Journal of Occupational and Environmental Hygiene

ISSN: 1545-9624 (Print) 1545-9632 (Online) Journal homepage: https://www.tandfonline.com/loi/uoeh20

Amphibole Asbestos in Tree Bark—A Review of Findings for This Inhalational Exposure Source in Libby, Montana

Tony J. Ward , Terry M. Spear , Julie F. Hart , James S. Webber & Mohamed I. Elashheb

To cite this article: Tony J. Ward , Terry M. Spear , Julie F. Hart , James S. Webber & Mohamed I. Elashheb (2012) Amphibole Asbestos in Tree Bark—A Review of Findings for This Inhalational Exposure Source in Libby, Montana, Journal of Occupational and Environmental Hygiene, 9:6, 387-397, DOI: 10.1080/15459624.2012.682217

To link to this article: https://doi.org/10.1080/15459624.2012.682217

Accepted author version posted online: 09 Apr 2012.
Published online: 11 May 2012.

Journal of Occupational and Environmental Hygiene, 9: 387–397
ISSN: 1545-9624 print / 1545-9632 online

DOI: 10.1080/15459624.2012.682217

Amphibole Asbestos in Tree Bark—A Review of Findings for This Inhalational Exposure Source in Libby, Montana

Tony J. Ward,[1] Terry M. Spear,[2] Julie F. Hart,[2] James S. Webber,[3] and Mohamed I. Elashheb[1]

[1]Center for Environmental Health Sciences, The University of Montana, Missoula, Montana
[2]Department of Safety, Health and Industrial Hygiene, Montana Tech of the University of Montana, Butte, Montana
[3]Wadsworth Center, New York State Department of Health, Albany, New York

In June 2009, the U.S. Environmental Protection Agency (EPA) designated the town of Libby, Montana, a public health emergency—the first and only time the EPA has made such a determination under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA). From about 1920 until 1990, the leading source of vermiculite ore for the United States and the world was from a mine near Libby. This vermiculite ore was contaminated with fibrous and asbestiform amphibole in veins throughout the deposit. Today, areas surrounding the abandoned vermiculite processing/mining facilities and much of the town of Libby are contaminated with these asbestos fibers, contributing to an outbreak of asbestos-related diseases in the Libby population. Trees in Libby and in forested areas surrounding the abandoned mine have accumulated amphibole asbestos fibers on their bark surface, providing for inhalational exposures. Several studies have been conducted to further understand this exposure pathway. To address exposures to the public, Libby amphibole (LA) was measured in personal breathing zone and Tyvek surface wipe samples collected during firewood harvesting simulations, as well as in the ash and emissions of woodstoves when amphibole-contaminated firewood was combusted. Occupational studies simulating wildland firefighting and routine U.S. Department of Agriculture (USDA) Forest Service activities have also been conducted in the forested areas surrounding the abandoned mine, demonstrating the potential for inhalational exposures during common regional workplace activities. We present a review of the findings of this emerging environmental health concern impacting not only the residents of Libby but applicable to other populations living near asbestos-contaminated areas.

Keywords asbestos, amphibole, exposure, Libby, tree bark

Correspondence to: Tony J. Ward, Center for Environmental Health Sciences, The University of Montana, Missoula, MT 59812; e-mail: tony.ward@umontana.edu.

INTRODUCTION

Libby is located in the northwest corner of Montana in Lincoln County. With a population of ∼2700, the economy of Libby historically has been supported by the use of natural resources, such as logging and mining. Vermiculite was originally discovered six miles northeast of Libby in 1881. In 1919, Dr. Edward Alley found that vermiculite expanded (or "popped") when heated, creating pockets of air that made the material suitable for use in building insulation and as a soil conditioner. In the early 1920s, Dr. Alley created the Zonolite Company in Libby and developed the mine and processing facility at Vermiculite Mountain (also known as Zonolite Mountain). W.R. Grace & Co. bought the mine in 1963 and continued operation of the facility until closing it in 1990. Although the vermiculite from the Libby mine has many beneficial uses, the ore removed from the Libby mine was contaminated with fibrous and asbestiform amphibole in veins throughout the deposit.(1)

In Libby, occupational exposure to Libby amphibole (LA) has been shown to be associated with significant increases in asbestosis, lung cancer, and pleural cancer compared with the U.S. population.(2) Elevated incidences of asbestos-related disease have been reported in former mine and mill workers.(3–6) Medical testing of individuals who lived or worked in the Libby area for at least 6 months before 1991 showed pleural abnormalities (calcifications, thickenings, or plaques) in 17.8% of 6668 participants.(7) Although the focus of the Peipins et al.(7) study was to describe lung abnormalities in the general Libby population, significant factors for predicting pleural abnormalities included occupational pathways.(8) Additional occupational and non-occupational mesothelioma cases have been identified since the end of the last follow-up,(9–11) and current mortality figures indicate one new case per year in Lincoln County, Montana. A positive association has also been demonstrated between activities resulting in exposure to vermiculite (frequent handling of vermiculite) and self-reported respiratory symptoms among individuals who were $\leq$ 18 years of age at the time of the mine closure.[12]

The prevalence of forests surrounding the town of Libby, the lack of a natural gas pipeline, and the rising cost of fuel oil for home heating continue to make wood burning the most prevalent source of residential heat in the region. In a recent survey, results showed that there were ~2190 wood burning households in Libby, with ~2264 wood burning devices burning >5000 tons of wood/year.[13] This equates to 33% of Libby households that use wood as their primary source of heat. The report did not specify the number of homes that used woodstoves as a secondary heat source, although this number is likely substantial. For comparison, the state of Vermont had the highest percentage of households (9.4%) that used wood as the primary heat source, followed by Idaho (7.7%) and Montana (7.5%).[14] It should also be noted that a large woodstove changeout program was recently conducted in Libby in an effort to reduce ambient levels of woodsmoke $PM_{2.5}$.[15] As part of this community-wide intervention, approximately 1200 old, inefficient woodstoves were replaced with new EPA-certified woodstoves.

In 2005, it was discovered that trees in areas surrounding the vermiculite mine and throughout Libby serve as reservoirs for LA.[16] In this article, we summarize the results from multiple independent studies that have been conducted in an effort to understand the impact of these findings on the Libby community. These studies include assessing the potential for inhalational exposures to the general public that disturb LA-contaminated trees through residential home heating activities (i.e., firewood harvesting and woodstove use), as well as studies designed to evaluate wildland firefighting and other routine occupational tasks conducted by the U.S. Department of Agriculture (USDA) Forest Service in Libby.

