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Fate of Libby Amphibole Fibers When Burning Contaminated Firewood

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In Libby, Montana, over 70 years of mining amphibole-contaminated vermiculite has led to amphibole contamination in areas surrounding the abandoned mine and in other areas throughout the town. In addition to contaminated soils, tree bark has also been found to be contaminated with amphibole fibers throughout the Libby area. As residential woodstoves are the main source of home heating in Libby, the purpose of this study was to determine if amphibole fibers become liberated into the ambient air when amphibole-contaminated firewood is combusted.

Amphibole-contaminated firewood was combusted in new, EPA-certified stoves during three trials. The results of these trials showed that the majority of the fibers remained in the ash following the combustion process, suggesting that additional potential exposures can occur within the homes to those that clean the ash out of woodstoves. The combustion trials also revealed that amphibole fibers can become liberated into the ambient air during the combustion process. Amphibole fibers were found impacted in the ductwork, as well as detected in wipe samples collected from an inverted container used to concentrate the woodsmoke emissions. These findings stress the need for identifying a clean fuel source for the inhabitants of Libby to prevent future exposures.

Introduction

Libby (population ~2,700, with nearly 12,000 in the surrounding area) is located in northwest Montana, and was once home to one of the world's largest vermiculite mines. In addition to the beneficial uses of vermiculite, the ore removed from the vermiculite mine (in operation ~1923−1990) was also contaminated with a toxic form of naturally occurring fibrous and asbestiform amphibole in veins throughout the deposit (*1*). Meeker et al. (*2*) conducted the first comprehensive study on the Libby fibers to determine the composition and morphology of both fibrous and nonfibrous amphiboles (double-chain silicates), verifying the previous results found by Wylie and Verkouteren (*3*) and Gunter et al. (*4*). The composition of the Libby amphiboles indicated the presence of both regulated (tremolite) and unregulated (primarily winchite and richterite) fibers (*2*), with the majority of the samples displaying morphologies between prismatic crystals and asbestiform fibers. These various fibers within Libby amphibole amphibole differ in their relative proportions of cations (Mg, Ca, Fe, Na, K). Results of the Meeker et al. (*2*) study also showed that (for the most part) all of the vermiculite samples produced amphibole fibers in a similar size range, and that the fibril diameter of the Vermiculite Mountain asbestiform amphibole ranged from approximately 0.1 to 1 μm, with approximately 40% of the fibers longer than 5 μm.

These amphibole fibers have caused a very high incidence of amphibole related diseases in not only the former mine and mill workers, but in the general Libby population (*5–8*). In addition to elevated rates of lung cancer and mesothelioma among Libby residents (*9, 10*), medical testing of persons who lived or worked in the Libby area for at least six months before 1991 showed pleural abnormalities (calcifications, thickenings, or plaques) in 17.8% of 6,668 participants (*11*). Occupational exposure to Libby vermiculite is also associated with significant increases in asbestosis, lung cancer, and pleural cancer compared to the rest of the U.S. population (*12*). A health consultation evaluating mortality in Libby from 1979 to 1998 revealed that when compared to Montana and U.S. mortality, there was a 20−40% increase in malignant and nonmalignant respiratory deaths. In addition, asbestosis mortality in Libby was 40−80 times higher than expected, and lung cancer mortality was 1.2−1.3 times higher than expected when compared to Montana and the United States (*10*).

Today, there is widespread amphibole fiber contamination within the soils in areas surrounding the vermiculite mine, much of which is an Environmental Protection Agency (EPA) Superfund site. In addition to the soils, the trees in proximity to the mine have been found to contain amphibole fibers, with substantial loadings in the outer layer of tree bark (*13*). Research has shown that fiber concentrations upward of 260 million fibers/cm^2 tree bark surface area have been measured in locations near the vermiculite mine site. All fibers found in these bark samples were typical of the Libby vermiculite amphibole, with standard elemental composition of Si > Mg > Ca > Fe > Na > K, mean length of ~3.5 μm, and width of ~0.4 μm (aspect ratio 3.5:0.4). Amphibole fibers likely came in contact with the bark surface through direct interception or impaction-type processes such as wind-blown dust, making the trees effective reservoirs for amphibole fibers.

