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Activity-based exposure assessments to Libby amphibole associated with public and occupational tasks in the Kootenai National Forest and weatherization of homes containing vermiculite attic insulation

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Libby Amphibole Activity-Based Exposure Assessments

Up to 80% of the world's supply of vermiculite was produced from a mine near Libby, MT. A common use for expanded vermiculite was loose fill insulation in attics and walls. Unfortunately, vermiculite from the Libby mine was contaminated with amphibole asbestos minerals referred to as Libby amphibole, LA. Fifteen years after the closure of the vermiculite mine, substantial (14 - 110 million structures/cm^2) LA contamination was discovered on the surface of tree bark in the forested areas surrounding the former mine.

Research was conducted to evaluate the potential for LA exposure associated with tree bark and vermiculite attic insulation (VAI) sources. The potential for airborne LA exposure and clothing contamination to Libby residents who harvest firewood for home heating and to United States Department of Agriculture Forest Service (USDA FS) employees who work in the in the Kootenai National Forest was evaluated through activity-based exposure assessments. In addition, research was conducted to evaluate the impact of weatherization activities in homes with VAI and/or other asbestos containing materials (ACM). Personal breathing zone (PBZ), high volume air and surface sampling were conducted throughout the weatherization of 37 homes.

Firewood harvest exposure assessments revealed a strong potential for exposure with LA detected in 100 % of the PBZ samples. The mean transmission electron microscopy (TEM) LA concentration for fibers $\geq$ 5 µm was 0.07 structures per mL (s/mL). Outside of the restricted zone, the PBZ samples from the FS occupational study revealed detectable LA in 25 % of the PBZ TEM samples. In addition to airborne exposure, LA was detected on wipe samples from all activities related to the firewood harvest and occupational assessment.

During the weatherization of homes containing VAI or other ACM, the majority (79% and 80% respectively) of high volume air and PBZ air samples did not reveal detectable concentrations of asbestos. However, airborne asbestos was detected in 76% of the homes. Airborne asbestos was detected during numerous weatherization measures, suggesting that weatherization practices as a whole, not single weatherization activities, may contribute to the disturbance and dispersal of asbestos fibers into the air.

While substantial cleanup of homes, yards, etc., has been conducted in Libby, MT, through Superfund activities, additional potential sources of LA exposure in this area, such as tree bark, are important to consider when assessing public and occupational health risks. Outside of Lincoln County, MT the number of homes and other structures containing VAI is unknown. The exposure potential associated with VAI, especially associated activities that may disturb this insulation, warrant attention from a U.S. public health standpoint as well.

Dedication

This thesis is dedicated to my sons, Zach and Eric. Thank you for your patience through the many days of being away from your activities while I worked on this research. May you never stop learning and remember to reach for the stars!

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Table of contents

List of Tables

List of Figures

Glossary of Terms

Term	
ACGIH	American Conference of Governmental Industrial Hygienists
ACM	Asbestos containing materials
AHERA	Asbestos Hazard Emergency Response Act
AIHA	American Industrial Hygiene Association
AR	Aspect ratio
ARD	Asbestos related diseases
AS	Analytical sensitivity
ATSDR	Agency for Toxic Substance and Disease Registry
DPHHS	Department of Public Health and Human Services
EDX	Energy dispersive x-ray analysis
f/mL	Fibers per milliliter of air
LA	Libby amphibole
LIHEAP	Low Income Home Energy Assistance Program
MCE	Mixed cellulose ester
MSHA	Mine Safety and Health Administration
NIOSH	National Institute for Occupational Safety and Health
NMAM	NIOSH Manual of Analytical Methods
NMRD	Non-malignant respiratory diseases
NVLAP	National Voluntary Laboratory Accreditation Program
OSHA	Occupational Safety and Health Administration
OU3	Libby Superfund Site Operable Unit 3
PBZ	Personal breathing zone
PCM	Phase contrast microscopy
PLM	Polarized light microscopy
RCC	Rainy Creek Complex
RfC	Reference Concentration
s/cm^2	Structures per square centimeter
s/gm	Structures per gram
SRM	Standard mortality rate
TEM	Transmission electron microscopy
TLV	Threshold limit value
TSI	Thermal system insulation
µm	Micron
USDA FS	United States Department of Agriculture Forest Service
VAI	Vermiculite Attic Insulation
WAP	Weatherization Assistance Program

1. Introduction

A vermiculite mine located seven miles northeast of Libby, MT (population ~2,700, with nearly 12,000 in the surrounding area) supplied up to 80% of the world's supply of vermiculite from the early 1900s through 1990 (USEPA, 2012a). Vermiculite expands or pops when heated, creating pockets of air that made the material suitable for use in building insulation and as a soil conditioner. Vermiculite from the Libby mine is contaminated with a toxic form of naturally-occurring fibrous and non-asbestiform amphibole in veins throughout the deposit (Pardee and Larsen, 1929). Approximately 30-40% of the amphiboles are asbestiform and include winchite, richterite, tremolite and magnesioriebeckite; differing in their relative proportions of cations (Mg, Ca, Fe, Na, K) (Meeker et al., 2003; Bandli et al., 2003; Gunter et al, 2003; Sanchez et a., 2008; Gunter and Sanchez, 2009). This amphibole mineral mixture is commonly referred to as Libby amphibole (LA).

Occupational exposure to LA is associated with significant increases in asbestosis, lung cancer, and pleural cancer compared to the rest of the U.S. population (Sullivan, 2007). High incidences of asbestos-related disease (ARD) have been reported in former mine and mill workers (McDonald, 1986; Amandus, 1987; Amandus and Wheeler, 1987). While ARD in the general Libby population has also been reported, risk associated with lower level exposures has not been as clearly defined. Current mortality figures for mesothelioma indicate one new case per year in Lincoln, County, MT (McDonald et al., 2004; Case, 2006; Whitehouse et al., 2008). Lincoln County has the third highest age-adjusted mesothelioma death rate in the nation with a rate of 56.1 per million population (NIOSH, 2008). In 2009, the U.S. Environmental Protection Agency (EPA) designated the town of Libby, Montana, a public health emergency (USEPA, 2009). This was the first and only time the EPA has made this determination under the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA).

Fifteen years after the closure of the vermiculite mine, substantial (14 - 110 million structures/cm^2) LA contamination was discovered on the surface of tree bark in the forested area surrounding the former mine (Ward et al., 2006; Webber et al., 2006). A comprehensive bark sampling program conducted by Region 8 EPA confirmed LA contamination on the bark surface (2.5 – 20 million structures/cm^2). Forest soil and duff samples were also collected at an approximate 5 foot radius from the same trees where the bark samples were collected. Libby amphibole asbestos was detected in a number of soil samples located relatively close to the former mine area, but was not detectable > approximately 2 miles from mine, whereas LA contamination on tree bark surfaces extended miles (>4) from the mine in all directions (USEPA, 2008a). Preliminary data revealed that LA was detected in duff samples as well; however, the extent of this contamination, in terms of distance from the former mine is not yet available (USEPA, 2008b; USEPA, 2008c). The tree bark LA findings are significant as this contamination extends miles beyond the original EPA Superfund Restricted Zone for Operable Unit 3 (OU3). The source of the LA observed at these locations is unknown, but has been postulated to include "a) naturally occurring outcrops of the LA-bearing ore body, b) deposition from historic airborne releases from the mine and mill, and c) water-based erosion from past and/or present materials at the mine site" (USEPA, 2008c).

According to USEPA (2011a), LA inhalation exposure potentials in OU3 include ambient air, and airborne emissions of LA from roadways, soil or duff, as well as tree bark sources. Considering these exposure potentials, a range of different human receptors that may potentially be exposed to LA in the OU3 forested area include commercial loggers, United States Department of Agriculture Forest Service (USDA-FS) employees, recreational visitors engaging in activities such as hiking, dirt bike or all-terrain vehicle riding, hunting, etc., and residential wood harvesters (USEPA, 2011a).

The majority of the land surrounding the former vermiculite mine is owned by the USDA FS and private logging companies. Timber sales have resulted in extensive logging within this area confirmed by interviews with FS personnel and Google Earth (Google.com) images. Forest Service personnel frequently travel on roadways and trails in this Kootenai forest region as a component of their daily activities. In addition, 33% of Libby households rely on wood burning as their primary source of heat (Ganesan et al., 2006). Prior to the research studies (described in this thesis), there had been no public or occupational activity-based exposure assessments associated with amphibole asbestos tree bark contamination in the Kootenai National Forest.

The shipment of vermiculite concentrate to exfoliation plants across the U.S. has extended the LA exposure potential beyond the borders of Lincoln County, MT. The precise number of U.S. homes insulated with Zonolite brand (from the Libby mine) vermiculite attic insulation (VAI) is unknown (Gunter et al., 2005; USEPA, 2011b; Zalac, 2003); however, vermiculite was widely distributed via processing plants throughout the country and may be present in millions of homes nationwide, including thousands of homes in MT (USEPA, 2011b). The fact that a very large estimated number of homes were insulated with Libby vermiculite represents an ongoing potential public health concern. As a component of the cleanup efforts under the Superfund program, The USEPA has removed VAI from hundreds of Lincoln county homes (USEPA, 2012a). This cleanup effort has not extended into other Montana counties and currently there are limited assistance programs for homeowners living in structures containing VAI outside of the Libby Superfund site.

In addition to VAI, many older homes contain serpentine asbestos in commercial products such as thermal insulation, floor tiles, roofing tiles or shingles, and siding (Dobson and Hammer, 2006). In the state of Montana, the Department of Public Health and Human Services, the Low Income Home Energy Assistance Program (LIHEAP), and the Weatherization Assistance Program participate in grant funded weatherization activities with the goal of increasing the energy efficiency of homes that meet various program qualification guidelines. An estimated 1,500 to 2,000 qualified homes per year are weatherized throughout the state. Unfortunately, weatherization services are denied to approximately 200 high-energy-burden LIHEAP recipient households due to the presence of asbestos containing materials (ACM) in their homes, either as loose-fill insulation in attics, in pipe or duct insulation, or in certain wall, ceiling and siding materials. Because of potential health and safety hazards to residents and agency workers, Department of Energy weatherization rules prevent agencies from weatherizing homes with VAI, or with other ACM that are friable or brittle and could potentially become airborne.

1.1. Research Objectives

The primary objective of this research was to evaluate the potential for LA exposure associated with tree bark and VAI sources. Research Aim 1 evaluated the hypothesis of the potential for airborne asbestos exposure and clothing contamination to Libby residents who harvest firewood for home heating in the Kootenai National Forest. Research Aim 2 transitioned into the occupational setting with an evaluation of the hypothesis that there was a potential for amphibole asbestos exposure to USDA FS employees while performing common day-to-day activities in the Kootenai National Forest. Research Aim 3 evaluated potential living space contamination and weatherization worker exposure through fiber dispersal resulting from weatherization activities in asbestos containing homes. The hypothesis for Research Aim 3 was that weatherization activities in homes with VAI and/or other ACM would result in detectable concentrations of amphibole asbestos in airborne or surface dust samples. A secondary component of Aim 3 was the development of asbestos-safe weatherization protocols. These research aims are presented in Section 5 (subsections 5.1, 5.2 and 5.3) of this dissertation, with refereed publications for Aims 1 and 2 provided in Appendix A.

2. Background

2.1. Libby Mining History

Mining history in Lincoln County, Montana dates back to the 1860s when early explorers prospected in the Libby district. This area was noted for its long-lived placer operations as well as several silver-lead mines. The largest of these was the Snowshoe

Mine which produced ore rich in lead, silver and gold from the 1890s to 1912 (LibbyMT, 2004).

An additional ore deposit, vermiculite, was also discovered seven miles northeast of Libby in the Rainy Creek complex (RCC) by gold miners. The dates pertaining to this original discovery are conflicting in literature and range from 1881 to 1913 (Grace, 2012; USEPA, 2013; Pardee and Larson, 1929). Edward N. Alley, a Libby businessman, is credited as the first individual to exploit vermiculite from the RCC in 1923 (Kriegel, 1940; Bandli and Gunter, 2006). The method by which Alley acquired an initial vermiculite sample has been disputed by historians (Peacock, 2003). Some reports describe Henry Brink, a local politician, providing a sample of a “rotten mica” like substance to Alley, who later acquired a mineral claim for the sample collection area (Peacock, 2003). Other accounts describe Alley holding his miner’s candle close to a wall to collect an ore sample while prospecting for vanadium minerals during World War I in the RCC, when the material swelled and assumed a golden color (Kiegel, 1940; Peacock, 2003; LibbyMT, 2004; Bandli and Gunter, 2006). Due to this unique property of expanding when heated, numerous uses were developed for vermiculite in the early to mid-20th century (Gooch, 1957). While additional vermiculite deposits were developed in Colorado prior to the Libby mine, the Libby mine became the largest producer.

Mr. Alley and his associates formed one of the earliest commercial operations of the RCC vermiculite deposits, the Zonolite Company. In 1925, the Company included a mine and plant at Libby and a plant and research laboratory in Detroit, Mich. (Kriegel, 1940). In addition to the Zonolite Company, the Vermiculite and Asbestos Company of Libby, Mont., operated properties on the west end of the deposits (Kriegel, 1940). This company was acquired by the Universal Insulation Company in 1934. The Universal Insulation Company had a distribution system similar to the Zonolite Company, with offices and a laboratory in Chicago, Ill. In 1939, the Zonolite and Universal Insulation Companies merged and were called the Universal Zonolite Company (Kriegel, 1940). In 1963, W.R. Grace Co. purchased the Universal Zonolite Company and continued to mine vermiculite until 1990, when the mine closed (Grace, 2012; USEPA, 2012a). It is estimated that the Libby mine produced up to 80% of the world's supply of vermiculite (USEPA, 2012a).

2.2. Libby Vermiculite Geology and Mineralogy

The Libby vermiculite mine was located in the Rainy Creek alkaline-ultramafic igneous complex (RCC) (Boettcher, 1966a; Meeker et al., 2003). The geology and mineralogy of the RCC has been described by numerous individuals; Pardee and Larson (1929), Kreigel (1940), Bassett (1959), Boettcher (1966a, 1966b), while Bandli and Gunter (2006) have provided a more recent literature review with updated geological terms.

Igneous complex rocks were formed by the Precambrian (Wallace Formation) intrusion into argillite, limestone, and dolstone (Boettcher, 1967; Bandli and Gunter, 2006). The main body of this complex is composed of biotite pyroxenite (20% of the intrusion), magnetite pyroxenite (40% of the intrusion), and biotitite (5% of the intrusion), with the remainder of the complex (approximately 35%) comprised of various alkaline rocks.