METHODS

Tree Bark Sampling—Libby, Montana

Tree bark samples were originally collected around the former W.R. Grace vermiculite mine and former processing structures on November 2, 2004, in support of a proposed commercial logging exposure study.[16] Samples were collected from three heavily forested locations to simulate an amphibole fiber concentration gradient emanating from the mine. During the bark sampling, a spatula was used to collect a piece (~200 grams, 50–150 cm^2 bark surface area) of bark approximately 1.2 m from the base of the tree, with samples then placed into labeled plastic bags. On the basis of unexpectedly high LA concentrations in the initial bark samples, a follow-up bark collection was conducted in June 2005, which included expanding the program to collecting tree bark from areas west of Libby along the railroad line and from within the town of Libby. At all sampling locations, bark was collected from representative coniferous tree types in the area, including lodgepole pine (*Pinus contorta*), ponderosa pine (*P. ponderosa*), larch (*Larix occidentalis*), and Douglas fir (*Pseudotsuga menziesii*).

Following the Webber method,[16] the original bark samples were analyzed by the Wadsworth Center, New York State Department of Health in Albany. Briefly, bark samples (or subsamples) of approximately 1 gram (and 10 cm^2 surface area) were weighed, dried to stable mass at 60 to 100°C, ashed at 450°C, and reweighed to determine percentage loss of organic material. Residue was suspended in deionized water and filtered through 0.4 μm polycarbonate filters before being prepared for transmission electron microscopy (TEM) analysis using carbon coating and ethylene-diamine dissolution onto TEM grids. TEM analysis was performed at a screen magnification of at least 15,000× on a Hitachi 7100 scanning transmission electron microscope (STEM; Hitachi, Mountain View, Calif.) interfaced to a PGT IMIX Image-Analyser/X-Ray Detector (Princeton Gamma Technologies, Princeton, N.J.) Identification and measurement of LA was conducted according to EPA's modified Asbestos Hazard Emergency Response Act's (AHERA) protocol,[17] with any asbestos structures and other asbestiform amphiboles identified by energy dispersive X-ray analysis (EDX) and selected area electron diffraction (SAED). All fibers counted were at least 0.5 microns long and had aspect ratios of at least 5:1.

Activity-Based Exposure Studies—Assessing Exposures to the Public

In an effort to address exposures to the public related to woodstove use, two studies were conducted that included a firewood harvesting study to assess exposures during the harvesting of firewood, and a woodstove combustion study to assess the potential for exposures when burning contaminated firewood in a woodstove.

Firewood Harvesting Exposure Study

A firewood harvesting study[18] was conducted in 2006, with potential exposures primarily assessed via personal breathing zone (PBZ) sampling and surface wipe sampling of the outer layer of Tyvek clothing. The firewood harvesting study consisted of three independent simulation trials conducted on Forest Service property in an area of the Kootenai Forest inside the EPA restricted zone (Figure 1). Another simulation was conducted near Missoula, Montana, (approximately 4 hr southeast of Libby) that served as a control. Four to five investigators participated in each trial and assumed specific roles related to firewood harvesting (chainsaw operator, chainsaw assistant, and wood stackers).

PBZ air samples collected in this study were analyzed for LA and other fibers per Method 7400, Asbestos and Other Fibers[19] in the *NIOSH Manual of Analytical Methods* by phase contrast microscopy (PCM) and per AHERA.[17] Wipe samples were analyzed for LA per the ASTM D 6480-05 Method, TEM Asbestos Analysis.[20]

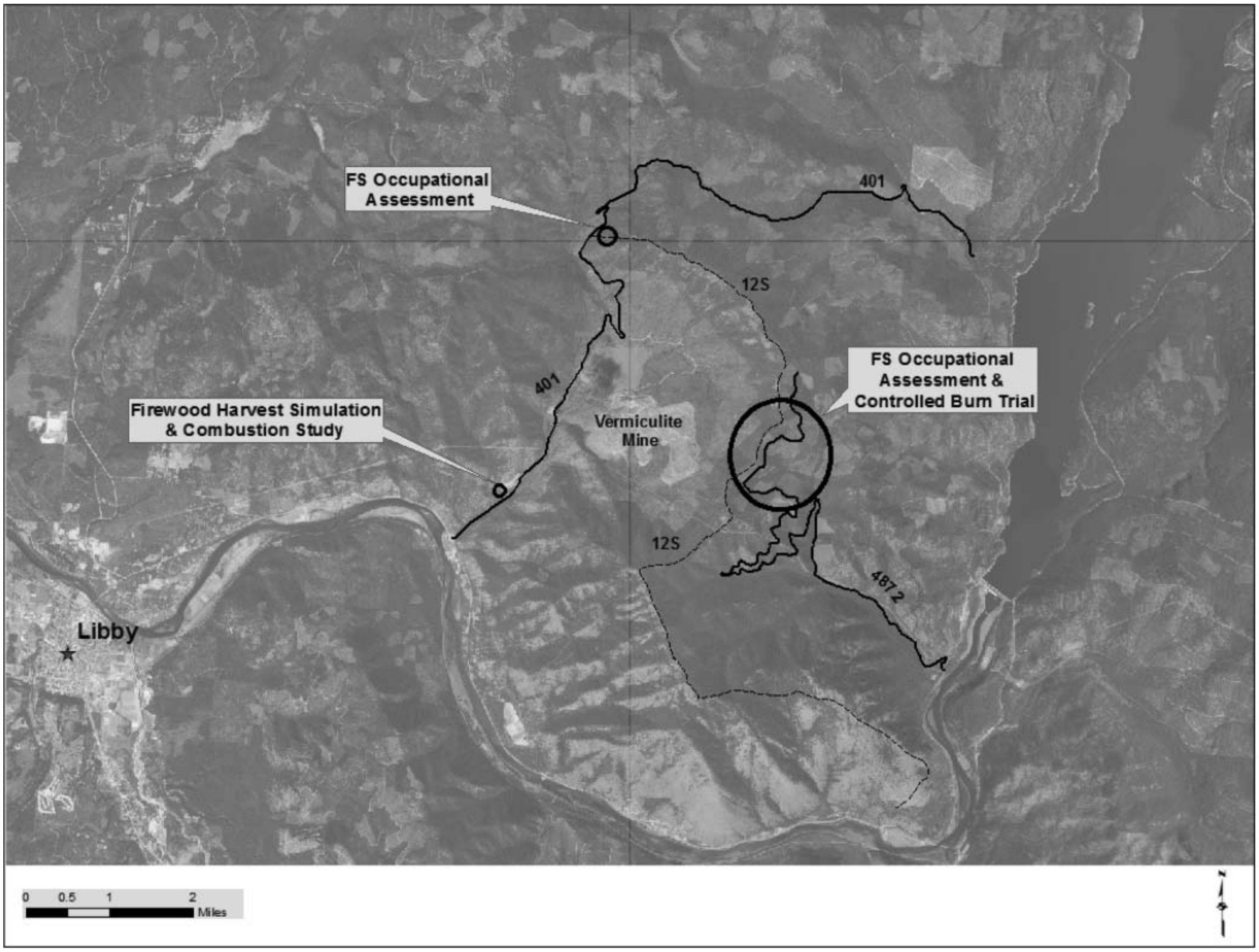

FIGURE 1. Location of firewood harvesting and Forest Service (FS) occupational trials in relation to the former vermiculite mine and the town of Libby, Montana