The Libby population is heavily dependent on woodstoves for residential home heating throughout the cold winter months. Given that some of the forested areas surrounding the mine (and outside of the Superfund boundaries) are contaminated with amphibole as a result of 70 years of processing vermiculite in the area, we hypothesize that there is the potential for exposure when using these trees as a fuel source. In an effort to determine if Libby inhabitants are exposed to amphibole during the burning of contaminated wood, the first step was to evaluate the potential for exposure during the firewood harvesting process. In 2006, a study was conducted by investigators from the University of Montana and Montana Tech to assess the potential for amphibole exposure while harvesting firewood from contaminated trees. Results from this study showed that amphibole fibers are liberated into the air when disturbing contaminated tree bark (i.e., cutting and stacking firewood). Fibers were measured in the breathing zone of the investigators during the study,

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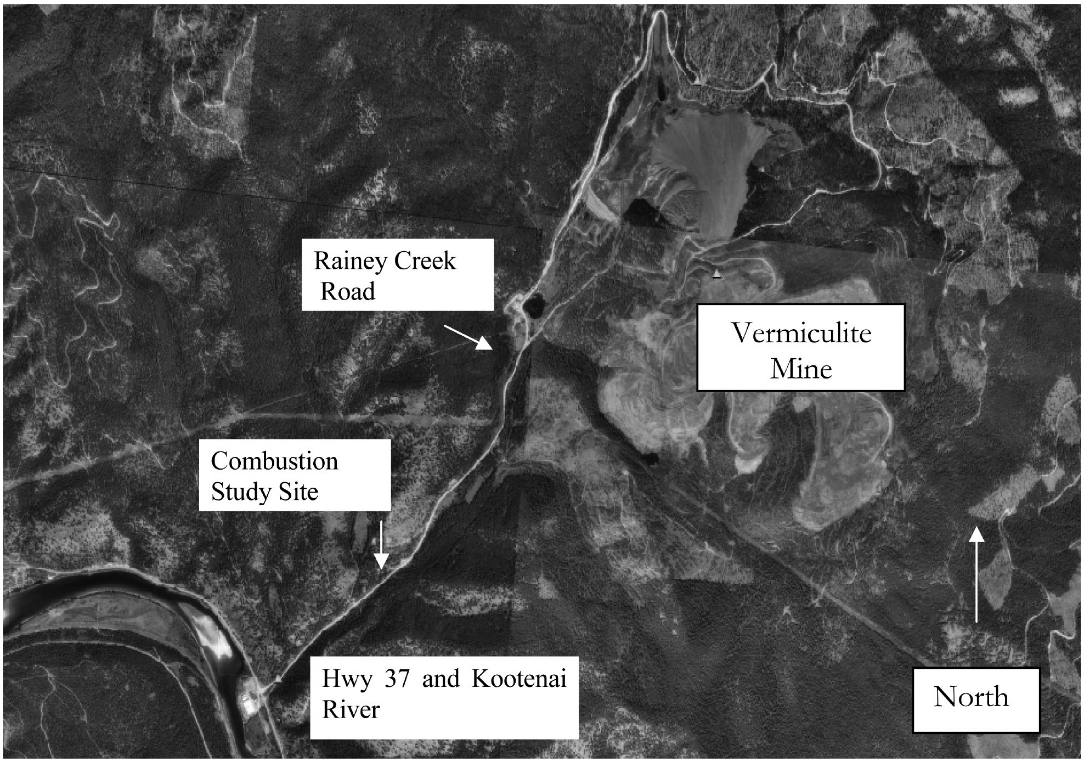

FIGURE 1. Combustion trial sites at the vermiculite mine in Libby, Montana.

with the majority of the amphibole fibers detected less than 5 μm in length. Perhaps more importantly, substantial amphibole fiber concentrations were also discovered on wipe samples collected from the Tyvek clothing worn by each of the investigators. Results from our firewood harvesting simulation suggest that not only is there an acute airborne exposure potential to Libby amphibole associated with disturbing contaminated trees, but fibers may also be transported on clothes into homes following the activity, potentially further exposing it is inhabitants (*14*).

After discovering that trees near the mine and other areas in and near Libby are contaminated with amphibole fibers (*13*), and that there is the potential for exposure during firewood harvesting activities in these areas (*14*), our next step was to determine what happens to the fibers when contaminated firewood is burned in woodstoves. In this manuscript, we present the results of a study where amphibole-contaminated firewood was burned in new, EPA-certified stoves to determine the fate of the fibers during residential home heating. We also present here an assessment of the potential for exposure when burning contaminated wood.

