combined soil exposure routes[a]
Dioxin	3.9 ng/kg
Arsenic	0.39 mg/kg
Chromium	30 mg/kg
Copper	3,100 mg/kg
Benzo[a]pyrene (PAH)	0.062 mg/kg

[a]Combined soil exposure includes inhalation, dermal absorption, and incidental ingestion.

The USEPA preliminary remediation goal (PRG) values are guidelines for remediating contaminated sites and are developed to be health protective for the general population, including particularly sensitive sub-populations (USEPA, 2006). PRG values are set for each potential exposure route for each hazardous environmental contaminant. The soil exposure PRG values for contaminants of interest at this site are listed in Table 1. The site is located within USEPA Region 4, which uses the PRG values adopted by USEPA Region 9 (USEPA, 2000).

The purpose of this study was to evaluate whether releases of persistent organic pollutants (POPs) increased exposure within the community. Our thesis is that the release of contaminants to the environment has resulted in an increase of body burden of POPs.

2. Methods

2.1. Blood sampling and analysis

Blood samples were collected from 21 current and past residents of Florala and Lockhart, AL and analyzed to evaluate levels of dioxins and furans. Each resident had been exposed to contamination from the facility during its operation for at least 18 years. Four of the sampled individuals worked at the facility at some point. There were 13 females and 8 males, ages ranged from 25 to 76 years. They were chosen because they exhibited illnesses that could potentially have resulted from exposure to contaminants released by the facility. The occurrence of diseases in the sampled individuals was evaluated using health surveys that had been filled out by the sampled individuals and verified by a physician. The whole blood samples were collected in March 2006. After collection, samples were shipped overnight on ice to Severn Trent Laboratories (STL) in Sacramento, CA.

For the blood sample analysis, STL used high-resolution (HR) gas chromatography (GC)/HR mass spectrometry (MS) in general accordance with US Environmental Protection Agency (USEPA) Method 8290. The serum was separated and each sample was spiked with $^{13}C^{12}$-labeled internal standards prior to extraction. The target analytes were chromatographically separated by a DB-5 capillary column. Each sample batch included a Method Blank (MB), a Laboratory Control Sample (LCS), and the unknown serum samples. The analytical data were reviewed using comprehensive multi-tiered quality assurance and quality control procedures. Blood lipid content was determined gravimetrically.

2.2. Attic sampling and analysis

Attic sampling was performed in March 2006, in 11 buildings (10 houses and 1 church) located within a 1-mile radius of the former wood treatment facility. The attic samples were not collected from homes of individuals from whom we collected blood samples. We could not sample attic dust from several of the sampled individuals' homes, so we focused on sampling from attics distributed across the community. Samples were collected using a High Volume Simplified Small Surface Sampler (HVS4) in general accordance with American Society for Testing and Materials method D5438, "Standard Practice for Collection of Floor Dust for Chemical Analysis." At least 52 g of attic dust were collected at each building, the minimum amount required to perform dioxin/furan, polycyclic aromatic hydrocarbon (PAH), chromium, copper, and arsenic analysis. Attic dust samples were shipped to STL for analysis. PAHs were evaluated in general accordance with Method 8270C using GC/MS SIM. Dioxins/furans were analyzed in general accordance with USEPA Method 8290 using HRGC/HRMS. Metals were analyzed in general accordance with Method 6020 using Inductively Coupled Plasma/MS. Internal standards were added to all samples and each sample batch included an MB, an LCS, and the unknown attic dust sample. Quality assurance and quality control measures were taken similar to the blood sample analysis.

2.3. Toxic equivalents (TEQs) calculations

The levels of dioxins and furans in the attic and blood samples were quantified using USEPA (1989) toxic equivalent factors (TEFs). Toxic equivalents (TEQs) were similarly used to evaluate the levels of PAHs in the attic dust samples. TEFs assigned by the USEPA (1993) relating the toxicity of carcinogenic PAHs to benzo[a]pyrene were used.

Non-detect concentrations of the analytes-of-interest in the blood and attic dust samples were addressed by considering the incidences as zero and as the limit-of-detection (LOD)/2, in analysis of the data. Considering the incidences as zero can be understood as an under-estimation of the actual levels.

2.4. NHANES analysis

To compare the results of our blood sampling efforts to the levels of dioxins and furans in the general US population, the NHANES data were analyzed using USEPA (1989) TEF values. The average concentrations reported by NHANES for individuals over the age of 20 for each dioxin and furan congener were combined using TEFs to determine the average TEQ dioxin concentration. A TEQ value was determined for each of the percentiles reported by NHANES (50th, 75th, 90th, and 95th). When non-detect concentrations were reported for particular congeners, the LOD/2 was used for the analysis.