Woodstove Combustion Study

While the firewood harvesting study was designed to assess the potential for exposure associated with the cutting of firewood, an additional study was necessary to evaluate the potential for exposures when contaminated wood was burned in EPA-certified woodstoves.[21] This study was conducted in the same geographical location as the firewood harvesting simulations (Figure 1). Wood generated from the firewood harvesting simulations was burned in noncatalytic Quadra-Fire 2100 Millennium woodstoves (Quadra-Fire, Colville, Wash.). Approximately 4.6 m of 38-cm-diameter steel ductwork was attached to the stove outlet to simulate flue ductwork within a house. In addition, a plastic tote (lined with aluminum foil) was inverted near the exhaust port of the ductwork in an effort to aggregate the smoke so that smoke samples could be collected exiting the ductwork. Bulk ash samples and wipe samples at varying points along the exhaust ductwork and within the tote (both before and following the trials) were also collected during each trial. Wipe samples were analyzed for LA per the modified ASTM D 6480-05 Method,[20] with bulk ash samples analyzed by TEM per EPA Method EPA/600/R-93/116.[22]

Activity-Based Exposure Studies—Assessing Occupational Exposures

To address occupational exposures, a Forest Service occupational exposure assessment study[23] and a Forest Service controlled burn were conducted. Potential exposures were primarily assessed in these studies via PBZ sampling and surface wipe sampling of the outer layer of Tyvek clothing.

Forest Service Occupational Exposure Study

The USDA Forest Service occupational exposure study was conducted during the summer of 2008 to assess the potential for Forest Service employee exposures while working near the abandoned vermiculite mine—but outside the EPA restricted zone (Figure 1). Investigators simulated the following four routine activities: (1) walking through forested areas,

(2) conducting tree measurement, (3) constructing a fireline, and (4) performing trail maintenance. In addition to PBZ and Tyvek clothing surface wipe sampling, pre- and post-vehicle wipes were collected on the rear bumper of the vehicle used to transport investigators and equipment to the research site. Wipe samples were also collected from the chainsaw used in post-activity of several trials. PBZ air samples were analyzed for LA using PCM per NMAM 7400[19] and using TEM per modified AHERA.[17] Wipe samples were analyzed for LA per the modified ASTM D 6480-05 Method.[20]

Forest Service Controlled Burn Exposure Study

While the Forest Service occupational exposure assessment provided some guidance into the exposure potential associated with common occupational activities, firefighting or controlled burn activities were not included in this assessment. To address this activity, a small-scale controlled burn was conducted in a (3.7 m × 3.7 m) plot in July 2009. The plot location (Figure 1) was within the same geographical area where several of the simulated Forest Service tasks were conducted in the occupational assessments described above.

The controlled burn consisted of three activities, including fireline construction, combustion, and mop-up. Sampling was performed independently for each controlled burn activity. In addition to PBZ and Tyvek clothing surface wipe sampling, high-volume ambient air sampling was performed during the controlled burn activities. This sampling consisted of four sampling stations positioned 1.2 m from the perimeter of the burn, one station positioned 3.7 m above the burn plot, and one station positioned within the prevailing wind direction. Following the controlled burn, three ash samples were collected from the burn plot. PBZ and high-volume air samples collected in these studies were analyzed per NMAM 7400[19] and modified AHERA.[17] Wipe samples were analyzed for LA per the modified ASTM D 6480-05 Method.[20] Bulk ash samples were analyzed using TEM following a modified ASTM D 5755-0.3 Method.[24]

RESULTS

Tree Bark Sampling—Libby, Montana

For the initial set of samples (November 2004) from the mine site, both core and bark samples were collected to investigate the potential mechanisms of amphibole fiber incorporation into trees. The lack of amphibole fibers in the tree core sample indicated that amphibole fibers were not taken up by the root system of the tree to be incorporated into the wood itself. Fibers found in the bark samples would support our hypothesis that fibers can become embedded on the outside of the trees by diffusion and/or impaction/interception-type processes. From the original samples that were collected near the abandoned mine in November 2004, concentrations ranged from 14 million asbestos structures per square centimeter bark surface area (s/cm^2) to 110 million s/cm^2.[16] These original results have been confirmed by our team in follow-up bark sampling programs throughout the mine site and through a more comprehensive bark sampling program conducted by Region 8 EPA.[25] The EPA program measured significant amphibole contamination in tree bark near the mine (2.5 to 20 million s/cm^2), with contamination extending out miles from the mine in all directions. Figure 2 presents a scanning electron micrograph of amphibole fibers on bark.

The composition of the LA measured in the bark samples indicated the presence of both regulated (tremolite) and unregulated (primarily winchite and richterite) asbestos, with the majority of the samples displaying morphologies between prismatic crystals to asbestiform fibers. All fibers identified were typical of the Libby vermiculite contaminants,[26–28] with standard elemental composition of Si>Mg>Ca>Fe>Na>K. The LA fibers measured from the bark samples (n = 14) averaged 0.39 μm in diameter, with a length of 3.4 μm (aspect ratio of 11.5).

Activity-Based Exposure Studies—Assessing Exposures to the Public

Firewood Harvesting Exposure Study

Table I presents the PBZ results of the firewood harvesting study. PBZ results (n = 14) from the TEM analyses showed that the majority of the LA fibers detected during the firewood harvesting simulations were <5 μm in length, which is consistent with the size fractions seen in the bark sample results collected near the vermiculite mine. The mean time-weighted average (TWA) concentration for fibers <5 μm long was 0.15 structures per milliliter (s/mL) (SD = 0.21), while the mean concentration for fibers >5 μm long was 0.07 s/mL (SD = 0.08). Even though the PBZ sample from the chainsaw operator's assistant revealed the highest mean total LA concentration (0.40 ± 0.51 s/mL), overall, no differences were observed in PBZ concentrations between tasks. Substantial LA concentrations were also revealed on Tyvek clothing wipe samples from each of the investigators (Table I). The mean LA concentration (n = 14) was 30,000 s/cm^2 (SD = 38,000), with 91% (27,000 s/cm^2) composed of fibers <5 μm long.