Altered biotite in the biotitite pyroxenite was the source of all mineable vermiculite and comprises approximately 40% of the biotite pyroxenite (Bandli and Gunter, 2006). Modification of biotite in the biotite pyroxenite resulted in the formation of vermiculite and hydrobiotite (Boettcher, 1966b; Bandli and Gunter, 2006). The chemistry of vermiculite in the RCC reveals that it was the result of leaching of biotite by ground water (Boettcher, 1966b), a process commonly described as a low temperature weathering (Bassett, 1959; Boettcher, 1966b). Hydrobiotite was formed, not by weathering, but due to a much higher temperature hydrothermal alteration (Boettcher, 1966b). While vermiculite and hydrobiotite have similar origins, vermiculite will expand upon heating, while biotites will not (Bandli and Gunter, 2006). Early miners relied on subtle color differences to distinguish vermiculite (neutral gray) from hydrobiotite (light brown) or biotite (gray-green) in selecting ore to avoid unaltered biotite (Bassett, 1959; Bandli and Gunter, 2006). The vermiculite content of vermiculite ore (primarily biotite, vermiculite and biotite) varies and was reported by Pardee and Larson (1928) as 30 to 84 percent.

Of primary interest from a human health perspective in the RCC are amphiboles, an additional mineral group formed through hydrothermal alteration processes of pyroxenes in the biotite pyroxenite, (Boettcher, 1966b; Bandli and Gunter, 2006). These amphiboles were described by Bassett (1959) as thin (approximately one inch) white veins that cut through the pyroxenite. Meeker et al. (2003) reported that amphiboles were the most abundant vein product, as well as a primary component of dikes and associated wall-rock alteration zones ranging in width from a few millimeters to meters. The amphibole content in these alteration zones of the deposit were estimated as 50 to 70% (Pardee and Larson, 1929).

Early geologists considered the amphibole component within the Libby vermiculite deposit as an accessory mineral; therefore, it was not studied in depth until the 21st century (Meeker et al., 2003; Bandli and Gunter, 2006). However, due to the health effects associated with LA asbestos, substantial work has been performed in an effort to characterize this mineral. This research is summarized in Section 2.5 of this thesis.

2.3. Libby Vermiculite Exfoliation

While numerous mineral varieties of vermiculite exist, its most distinguishable feature is its ability to expand when heated (Kriegel, 1940; Gooch, 1957). When heated to temperatures of 1,500 to 2,000 degrees Fahrenheit, the water present in the vermiculite structure rapidly vaporizes into steam (Kriegel, 1940; Bandli and Gunter, 2006). This forces layers of the sheet silicate apart, resulting in low density (< 6 lb/ft^3) product that expands 6 to 20 times its original size (Kriegel, 1940; Gooch, 1957). This expansion process is referred to as “exfoliation” or “popping.”

The primary commercial product produced from the Libby mining operation was vermiculite concentrate. Ore was mined via a truck/shovel operation and transported to a screening area where coarse fractions were removed (USEPA, 2001). The ore was then concentrated (beneficiation) and screened into five size ranges or grades (Atkinson et al. 1982). A portion of the vermiculite concentrate was sent to an exfoliating and export plant in Libby and either used locally or packaged and shipped elsewhere (USEPA, 2001). The majority of the vermiculite concentrate was shipped by rail to expansion facilities across the United States (USEPA, 2001). From 1964-1990, over six million tons of vermiculite concentrate were shipped to over 200 expansion facilities (ATSDR, 2008).

2.4. Commercial Uses for Vermiculite

Expanded vermiculite was used in numerous commercial products throughout the 20th century. The largest size or grade 1 was used as a loose-fill insulation (attic and wall cavities) in dwellings and buildings and in refrigerators and furnaces (Keigel, 1940). In addition, expanded vermiculite has been used in potting soils and other horticultural products (Potter, 1997, USEPA, 2000). Other uses include an aggregate for insulation plasters, manufacture of fire-resistant, insulating wall-boards and acoustic tile. Very fine sizes were reported to be used as extenders in gold and bronze paints and inks due to their silver to gold colors that were produced upon expansion (Keigel, 1940).

Commercial names for expanded vermiculite include Unifil, Porosil, Zonolite®, and Monokote® (Keigel, 1940; Bandli and Gunter, 2006).

2.5. Mineralogy of Libby Amphibole

The amphibole minerals within the RCC have been referred to under a variety of names. They were initially classified as tremolite, tremolite/actinolite, or soda-rich tremolite by early geologists (Pardee and Larsen, 1929; Bassett, 1959; Boettcher, 1966b), with Larsen (1942) and Deer et al., (1963), further characterizing the amphibole mineral as richterite. Langer et al. (1991) and Nolan et al. (1991) classified the RCC amphibole as tremolite and richterite, while Wylie and Verkouteren (2000) and Gunter et al. (2003) identified the RCC amphiboles as primarily winchite (once considered a subset of richterite). Wylie and Verkouteren (2000) further postulated that the amphibole composition may range from winchite to richerite.

An extensive systemic evaluation of the RCC amphibole minerals was conducted by Meeker et al. (2003) which included 30 sample locations from the former mine area. Analytical techniques to characterize composition, mineralogy, and morphology of both fibrous and non-fibrous components of RCC amphiboles included X-ray diffraction, electron probe microanalysis using wavelength dispersive spectroscopy, and scanning electron microscopy combined with energy dispersive X-ray analysis, respectively. Amphiboles were classified based on the Leake et al. (1997) system which is based on site assignments for each cation in the structure, including the oxidation state of iron. Meeker et al. (2003) approximated the respirable fraction of RCC amphiboles as winchite (84%), richterite (11%) and tremolite (6%), with possible magnesioriebecktite, edenite, and magnesio-arfvedsonite components. Meeker et al. (2003) further reported that the Vermiculite Mountain amphibole minerals displayed a range of morphologies from prismatic to asbestiform, with fibril diameters ranging from 0.1 to 1 µm.

The discrepancy in the RCC amphibole mineral classification may be due to several factors. These include 1) amphiboles were viewed as a secondary mineral by early geologists and received little attention (Bandli and Gunter, 2006), 2) there have been modifications in the International Mineralogical Association classification systems (Wylie and Verkouteren, 2000), 3) naming of amphibole species is complex because of the variations in chemistry and the substitutions that occur in this mineral group (Gunter et al. 2003), 4) the optical properties of winchite from the RCC are very similar to tremolite (Bandli and Gunter, 2006), and 5) many techniques and methods available for analysis and classification of asbestos are not capable of adequately identifying or distinguishing these minerals according to current International Mineralogical Association guidelines (Meeker et al., 2003).

2.6. Amphibole Occurrence in Vermiculite

An EPA sponsored study (Atkinson et al. 1982) was conducted to determine the amount of asbestiform minerals in vermiculite from three U.S. mining operations; Zonolite Mountain and two South Carolina mines. The estimated content of LA (referred to as tremolite/actinolite) in Libby raw ore at the head feed was reported as 21-26% (by weight) and in processed ore (beneficiated and sized) was reported as 0.3 to 7 % (by weight) (Atkinson et al. 1982). Amphibole asbestos, reported as temolite/actinolite and anthophyllite, was also detected in South Carolina processed and unprocessed vermiculite, but in “substantially lower” concentrations (<1% by weight) for the fibrous phases (Atkinson et al. 1982). Amandus et al. (1987) reported LA concentrations from internal company (W.R. Grace Co.) sampling in raw ore at the head feed and concentrate as 3.5 – 6.4 % and 0.4 – 1.0 % (by weight), respectively.

The amphibole content has also been studied in expanded vermiculite. Moatamed et al. (1986) reported amphibole asbestos concentrations of 0.8 – 2 % and 0.6% for two samples of grade 2 and 3 unexpanded and expanded Libby vermiculite, respectively. A study conducted by the USEPA (2000) revealed asbestos concentrations of 0 to 2.8 % (by weight) in Zonolite brand expanded vermiculite sold in Seattle area retail stores for lawn and garden care use.

Bulk vermiculite samples collected from attic spaces as a component of attic insulation disturbance based studies have revealed amphibole asbestos concentrations from non-detect to 10% (Cowan, 1997; USEPA, 2003A; Ewing et al., 2010: Spear et al., 2012). Forty Montana homes (outside of Lincoln County, MT) containing VAI all revealed the presence of LA asbestos in bulk samples (Spear et al., 2012). These studies are discussed in more detail in Section 5.3 of this dissertation.

2.7. Asbestos Exposure Limits

The earliest account of asbestos related health hazards was reported by Lucy Deane, one of first Women Inspectors of Factories in the United Kingdom (Gee and Greenberg, 2002). Deane included asbestos work as one of the four dusty occupations under investigation in 1898 due to "injury to bronchial tubes and lungs medically attributed to employment" (Deane, 1898 as reported by Gee and Greenberg, 2002). The term pulmonary asbestosis was first used in 1927 by W.E Cooke to describe the fibrotic lung disease caused by inhalation of asbestos fibers (Cooke, 1927). Even prior to this, an early account linking asbestos with pulmonary injury was a statement made by the chief actuary of the Prudential Life Insurance Company in 1918: "In the practice of American and Canadian life insurance companies, asbestos workers are generally declined on account of the assumed health-injurious conditions of the industry" (Dodson and Hammer, 2006; Michaels, 2008). Twenty years later, an epidemiological study of three asbestos textile plants in North Carolina revealed that 68 % of the workers in the higher asbestos exposure group (5 – 10 million particles per cubic foot) had asbestosis (Dreessen et al., 1938; Michaels, 2008). This led to the first federally proposed recommended exposure limit to asbestos of 5 million particles per cubic foot. Although these early historical reports existed, the Occupational Safety and Health Act, Federal Mine Safety and Health Act and associated Occupational Safety and Health Administration (OSHA) and Mine Safety and Health Administration (MSHA) were not created until decades later (USDOL, 1970; MSHA, N.D.).

The American Conference of Governmental Hygienists (ACGIH) was the first to establish an occupational exposure limit for asbestos (Table 1.) (ACGIH, 2001). It is important to note, however, that ACGIH is not, and was not at the time of this first asbestos exposure limit, a regulatory agency. The Occupational Safety and Health Administration (OSHA) published the first general industry U.S. regulatory occupational asbestos standard in 1971 (Table 1.) (OSHA, 1994a).

The current National Institute for Occupational Safety and Health (NIOSH), OSHA, and ACGIH time weighted average (TWA) occupational exposure limit of 0.1 fiber/mL is specific to chrysotile and the asbestiform habit of five amphiboles: amosite, crocidolite, tremolite, anthophyllite, and actinolite (ACGIH, 2001; CDC, 2010; OSHA, 1994b). In addition, United States EPA regulations pertaining to asbestos also specify these minerals as regulated varieties (Title 40, CFR, Part 61 and Part 763).

Table I: Chronology of U.S. Asbestos Exposure Limits

	Year	**Exposure Limit** particles per cubic foot (p/ft^3) fibers per milliliter (f/mL)
ACGIH	1946	5×10^6 p/ft^3
ACGIH	1968[a]	12 f/mL or 10^4 p/ft^3
ACGIH	1970[a] 1974[b]	5 f/mL
OSHA	1971	12 f/mL
OSHA	1972	5 f/mL
NIOSH	1976	0.1 f/mL
MSHA	1977	2 f/mL
ACGIH	1978[a] 1980[b]	0.2 f/mL crocidolite 0.5 f/mL amosite 2.0 f/mL chrysotile and other forms
OSHA	1986	0.2 f/mL
OSHA	1994	0.1 f/mL
ACGIH	1997[b] 1998[c]	0.1 f/mL

[a] Notice of intent
[b] Adopted as threshold limit value (TLV)

In the documentation of the threshold limit value (TLV) for asbestos, the ACGIH (2001) defined asbestos in two ways. The first referred to the Campbell et al. (1977) mineralogical definition of "a collective mineralogical term encompassing the asbestiform varieties of various minerals", while the second definition was "an industrial product obtained by mining and processing primarily asbestiform minerals." However, ACGIH (2001) further clarified that "for the purpose of considering a TLV recommendation for asbestos dust in the workplace, only the second definition is possible". At the time guidelines and regulations were written, exposure limits were established for minerals known to have been mined commercially as asbestos.

Considering that the amphibole habit from the RCC are not listed in these exposure limits, with the exception of tremolite, and that exposure to these amphiboles has presented a substantial health threat, suggestions have been presented by several mineralogists to revise the regulated asbestos language to include the broader definition of “asbestiform amphiboles” (Wylie and Verkouteren, 2000; Meeker et al., 2003; Bandli and Gunter, 2006).

In addition to the occupational exposure limits specifying mineral species, counting rules for asbestos apply when comparing air concentrations to occupational exposure limits. Fibers equal to or longer than 5 microns (µm) with a length-to-width ratio (aspect ratio) (AR) of 3:1 or greater are counted (ACGIH, 2001; CDC, 2010; OSHA, 1994b). This counting rule has been questioned by epidemiologists and others in the environmental health community (Dodson et al., 2003; Stayner et al., 2008).

Stayner et al. (2008) emphasized that the counting rule was based largely on accuracy and reproducibility limitations associated with phase contrast microscopy (PCM) counting versus a toxicological basis. Libby amphibole studies which revealed similar inflammatory potencies in respirable size fractioned and non-size fractioned LA, as discussed in section 3.2 of this thesis, strengthen this discussion (Duncan et al., 2010).

A common toxicological justification for the counting rule is that short fibers are cleared more readily from the lungs (Dodson et al., 2003) and that longer fibers impair the phagocytic process (Stanton et al., 1981). Longer fibers have a greater potential than short fibers to generate an inflammatory response and stimulate release of Il-1B from macrophages (Kane, 1992; Donaldson et al., 2010, Palomaki et al., 2011). However, as in any toxicological assessment, the dose and dosing frequency are critical factors to consider in the long versus short fiber toxicity discussion (Kane et al., 1992; Castranova et al., 2000, and Dodson et al., 2003).

In the Dodson et al. (2003) review of fiber length and pathogenicity, the conclusions drawn from Castranova et al. (2000), of “constant infusions of short fibers and a resultant eventual dust overload, can greatly compromise clearance" was cited as the main reason to underscore the short fiber clearance reasoning. A similar hypothesis regarding particle overload and the potential for short crocidolite asbestos fibers to generate substantial inflammatory responses was discussed by Kane (1992). Dodson et al. (2003) further emphasized that when appropriate techniques are used to analyze asbestos fiber tissue burden, in most tissues, a substantial majority of asbestos fibers are less than 5 µm in length. These observations may be due to increased deposition of shorter fibers and/or breaking of longer fibers over time.