3. Results

3.1. Blood samples

The results of the dioxin and furan analysis of blood samples are presented below in Table 2. Blood samples often contained non-detectable concentrations for some of the congeners, so there was a large difference between considering non-detects as zero as opposed to considering them as LOD/2. Using the more conservative method, considering the non-detects as zero, the average total dioxin TEQ concentration in the residents' samples was 36.6 pg/g lipids. The average level in females ($n = 13$) was slightly greater than in males ($n = 8$), 37.2 and 35.8 pg/g lipid, respectively. The average dioxin TEQ concentration increased with age. Residents ages 20–29 averaged 13.3 pg/g lipids ($n = 2$), ages 44–59 averaged 37.0 pg/g lipids ($n = 14$), and ages 60+ averaged 45.0 pg/g lipids ($n = 5$). There were no individuals sampled between the ages of 29 and 44. When non-detectable concentrations are considered as

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LOD/2, the average total dioxin TEQ concentration in the residents' samples was 108.0 pg/g lipids.

The blood dioxins and furan concentrations in individuals that may have been exposed occupationally were found near the center of the range of concentrations. The highest concentration among those occupationally exposed was 50.3 pg/g lipids (using zero for non-detectable concentrations). The pattern of congeners observed in the blood samples is similar to the pattern expected from exposure to burning PCP-treated wood. The congeners with more chlorines were more prevalent than those with fewer chlorines. Among the residents of the households sampled, 13 separate health conditions possibly related to dioxin, arsenic, or PAHs exposure were reported. The most common conditions reported were hypertension, followed by diabetes, breast cancer, and lung cancer.

Table 2
Total dioxin TEQ concentration in blood samples collected from the community surrounding the former wood treatment facility (pg/g lipids)

Samples	Sample size	Total dioxin TEQ concentration	
		Method 1	Method 2
All	21	36.6	108.0
Males	8	35.8	101.7
Females	13	37.2	111.9
Ages 20–29	2	13.3	129.5
Ages 44–59	14	37.0	111.2
Ages 60+	5	45.0	90.4
Males ages 20–29	0	na	na
Males ages 44–59	4	32.8	114.5
Males ages 60+	4	38.7	88.8
Females ages 20–29	2	13.3	129.5
Females ages 44–59	10	38.6	116.7
Females ages 60+	1	na	na
50th percentile		35.5	100.9
75th percentile		53.6	137.8
90th percentile		65.0	143.6
95th percentile		70.0	153.0

Method 1: non-detects = zero.
Method 2: non-detects = LOD/2.
na, not applicable.

3.2. Attic dust samples

The results of the attic sampling dust analysis are presented in Table 3. The PAH analyses were performed twice (Run 1 and Run 2) due to the high values detected in the samples by the laboratory. Both runs were performed using the same methods. The average total dioxin TEQ concentration was 145 ng/kg. The average levels of arsenic, chromium and copper were 29.8, 21.7, and 33.9 mg/kg, respectively. The average level of total benzo[a]pyrene TEQs detected in the attic samples was 0.98 mg/kg. There were very few instances of non-detects for particular congeners in the attic dust samples, so there was very little difference between results using zero for non-detects and using LOD/2 for non-detects. The pattern of dioxin/furan congeners found in the attic dust is very similar to the pattern expected from a source that was burning PCP-treated wood. The higher chlorinated congeners were found at higher TEQ concentrations in the attic dust samples than congeners with fewer chlorines.

Concentrations of dioxin, arsenic, and PAHs found in the attic samples exceeded the USEPA Region 4 soil combined exposure cancer risk PRG values (Table 3) (USEPA, 2006). The total dioxin TEQ concentrations in the attic dust exceed the PRG values by approximately 2–128 times. Arsenic concentrations exceed the PRG values by about 5–653 times. Nine of the 11 attic sample concentrations of benzo[a]pyrene TEQs are above the PRG for benzo[a]pyrene, exceeding the value by approximately 2–123 times.