Woodstove Combustion Study

The majority of the LA fibers measured following each of the three combustion trials were found in the ash, with measured concentrations of 137, 84, and 18 million s/gram, respectively. Results from the first two combustion trial wipe samples showed that LA fibers <5 μm were detected from within the ductwork and inside the tote following the burns (ranging from none detected (ND) to 20,476 s/cm^2). Given that LAs were not detected in pre-combustion wipe samples, these results suggest that LA fibers can be liberated into the ambient air when combusting contaminated firewood. No LA fibers were detected in the wipes from the final combustion trial ductwork, possibly due to the short duration of the trial (only 29 min). Consistent with tree bark samples in the area, the majority of the fibers detected from the ash and wipe samples were <5 μm in length. It should also be noted that LA fibers were detected in wipe samples only from the elbows and the

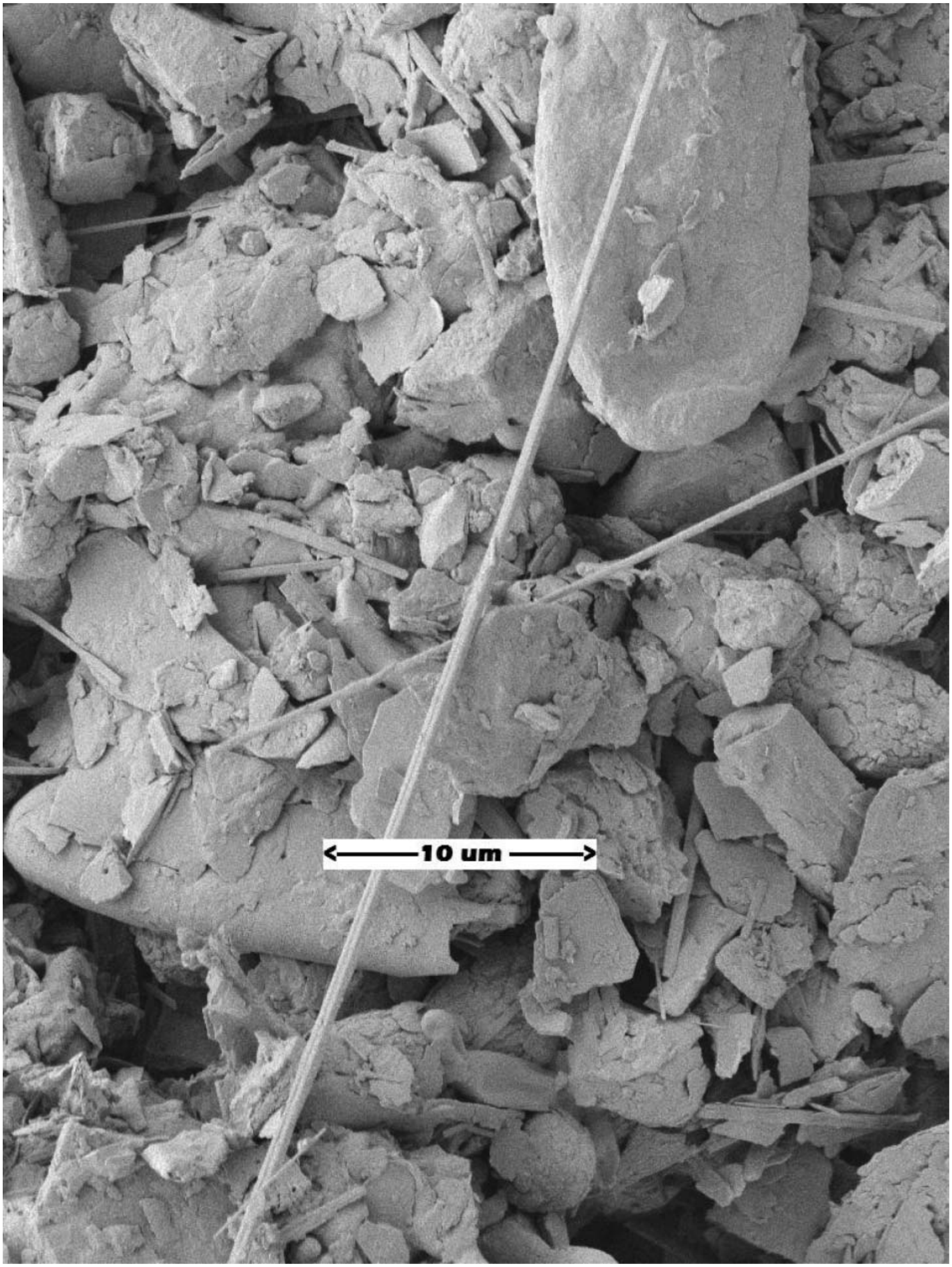

FIGURE 2. Scanning electron micrograph of amphibole fibers on bark

TABLE I. PBZ Results from the Public and Occupational Activity-Based Exposure Studies

	No. of Samples (n)	Mean Sample Time-Weighted PCM Concentration (f/mL)	% of Samples with Detectable Asbestos when Analyzed by TEM	TEM Sample Time-Weighted Concentration Range (s/mL) for LA <5 μm	TEM Sample Time-Weighted Concentration Range (s/mL) for LA >5 μm
Firewood harvesting trials	14	0.29	100	0.01–0.70	<AS–0.30
Forest Service occupational trials	24	0.04	25	<AS–0.05	<AS–0.07
Forest Service controlled burns[A]	12	0.06	75	<AS–0.04	<AS–0.06

Note: AS = 0.006–0.271 s/mL.
[A]Forty-four percent of controlled burn samples contained excessive material on the filter and required an indirect prep. Samples may have a negative bias because it was not possible to transfer all of the material from the air filter to an indirect prep.

plastic tote, and not from the straight sections of the ductwork (i.e., only from points where the airflow was redirected or impacted). Results from the air (smoke) samples collected inside the tote were less conclusive. Because of the volume of smoke inside the tote, the loading on the cartridges was significant, resulting in pump flow failures within minutes of the start of sample collection. Therefore, no air (smoke) samples were reported.