Additional counting rules other than those specified by OSHA are used for ambient and indoor asbestos monitoring to provide a more detailed quantification of asbestos structures. Two that have been used in studies assessing exposure to LA are the Asbestos Hazard Emergency Response Act (AHERA) and International Standards Organization 10312 methods (AHERA, 1987; ISO, 1995). The AHERA method was derived for clearance sampling in school buildings following asbestos abatement. Under the AHERA method, an asbestos fiber is defined as a structure greater than or equivalent to 0.5 µm in length and a diameter > 0.002 µm with an AR of 5:1 or greater. Fibers are classified as 0.5 – 5 µm and > 5 µm in length (AHERA, 1987). The ISO 10312 method applies the same minimum length and diameter criteria as AHERA, however, 3:1 or 5:1 AR may be used. From an ISO 10312 analysis, several different airborne asbestos structure concentration values based on a number of fiber size classifications may be obtained (ISO, 1995).

Analytical techniques that count only fibers greater than 5 µm may substantially under report inhalation exposures. Fiber lengths reported for LA range from less than 1 µm to greater than 20 µm with thicknesses ranging from 0.1 to 1 µm. If PCM counting rules are applied to LA, only one third of the fibers observed would be counted (Weis, 2001). Because the health effects associated with asbestos are not confined to fibers in the regulatory size fraction of greater than 5 µm, AHERA counting rules were employed for the research Aims discussed in section 5 of this thesis. In order to provide meaningful measurements of exposure, it is important to thoroughly characterize the fiber concentration and morphology and not limit this characterization to a counting rule that exists primarily because of an analytical method limitation.

2.8. Libby Amphibole Proposed Reference Concentration

In addition to the occupational exposure limits described in the previous section, as of the date of this thesis, a reference concentration (RfC) for non-cancer risk models has been proposed for LA. This is significant, in that there are no other proposed or published RfCs for other asbestos minerals. The proposed RfC of 2×10^{-5} f/mL was based on exposure-response data for the relationship between LA exposure and the risk of localized pleural thickening derived from a Marysville, OH worker cohort data set. The Marysville cohort was selected because the workers were exposed to lower LA concentrations relative to Libby cohorts and workers showered and changed at the conclusion of the work shift, resulting in minimal non-occupational exposures.

Epidemiology studies from the Marysville cohort are discussed further in section 3.1 of this thesis. In summary, the prevalence of pleural abnormalities for this cohort was 2% in 1984 (Lockey et al., 1984) and 28.7% in 2004 (Rohs et al., 2008). This increase in prevalence is most likely due to the additional time for the abnormalities to be detected with conventional X-ray techniques. Twenty % of participants with low lifetime cumulative fiber exposures of 2.21 fiber/cc years revealed pleural changes. The Rohs et al. (2007) study also revealed a significant ($p < 0.001$) exposure-response relationship with increasing cumulative exposure to LA. Pulmonary function testing was not reported in the Rohs et al. (2007) study. However, documentation for the RfC reported the radiographic classification of localized pleural thickening to include "pleural lesions associated with chronic chest pain, decreased lung volume, and decreased measures of lung function. Therefore, localized pleural thickening was considered an adverse effect and an endpoint adequate for RfC derivation."

The RfC is derived from the lower 95% confidence limit of the lifetime benchmark concentration level divided by an uncertainty factor of 100. The uncertainty factor of 100 is based on an intraspecies uncertainty factor of 10, applied to account for human variability and potentially susceptible individuals and a database uncertainty factor of 10, applied to account for database deficiencies in available literature.

It must be stressed that the proposed RfC for Libby amphibole is currently under review and "it does not represent and should not be construed to represent any USEPA determination or policy. The final RfC, if adopted, will assist USEPA in conducting human health risk assessments to evaluate the potential health risks from exposure to Libby Amphibole asbestos (LA) through possible exposure from a variety of daily activities. Data from the risk assessment will be also used, in part, to make cleanup decisions for Libby" (USEPA.2012b).

2.9. Asbestos Analytical Techniques

Having accurate techniques for measuring asbestos levels is critical in determining the extent of asbestos contamination and the health risks for humans. There are a number of microscopy techniques for asbestos detection that have been developed. The most important and widely used microscopy techniques are phase contrast microscopy (PCM), transmission electron microscopy (TEM), and polarized light microscopy (PLM).

2.9.1. Phase Contrast Microscopy

Phase contrast microscopy (PCM) is an optical microscopy analytical technique used to measure asbestos levels in air. Regulations issued by OSHA require the use of PCM to determine indoor asbestos air levels for occupational settings to ensure a safe working environment. PCM uses a compound light microscope to illuminate the fibers with a hollow cone of light. The lens induces a phase shift of a wavelength of light that causes minute variations of the refractive index of the specimen. The magnification is 400 times. The change in the phase contrast allows fibers as thin as 0.25 μm in diameter to become visible but prevents fiber identification. Therefore, PCM is used to identify fibers, but cannot distinguish between asbestos fibers and non asbestos fibers. Only fibers that are greater than 5 μm in length and have an aspect ratio of 3:1 or greater are counted in this method (Dodson and Hammar, 2006). Air sample analytical techniques that utilize PCM methods include NIOSH 7400, asbestos by PCM where samples are mounted on a slide, immersed in acetone and counted to yield total fiber counts per sample (NIOSH, 1994a).

The advantages of the PCM method for determining asbestos in air is that it is inexpensive and analysis can be performed on-site (DeMalo, 2004), which makes it a convenient technique for monitoring asbestos exposure in the workplace. Also, PCM has been used in historical epidemiological studies (OSHA, 1997) so the results from a PCM analysis can be compared to health studies used to estimate the risk of acquiring an asbestos-related disease (Chesson et al., 1990; Verma and Clark, 1995). This makes the results from a PCM analysis more applicable in assessing risk than a transmission electron microscopy (TEM) or scanning electron microscopy (SEM) analysis.

The main disadvantage with PCM is that it cannot distinguish between asbestos and non-asbestos fibers, which causes great uncertainty about the actual asbestos fiber concentration for a given area. Another disadvantage of PCM, compared to TEM, is its lower resolution. PCM analysis misses many smaller fibers during fiber counting that can be quantified using other techniques (OSHA, 1997; NIOSH, 1994a; Mossman et al., 1990; Verma and Clark, 1995; Karaffa et al., 1987). Using PCM, the smallest fibers that are visible have diameters of about 0.20 to 0.25 μm (OSHA, 1997; NIOSH, 1994a; Harper and Bartolucci, 2003; Karaffa et al., 1987) or 0.3 μm (Verma and Clark, 1995), while the finest asbestos fibers may have diameters as small as 0.02 μm (OSHA, 1997; NIOSH, 1994a). Because of its poor resolution, PCM can result in a significant underestimation of the asbestos fiber concentration in air.

2.9.2. Transmission Electron Microscopy

Transmission electron microscopy (TEM) is used as an analytical technique for air and surface samples when specific identification of individual asbestos fibers is required. This technique relies on electron microscopy rather than optical microscopy. TEM uses electromagnetic coils as lenses to form magnified images from an electron beam to form images. TEM allows for magnification of about 100,000 with a resolution greater than 10 nm. Fibers as small as 0.02 μm in diameter can be identified. TEM classifies fibers as non-asbestos or asbestos, identifies fiber morphology (type of asbestos), and reports the concentration of structures (Dodson and Hammar, 2006). Air sample analytical techniques that utilize TEM methods include NIOSH 7402, asbestos by TEM and EPA AHERA (NIOSH, 1994b). Surface sample analytical techniques that utilize TEM analysis include ASTM D 6480-05 (ASTM, 2006) and ASTM D 5755-03 (ASTM, 2003).

TEM is considered a superior technique to PCM for several reasons. First, transmission electron microscopes have greater resolution and thus can better detect smaller fibers (Mossman, et al., 1990; Kauffer et al., 1996; Karaffa et al., 1987) and better examine a particulate's morphology. Secondly, TEM methods for analyzing airborne asbestos use EDXA to determine the elemental makeup of a fiber, which enables this technique to be able to determine if a fiber possesses a chemical composition characteristic of asbestos or not (DeMalo, 2004) (USEPA, 1987).

2.9.3. Polarized Light Microscopy

Bulk samples of suspect ACM are commonly analyzed by polarized light microscopy (PLM). PLM utilizes a compound light microscope containing a polarized material in the light path below the sample and another in the light path above the sample to identify the fibers among the binders and fillers. Bulk analysis of asbestos using PLM methods involve identifying the type of asbestos present based on optical properties and then estimating the relative amount of asbestos in relation to the rest of the sample PLM identification of asbestos fibers is limited to fibers approximately1 μm in diameter (Dodson and Hammar, 2006).

Polarized light microscopy is frequently used for determining the asbestos content of bulk samples of insulation or other building materials (NIOSH Method 9002 [NIOSH, 1989] and OSHA method ID-191 [OSHA, 1994]). This method also enables qualitative identification of asbestos types using morphology, color, and refractive index.

3. Libby Amphibole Health Studies

Exposure to LA is associated with nonmalignant and malignant ARDs including; asbestosis, lung cancer, mesothelioma and pleural plaques (McDonald, 1986; Amandus et al., 1987; Amandus and Wheeler, 1987; McDonald et al., 2004; Case, 2006; Sullivan, 2007; Whitehouse et al., 2008).

3.1. Epidemiology Studies

Numerous studies have demonstrated excess mortality related to ARDs among sub-cohorts of Libby mine and mill workers. An interesting observation in terms of epidemiology studies is that the first publication associating Libby vermiculite with pulmonary changes considered a working population from a Marysville, Ohio fertilizer plant that had processed vermiculite from the Libby mine and South Africa (Lockey et al., 1983). This cohort became the basis for the proposed RfC discussed in section 2.8 of this thesis. Significant correlations were observed with respiratory symptoms (shortness of breath and pleuritic chest pain) and cumulative fiber exposures (Lockey et al., 1984). Studies focusing on Libby workers soon followed.

McDonald et al. (1986) included a cohort of 406 men employed at the mine for at least one year prior to 1963 and followed them until 1983. Compared with white men in the U.S., the cohort experienced excess mortality, with standard mortality rates (SMR) of 2.45, 2.55, 2.14 for respiratory cancer, non-malignant respiratory disease (NMRD), and accidents, respectively. The proportional mortality for four mesothelioma deaths was 2.4%. Data collection for a parallel NIOSH sponsored study was initiated at approximately the same time (Amandus et al. 1987; Amandus and Wheeler, 1987) and included 575 men employed at the mine for a minimum of one year prior to 1970. Similar to the McDonald et al. (1986) study, SMRs were 2.23, 2.43, and 1.44 for respiratory cancer, NMRD and accidents, respectively (Amandus and Wheeler, 1987). These early occupational-based studies demonstrated strong exposure years/response relationships (McDonald et al., 1986; Amandus and Wheeler, 1987; Antao et al., 2012).

McDonald published additional work in 2004 in which he updated epidemiology data for his original 406 man cohort, following them until 1999 (McDonald et al., 2004). The SMRs reported in this update for lung cancer and NMRD were 2.40 and 3.09, respectively. The proportional mortality for 12 mesothelioma deaths was 4.21%. An all-cause linear model implied a 14% increase in mortality for mine workers exposed occupationally to 100 f/mL/yr and approximately 3.2% increase for the general population exposed to 0.1f/mL for 50 years (McDonald et al., 2004).

An additional NIOSH sponsored study included a cohort of 1,672 Libby miners, millers, and processors in 1982 and followed subjects through 2001 (Sullivan, 2007).

Compared with U.S. white men, SMRs for asbestosis, lung cancer, and cancer of the pleura, were 165.8, l.7, and 23.3, respectively, with observed dose related increases in asbestosis and lung cancer. An update of the Sullivan (2007) cohort was published recently (Moolgavkar et al., 2010), revealing similar SMRs to Sullivan. In addition, estimates of relative risk for lung cancer, NMRD, and total mortality were 1.2, 1.4, and 1.06, respectively, with 95% confidence intervals of [(1.06, 1.17), (1.09, 1.18), and 1.04, 1.08)] (Moolgavkar et al., 2010).

One of the latest updates regarding vermiculite worker mortality (Larson et al., 2010), with a cohort of 1862 Libby miners, demonstrated a clear exposure response relationship between cumulative LA fiber exposure and asbestosis, lung cancer, mesothelioma, and NMRD mortality. A limitation noted for earlier epidemiology studies evaluating lung cancer SMRs in Libby mine and mill workers was the lack of control for cigarette smoking, bias analysis revealed that cigarette smoking had minimal impact on the exposure response relationships reported in this study (Larson et al., 2010; reviewed by Antao et al., 2012). An additional conclusion from this study was the association between LA fiber exposure and cardiovascular mortality based on a rate ratio of 1.5 with a 95% confidence interval of 1.1 to 2.0 (Larson et al., 2010).

A follow-up to the Lockey et al. (1984) Marysville, Ohio fertilizer plant study revealed pleural changes in 28.7% of the cohort (Rohs, et al., 2008). As noted previously, this cohort was the basis for the proposed LA RfC. Pleural changes were originally reported in 2.2% of the overall cohort and 8.4% of the highest cumulative fiber exposure group (Lockey et al., 1984). The study is significant in that the cohort was based on exfoliation plant workers outside of Libby, MT, with relatively low cumulative fiber exposure levels compared to those described in the Libby mine and mill worker studies.

In addition to epidemiology studies that considered Libby mine and mill workers, research has also included studies evaluating ARD mortality among Libby community members. A cross-section interview and medical testing of 7,307 persons who had lived, worked or played in Libby for at least six months prior to 1991 was conducted in 2000 and 2001 by Agency for Toxic Substance and Disease Registry (ATSDR) investigators (Peipins et al., 2003). Of the 6,668 participants $\geq$ 18 years of age who received chest radiographs, pleural abnormalities and interstitial abnormalities were observed in 17.8% and < 1% of the participants, respectively. Participant interviews revealed that the factors most strongly associated with pleural abnormalities were being a former vermiculite mine or mill worker, age, having been a household contact of a former vermiculite mine or mill worker, and being male (Peipins et al., 2003).