Experimental Section

In this study, EPA-certified woodstoves were used to combust amphibole-contaminated firewood to determine the fate of the fibers. Four separate trials were conducted during this project, including one trial in which noncontaminated wood was burned (control), and three trials where amphibole-contaminated firewood was burned within the Libby Superfund site. During each of the trials, multiple samples were collected. These included bulk samples of the ash remaining after each of the trials, wipe samples at varying points along the exhaust ductwork, and air samples of the smoke exhaust exiting the woodstoves. In addition, personal breathing zone (PBZ) samples were collected during each of the trials to assess personal exposures.

Each of the four trials was conducted during spring 2007. The control trial burn was conducted on private land near Missoula, Montana. The three Libby trials were performed outdoors on U.S. Forest Service (USFS) property inside the EPA restricted zone surrounding the mine in the Kootenai Forest. These trials, referred to as combustion trial A, combustion trial B, and combustion trial C, were conducted approximately 1.5 km (km) up Rainy Creek road from Highway 37 (see Figure 1) within the Superfund site.

The tree species used in this study are representative of the types of trees used for firewood fuel in Libby, and throughout western Montana. In addition, the same wood that was cut and stacked during our firewood harvesting study (*14*) was also used in the combustion study. For our control trial burn in Missoula, lodgepole pine (*Pinus contorta*) was utilized. For combustion trials A−C at the mine site, tamarack (*Larix laricina*), Douglas fir (*Pseudotsuga menziesii*), lodgepole pine (*Pinus contorta*), and ponderosa pine (*Pinus ponderosa*) wood were combusted in the stoves.

In this research project, EPA-certified stoves were utilized instead of older model, higher emission woodstoves. EPA-certified stoves are those that meet PM emissions limit of 7.5 g per hour (for noncatalytic stoves only) as certified by an independent laboratory. Beginning in 2004, a large woodstove changeout program was conducted in Libby in an effort to lower the ambient concentrations of $PM_{2.5}$ during the winter months. Prior to the revised 24-h National Ambient Air Quality Standards (NAAQS), Libby was the only nonattainment $PM_{2.5}$ airshed west of the Mississippi River, and outside of southern California. Results from a 2003/2004 source apportionment study showed that woodstoves were the main source of $PM_{2.5}$, contributing over 80% of the measured ambient fine fraction (*15*). As a result, nearly 1,200 old, polluting woodstoves were replaced with new, EPA-certified stoves. Today, the majority of the stoves in Libby are EPA-certified. In this study, three noncatalytic Quadra-Fire 2100 Millennium woodstoves (EPA-certified) were used in the combustion trials.

Three separate wood stoves were used for combustion trials A, B and C, respectively, in order to avoid cross-contamination. One of the stoves was used in both the control

FIGURE 2. Woodstove combustion study setup.

trial and for combustion trial C. Approximately 15 ft. of 6-in. diameter steel ductwork was attached to the stove outlet to simulate flue ductwork within a house (see Figure 2). The ductwork included a 90° elbow that was attached to the stove outlet (therefore extending the ductwork parallel to the ground), and then four sections (3 ft. long) of straight ductwork. This straight section was then connected to a second 90° elbow, which directed the smoke exhaust up into an aluminum foil-lined plastic tote used to concentrate the smoke for sample collection. Duct tape was used to seal all seams between sections of ductwork to prevent leakage.

During the combustion trials, researchers were equipped with personal protective equipment (PPE), including Tyvek suits and full-face air purifying respirators. The combustion duration of the control trial, combustion trial A, and combustion trial B ranged from 79 to 82 min. The duration of combustion trial C was 29 min, and was terminated earlier than the previous trials A and B to allow time for decontamination procedures during daylight hours. For each trial, approximately 1.6 kg of wood was burned. Temperatures inside the stove and ductwork (recorded with a Digi-Sense type K thermocoupler) had a mean stove temperature of 454 °C. The meteorological conditions during the combustion trials included cool temperatures (approximately mid-60s °F), no rain, and light winds.