Activity-Based Exposure Studies—Assessing Occupational Exposures

Forest Service Occupational Exposure Study

For individual PBZ samples with LA >5 μm detected (Table II), 10 of 24 samples (42%) exceeded the OSHA permissible exposure limit (PEL) of 0.1 f/mL (assuming an 8-hr exposure duration) when analyzed by PCM. These 10 PBZ samples were all collected during the fireline construction simulation activity. When analyzed by TEM (and therefore excluding cellulose fibers from the analyses), 25% of the PBZ samples revealed asbestos concentrations greater than the analytical sensitivity (AS). These samples were collected during the fireline construction and tree measurement simulation activities. The mean (n = 4) PBZ sample TWA concentration for fireline construction activity samples was 0.08 s/mL (SD = 0.03), while the mean PBZ sample TWA concentration for tree measurement activity was 0.01 s/mL (SD = 0.00). AHERA TEM analyses for LA >5 μm revealed that none of the samples collected exceeded the OSHA PEL, assuming an 8-hr exposure duration. The specific tasks that measured PBZ concentrations greater than the AS for the fireline construction activity were brush clearer and Pulaski tool operator.

LA was detected on wipe samples collected from all of the activities evaluated. Fifty-two percent of post-activity wipe samples (n = 10) revealed concentrations greater than the detection limit, with mean concentrations of 941 s/cm^2 (SD = 899). The most elevated wipe concentrations were associated with the fireline construction activity, with a mean (n = 4) of 1456 s/cm^2 (SD = 1120). Similar to the PBZ samples, the tasks that generated wipe sample concentrations greater than the AS for the fireline construction activity were brush clearing, combi tool operating, and Pulaski tool operating. Other activities that generated LA (as detected by the wipes) were tree measurement activities, trail maintenance (brush clearer and chainsaw operator), and hiking activities. Vehicle wipes collected for one of the roads evaluated near the mine revealed concentrations below the AS, while results from

TABLE II. Tyvek Clothing Surface Wipe Results from the Public and Occupational Activity-Based Exposure Studies

	No. of Samples (n)	% of Samples with Detectable Asbestos when Analyzed by TEM	TEM Concentration Range (s/cm^2) for LA <5 μm	TEM Concentration Range (s/cm^2) For LA >5 $\mu\mu$m
Firewood harvesting trials	14	100	5000–109,000	400–5000
Forest Service occupational trials	23	52	<AS–1800	<AS–1800
Forest Service controlled burns	4	100	600–2500	<AS–1900

Note: AS = 400 – 5,270 s/cm^2.

TABLE III. High-Volume Air Sample TWA Results from the Forest Service Controlled Burns

Activity	PCM (f/mL)	TEM <5 μm (s/mL)	TEM >5 μm (s/mL)	TEM Total (s/mL)
Fireline const	0.02	0.01	0.01	0.02
Fireline const	<LOD	<AS	<AS	<AS
Fireline const	0.03	<AS	0.02	0.02
Fireline const	0.03	0.02	0.01	0.02
Fireline const	0.02	0.01	0.01	0.02
Combustion	<LOD	<AS	<AS	<AS
Combustion	<LOD	<AS	<AS	<AS
Combustion	0.05	<AS[A]	<AS[A]	<AS[A]
Combustion	0.01	<AS	0.01	0.01
Combustion	0.06	0.08	<AS[A]	0.08[A]
Combustion	0.26	<AS[A]	<AS[A]	<AS[A]
Combustion	0.26	<AS[A]	<AS[A]	<AS[A]
Mop-up	<LOD	<AS	<AS	<AS
Mop-up	<LOD	<AS	0.02	0.02
Mop-up	0.06	<AS[A]	0.02[A]	0.02[A]
Mop-up	0.02	0.01	0.01	0.02
Mop-up	0.04	<AS[A]	0.01[A]	0.01[A]
Mop-up	0.02	<AS	0.02	0.02

Notes: AS = 0.006–0.271 s/mL; Limit of Detection (LOD) 0.006–0.035 f/mL.
[A]Samples contained excessive material on the filter and required an indirect prep. Samples may have a negative bias because it was not possible to transfer all of the material from the air filter to an indirect prep.

another roadway evaluated measured LA concentrations of 17,917 s/cm^2. It should also be noted that LA was detected from post-activity chainsaw bar wipe samples. In terms of structure counts reported by the laboratory, 12 of 15 fibers were <5 μm long. No LA fibers were measured in the PBZ or wipe blanks.

Forest Service Controlled Burn Exposure Study

Nine of 12 (75%) of the PBZ samples revealed concentrations greater than the AS when analyzed by modified AHERA TEM (Table I), with the majority (64%) of structures detected >5 μm. TEM results for LA fibers >5 μm showed that none of the samples collected exceeded the OSHA PEL, assuming an 8-hr exposure duration. Tyvek clothing wipe samples collected from each investigator showed TEM total LA structure concentrations ranging from ND to 2500 s/cm^2 (Table I), with the majority (62%) of LA <5 μm.

Sixty-two percent of the high-volume ambient air samples revealed LA concentrations greater than the analytical sensitivity when analyzed by AHERA TEM (Table III), with LA identified in samples collected during all three activities (fireline construction, combustion, and mop-up). The mean high-volume TEM air concentrations for LA <5 μm and >5μm were 0.01 and 0.01 s/mL, respectively. In terms of fiber counts, 70% of the LA fibers identified in high-volume air samples were >5 μm long. Bulk ash LA concentrations collected above mineral soil ranged from 8 to 19 million s/gm, with 61% of LA <5 μm.

DISCUSSION

Tree Bark Sampling

Tree bark has been used since the late 1980s as a passive sampling media for both inorganic and organic pollutants.[29] Using the tree bark media, polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/PCDFs),[30] polyaromatic hydrocarbons (PAHs),[31] polychlorinated biphenyls (PCBs),[32–34] and organochlorine pesticides[35–36] have all been studied. Tree bark has also proved to be a good collection media for LA in Libby, with results collected near the abandoned vermiculite mine measuring concentrations from 14 to 260 million s/cm^2. Comparison of the Libby tree bark results to asbestos measured in settled dust in the United States underscores the significance of the Libby bark contamination. Ewing[37] discusses concentrations of surface dust found in a variety of settings and suggests that a concentration of 1000 asbestos/cm^2 may be considered clean, whereas concentrations >100,000 s/cm^2 indicate contamination. Concentrations on Libby bark near the vermiculite mine were in the *millions* of s/cm^2 range, concentrations that were seldom measured in settled dust within buildings, and only on surfaces under exposed asbestos-containing fireproofing.

Implications of Activity-Based Exposure Study Findings

Figure 3 presents a comparison of the PBZ results from the firewood harvesting and Forest Service occupational trials.