In 2008, a clinical and exposure summary report for 11 individuals diagnosed with mesothelioma who were not Libby mine or mill employees was published (Whitehouse et al., 2008). All cases were non-occupationally exposed individuals. The authors concluded that exposure most likely resulted from LA contamination in the community, the surrounding forested area, and areas in proximity to the Kootenai river and railroad tracks that were used to transport vermiculite concentrate (Whitehouse et al., 2008). The mean LA occupationally related mesothelioma latency period has been reported as 35 years (Case, 2006). The latency period reported for these non-occupational cases was 13 – 67 years from the first known exposure (Whitehouse et al., 2008).

In terms of both occupational and non-occupational mesothelioma cases, current mortality figures indicate one new case per year in Lincoln, County, Montana (McDonald et al., 2004; Case, 2006; Whitehouse et al., 2008). Lincoln County has the third highest age-adjusted mesothelioma death rate in the nation with a rate of 56.1 per million population (NIOSH, 2008).

A community study of Libby residents who were children ($\leq$18 years) when the vermiculite mine closed in 1990 revealed a positive association between self-reported respiratory outcomes and certain activities with potential LA exposure pathways (Vinikoor et al., 2012). Of the 1,003 study participants, 10.8% reported usually having a cough, 14.5% reported experiencing shortness of breath when walking up a slight hill or hurrying while on level ground, and 5.9% reported having coughed up bloody phlegm in the past year. Handling vermiculite insulation was positively associated with three of the four outcomes examined compared with never handling vermiculite insulation. No association was found between vermiculite insulation in the home and respiratory symptoms and no association was found between any of the activities and abnormal spirometry (Vinikoor et al., 2012).

A community study was conducted in a densely populated urban residential neighborhood in Minneapolis, Minnesota where an expansion facility processed Libby vermiculite ore from 1938 to 1989 (Alexander et al., 2012). In addition to commercial vermiculite products such as Zonolite® insulation and Monokote® fireproofing, the facility produced a waste material reported by the Minnesota Department of Health to contain 10% amphibole asbestos (Alexander et al., 2012). The waste product was piled on the property and offered to the community for use in gardening, driveway fill materials, etc. The prevalence of pleural abnormalities obtained for the 461 participants was 10.8%. The odds ratio associated with direct contact with vermiculite ore waste or ever playing in waste piles and pleural abnormalities was 2.78 (95% CI: 1.26, 6,10) and 2.17 (95% CI: 0.99, 4.78) when adjusted for background exposure. Although this study was conducted outside of Libby, MT, the results suggest that community exposure to Libby vermiculite is associated with measurable effects (Alexander et al., 2012).

In addition to pulmonary based ARDs, rates of systemic autoimmune diseases (SAIDs) have been evaluated in the Libby community. A follow-up case-control study was conducted among the participants in the 2000/2001 ATSDR study (Peipins et al, 2003) with cases including subjects that reported one of three (SAIDs) in the initial screening; systemic lupus erythematosus, scleroderma, or rheumatoid arthritis, and controls including subjects in the initial screening that responded negatively to questions regarding SAIDs (Noonan et al., 2005). Odds ratios among former Libby mine and mill workers $\geq$ 65 years of age of 2.14 (95% CI, 0.9 – 5.1) for all SAIDs and 3.23 (95% CI, 1.31 7.96) for rheumatoid arthritis, suggest that LA exposure is associated with SAIDs (Noonan et al., 2005). Increasing SAIDs risk estimates were reported for participants with relative increases in reported vermiculite exposure pathways.

These epidemiology studies demonstrate clear and significant increases in ARD, including asbestosis, lung cancer, and mesothelioma among former mill and mine workers. In addition, ARD has been observed in area residents with no direct occupational exposures. The most common health outcome among Libby residents and others with low lifetime cumulative fiber exposure levels are pleural changes. Research has been performed within the past decade to gain further insight into the toxicological mechanisms of these LA ARDs. Outcomes of these studies are summarized in the following section.

3.2. Toxicity Studies

The mechanisms by which asbestos mineral fibers interact with cells of the lung and pleura, resulting in ARDs, are not clearly defined (Manning et al., 2002;). Proposed factors influencing toxicity include structural and surface properties of fibers, fiber dose, uptake of fibers by epithelial cells, generation of reactive oxygen species, DNA damage, cytokine production and pro-inflammatory mediators (Manning et al., 2002).

While research evaluating the toxicity of the regulated varieties of asbestos has been conducted for decades, research aiming to describe the mechanisms of LA toxicity is relatively new (Hamilton et al., 2004; Blake et al., 2007; Putnam et al., 2008; Smartt et al., 2009; Duncan et al., 2010; Hillegass et al., 2010; Padilla-Carlin et al., 2011; Shannahan et al., 2011; Cyphert et al., 2012; Li et al., 2012; Marchand et al., 2012;). A summary of recently published investigations regarding LA toxicity is presented below. Many of these studies include comparisons with more widely studied serpentine chrysotile or other amphibole species as positive controls.

Libby amphibole samples commonly used in toxicological assessments are referred to as LA2000 or the “six-mix” (Lowers et al., 2012). This mineral sample set represents 6 of the original 30 samples collected by Meeker and others (2003) and characterized by Bellamy and Gunter (2008). The LA2000 material was ground to approximate the fiber size (length, width, and aspect ratio) of fiber dimensions reported for air data (Lowers et al., 2012).

A primary murine alveolar macrophage study revealed that LA asbestos internalized in the cells and induced oxidative stress through increasing reactive oxygen species (ROS) levels and decreased superoxide dismutase activity (Blake et al., 2007). Both LA and crocidolite decreased intracellular glutathione levels, however, crocidolite generated significant DNA damage, while LA did not. The authors postulated that differences observed in DNA damage between crocidolite and LA may be due to the chemical composition of the fibers (crocidolite has greater iron content) and/or the activation of distinct cellular pathways.

In vivo studies were conducted by a single intratracheal installation of LA or crocidolite asbestos to C57B1/6 mice (Putnam et al., 2008: Smartt et al., 2010). Alterations in lung gene expression, primarily involving plasma membrane-associated genes, were observed six months after exposure (Putnam et al., 2008) with both amphibole minerals. Both amphibole species induced increased collagen types I and III mRNA expression and collagen deposition compared to the control; however, crocidolite produced more collagen than LA (Putnam et al., 2008; Smartt et al., 2010). The most significant collagen deposition was observed one month after exposure.

An additional gene profiling study (Hillegass et al., 2010) was performed to characterize alterations in gene expression in human mesothelial cells induced by LA, with glass beads and crocidolite asbestos serving as negative and positive controls, respectively. At 8 hours LA induced significant ($p < 0.05$) upregulation of one gene, superoxide dismutase 2 (SOD2) (4-fold) and at 24 hours, 111 gene changes were observed, including upregulation of SOD2 (5-fold). Crocidolite also induced significant upregulation of SOD2 (6-fold at 8 hrs) and changes in 205 genes at 24 hours. Increased production of oxidants and decreased glutathione levels were observed with both amphibole minerals compared with the negative control.

Three studies were performed assessing in vitro (Duncan et al., 2010) and in vivo (Padilla- Carlin et al., 2011; Cyphert et al., 2012) cellular responses to size fractionated LA and amosite asbestos. Asbestos particles with aerodynamic fiber diameters (d_{ef}) ≤ 2.5 µm were isolated from a portion of the Libby six mix and amosite with the Webber et al. (2008) water elutriation method to represent the fraction most likely to deposit in the alveolar region (Dai and Yu, 1998).

Human airway epithelial cells exposed to four separate doses of unfractioned LA and amosite revealed comparable interleukin 8 (IL-8), cyclooxygenase 2 (COX 2) and heme oxygenase 1 mRNA transcript levels following a 24 hour exposure (Duncan et al., 2010). In terms of the fractioned ($PM_{2.5}$) form of each mineral species, LA $PM_{2.5}$ revealed similar potency to the unfractionated forms in the production of pro-inflammatory cytokines (Duncan et al., 2010). The largest pro-inflammatory response was observed with the amosite $PM_{2.5,}$ which demonstrated a 4 and 10 fold increase in IL-8 and COX-2 mRNA expression, respectively, compared to LA $PM_{2.5}$ after a 24 hour exposure (Duncan et al., 2010).

Single, intratracheally instilled doses of LA (0.65 mg/rat or 6.5 mg/rat) or amosite (0.65 mg/rat) $PM_{2.5}$ fractions were used in a male F344 rat model to assess inflammatory and fibrotic effects over a 3 month (Padilla-Carlin et al., 2011) and 2 year (Cyphert et al., 2012) period. One day post exposure, total bronchial alveolar lavage fluid revealed significantly elevated ($p < 0.05$) cell numbers in the amosite and high dose LA compared with the saline control (Padilla-Carlin et al., 2011). A similar pattern was observed with bronchial alveolar inflammatory cytokines, with the exception of IL-1*B*, which decreased in the high-dose LA group. In terms of equal mass dosing, the $PM_{2.5}$ amosite presented a greater inflammatory and fibrotic response; however, the higher dose LA resulted in prolonged inflammation similar to the amosite (Padilla-Carlin et al., 2011).

At one year post intratracheal instillation, no differences were observed in BALF total cell numbers and inflammatory cytokines in the amphibole treated groups compared to controls (Cyphert et al., 2012). Elevated BALF total cell numbers were observed in all groups at 2 years post instillation compared to one year post exposure cell numbers; however, this was attributed to age. Lung inflammation and fibrosis progressed in a time and dose dependent manner for LA exposed groups, although the severity of inflammation and fibrosis was greater in amosite exposed groups (Cyphert et al., 2012). The gene expression changes of collagen markers Col 1A1, Col 1A2 and Col 3A1 were unchanged in asbestos treated groups compared to saline controls at one year post installation, while all increased in the asbestos exposed groups at two years post installation, with the low dose LA group revealing statistical significance ($p < 0.05$) over saline treated controls (Cyphert et al., 2012). At one year post installation, expression of Wilms' tumor gene 1 (WT1) was significantly increased in the low dose LA relative to saline-treated controls, while gene expression of WT1, Msln, and EGFR were increased in LA exposure groups at two years.

Toxicological differences observed in PM2.5 fractionated LA and amosite studies may be attributable to surface chemistry characteristics, fiber size, and/or aspect ratios (AR) (Duncan et al., 2010; Padilla Carlin et al., 2011; Cyphert et al., 2012). LA samples contained a lower concentration of ionizable iron compared to amosite. In the fractionated amphiboles, the surface-complexed iron correlated with hydroxyl radical ions and pro-inflammatory responses. However, in the unfractionated amphiboles, differences in oxidant production did not correlate with pro- inflammatory responses (Duncan et al., 2010).

The mean length and AR of PM $_{2.5}$ fractionated amosite (6.4 µm, AR 22.5) was greater than that measured for PM $_{2.5}$ fractionated LA (1.9 µm, AR 6.7) (Lowers and Bern, 2009; Duncan et al., 2010). Increased inflammatory responses observed with fractionated amosite compared with fractionated LA may be associated with these differences in fiber lengths and ARs.

To further examine the role that iron may play in amphibole fiber toxicity, Shannahan et al. (2011) assessed the ability of PM2.5 fractionated LA to bind to exogenous iron and then evaluated the role of iron in related ROS production. Further in vitro and in vivo assessments were performed with human bronchial epithelial cells and SH rats, respectively. LA fibers contained relatively low levels of surface iron (6.04 ng/mg of LA), but when incubated with $FeCl_3$, substantial iron (approximately 16.6 µg Fe/mg of fiber) bound to the fibers and was not dissociable after 3 distilled water washes. In an acellular system, loading of LA with iron significantly ($p < 0.05$) increased ROS production and this increased ROS production was significantly decreased when deferoxamine, an iron chelator, was added. However, in vitro assessments revealed that iron loading on LA decreased IL-8 expression, which was increased by the chelator. In vivo responses of iron loading on LA revealed decreased neutrophil inflammation and inflammatory gene expression, while the chelator increased these responses. While the iron loading on LA decreased the inflammatory response potential, which was contrary to other asbestos studies, Shannahan et al. (2011) postulated that opened binding sites on LA surfaces promotes tightly bound iron to be released from proteins, resulting in their activation and signaling.

A recent in vitro study (Li et al., 2012) evaluated the inflammatory responses of THP-1 macrophage cells and epithelial BEAS-2B cells exposed to 2 separate doses (20 µg/mL and 40 µg/mL) of chrysotile asbestos and LA six mix for 24 hours. Supernatant of the culture medium, referred to as a conditioned medium (containing no asbestos fibers), from the THP-1 treatment was used in an additional assessment to evaluate signal transduction from macrophages to bronchial epithelial cells. Exposure of THP-1 cells to chrysotile and LA six mix resulted in caspase-1 mediated inflammasome activation and production of IL-1*B*. Although both minerals resulted in the production of IL-1*B*, this secretion was most pronounced in chrysotile exposed macrophages. Separate pathways for inflammasome activation were proposed for chrysotile versus LA. Chrysotile activates the caspase – 1 Nod like receptor protein NLRP3 inflammasome while IL-1B production induced by LA is more ROS dependent (Li et al., 2012). These differences may be related to fiber length and surface properties. The secretion of IL-1B by the THP-1 macrophages stimulated a BEAS-2B immune response, implying a secondary response.

While the mechanisms of LA pulmonary/pleural toxicity are not clearly defined, this research has advanced the scientific knowledge. The inflammatory and cytotoxic effects of LA are similar to those observed with other forms of asbestos and the generation of ROS appears to be a key factor. Further research is needed to characterize mechanisms.

Considering the rates of ARD clearly defined among former mine and mill workers, the fact that ARD has been observed in Libby residents with no direct occupational exposures, and the prevalence of pleural changes among those exposed to relatively low lifetime cumulative fiber exposure levels, it is critical to identify potential LA exposure pathways. Data from the ATSDR community health screening study (Peipins et al., 2003) suggest that the prevalence of pleural abnormalities increases substantially with increasing exposure pathways. Research quantifying potential LA exposure pathways are presented in sections 4 and 5. The studies summarized in section 4 are provided as related background information, while the three research Aims are presented in section 5.

4. Related Studies

After the discovery of LA on the surface of tree bark in the forested areas surrounding the former vermiculite mine, studies were performed to assess the human exposure potential related to this newly discovered source of contamination. Two of these studies are discussed in detail as Research Aims 1 and 2 of this dissertation (Hart et al., 2007; Hart et al, 2009). Additional related studies, including exposure potential evaluations related to combustion activities (Ward et al., 2009; Ward et al., 2012; USEPA 2012c) are summarized below. In addition, a study evaluating potential bark contamination in trees near historical vermiculite processing facilities is discussed (Elashheb et al., 2011).