Bulk ash samples were collected approximately 30 min after the conclusion of each trial, where a small metal shovel was used to collect approximately 600 g of ash from the bottom of each stove. The shovel was wiped with wet wipes after each sample collection to avoid cross-contamination. Bulk ash samples were analyzed by transmission electron microscopy (TEM) per EPA Method EPA/600/R-93/116 by DataChem Laboratories (Cincinnati, OH) (*16*). DataChem is accredited by the American Industrial Hygiene Association (AIHA) (PCM), the National Voluntary Laboratory Accreditation Program (NVLAP) (TEM), and the New York State Department of Health Environmental Laboratory Approval Program (PCM and TEM). Amphibole fibers in the ash samples were later verified at the Wadsworth Center using a Hitachi 7100 STEM interfaced to a PGT IMIX Image-Analyzer/X-Ray Detector. Fibers were identified using selected area electron diffraction (SAED) and energy dispersive X-ray (EDX).

Prior to the combustion process for each of the trials, precombustion wipe samples were collected inside the ductwork and the plastic tote to ensure that the inner surface areas were amphibole-free. At the conclusion of each combustion trial, wipe samples were again collected at five locations inside the combustion ductwork. These included (1) the first elbow exiting the stove, (2) the first 3 ft of ductwork past the first elbow, (3) the last 3 ft of ductwork past the first elbow, (4) the last elbow in the system, and (5) inside the plastic tote used to aggregate the emitted smoke. The wipe sampling protocol (DataChem Laboratories) followed the American Society for Testing and Materials (ASTM) D 6480-05 procedures, Wipe Sampling for Settled Amphibole (*17*). Wipes were collected with SKC Ghost wipes premoistened with deionized water and 5 mL of acetone. A 10 × 10 cm SKC disposable manila paper template was used for each wipe.

During each of the trials, smoke samples were collected from inside the tote. In addition, personal breathing zone (PBZ) samples were collected to evaluate the potential inhalation exposure associated with this research. SKC Aircheck 224 sampling pumps (calibrated before and after each trial with a Gilian Gilibrator 2 primary flow meter at a flow rate of 4 L per min) were used to collect air samples using nonconductive three-piece amphibole sampling cassettes containing 25 mm (mm) 0.8 μm (μm) pore size mixed cellulose ester membrane filters. Both the smoke and the PBZ air samples were analyzed by DataChem per National Institute for Occupational Safety and Health's Manual of Analytical Method (NMAM) 7400, Asbestos and Other Fibers by phase contrast microscopy (PCM) (*18*) and per EPA's Asbestos Hazard Emergency Response Act's (AHERA), Airborne Asbestos by TEM (*19*).

Results

In this study, we combusted the same firewood that was originally cut and stacked during our firewood harvesting simulation study (*14*). As part of this original study, tree bark samples were collected from standing dead or fallen trees

TABLE 1. Bulk Ash Sample Results Presented in Structure Counts and Structures Per Gram of Ash (s/g)

combustion trial	amphibole structures <5 μm	amphibole concentration <5 μm (s/g)	amphibole structures >5 μm	amphibole concentration >5 μm (s/g)
A	7[a]	136,670,640	0	<AS
B	5	84,004,8444	0	<AS
C	1	17,519,735	0	<AS
control	0	<AS[b]	0	<AS

[a] 1 of 7 fibers identified was a chrysotile fiber. [b] Analytical Sensitivities (AS) defined as one fiber per filter area analyzed. AS range= 14,219,577−19,524,377 s/g.

TABLE 2. Ratio of Ca + Fe Divided by Mg for Fibers in Bark (This Study) versus Ratio from Elutriated (Unashed) Libby Amphiboles (*22*) (All Elements Were k-Factor Corrected)

	(Ca + Fe)/Mg	
	ashed	unashed
mean	0.76	0.84
minimum	0.53	0.65
maximum	1.29	1.41

prior to harvesting, and analyzed for amphibole. As reported previously, amphibole fibers were not detected in bark samples collected from the Missoula trees (control). Results from the eight bark samples collected and analyzed from the trees used during both the firewood harvest study and combustion trials (Libby) showed significant amphibole fiber concentrations, ranging from 7 million to 97 million fibers/cm^2 of bark surface area. Fibers also exhibited mineral characteristics consistent with Libby amphiboles.