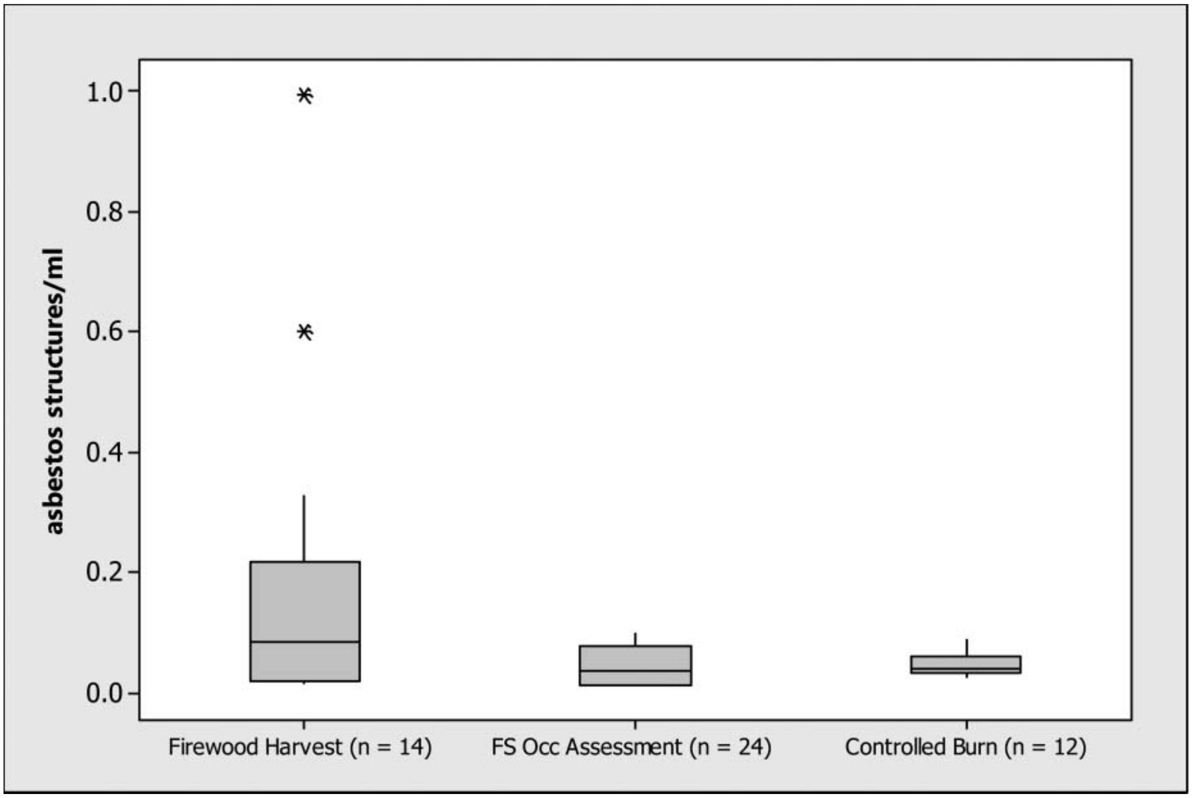

FIGURE 3. Comparison of Total LA (<5 μm and >5 μm) results from PBZ sampling, as measured by TEM

These results suggest there is an acute airborne exposure potential to LA associated with disturbing contaminated trees—both through common public and occupational activities. When analyzed by TEM, 100% of the firewood harvesting samples, 25% of the Forest Service occupational assessment samples, and 75% of the controlled burn samples revealed detectable concentrations of LA. PBZ results showed that the majority of the fibers detected were <5 μm in length, which is consistent with the size fractions seen in our bark sample results measured in the areas surrounding the abandoned vermiculite mine.

LA concentrations as measured by PBZ sampling were consistently higher in the firewood harvesting simulation samples compared with samples collected during the Forest Service occupational assessment and controlled burn trials (Figure 3). It is unclear whether the firewood harvesting activity is more likely to contribute to inhalation and clothing contamination or whether the higher concentrations observed were due to elevated concentrations of LA in tree bark. Since two of the Forest Service occupational activities evaluated also employed the use of a chainsaw (fireline construction and trail maintenance), this supports the hypothesis that the higher PBZ and wipe concentrations are most likely associated with elevated tree bark (source) concentrations.

In addition to respiratory exposures, substantial LA concentrations were also revealed on Tyvek clothing wipe samples from each of the investigators and from occupational trials. In the Forest Service occupational exposure assessment study, clothing contamination was revealed in 52% of the wipe samples, with each occupational activity having at least one positive wipe sample for LA. Consistent with the PBZ samples, the highest LA clothing wipe concentrations were found within the firewood harvesting samples. Equipment contamination was also measured following the occupational trials. Cross-contamination of vehicle cabs, vehicle boxes, equipment storage areas, and offices may occur as a result of clothing and equipment contamination.

We also discovered a seasonal influence to the exposures. When carrying out the firewood harvesting trials, one trial was conducted during July, while the remaining two trials were conducted in October. Results from the firewood harvesting trials showed that there were much higher levels of LA measured from the wipes during the hot, dry July trials (for each of the participants) when compared with the two trials conducted during the cooler, wetter month of October. These findings support the hypothesis that there is a greater potential for exposure during summer compared with late fall and early spring. From a public health point of view, this is an important finding.

Potential In-Home Exposures from Burning Contaminated Wood

Results from the woodstove combustion study suggest that there are also potential exposures associated with burning contaminated firewood during "typical" residential home heating practices. Results of the combustion trials showed that the majority of the LA fibers remained in the ash following the combustion process.[21] This suggests that disturbing the contaminated ash (as part of routine cleaning of the stove) is an additional (and ongoing) source of exposure within the home. The detection of LA in the elbows and tote (i.e., where the emission stream was either redirected or concentrated) supports the hypothesis that fibers can become impacted or intercepted onto the ductwork during combustion. When burning firewood with low-level contamination over a long period, it should be considered that fibers may build up within the creosote inside the ductwork. This could have implications when cleaning and/or changing flue ductwork.

Potential for Asbestos Contamination in Areas Outside Libby

Similar to the areas surrounding the Libby mine (a vermiculite mine contaminated with asbestos), it is possible that there is asbestos contamination in areas surrounding asbestos mines as a result of fugitive releases and downwind deposition. In addition to active/abandoned mines located in Canada, China, Brazil, South Africa, Italy, Zimbabwe, India, Australia, South Korea, and in the former Soviet Union, natural asbestos occurrences are found throughout the world. In the United States, asbestos is naturally occurring (with some areas mined) in 17 states, including the eastern United States, Appalachian region, California, Arizona, and Oregon.[38–41] Historically, chrysotile has accounted for more than 90% of the world's mined asbestos.[42]

We also hypothesize that areas surrounding historical Libby vermiculite-processing facilities are contaminated with LA. Raw vermiculite was hauled from Libby to over 100 processing (exfoliation) facilities across the United States for 70 years. Findings from the Agency for Toxic Substances and Disease Registry (ATSDR) study suggest that in addition to 28 historical exfoliation sites evaluated, many of the other abandoned exfoliation facilities throughout the United States may have residual Libby amphibole contamination in indoor settled dust, as well as in exterior soil.[43] This hypothesis is supported by results from bark sampling programs conducted in residential areas surrounding historical vermiculite processing facilities in Spokane (Wash.), Santa Ana (Calif.), Newark (Calif.), and Phoenix (Ariz). Even though these facilities last processed Libby vermiculite more than 20 years ago, LA was still detected from a subset of the bark samples collected from each of the sites, with concentrations approaching 3 million s/cm^2.[44]

CONCLUSION

On June 17, 2009, EPA Administrator Lisa P. Jackson announced that a public health emergency exists in Libby. This is the first time that the EPA has made a determination under CERCLA that conditions at a site constitute a public health emergency. This designation recognizes the serious public health impacts on the residents in the area and underscores the need for further action and health care for area residents who have been or are currently being exposed to LA.