4.1. Combustion Studies

Libby amphibole contaminated firewood obtained from the Aim 1 firewood harvest simulations (Hart et al., 2007) conducted near the former vermiculite mine was combusted in new, EPA certified wood stoves (Ward et al., 2009). Approximately 4.6 m of 38 cm diameter galvanized steel ductwork was attached to the stove outlet to simulate ductwork within a home. In addition, an inverted plastic tote lined with aluminum foil was placed near the outlet of the ductwork exhaust port. During each combustion trial, air sampling was conducted within the tote and at the conclusion of each trial, surface wipe sampling was conducted within the ductwork and bulk ash samples were collected from the stoves.

The majority of LA fibers remained in the ash, with concentrations quantified from each combustion trial as 137, 84, and 18 million asbestos structures per gram (s/gm) (Ward et al., 2009). Wipe sample concentrations ranged from non-detect to 20 thousand structures per square centimeter (s/cm^2). Post combustion ductwork wipe samples revealed LA contamination in locations where bends or turns occurred in the ductwork or within the inverted tote at the exhaust outlet. Libby amphibole was not detected in pre-combustion ductwork wipes, suggesting that LA can be liberated into ambient air when contaminated firewood is combusted. Detection of LA in ductwork where bends occurred only, and not in straight sections of ductwork, implies that the fibers were liberated into the combustion airstream and were most likely impacted or intercepted on the surface of the ductwork. Air sample results from within the tote were inconclusive. Because of the large amount of smoke within the tote, air sampling filters became heavily loaded within minutes of each trial, resulting in air sampling pump flow resistance failures. Therefore, air sampling data was not reported.

A small scale (3.7 m x 3.7 m) controlled burn was conducted in the same geographic location as the Aim 2 FS occupational study in an effort to assess the LA exposure potential to USDS FS personnel performing firefighting activities (Ward et al., 2012). The controlled burn consisted of three activities, including construction of a fire line around the perimeter of the burn plot, combustion, and mop-up. High volume air and personal breathing zone air sampling was conducted independently throughout each activity and bulk ash and Tyvek® clothing wipe sampling was conducted at the conclusion of the mop-up. High volume air sampling consisted of four sampling stations positioned 1.2 m from the perimeter of the burn, one station positioned 3.7 meters above the burn plot and a final station positioned within the prevailing wind direction.

As with the wood stove combustion study discussed above, the majority of fibers remained in the ash with bulk ash concentrations ranging from 8 to 19 million s/gm. However, the potential for inhalation exposure exists as seventy-five and 62 percent of the PBZ and high volume air samples, respectively revealed detectable LA concentrations when analyzed by AHERA TEM (Ward et al., 2012).

The potential emissions of LA from duff collected in the forested area near the former vermiculite mine were evaluated in a laboratory simulation of a wildfire (USEPA, 2012c). The combustion was conducted in a burn chamber placed in an enclosed shed. Two experimental conditions were evaluated; 1) a high temperature condition (1800 ^{o}F) intended to simulate the rapid combustion associated with a wildfire and 2) a low temperature condition (800 ^{o}F) intended to simulate smoldering conditions post wildfire.

The exhaust plume was sampled with both a mixed cellulose ester (mixed cellulose ester) filter method and an impinger method.

As reported with the Ward et al. (2006, 2009) studies, the majority of LA remained in the ash during the low and high temperature trials (USEPA, 2012c). However, LA was liberated in the flue gas as well. Poor collection efficiency was observed with the MCE filter air sample collection, which was postulated to be associated with the high temperature and water vapor concentrations in the flue gas. Therefore, only impinger results were used. The mean total and phase contrast microscopy equivalent (PCME) concentrations of LA in the high temperature trial flue gas were 8.2 and 1.7 f/mL, respectively. While the mean total and PCME concentrations of LA in the low temperature trial flue gas were 1 and 0.16 f/mL, respectively.

4.2. Tree Bark Contamination Near Historical Processing Facilities

To further assess the role of trees serving as reservoirs for LA, tree bark samples were collected near four former vermiculite processing (exfoliation) facilities (Elashheb et al., 2011). These historical facilities were located in Spokane, WA, Newark, CA, Santa Anna, CA and Phoenix, AZ. The Spokane site was selected due to its geographic proximity to Libby, MT. The remaining sites were selected based on a ranking system which considered 1) the tonnage of vermiculite concentrate processed, 2) the year exfoliation activities were terminated at the site, 3) the population density within one mile of the site, and 4) the total duration of site operation.

Of the 22 samples collected near the Spokane site, LA was detected in one sample and chrysotile in another. Funding restraints limited analyses at the remaining sites to 3-5 samples per site. Of these, LA and/or actinolite tremolite amphibole was detected in a subset of samples from all locations. It is interesting to note that serpentine chrysotile asbestos was detected more frequently than amphibole asbestos. It is plausible that chrysotile's wide spread usage in industrial as well as residential applications has resulted in its ubiquity in the ambient environment. This study revealed that tree bark may serve as reservoirs for asbestos, and indicators of past and current contamination in Libby, MT as well as other locations.

5. Research Aims

As discussed previously in the Introduction of this thesis, LA was detected in substantial (2.5 -110 million structures/cm^2) concentrations on the surface of tree bark in the forested area surrounding the former vermiculite mine (Ward et al, 2006; Webber et al., 2006; USEPA, 2008a). Since a relatively large fraction (33%) of Libby residents rely on wood burning as their primary source of heat (Ganesan et al., 2006), the LA exposure potential associated with firewood harvesting activities was the first evaluation performed. The introduction, background, methodology, results, and conclusion for this work were published in the *Annals of Occupational Hygiene* (Hart et al., 2007) and are presented in section 5.1 of this dissertation with a copy of the refereed publication provided in Appendix A.

The firewood harvesting assessments presented in section 5.1 were performed within the USEPA OU3 Superfund site restricted zone. At the conclusion of this work, tree bark sampling was performed in the forested area surrounding the former vermiculite mine outside of the restricted zone, an area accessible to the public for recreational activities and an area where USDA FS personnel commonly worked. Libby amphibole concentrations in tree bark, ranging from 37,000 – to 15,000,000 structures/cm^2 (Hart et al., 2009), were revealed through this preliminary sampling. This substantial LA source in the Kootenai National Forest warranted further evaluation and the USDA FS was the most logical group to include in this exposure assessment due to the frequency of their work in this area. The introduction, background, methodology, results and conclusion for this FS occupational exposure assessment were published in the *Journal of*

Environmental and Public Health (Hart et al., 2009) and are presented in section 5.2 of this dissertation with a refereed copy of the publication provided in Appendix A.

The LA exposure potential associated with vermiculite from the Libby mine extends throughout the U.S. due to vermiculite concentrate shipments to over 200 exfoliation facilities (Horton et al., 2006). The rates of ARDs among people who worked at vermiculite processing facilities (outside of Libby, MT) or came into contact with vermiculite have been evaluated at several sites (Lockey et al., 1983; Horton et al., 2006; Rohs et al., 2008; Alexander, 2012; Vinikoor et al., 2012).

A common application for expanded vermiculite was loose fill insulation in attics and homes. The precise number of homes insulated with Zonolite® brand insulation derived from the Libby mine, is unknown (Gunter et al., 2005; USEPA, 2011b; Zalac, 2003) and limited research has been performed to assess LA exposure potentials associated with VAI (Cowan, 1997; USEPA, 2003A; Ewing et al., 2003: Spear et al., 2012).

A three year study evaluating the exposure potential associated with LA from insulation sources and chrysotile from commercial building materials was conducted while weatherization activities were performed in homes. The study was divided into two phases. The first phase consisted of living space high volume air and surface sampling to evaluate potential asbestos pathways into the living space (Spear et al., 2012) prior to weatherization. Phase I of this study was also used to develop sampling strategies, personal protective equipment (PPE) selections, and exposure control strategies for the Phase II, weatherization component. Phase II of this study evaluated the potential for LA or chrysotile exposure associated with weatherization activities. The background, methodology, results and conclusion for this exposure assessment will be submitted to *Environmental Health Perspectives* and are presented in section 5.3 of this dissertation.

Research Aims 1 and 2 were conducted in the Kootenai National Forest 7 miles northeast of Libby, MT as illustrated in Figure 1 below. Research Aim 3 was conducted in 37 homes, throughout the state of MT, excluding homes in Libby as illustrated in Figure 2 below.

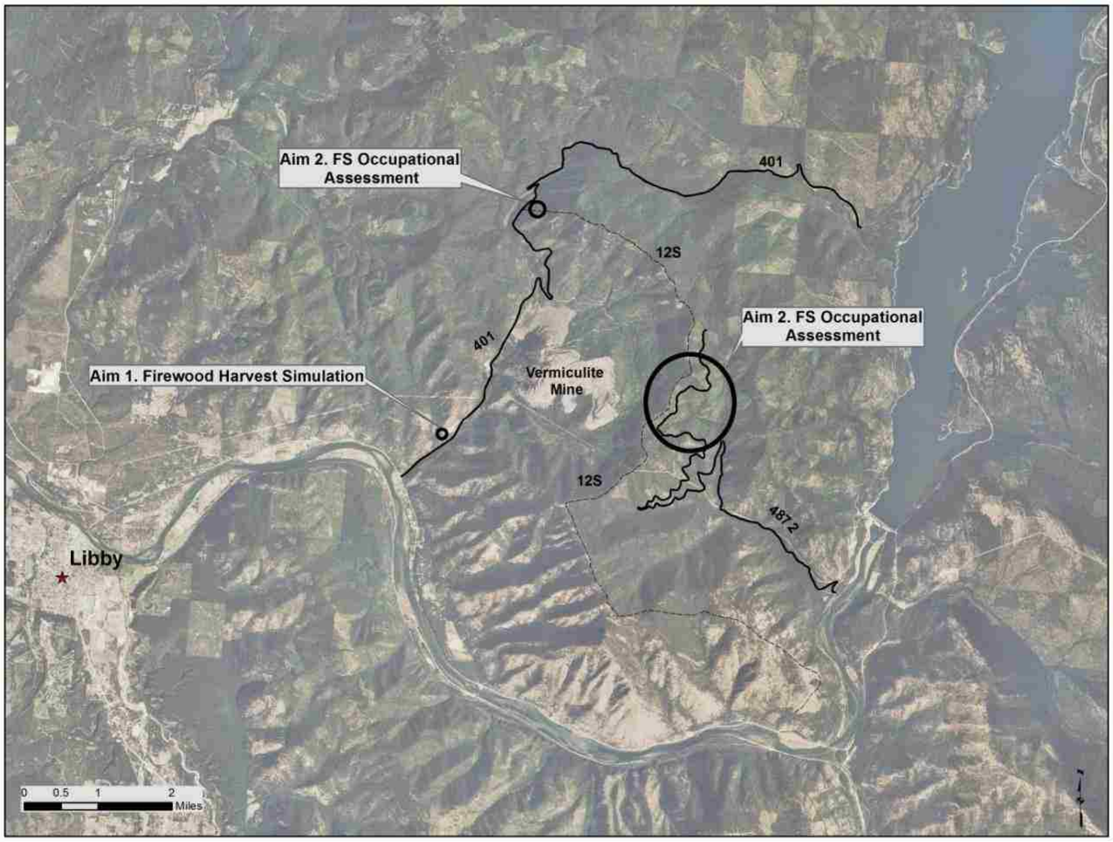

Figure 1. Location of Aim 1 and 2 activity based sampling in relation to the former vermiculite mine and the town of Libby, MT.

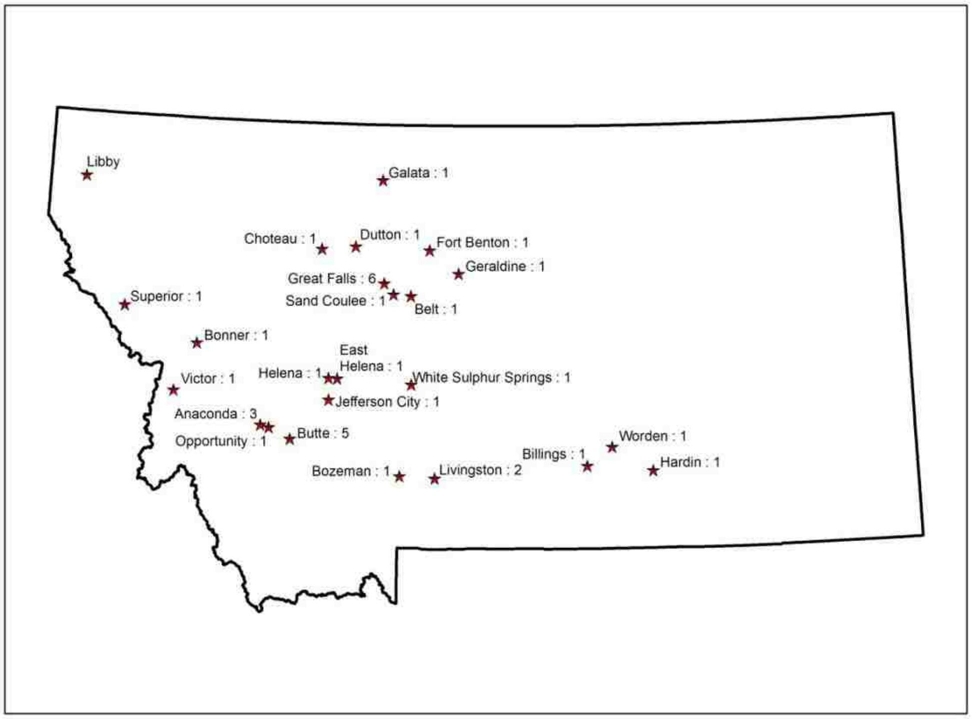

Figure 2. Location of Aim 3 weatherization sampling in relation to Libby, MT.
The number of homes during weatherization activities in each town is indicated numerically.

5.1. Research Aim 1: Firewood Harvest Exposure Assessment

5.1.1. Introduction

It has recently been discovered that tree bark samples collected within the town of Libby, within the EPA restricted mine area, and along the railroad corridor west of town contain varying levels of amphibole contamination (Ward et al., 2006). Analyses to date have yielded substantial amphibole fiber concentrations ranging from 41 million to 530 million fibers per gram of bark, while a bark sample collected approximately seven miles west of town along the railroad line had concentrations of 19 million fibers per gram. A conversion of these mass-based concentrations to areal concentrations (to reflect surface area contamination) revealed concentrations in excess of 100 million amphibole fibers per cm^2.