One bulk ash sample was analyzed from each of the combustion trials, with the results presented in Table 1. Libby amphiboles were not detected in the Missoula control sample. However, significant amphibole fiber concentrations (units in structures per gram of ash, s/g) were detected in the ash following each of the three combustion trials at the Libby Superfund site. All of the fibers identified were less than 5 μm in length. Gunter et al. (*20*) have reported that Libby amphiboles heated at 1000 °C for 10 min are converted to pyroxenes, and speculated that woodstove combustion might likewise convert amphibole fibers to pyroxene fibers. When amphiboles are converted to pyroxenes, Mg generally decreases while Ca and Fe increase (*21*). When the chemistry of the fibers in the bark ash is compared to the chemistry of (unashed) Libby amphiboles elutriated in an earlier study (*22*), no major differences are apparent (see Table 2). One explanation might be that fibers on bark are not exposed to conversion temperatures for long periods of time, if at all. As noted earlier, temperatures inside the stove had a mean stove temperature of 454 °C.

Wipe sampling results are presented in Table 3. Amphibole fibers were not detected in wipe samples collected during the Missoula control burn. In addition, no fibers were detected from wipes collected from within the ductwork prior to the combustion trials, establishing that precontamination was not an issue.

Results from the combustion trial A and B wipe samples showed that amphibole fibers were detected from within the ductwork and from within the tote following the burns. Given that amphibole fibers were not detected in precombustion wipe samples, these results suggest that amphibole fibers can be liberated into the ambient air when combusting amphibole-contaminated firewood. No amphibole fibers were detected in the wipes from the combustion trial C ductwork, likely due to the short duration of the trial (only 29 min). Consistent with the tree bark and the ash samples, the majority of the fibers detected from the wipe samples were shorter than 5 μm in length (though one fiber longer than 5 μm was detected in the last elbow in trials A and B). It should also be noted that amphibole fibers were detected in wipe samples only from the elbows and the plastic tote, and not from the straight sections of the ductwork. Amphibole fibers released from tree bark during the combustion process were most likely impacted or intercepted on the surface of the ductwork as the airflow pattern changed.

Results from the air (smoke) samples collected within the tote were less conclusive. Due to the volume of smoke inside the tote, an accurate measurement of the amphibole concentrations within the liberated smoke was not feasible. Even over a short duration of sample collection, the loading on the cartridges was significant, resulting in pump flow failures within minutes of the start of sample collection. Therefore, the data from these samples were not useable.

PBZ sample results were intended to evaluate the potential amphibole inhalation hazard associated with this research and to ensure that the appropriate PPE was selected for this study. Fibers were observed on all PBZ samples analyzed by PCM, excluding field blanks. The sample time weighted average PBZ exposures ranged from 0.02 to 0.08 fiber per mL when analyzed by PCM. AHERA TEM analyses were further performed to identify structure morphologies. In terms of fiber counts reported by the laboratory, more than 5 nonasbestos fibers (organic, gypsum) were identified on all PBZ AHERA TEM samples. AHERA TEM analyses for asbestos fibers <5 μm long revealed concentrations from less than the analytical sensitivity (AS, 0.004−0.02 structures per mL) to 0.05 structures per mL. AHERA TEM analyses for the concentration of asbestos fibers >5 μm revealed concentrations less than the analytical sensitivity for all of the PBZ samples. PBZ results should not be used to determine if amphibole fibers were released during the combustion trials, as it is impossible to distinguish between amphibole fibers liberated during the trials versus exposure to amphibole fibers from simply being within the Superfund site.

Discussion

Amphibole fibers have small aerodynamic diameters, and can remain airborne following liberation for hours or even days before settling into soil, sediment, or other indoor materials such as carpet. Since amphibole fibers are insoluble and do not decompose in the environment, the airborne amphibole fibers released and dispersed from the Libby mine and processing facilities over 70 years of operation have likely deposited and accumulated in areas surrounding point and area sources in Libby. Due to the fact that much of the areas surrounding the Vermiculite Mine are forested areas, it is a concern that some of these areas where the inhabitants of Libby harvest firewood are today heavily contaminated with amphibole.