Recently, it has been discovered that trees in Libby serve as reservoirs for LA asbestos. Fugitive fibers that were generated from the abandoned vermiculite mine near Libby during 70 years of mining have settled and accumulated in the surrounding forested areas, with fibers traveling miles from the point source once liberated into the air. It has also been demonstrated that LA can become liberated when the contaminated trees are disturbed through common local activities, such as firewood harvesting, and that occupational exposures can occur when working in these contaminated areas. In addition, LA detected in high-volume air sampling during a controlled burn suggests that fibers can become liberated during a wildland fire event.

Residential wood combustion (woodstoves) is the predominant method of home heating in Libby and many areas throughout northwest Montana. Through residential woodstove combustion studies, results showed that although fibers can be released into the ambient air during the combustion process, the majority of the fibers remain in the ash post-combustion, leading to further in-home exposures. These findings have serious implications for the Libby cleanup, where millions of dollars have been spent cleaning up the homes. Burning of contaminated firewood may be an ongoing source of re-exposure within these homes, thereby negating the impact of the in-home cleanups.

Cumulatively, the findings from these studies demonstrate the potential for inhalational exposures resulting from common activities that disturb LA-contaminated trees in Libby. We believe that additional activity-based sampling in contaminated areas of interest, coupled with risk assessment modeling focused on the sizes of fibers typically measured in the Libby vermiculite product (<5 μm in length), should be conducted in an effort to further understand this exposure pathway and, subsequently, prevent future exposures in Libby. Finally, the findings from these studies also provide insight into inhalational exposure risks resulting from disturbing trees contaminated with other pollutants (i.e., organochlorine pesticides, metals, PCBs, and so on) in other areas throughout the United States and the world.

ACKNOWLEDGMENTS

The authors thank U.S. EPA Region 8 and the Forest Service Kootenai District for granting access to the restricted mine area. The majority of the work was supported by NIH COBRE grant P20-RR017670, with additional funding provided by The Rocky Mountain Center for Occupational and Environmental Health, the University of Utah School of Medicine (with NIOSH support) the Forest Service Kootenai District, and W.R. Grace & Co.