In addition to vermiculite mining, much of the economy in Libby has historically been supported by the harvesting and processing of timber. Western Montana logging companies own approximately 315,000 acres of land surrounding the Libby mine that could potentially be harvested. Because firewood is the cheapest source of fuel in the Libby area, it is the most common source of residential heating during the cold Libby winters. There are an estimated 1,300 wood stoves in use in Libby, with at least some of the firewood being harvested within the Libby valley and surrounding forests.

Previous results from tree bark sampling suggest a potential for asbestos exposure to those who harvest or disturb contaminated wood within the Libby area (Ward et al., 2006). Despite the reliance on local timber resources in Libby, currently no definitive efforts exist to evaluate the potential for asbestos exposure during the common practice of harvesting firewood for residential home heating. The research within this study presents preliminary data that evaluates the potential of amphibole exposure associated with firewood harvesting within a known asbestos-contaminated area. Research trials were conducted inside the Libby EPA restricted zone where amphibole contamination in tree bark was previously demonstrated (Ward et al., 2006).

5.1.2. Methodology

During the summer and fall of 2006, three separate firewood harvesting simulations were conducted on U.S. Forest Service property in an area of the Kootenai Forest inside the EPA restricted zone surrounding the former W.R. Grace vermiculite mine. These trials were conducted approximately 30 to 35 meters (m) off of Rainy Creek road approximately 1.5 kilometers (km) up Rainy Creek road from Highway 37 (Figure.1). Another simulation was conducted near Missoula, Montana (approximately 4 hours southeast of Libby) to serve as a control.

All of the investigators participating in this study completed a 40-hour OSHA Hazardous Waste Operations and Emergency Response course, or demonstrated competency via education and/or professional certifications (industrial hygiene Ph.D., certified industrial hygienist). In addition, investigators participated in a training/planning session developed specifically for the harvest simulations. A site safety and health plan was also written and submitted to the Libby EPA supervisor for approval. All investigators obtained medical clearance to wear negative pressure respiratory protection and passed quantitative fit tests within the past year.

Trees selected for the harvesting simulation at each site consisted of three to four standing dead and three to four downed trees. The location of each simulation site was identified and recorded using a Garmen Etrex 12 channel global positioning system

(GPS). Tree species were identified and documented. Prior to harvesting, a minimum of one 200 gram bark sample was collected from two sides of each tree approximately 1.2 m from the base. Additional bark samples were collected from randomly selected harvested trees at 1.2 m intervals from the base to the tree top. The bark was collected by prying off sections with a small pry bar and placing them in labeled plastic bags. The bags were then sealed and labeled and the pry bar was cleaned with a wet wipe after each collection. The bark samples were preserved for later analysis by TEM.

New Poulan® model 3416 gas chainsaws were used for each research simulation trial. The chainsaw was replaced prior to each trial in order to avoid cross contamination and to ensure that the condition of the chain (sharpness) remained consistent. The harvesting simulation process at each site consisted of downing the tree, removing tree branches, and sawing the log into 30 centimeter (cm) long blocks. The blocks were then gathered and stacked in a pile approximately 20 m away. Four to five investigators participated in each simulation trial, with the work practices employed by each investigator remaining consistent throughout each of the trials. One investigator operated a chainsaw, while a second investigator assisted the chainsaw operator by clearing debris, moving downed trees, and holding downed trees steady while being sawed. Two investigators gathered the wood blocks and stacked them into piles. An additional investigator was present for Trials 2 and 3 and served as a data recorder.

Personal breathing zone (PBZ) samples were collected during the trials using conductive three piece asbestos sampling cassettes. The cassettes contained 25 millimeter (mm) 0.8 micron (µm) pore size mixed cellulose ester membrane filters. SKC Aircheck 224 sampling pumps were calibrated before and after each trial with a Gilian®

Gilibrator 2 primary flow meter at a flow rate of four liters per minute. Throughout each trial, each investigator wore a sampling pump with the asbestos cassette placed in the breathing zone. PBZ samples were analyzed for asbestos per NMAM 7400, Asbestos and Other Fibers by PCM (NIOSH, 1994a) and for asbestos per EPA's AHERA, Airborne Asbestos by TEM, EPA Enhanced Protocol (Level III) (USEPA, 1987). All air samples were analyzed by DataChem Laboratories (Cincinnati, OH), a National Voluntary Laboratory Accreditation Program (NVLAP) certified and an American Industrial Hygiene Association (AIHA) accredited laboratory. PBZ samples submitted included ten percent field blanks.

In addition to PBZ sampling, surface wipe sampling of the outer layer of Tyvek® clothing was conducted at the conclusion of each trial. The wipe sampling protocol followed the American Society for Testing and Materials (ASTM) D 6480-05 procedures, Wipe Sampling for Settled Asbestos (ASTM, 2006). Wipes were collected with SKC Ghost wipes pre-moistened with deionized water. A 10 by 10 cm SKC disposable manila paper template was used for each wipe. A wipe sample was gathered on each investigator's chest and upper thigh. The site of the chest wipe sample and thigh sample (right/left) was randomly selected. The two wipe samples collected for each investigator were submitted for analysis as a composite sample. In addition to the post-harvest wipes collected, pre harvest wipes, inner layer Tyvek® wipes and ten percent field blanks were analyzed. The wipe samples were analyzed for asbestos per ASTM's D 6480-05 Method, TEM Asbestos Analysis (ASTM, 2006) by DataChem.

The average duration of each firewood harvest simulation was 89 minutes, with 45 to 50 minutes dedicated to the harvest simulations and the remaining time associated with bark collection. The harvest duration was limited to minimize the potential to overload the sample media.

5.1.3. Results and Discussion

Multiple tree bark samples were collected from standing dead or fallen trees selected for harvesting during both the control harvest in Missoula and the firewood harvesting simulations conducted within the Libby restricted zone. The samples were collected from common coniferous tree types (lodgepole pine (*Pinus contorta*), ponderosa pine (*P. ponderosa*), larch (*Larix occidentalis*), and Douglas fir (*Pseudotsuga menziesii*)) found within the area, and are representative of the types of trees typically burned during residential home heating in western Montana. Amphibole fibers were not detected in bark samples collected from Missoula, Montana. Eight bark samples analyzed to date from the Libby EPA restricted zone (collected in the same area where the firewood harvesting simulations were conducted) revealed substantial amphibole fiber concentrations ranging from 7 million to 97 million fibers per cm^2 of bark surface area. These concentrations are consistent with amphibole contamination in tree bark previously reported by Ward et al., 2006. Fiber dimension analyses of the bark samples revealed that the majority of the asbestos fibers detected were less than 5 microns in length. Results from the bark samples collected in these trials showed that all identified fibers were typical of the Libby vermiculite amphibole contaminants, with typical elemental composition of Si>Mg>Ca>Fe>Na>K (Meeker, et al., 2003).

PBZ samples collected during the firewood harvesting trials were analyzed for asbestos by both PCM and by AHERA TEM. Fibers were observed on all samples analyzed by PCM, excluding field blanks. The PCM fiber concentrations from the control (Missoula) trial ranged from 0.014 to 0.019 fibers per milliliter (mL). The NIOSH PCM method cannot identify fiber types (Dodson, Hammar, 2006), but AHERA TEM analysis revealed fibers in the control samples to be organic, non-asbestos (cellulose), with no asbestos concentrations above the AHERA TEM analytical sensitivity (AS) of (0.009 - 0.01 structures per mL).

Table 2 presents PBZ air sampling results, including the mean PBZ asbestos concentrations (measured by PCM and AHERA TEM, respectively) and the standard deviation (SD) from the three harvest trials per task (chainsaw operator, operator assistant, wood stackers 1 and 2). While the PBZ sample from the chainsaw operator's assistant revealed the highest mean total asbestos concentration (Column 5, Table 2), overall no significant differences were observed in PBZ asbestos concentrations between tasks.

Differences were observed in the concentration of shorter fibers (< 5 µm long) compared to the concentration of longer fibers (> 5 µm long) ($p = 0.055$) for PBZ air samples. The mean concentration of asbestos fibers less than five µm long for all samples gathered from the Libby EPA restricted zone trials was 0.1526 fibers per mL, SD = 0.2136, while mean concentration of asbestos fibers greater than five µm long for all samples gathered from the Libby EPA restricted zone trials was 0.0681 fibers per mL SD = .0828 (Row 6, Table 2). Three of twelve analyses for fibers greater than five µm long from the Libby EPA restricted zone trials revealed concentrations that were less than the AS of .0068, .0145 and .0148 fibers per mL respectively. In order to perform statistical analysis on concentrations that were less than the AS, a value of 70% of the AS was used (Mulhausen and Damiago, 1998).

Table 2. Firewood harvest personal breathing zone (PBZ) air sampling results
Mean concentrations reported in fibers per mL (f/mL) and standard deviations (sd) from three firewood harvest simulation trials conducted in the EPA restricted zone near Libby, MT. Results are reported by task performed (chainsaw operator, operator assistant, and wood stackers 1 and 2). Phase contrast microscopy (PCM) and AHERA transmission electron microscopy (TEM).

Task Performed	Mean PCM sample time weighted average (f/mL) > 5 µm [A](sd)	Mean TEM sample time weighted average (f/mL) < 5 µm [A](sd)	Mean TEM sample time weighted average (f/mL) > 5 µm [A](sd)	Mean TEM sample time weighted average (f/mL) Total Asbestos [A](sd)
Chainsaw Operator n= 3	0.723 (1.059)	0.0726 (0.0306)	0.0411 (0.0264)	[B]0.1137 (0.0568)
Operator Assistant n = 3	0.263 (0.316)	0.265 (0.374)	0.1369 (0.1398)	[B]0.402 (0.513)
Wood Stacker 1 n = 3	0.0730 (0.0574)	0.0867 (0.1199)	[C]0.0396 (0.0558)	0.126 (0.176)
Wood Stacker 2 n = 3	0.1173 (0.1015)	0.186 (0.244)	[D]0.0549 (0.0683)	0.241 (0.312)
Total Mean for all Tasks n = 12	0.294 (0.545)	0.1526 (0.2136)	0.0681 (0.0828)	0.2207 (0.2990)

[A] standard deviation (sd)

[B] One of three samples had loose material on the filter and was prepared using an indirect prep method

[C] Two samples were less than the AS of 0.0145 and 0.0148 structures/mL respectively. 70% of the AS was used to calculate mean concentration.

[D] One sample was less than the AS of 0.0148 structures/mL. 70% of the AS was used to calculate mean concentration.

In terms of fiber counts reported by the laboratory (not shown), sixty-nine percent of the fibers collected on PBZ samples were < 5 µm long. This is consistent with ambient air sampling trends reported for Libby (ATSDR, 2003).

Due to the lack of public exposure limits for asbestos applicable to this situation, PBZ concentrations were compared with occupational exposure limits. For individual PBZ harvest trial samples for fibers > 5 µm (not shown in Table 2), two of three samples from both the chainsaw operator and the operator’s assistant exceeded the OSHA exposure limit of 0.1 fiber per mL, assuming an eight hour exposure duration, while one of three PBZ samples from both of the wood stackers exceeded the OSHA exposure limit assuming an eight hour exposure duration when analyzed by PCM.

A substantial portion of cellulose (from sawdust) fibers was expected in PCM analyses. AHERA TEM analyses were performed to describe the fiber population. In terms of fiber counts reported by the laboratory (not shown in Table 2), more than 5 non-asbestos fibers (organic, gypsum) were identified on all PBZ AHERA TEM samples. AHERA TEM analyses for the concentration of asbestos fibers > 5 µm revealed that 3 of twelve samples exceeded the OSHA PEL, assuming an 8 hour exposure duration (not shown in Table 2). These samples were collected on the chainsaw operator’s assistant, and wood stackers 1 and 2 during the firewood harvest trial 3.

The current regulatory methods of counting fibers based on fiber length and aspect ratio may not adequately describe the risk of asbestos related health effects. Fiber size, shape, and composition contribute collectively to health risks in ways that are currently being evaluated (ATSDR, 2003). Although we compared concentrations of asbestos > 5 µm to occupational exposure limits, the concentrations of fibers < 5 µm may contribute to health risks.

Surface wipe sampling of the outer layer of Tyvek® clothing was conducted at the conclusion of each trial. These wipe samples were analyzed for asbestos fibers by TEM, with results presented in Table 3 and Figure 3. All of the field blank, inner layer Tyvek®, and pre-harvest outer layer Tyvek® wipe samples showed no asbestos contamination, and were below the AS (878 structures per cm^2) for the D 6480-05 TEM method. There was a striking difference between the sizes of the asbestos fibers (length) measured from the suits following the firewood harvesting simulations, with significant concentrations of the shorter fibers (< 5 µm) found when compared to longer fibers (> 5 µm in length) ($p = 0.038$). The mean concentration of asbestos fibers < 5 µm long for all Libby restricted zone post harvest wipe samples was 27,192 fibers per cm^2, SD = 36,749. The mean concentration of fibers > 5 µm in length for all Libby restricted zone post harvest wipe samples (and for each job description) was more consistent compared to the smaller fibers detected, with 2,631 fibers per cm^2 (SD = 1,982) measured. One of twelve wipe sample analyses for fibers > 5 µm long revealed concentrations that were less than the AS of 5,270 fibers per cm^2.   In order to perform statistical analysis on concentrations that were less than the AS, a value of 70% of the AS was used (Mulhausen and Damiago, 1998).

Table 3. Firewood harvest wipe sampling results
Transmission electron microscopy (TEM) results from three simulation trials conducted in the Libby EPA restricted zone near Libby, MT. Results are reported by task performed (chainsaw operator, operator assistant, and wood stackers 1 and 2).

Task Performed	Harvest Trial	TEM (f/cm^2) < 5 µm	TEM (f/ cm^2) > 5 µm	TEM (f/ cm^2) Total Asbestos
Chainsaw Operator	1	100,123	[A]3, 689	103,812
	2	4,830	878	5,708
	3	15,848	4,953	20,801
Operator Assistant	1	108,905	3,513	112,418
	2	5,709	439	6,148
	3	14,134	2,827	16,961
Wood Stacker 1	1	16,863	2,108	18,971
	2	5,709	439	6,148
	3	14,135	3,392	17,527
Wood Stacker 2	1	6,324	2,108	8,432
	2	6,587	439	7,026
	3	27,140	6,785	33,925
Total Mean for all Tasks n = 12		27,192 (36,749)	2,631 (1,982)	29,823 (37,548)

[A] One sample was less than the AS of 5,270 structures/ cm^2. 70% of the AS was used to calculate mean concentration.