3.3. NHANES analysis

The average TEQ concentrations of dioxins and furans in the NHANES data from people over the age of 20 were 9, 16, 30, and 39 pg/g lipid for the 50th, 75th, 90th,

Table 3
Contaminants detected in attic dust samples collected from the community surrounding the former wood treatment facility

Site number	Total dioxin TEQ (ng/kg)	Arsenic (mg/kg)	Chromium (mg/kg)	Copper (mg/kg)	Total PAH TEQ (mg/kg)	
					Run 1	Run 2
1	**59.9**	**6.0**	**32.8**	22.8	**0.64**	**0.77**
2	**84.9**	**8.0**	**39.6**	33.2	**0.57**	**0.57**
3	**8.4**	**2.0**	18.5	8.6	**0.09**	0.05
4	**47.7**	**7.0**	15.6	28.8	**0.76**	**0.76**
5	**84.3**	**7.8**	16.6	26.5	**0.18**	**0.10**
6	**29.3**	**3.1**	11.9	26.4	**1.07**	**1.18**
7	**378.8**	**261.0**	25.2	44.7	**0.35**	**0.38**
8	**501.5**	**14.0**	27.7	122.0	**2.25**	**0.20**
9	**30.0**	**3.0**	17.8	13.3	0.04	0.01
10	**286.5**	**5.0**	17.4	18.3	**0.22**	**0.25**
11	**88.5**	**10.7**	15.7	28.4	**7.62**	**3.60**

Bold values: concentrations that exceed the US EPA Region 4 PRG value.

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and 95th percentiles, respectively, using USEPA TEF values (USEPA, 1989).

4. Discussion

4.1. Blood samples

The level of total dioxin TEQs the sampled residents' blood suggests that these individuals experienced an average level of dioxin exposure that is greater than the level of exposure that 95% of the US adult population experiences. The TCDD TEQ concentration are presented in Table 2. The serum half-life for TCDD in humans was estimated to be 7.1 years (range of 2.9–26.9 years) for TCDD in a group of 36 Vietnam veterans (Pirkle et al., 1989). It is important to consider that the observed levels were present-day average concentration in the blood samples. Considering the half-life of dioxin and the long history of dioxin release, the historical concentrations of dioxin in the sampled residents' blood were likely more significant in the past.

4.2. Attic dust samples

By comparing the levels of dioxins, furans, arsenic, and PAHs found in the attic dust to USEPA Region 4 PRG values, the levels can be considered unsafe for the general population, particularly for sensitive subpopulations. The present-day concentrations of these hazardous contaminants demonstrate that, although the contaminants are no longer being released from the facility, the residents' exposure is potentially ongoing as they are exposed to resuspended, contaminated dust. Although sampling outdoor dust would be informative of present day exposure via dust, attic dust can be more suggestive of historical exposure due to attics' time capsule-like qualities. The variability between sample locations could be a result of the construction of the homes. Some attics may be built in such a way that allows less outdoor dust to infiltrate than other attics. Prevailing wind direction should also be considered, but most of the emissions from the facility occurred at night according to former employees. This suggests that due to stable air conditions, the emission could have blown in many directions.

4.3. Potential health risks

Many epidemiological and experimental studies have demonstrated that exposure to chemicals such as dioxins, PAHs, and arsenic, cause a variety of diseases. The effects of dioxin exposure in humans include, but are not limited to, endocrine disruption, reproductive and developmental defects, immunotoxicity, hepatotoxicity, neurotoxicity, and a variety of cancers (Mandal, 2005; Schecter and Gasiewicz, 2003). Effects of exposure to PAHs can include breast, lung, and skin cancer, leukemia, respiratory toxicity, and reproductive toxicity (ATSDR, 1995; Bostrom et al., 2002). Arsenic exposure can lead to skin disease, cardiovascular disease, neurotoxicity, hepatic toxicity, hematological toxicity, endocrine disruption, and renal toxicity (Hughes, 2002; ATSDR, 2005). TCDD and arsenic are known human carcinogens according to the IARC and the NTP. The IARC and the USEPA consider several PAHs as probable human carcinogens and the NTP regards benzo[a]pyrene as reasonably anticipated to be a human carcinogen. The USEPA considers arsenic as carcinogens and dioxins as likely carcinogens (USEPA, 1998). Exposure to any combination of these compounds could very likely increase the risk of cancer or other diseases. These chemicals may also act as disease-promoting agents in an additive or synergetic manner (Carpenter et al., 2002).

4.4. Conclusions

The levels of dioxins, arsenic, and PAHs in the attic dust and blood samples indicate a very significant potential for related health effects in these communities. Because the attic dust samples showed elevated levels of dioxins, arsenic, and PAHs, there is evidence that residents have been and are being exposed to dust containing potentially unsafe levels of these contaminants. The present-day average total dioxin TEQ concentrations in the blood samples demonstrate that the residents were exposed to greater dioxin levels than 90% of the general US adult population. Historically, these concentrations were even more significant, considering dioxin's long half-life in blood. These results illustrate the need for biomonitoring and further investigation of the potential risks to the community surrounding this former wood treatment facility.

Acknowledgments

Thanks go to Florala Memorial Hospital for helping us collect and ship the blood samples and Environmetrics, Inc., for providing the HVS4 sampling equipment for the attic dust collection.

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