REFERENCES

1. **Pardee, J.T., and E.S. Larsen:** Deposits of vermiculite and other minerals in the Rainy Creek District, near Libby, Montana. *USGS Bulletin 805*:17–28 (1929).
2. **Sullivan, P.A.:** Vermiculite, respiratory disease and asbestos exposure in Libby, Montana: Update of a cohort mortality study. *Environ. Health Perspect. 115*:579–585 (2007).
3. **McDonald, J.C., A. McDonald, B. Armstrong, and P. Sebastien:** Cohort study of mortality of vermiculite miners exposed to tremolite. *Br. J. Ind. Med. 43*:436–444 (1986).
4. **Amandus, H.E., and R. Wheeler:** The morbidity and mortality of vermilite miners and millers exposed to tremolite-actinolite: Part II. *Mortality. Am. J. Ind. Med. 11(1)*:15–26 (1987).
5. **Amandus, H.E., R. Althouse, W. Morgan, E. Sargent, and R. Jones:** The morbidity and mortality of vermiculite minters and millers exposed to tremolite-actinolite: Part III. Radiographic findings. *Am. J. Ind. Med. 11(1)*:27–37 (1987).
6. **Amandus, H.E., P. Wheeler, J. Jankovic, and J. Tucker:** The morbidity and mortality of vermilite miners and millers exposed to tremolite-actinolite: Part I. Exposure estimates. *Am. J. Ind. Med. 11(1)*:1–14 (1987).
7. **Peipins, L.A., M. Lewin, S. Campolucci, et al.:** Radiographic abnormalities and exposure to asbestos contaminated vermiculite in the community of Libby, Montana, USA. *Environ. Health Perspect. 111*:1753–1759 (2003).
8. **Price, B.:** Correspondence: Radiographic abnormalities and asbestos exposure: Libby, Montana. *Environ. Health Perspect. 112(2)*:A82–A83 (2004).
9. **McDonald, J.C., J. Harris, and B. Armstrong:** Mortality in a cohort of vermiculite miners exposed to fibrous amphibole in Libby, Montana. *Occup. Environ. Med. 61*:363–366 (2004).
10. **Case, B.:** Mesothelioma update for Libby, Montana: Occupational and non-occupational. *Lung Cancer 54*:S1–S10 (2006).
11. **Whitehouse, A.C., C. Black, M. Heppe, J. Ruckdeschel, and S. Leven:** Environmental exposure to Libby asbestos and mesotheliomas. *Am. J. Ind. Med. 51(11)*:877–880 20620 (2008).
12. **Vinikoor, L.C., T. Larson, T. Bateson, and L. Birnbaum:** Exposure to asbestos-containing vermiculite ore and respiratory symptoms among individuals who were children while the mine was active in Libby, Montana. *Environ. Health Perspect. 118(7)*:1033–1038 (2010).
13. **Ganesan, K., C. Cain, and J. Carlin:** *$PM_{2.5}$ Emissions from Residential Wood Burning in Libby, Montana*. Helena, Mt.: Montana Department of Environmental Quality, 2006.
14. "Historical census of housing tables, house heating fuel." [Online] Available at http://www.census.gov/hhes/www/housing/census/historic/fuels.html (accessed January 6, 2008).
15. **Ward, T.J., L. Rinehart, and T. Lange:** The 2003/2004 Libby, Montana $PM_{2.5}$ Source Apportionment Research Study. *Aerosol Sci. Technol. 40*:166–177 (2006).
16. **Ward, T.J., T. Spear, J. Hart, et al.:** Amphibole fibers in tree bark. *Sci. Tot. Environ. 367(1)*:460–465 (2006).
17. "Asbestos," *Code of Federal Regulations Title 40*, Part 763. 1987.
18. **Hart, J.F., T. Ward, T. Spear, K. Crispen, and T. Zolnikov:** Evaluation of asbestos exposures during firewood harvesting simulations in Libby, Montana. *Ann. Occup. Hyg. 51(8)*:1–7 (2007).
19. **National Institute for Occupational Safety and Health (NIOSH):** Method 7400, Asbestos and Other Fibers by PCM. In *NIOSH Manual of Analytical Methods (NMAM)*, 1994.
20. **American Society for Testing and Materials Standards (ASTM):** *Standard Test Method for Wipe Sampling of Surfaces, Indirect Preparation, and Analysis for Asbestos Structure Number Concentration by Transmission Electron Microscopy* (ASTM D6480–05). [Standard] West Conshohocken, Pa.: ASTM, 2006.
21. **Ward, T.J., J. Hart, T. Spear, B. Meyer, and J. Webber:** Fate of Libby amphibole fibers when burning contaminated firewood. *Environ. Sci. Technol. 43(8)*:2878–2883 (2009).
22. **U.S. Environmental Protection Agency (USEPA):** *Method for Determination of Asbestos in Bulk Building Materials* (EPA/600/R-93/116). Washington, D.C.: USEPA, 1993.
23. **Hart, J.F., T. Spear, T. Ward, C. Baldwin, M. Salo, and M. Elashheb:** An evaluation of the potential exposure to asbestiform amphiboles near a former vermiculite mine. *J. Environ. Public Health* doi:10.1155/2009/189509 (2009).
24. **American Society for Testing and Materials Standards (ASTM):** *Standard Test Method for Microvacuum Sampling and Indirect Analysis of Dust by Transmission Electron Microscopy for Asbestos Structure Number Surface Loading* (Method D5755–03). [Standard] West Conshohocken, Pa.: ASTM International, 2003.
25. **U.S. Environmental Protection Agency (USEPA):** *Asbestos Levels in Tree Bark*. Project number 0100-008-900. Washington, D.C.: USEPA (2008).
26. **Meeker, G.P., A. Bern, I. Brownfield, et al.:** The composition and morphology of amphiboles from the Rainy Creek Complex, near Libby, Montana. *Am. Mineral 88*:1955–1969 (2003).
27. **Wylie, A.G., and J. Verkouteren:** Amphibole asbestos from Libby, Montana, aspects of nomenclature. *Am. Mineral 85*:1540–1542 (2000).
28. **Gunter, M.E., D. Dyar, B. Twamley, F. Foit, and S. Cornelius:** Composition, Fe^{3+}/Fe, and crystal structure of non-asbestiform and asbestiform amphiboles from Libby, Montana, U.S.A. *Am. Mineral 89*:1579–1579 (2003).
29. **Schulz, H., P. Popp, G. Huhn, H-J. Stark, and G. Schuurmann:** Biomonitoring of airborne inorganic and organic pollutants by means of pine tree barks. I. Temporal and spatial variations. *Sci. Total Environ. 232*:49–58 (1999).
30. **Clarkson, P.J., D. Larrazabal-Moya, I. Staton, et al.:** The use of tree bark as a passive sampler for polychlorinated dibenzo-*p*-dioxins and furans. *Int. J. Environ. Anal. Chem. 82*:843–850 (2002).
31. **Sturaro, A., G. Parvoli, and L. Doretti:** Plane tree bark as a passive sampler of polycyclic aromatic hydrocarbons in an urban environment. *J. Chromatogr. 643*:435–438 (1993).
32. **Meredith, M.L., and R. Hites:** Polychlorinated biphenyl accumulation in tree bark and wood growth rings. *Environ Sci. Technol. 21*:709–712 (1987).
33. **Hermanson, M.H., and R. Hites:** Polychlorinated biphenyls in tree bark. *Environ. Sci. Technol. 24*:666–671 (1990).
34. **Hermanson, M.H., and G. Johnson:** Polychlorinated biphenyls in tree bark near a former manufacturing plant in Anniston, Alabama. *Chemosphere 68*:191–198 (2007).
35. **Simonich, S.L., and R. Hites:** Relationships between socioeconomic indicators and concentrations of organochlorine pesticides in tree bark. *Environ. Sci. Technol. 31*:999–1003 (1997).
36. **McDonald, J.C., and R. Hites:** Radial dilution model for the distribution of toxaphene in the United States and Canada on the basis of measured concentrations in tree bark. *Environ. Sci. Technol. 37*: 475–481 (2003).
37. **Ewing, W.M.:** Further observations of settled asbestos dust in buildings. In *Advances in Environmental Measurement Methods for Asbestos*, M.E. Beard and H.L. Rook (eds.). West Conshohocken, Pa.: ASTM, 2000.
38. **U.S. Geological Survey (USGS):** *Reported Historic Asbestos Mines, Historic Asbestos Prospects, and Natural Asbestos Occurrences in the Eastern United States* by B.S. Van Gosen. Open-File Report 2005–1189. Reston, Va.: USGS, 2005.
39. **U.S. Geological Survey (USGS):** *Reported Historic Asbestos Prospects and Natural Asbestos Occurrences in the Central United States* by B.S. Van Gosen. Open-File Report 2006–1211. Reston, Va.: USGS, 2006.
40. **U.S. Geological Survey (USGS):** *Reported Historic Asbestos Mines, Historic Asbestos Prospects, and Natural Asbestos Occurrences in the Rocky Mountain States of the United States (Colorado, Idaho, Montana, New Mexico, and Wyoming)* by B.S. Van Gosen. Open-File Report 2007–1182. Reston, Va.: USGS, 2007.
41. **Milano, M.:** "Where is asbestos found?" [Online] Available at http://www.asbestos.net (accessed January 6, 2008).

42. **Ross, M., and R. Virta:** Occurrence, production and uses of asbestos. In *The Health Effects of Chrysotile Asbestos: Contribution of Science to Risk-Management Decisions*, R.P. Nolan, A.M. Langer, F.J. Wicks, and R.F. Martin (eds.). *Can. Mineral.* (Special Publication) 5:79–88 (2001).
43. **Agency for Toxic Substances and Disease Registry (ATSDR):** *Summary Report: Exposure to Asbestos-Containing Vermiculite from Libby, Montana, at 28 Processing Sites in the United States*. Atlanta, Ga.: ATSDR, 2008.
44. **Elashheb, M.I.**, **T. Spear**, **J. Hart**, **J. Webber**, **and T. Ward:** Libby amphibole contamination in tree bark surrounding historical vermiculite processing facilities. *J. Environ. Protection 2*: 1062–1068 (2011).