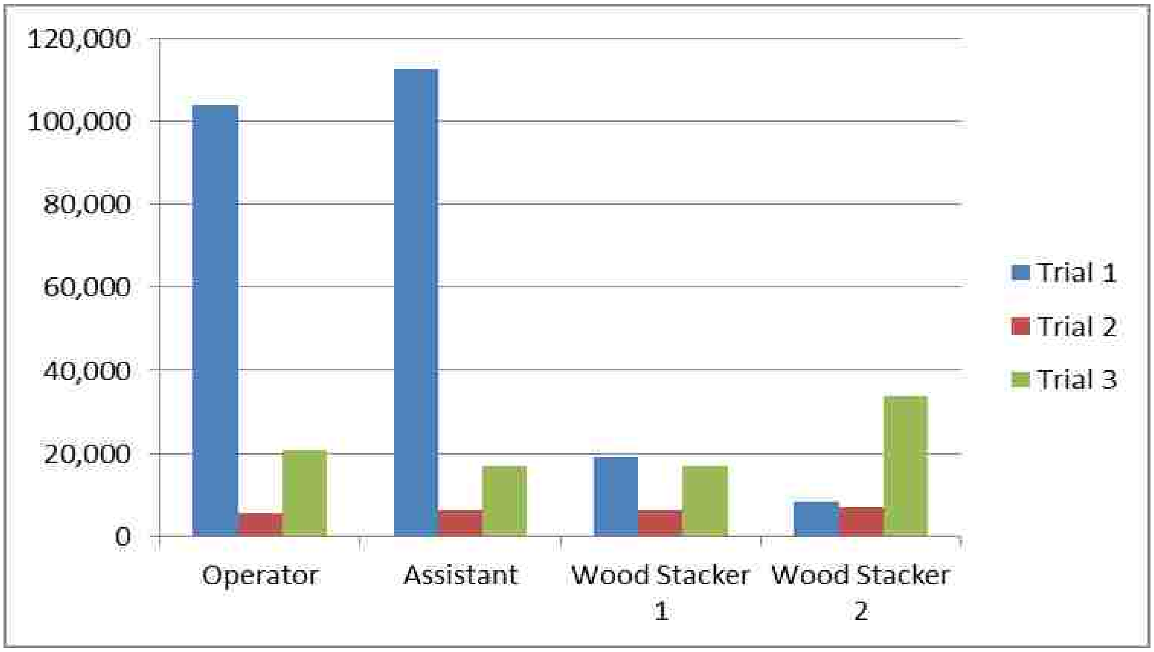

Figure 3. Firewood harvest wipe sample results presented as total asbestos fibers (s/cm2) for 3 trials and 4 activity tasks.

Wipe samples collected from the chainsaw operator and the chainsaw operator's assistant after harvest trial 1 showed concentrations of asbestos fibers < 5 µm long at least six times the asbestos wipe concentrations measured from the two wood stackers (Column 3, Table 3 and Figure 3). However, this same trend was not observed for harvest trials 2 and 3 and this trend was not observed in the breathing zone sampling results. There were no statistically significant differences observed in wipe asbestos concentrations between the four investigators. It is postulated that this spike in wipe sample concentrations observed during trial 1 for the chainsaw operator and the chainsaw operator assistant may be due to seasonal differences or increased dispersion related to the condition of the trees harvested. Trial 1 was conducted in July while trials 2 and 3 were conducted in October. Increased precipitation in October may have contributed to lower wipe sample concentrations. During the July trial, one of the downed trees selected for harvesting displayed signs of increased rot and decay compared to trees in other trials.

When the chainsaw bar penetrated the tree, substantially more debris (bark, wood chips) was observed being dispersed from the harvest process.

5.1.4. Conclusion

Results from the firewood harvesting simulations conducted within this study indicate that amphibole fibers can become liberated from trees when harvesting firewood in asbestos contaminated areas. Bark samples collected in the same area where the firewood harvesting simulations were conducted revealed substantial amphibole fiber concentrations ranging from 7 million to 97 million fibers per cm^2 of bark surface area. One hundred percent of the PBZ samples collected during the EPA restricted zone harvest simulations revealed airborne concentrations above analytical sensitivities for fibers < 5 µm, while 75% of the PBZ samples revealed detectable LA concentrations for fibers > 5 µm. It should be noted, that in the Hart et al., (2007) firewood harvest publication provided in Appendix A, the conclusion stated that the majority (21 of 24) PBZ samples revealed LA. Upon further review, since the <5 µm and > 5 µm concentrations values were derived from single air samples, it is more accurate to present these statistics as 12 samples with two different fiber counting methods. Personal breathing zone samples collected during a control harvest simulation near Missoula, MT did not detect asbestos fibers above TEM analytical sensitivities. A higher concentration of shorter fibers (< 5 µm) was observed on the PBZ air samples compared to longer fibers (> 5 µm), and the task performed by each investigator was not a factor in their PBZ exposures. The wood stackers had PBZ exposures comparable to the investigators much closer to the source; i.e., the chainsaw operator and the chainsaw operator’s assistant.

The lack of difference in exposure between the investigators indicates that the plume was not narrowly localized.

In addition to the airborne exposure potential associated with harvesting amphibole contaminated trees, there is also a strong potential for clothing contamination. One hundred percent of the wipe samples collected during the EPA restricted zone harvest simulations revealed detectable amphibole asbestos for fibers < 5 µm, while 92% of the wipe samples revealed detectable LA concentrations for fibers > 5 µm. As noted in the PBZ sample result discussion above, in the Hart et al., (2007) firewood harvest publication provided in Appendix A, the conclusion stated that the majority (23 of 24) of wipe samples collected from the investigators' chest and thigh revealed asbestos fiber contamination above the analytical sensitivity. Upon further review, it is most accurate to present these statistics as 12 samples with two different fiber counting methods.

Clothing contamination may serve as a secondary source of exposure to those that harvest amphibole contaminated wood. In addition, family members, etc., not directly exposed to asbestos during firewood harvests, may be exposed while laundering contaminated clothing. As noted with PBZ samples, there were no significant differences in wipe sample concentrations between the four investigators. And, consistent with the PBZ samples, a higher concentration of fibers < 5 µm was observed on the wipe samples compared to longer fibers (> 5 µm).

The authors recognize that the firewood harvesting simulations presented in this study represent near worst-case scenarios. The study was conducted on U.S. Forest Service land within the EPA restricted zone. This area is currently secured and not available to the public for firewood harvesting. However, areas within the Libby EPA restricted zone have historically been utilized for pubic firewood harvesting and commercial logging. Amphibole contamination in tree bark has been demonstrated in areas near Libby that are outside of the Libby EPA restricted zone. The results of this study suggest that similar exposure potentials may exist to members of the public when harvesting firewood or to commercial loggers working in the Libby area. Further studies are needed to address the degree of amphibole contamination in tree species outside of the Libby EPA restricted zone, and the related risk to members of the public as well as occupational exposure groups.

5.2. Research Aim 2: Occupational Exposure Assessment to Libby Amphibole

5.2.1. Introduction

Over 70 years of mining amphibole-contaminated vermiculite has led to LA contamination in areas surrounding the abandoned mine and in other areas throughout the town. Libby was added to the EPA's National Priorities List in October 2002. In 2005, researchers discovered that trees surrounding the former vermiculite mine served as reservoirs for AA (Ward et al., 2006). Transmission electron microscopy (TEM) analysis of bark samples from trees near the vermiculite mine yielded amphibole fiber concentrations in excess of 100 million amphibole structures per square centimeter of bark surface (s/cm^2). Contamination has also been identified in trees near transportation corridors where vermiculite was transported from Libby to processing facilities around the country (Ward, et al., 2006).

In 2006, research was conducted to assess potential exposure to LA associated with harvesting firewood within the EPA restricted zone (Hart et al., 2007). Personal breathing zone (PBZ) and Tyvek® clothing wipe samples revealed that LA was liberated from tree bark during harvesting tasks and that a potential exists for direct inhalation exposure and clothing contamination.

In September, 2007, EPA and W.R. Grace entered into an agreement to determine the nature and extent of contamination and any threat to the public health, welfare, and the environment caused by the release or threatened release of hazardous substances, pollutants, or contaminants at or from the former mine site. In 2007/2008 EPA contractors collected bark samples from forested areas surrounding the former mine site and found LA bark contamination ranging from less than the limit of detection (LOD) to

20 million s/cm^2. LA contamination on tree bark extends several kilometers (km) from the mine site outside of the EPA restricted zone (EPA, 2008,b).

Much of the land surrounding the former vermiculite mine is owned by the USDA and private logging companies. USDA Forest Service (FS) personnel frequently travel on roadways and trails in the Kootenai forest. To date, there have been no occupational exposure assessments of FS employees pertaining to LA. The purpose of this research was to evaluate the potential for occupational LA exposure as a result of FS activities in the Kootenai National Forest. The potential for LA exposure was evaluated through the analysis of PBZ samples and Tyvek® clothing wipe samples collected during and immediately after trials that simulate FS tasks.

5.2.2. Materials and Methods

5.2.2.1. Preliminary Work

Preliminary work for this research was conducted in the fall of 2007. Investigators met with FS personnel and discussed tasks typically performed (and roadways and trail systems most commonly used) in areas within an 8 km radius of the former vermiculite mine. In addition, prevailing wind data via a Windrose was obtained (MesoWest, 2008).

Tree bark samples were also collected during this time to determine if LA contamination was present in areas frequented by FS personnel near the former vermiculite mine, but outside of the EPA's restricted zone, and within prevailing wind locations from the mine. Bark samples were collected from several tree species; Tamarack (Larix laricina), Douglas fir (Pseudotsuga menziesii), and Ponderosa pine

(Pinus ponderosa) employing (Ward et al., 2006) methods. The location of each tree sampled was identified and recorded using a Garmen Etrex 12 channel global positioning system (GPS). A minimum of one 200 gram bark sample was collected from two sides of each tree approximately 1.2 m from the base. The bark was collected by prying off sections with a small pry bar and placing them in labeled plastic bags. The bags were then sealed and the pry bar was cleaned with a wet wipe after each collection. The bark samples were preserved for later analysis by TEM.

The activities selected for evaluation included: driving on roadways, walking through forested areas, performing tree measurement activities, performing trail maintenance and constructing a fire line. Tree measurement, trail maintenance and fire line construction activities were demonstrated by FS personnel in an area with no known LA contamination. Tree measurement tasks are typically performed by at least two foresters in a plot of 10 – 12 trees. Tree diameter is measured with diameter tape. Tree height is then measured by securing loggers tape to the tree surface approximately 1.2 m from the ground and walking while unrolling the tape 9 - 15 m away from the tree. A clinometer is then used to indirectly measure tree height. Along with tree diameter and tree height, tree measurement activities usually include visually evaluating all the trees in the plot for disease.

Fire line construction is performed by a minimum of four foresters. The objective of the fire line is to construct a 1 – 2 m fuel break with combustible materials cleared to a mineral soil base. The type of fire line constructed; flat scrape or cup trench, is dependent on the slope grade. The first task performed in fire line construction is removal of trees and brush. This is performed by a chainsaw operator and a brush clearer. A Pulaski tool, a comby (combination) tool and/or a Rogue hoe are then used to clear vegetation approximately 30 – 35 cm to mineral soil.

Trail maintenance activities are similar to fire line construction in that a chainsaw operator and brush clearer remove vegetation growth from the trail; however, the trail is not cleared to mineral grade soil. Trail maintenance also involves a wider corridor 2 – 3 m, and trees are limbed with the chainsaw to a height of 2.4 m to allow for transportation by horseback.

FS personnel do not currently employ the use of personal protective equipment (PPE) beyond level D when performing field tasks in the Kootenai forest. Therefore, the tasks typically conducted by FS personnel were simulated by the research team. In an effort to minimize risks associated with the task simulations, FS personnel provided training on vehicle safety procedures, emergency radio communication, procedures for minimizing hunting related risks and procedures for wild animal encounters. The investigators were also issued a radio for emergency communication. The investigators were suited in level C PPE while performing task simulations. This PPE consisted of hooded Tyvek® coveralls, neoprene gloves, Tyvek® booties, a half mask air purifying respirator with P100 filters, work boots, hard hat and orange reflective vests (during hunting season only). All investigators obtained medical clearance to wear negative pressure respiratory protection and passed quantitative fit tests within the past year. This project was approved by University of Montana’s Institutional Review Board for the Use of Human Subjects in Research.

The PPE selected for this research presented a potential heat stress risk to the investigators. This risk was minimized by conducting the task simulations in the early morning and evening hours. In addition, task durations associated with the most physical simulation, fire line construction, were minimized and adequate fluid intake and work breaks were emphasized.

5.2.2.2. Research Methods

Simulations were performed in July of 2008. The meteorological conditions during the sampling period included temperatures from 15.8 to 25.5 ^{o}C, 20-24% humidity and wind speeds from 8-18 km per hour. Morning dew condensation on vegetation was observed during early morning trials, but no measured precipitation was reported.

Two simulation trials each were performed for the following tasks: 1) driving on FS roads, 2) walking through forested areas, 3) tree measurement, and 4) fire line construction activities. In addition, one trail maintenance activity was performed. One driving simulation was also conducted in November of 2007, when preliminary data collection necessitated roadway driving. All of the simulations were conducted on FS land north and east of the former mine and EPA restricted zone (Fig. 1).

Potential LA exposure was assessed via PBZ sampling and Tyvek® clothing wipe sampling for all tasks with the exception of roadway driving. The roadways selected for the roadway driving task include FS Roads 4872 and 401 (Figure. 1). Prior to driving up these roadways from paved access ways, a 10 x 10 cm disposable manila template was secured to the rear vehicle bumper with duct tape. The template was then wiped three times with SKC Ghost wipes pre-moistened with de-ionized water. These wipes were then discarded and a 4th wipe was used to gather a pre-travel vehicle wipe. The wipe sampling protocol followed the American Society for Testing and Materials (ASTM) D

6480-05 procedures, Wipe Sampling for Settled Asbestos (ASTM, 2006). This 4th wipe was placed in a labeled plastic bag and sealed. The vehicle was then driven to the terminal destination (Figure. 1) and parked while the investigators got out of the vehicle and performed other task simulations. Other task simulations were conducted at least eight meters from the vehicle. Investigators then returned to the vehicle and drove down the roadways to the same location where the pre-travel vehicle wipe was collected. A post-travel vehicle wipe sample, employing the methods described above, was then collected and placed in a labeled plastic bag and sealed. The wipe samples were analyzed for asbestos per ASTM's D 6480-05 Method, TEM Asbestos Analysis (ASTM, 2006) by ALS Laboratories (Cincinnati, OH), a laboratory 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). Wipe samples submitted included ten percent field blanks.