In this manuscript, we present the results of a research study that determined what happens to amphibole fibers when contaminated firewood is burned within EPA-certified stoves. The results of this study revealed that the majority of the fibers remained in the ash following combustion, however, fibers did become liberated into the ambient air when combusting the amphibole-contaminated firewood. The majority of the amphibole fibers detected in the ash and wipe samples were all less than 5 μm long. These results are consistent with our findings from the original bark samples collected from the mine and within the town of Libby (*13*), as well as the samples (bark, PBZ, and wipe) collected during our firewood harvesting simulations (*14*).

The findings that the majority of the fibers remain in the ash suggest that disturbing the contaminated ash (as part of

TABLE 3. Wipe Sample Results from Combustion Trials A and B Presented in Structure Counts and Structures Per Square Centimeter (s/cm^2)

combustion trial	sample location	amphibole structures <5 μm	amphibole concentration <5 μm (s/cm^2)	amphibole structures >5 μm	amphibole concentration >5 μm s/cm^2)
A	tote	2	1,024	<AS	<AS
A	first elbow	<AS	<AS	<AS	<AS
A	last elbow	1	10,238	2	20,476
A	last section of ductwork	<AS	<AS	<AS	<AS
B	tote	1[a]	2,560	<AS	<AS
B	first elbow	2	20,476	<AS	<AS
B	last elbow	7	14,333	1	2,048
B	last section of ductwork	<AS	<AS	<AS	<AS

[a] Analytical Sensitivities (AS) defined as one fiber per filter area analyzed. AS range = 341−10,238 s/cm^2. One fiber identified was a chrysotile fiber.

routine cleaning of the stove) is an additional potential source of exposure within the home. Detecting amphibole in the elbows and tote (i.e., where the air stream was either redirected or concentrated) supports that fibers can become impacted or intercepted onto the ductwork during combustion. When burning firewood with low-level amphibole contamination over a long period of time, it should be considered that amphibole would be allowed to build up within the creosote inside the ductwork. This could have implications when cleaning and/or changing out flue ductwork. The findings from the tote wipe samples showed that amphibole fibers can be liberated into the ambient air, thereby re-entraining amphibole into the Libby airshed. As the trials were conducted with lower emission EPA-certified woodstoves, we feel that non EPA-certified stoves (higher emission stoves) would have an even greater potential for releasing amphibole fibers into the ductwork and/or ambient air.

Due to the nature and logistics of this research project, there were several limitations to this study. One limitation was the duration for each of the burns. The duration of the control trial, combustion trial A, and combustion trial B ranged from 79 to 82 min. Combustion trial C lasted for 29 min, and was cut short as a result of decreasing daylight. The shortened duration of the trial C burn could explain why no amphibole fibers were found in the ductwork or tote during this trial. For comparison, all of the trial burns were much shorter in duration than a typical burn used for residential home heating.

Another limitation of this study was that the amphibole concentrations in the contaminated firewood used in our combustion trials were likely much more elevated compared to concentrations used during "typical" home heating in Libby. The goal of this study was to determine the fate of fibers when combusting amphibole-contaminated firewood. The authors recognize that the combustion research presented in this study represents a near worst-case scenario, as the study was conducted on U.S. Forest Service land within the EPA restricted zone (Superfund site).

It is not known why chrysotile was detected in some of our samples in this study, as it is not known to be one of the prevalent fiber types in Libby. Of the seven amphibole fibers found in the combustion trial A bulk ash sample, one was a chrysotile fiber. One chrysotile fiber was also detected in the wipe samples collected from the tote in combustion trial B. Chrysotile asbestos has been used as an insulator in high temperature equipment such as wood burning stoves. The stoves were not disassembled prior to the study for the presence of asbestos insulation. If chrysotile was used as an insulator for these stoves the possible contamination from their assembly could account for the single fiber noted. Chrysotile was not detected in any of our prewipe samples from any of the ductwork, however we did not take pre-trial wipes from within the stoves. Another possibility could be laboratory contamination, although the contracted laboratory used for this study has a comprehensive Quality Assurance/Quality Control program in place, and all field blank samples revealed no asbestos contamination.

The results from this study should be viewed with caution. As the goal of this study was to determine the fate of amphibole fibers during the combustion process, the trials were conducted under worst-case scenario conditions (i.e., using firewood heavily contaminated with amphibole fibers). However, these results do show that the potential for amphibole exposure does exist when burning contaminated firewood, and stresses the need for identifying a clean fuel source for the inhabitants of Libby who rely on woodstoves as their primary source for home heating.

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