The total distance driven for the FS Roadway 4872 and 401 activities were 25 and 21 km respectively. The average vehicle speed was 16-24 km per hour. Other vehicle traffic, ahead of the test vehicle, was noted for the November roadway driving assessment, and no other vehicle traffic was observed during the remaining roadway driving activities.

PBZ samples were collected during the walking, tree measurement, fire line construction, and trail maintenance simulation trials using conductive three piece asbestos sampling cassettes. The cassettes contained 25 mm 0.8 micron (µm) pore size mixed cellulose ester membrane filters. SKC Aircheck 224 sampling pumps were calibrated before and after each trial with a Bios Defender 520 primary flow meter at an average flow rate of three liters per minute (L/min). Throughout each trial, each investigator wore a sampling pump with the asbestos cassette placed in the breathing zone. PBZ samples were analyzed for fibers per National Institute for Occupational Safety and Health's Manual of Analytical Method (NMAM) 7400, Asbestos and Other Fibers by phase contrast microscopy (PCM) (NIOSH, 1994) and for asbestos per EPA's Asbestos Hazard Emergency Response Act 's (AHERA), Airborne Asbestos by TEM (EPA, 1987). AHERA requires selected area electron diffraction and energy dispersive x-ray analysis to determine mineral type and elemental composition (asbestos types). Fibers classified as "actinolite/tremolite" also included the winchite/richterite fibers characterized by Meeker et al, 2003. Asbestos structures greater than 0.5 µm long with an aspect ratio (length:width) greater than or equal to 5:1 are recorded in the AHERA analysis. Data were reported as the concentration of asbestos structures less than (<) 5 µm long and the concentration of asbestos structures greater than or equal to (≥) 5 µm long. All air samples were analyzed by ALS Laboratories. PBZ samples submitted included ten percent field blanks.

In addition to PBZ sampling, surface wipe sampling of the outer layer of Tyvek® clothing was conducted at the conclusion of each walking, tree measurement, fire line construction, and trail maintenance simulation trial. The wipe sampling protocol followed the American Society for Testing and Materials (ASTM) D 6480-05 procedures, Wipe Sampling for Settled Asbestos (ASTM, 2006). Wipes were collected with SKC Ghost wipes pre-moistened with deionized water. A 10 by 10 cm SKC disposable manila paper template was used for each wipe. A wipe sample was gathered on each investigator’s chest, forearm, and shin. The site of the chest, forearm and shin sample (right/left) was randomly selected. The three wipe samples collected for each investigator were submitted for analysis as a composite sample. In addition to the post simulation trial wipes collected, pre simulation trial wipes and ten percent field blanks were collected and analyzed. The wipe samples were analyzed for asbestos per ASTM’s D 6480-05 Method, TEM Asbestos Analysis (ASTM, 2006) by ALS.

The average duration of each activity simulation was 66 min. The fire line construction activities were conducted for 31-42 min and the remaining task durations were 70-90 min. The fire line activity was shorter in duration simply because of the physical nature of the task. An effort was made to minimize potential overloading of the PBZ filters and, as described above, a shorter duration was selected for the fire line construction activities to minimize potential heat stress hazards to the investigators.

FS personnel loaned the research team equipment in order to perform task simulations. The tools included a new Stihl Model MS361 chainsaw, Pulaski tool, comby tool, diameter tape, clinometer and forester tape. These tools were wiped with wet wipes prior to and immediately after each simulation trial. At the conclusion of the fire line construction and trail maintenance trials, and prior to equipment cleaning, one wipe sample was collected on the chainsaw bar. The wipe samples were collected using the methods described above and placed in labeled bags and sealed. The wipe samples were analyzed for asbestos per ASTM’s D 6480-05 Method, TEM Asbestos Analysis (ASTM, 2006) by ALS.

A minimum of two investigators conducted the walking simulation trials. The tree measurement simulation trials were conducted by three investigators; two investigators conducted tree diameter and height measurements, while the third investigator served as the data recorder. Fire line construction simulation trails were conducted with five investigators; one investigator each served as a chainsaw operator, brush clearer, Pulaski tool operator, comby tool operator, and data recorder. Five investigators conducted the trail maintenance simulation trials; one served as a chainsaw operator, three served as brush clearers, and one was the data recorder.

All simulation activities were performed within a 4.8 km radius of the former vermiculite mine. Fire line construction simulations were conducted near the Rainy Divide stock trail head (12S), and in a forested area northwest of FS roadway 4872. Tree measurement simulation activities were performed in the Alexander Test Site and in a forested area northwest of FS roadway 4872. Trail maintenance simulation activities were performed on the Rainy Divide Trail (12S). Walking activities were performed in the forested area northwest of FS roadway 4872 and Rainy Divide Trail (12S). The location of each simulation trial in relation to the former vermiculite mine is illustrated in Figure 1. The area selected for the majority of the simulations, near roadway 4872, is accessible by vehicle travel for approximately 8 km up roadway 4872 from the paved roadway (228) (Figure 1). Past this point, the roadway is currently restricted to general public vehicle traffic, but may be accessed by non-mechanized means or FS vehicles. The Rainy Divide stock trail head (12S) is available for general public and FS travel from the northern section of roadway 401 (Figure 1).

5.2.3. Results

5.2.3.1. Tree Bark Sampling Results

Seven bark samples collected from trees northeast of the former vermiculite mine showed substantial LA contamination, ranging from 37 thousand to 15 million structures/cm^2 of bark surface area (Table 4). These concentrations are consistent with LA contamination in tree bark previously reported by Ward et al., 2006. Fiber dimension analyses of the bark samples revealed that the majority of the asbestos fibers detected were < 5 μm long. Fibers exhibited mineral characteristics consistent with Libby amphiboles. Amphibole fibers were not detected in bark sample collected from the Missoula, MT tree (control).

Table 4. Tree bark sample results –Forest Service land northeast of the former vermiculite mine.

Location n = (8)	Tree Species	Amphibole structures / cm^2
Alexander Test Site	Tamarack	36,898
Alexander Test Site	Tamarack	158,583
Alexander Test Site	Tamarack	112,336
Rainy Divide Trail 12S	Ponderosa pine	568,137
Rainy Divide Trail 12S	Douglas Fir	12,356,979
Rainy Divide Trail 12S	Douglas Fir	15,383,941
Rainy Divide Trail 12S	Douglas Fir	13,377,926
Bark sample collected in Missoula, MT. Serves as control sample	Ponderosa pine	[a]ND

[a]ND = Non detect

5.2.3.2. Personal Breathing Zone Sampling Results

PBZ samples collected during the FS simulation activities were analyzed for asbestos by both PCM and by AHERA TEM. Table 5 and Figure 4 present individual sample and Mean PBZ air sampling results, reported for each simulation activity (fire line construction, tree measurement, trail maintenance and walking) as well as by the task(s) associated with the activity. Mean concentrations were calculated by using a value of zero for non-detect concentrations. In terms of TEM mean concentrations, this method may reflect an uncertain estimate of true mean and actual risks may be higher or lower (EPA, 2008c). Fibers were observed on all samples analyzed by PCM, excluding field blanks.

For individual PBZ FS simulation trial samples for fibers > 5 µm, 10 of 24 samples (forty-two percent) exceeded the OSHA exposure limit of 0.1 fiber per mL, assuming an eight hour exposure duration, when analyzed by PCM. These 10 PBZ samples were all collected during the fire line construction simulation activity.

A substantial portion of cellulose (from forest vegetation) fibers was expected in PCM analyses, therefore, AHERA TEM analyses were performed to describe the fiber population. In terms of fiber counts reported by the laboratory (not shown in Table 5 or Figure 4), one to five non-asbestos fibers (organic, gypsum) were identified on all PBZ AHERA TEM samples. Twenty-five percent of the PBZ samples revealed concentrations greater than the analytical sensitivity (AS) when analyzed by AHERA TEM. These samples were collected during the fire line construction and tree measurement simulation activities. AHERA TEM analyses for the concentration of asbestos fibers > 5 µm revealed that none of the samples collected exceeded the OSHA PEL, assuming an 8 hour exposure duration (not shown in Table 5).

Table 5. Forest Service occupational assessment PBZ results reported by activity performed, task associated with activity, PCM and TEM concentrations.

Test Activity	Test Activity Task	Number Of Samples	[a]Number Of Detects (PCM)	PCM Conc. (fibers/mL)	[b]Number Of Detects (TEM)	TEM Conc. < 5 µm (structure/mL)	TEM Conc. > 5 µm (structure/mL)
Fire Line		10	10		4		
	Brush Clearer			0.354		0.0277	0.0277
				0.249		ND	0.0367
	Chainsaw Operator			0.384		ND	ND
				0.242		ND	ND
	Pulaski Operator			0.438		0.0524	0.0262
				0.410		0.0332	0.0664
	Comby Operator			0.446		ND	ND
				0.238		ND	ND
	Data Recorder			0.220		ND	ND
				0.117		ND	ND
	Mean Concentrations			0.302		0.011	0.016
Trail Maintenance		5	5		0		
	Brush Clearer			0.059		ND	ND
				0.045		ND	ND
				0.024		ND	ND
	Chainsaw Operator			0.063		ND	ND
	Data Recorder			0.015		ND	ND
	Mean Concentrations			0.041		ND	ND
Tree Measurement	Tree Measurer	5	5		2		
				0.021		ND	ND
				0.035		ND	0.0162
				0.015		ND	ND
				0.038		0.0134	ND
				0.062		ND	ND
	Mean Concentrations			0.034		0.003	0.003
[c]**Walking**	Walking	4	4		0		
				0.021		ND	ND
				0.043		ND	ND
				0.011		ND	ND
				0.019		ND	ND
	Mean Concentrations			0.024		ND	ND

[a] PCM Analytical Limit of Detection = (0.009 – 0.19 f/mL)

[b] ND = Non detect, TEM analytical sensitivity = (0.0123 – 0.0367 s/mL)

[c] One walking activity PBZ sample (not reported) revealed chrysotile asbestos, not amphibole asbestos.

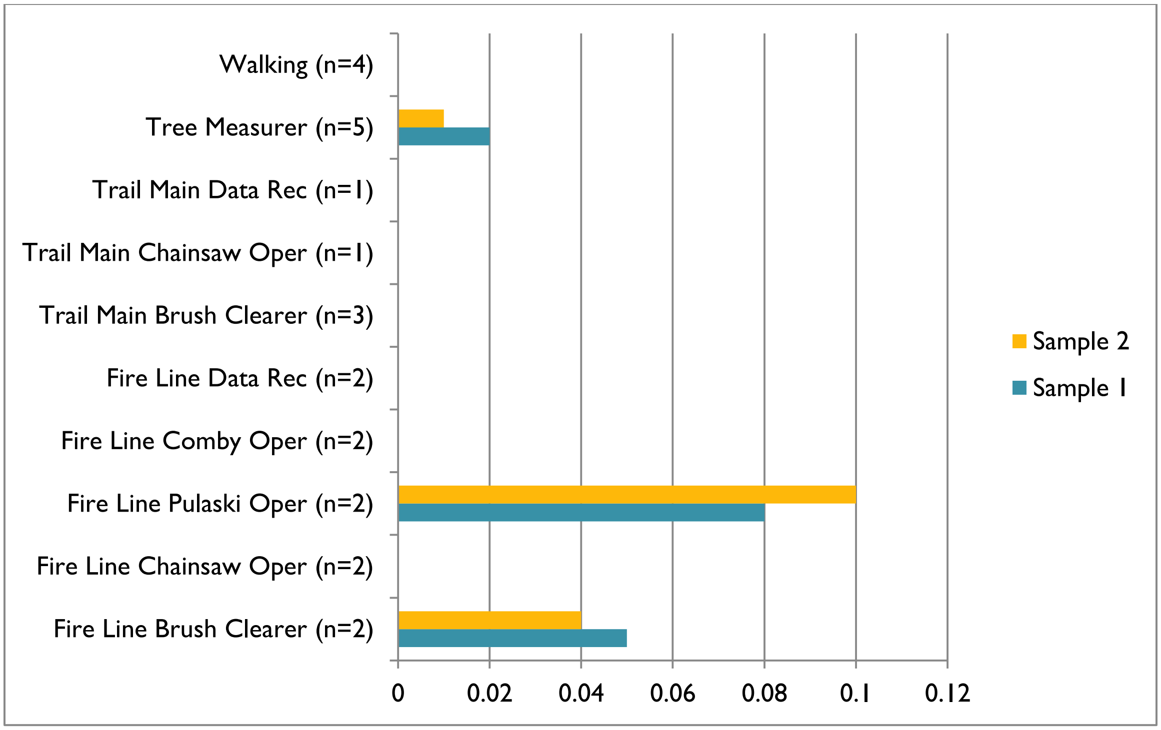

Figure 4. Forest Service occupational study individual personal breathing zone sampling results presented as total asbestos (s/mL). TEM analytical sensitivity = (0.0123 – 0.0367 s/mL)

Although the simulations for each task were conducted in two separate geographical areas (Figure 1), no differences in PBZ concentrations were observed for each simulation based on the area that the simulation activity was conducted (not shown in Table 5 of Figure 4).

The tasks that revealed PBZ concentrations greater than the AS for the fire line construction activity were brush clearer (2 of 2 samples) and Pulaski tool operator (2 of 2 samples). Two of five tree maintenance activity samples revealed concentrations greater than the AS. One of the walking activity PBZ samples revealed chrysotile asbestos (not shown in Table 5). Chrysotile asbestos is not part of the amphibole family, and this PBZ sample contamination may have been derived from sources other than the vermiculite mine.

A review of the scanning electron microscope energy dispersive x-ray spectroscopy spectra (not shown) for PBZ samples with detectable amphibole asbestos revealed measurable amounts of sodium and potassium in 100% of the samples. Recent research has demonstrated that amphiboles originating from the vermiculite deposit contain sodium and potassium that can be observed in the scanning electron microscope spectra (Gunter and Sanchez, 2009).

5.2.3.3. Wipe Sampling Results

Surface wipe sampling of the outer layer of Tyvek® clothing was conducted at the conclusion of each activity simulation trial. These wipe samples were analyzed for asbestos fibers by TEM, with summary results presented in Table 6 and Figure 5. All of the field blank and pre-activity Tyvek® wipe samples showed no asbestos contamination, and were below the AS (448 structures per cm^2) for the D 6480-05 TEM method. Fifty-two percent of post activity wipe samples revealed concentrations greater than the AS. While the concentrations of LA were associated with the fire line construction activity, LA was detected on wipe samples collected from all of the activities evaluated.

Table 6. Forest Service occupational assessment clothing wipe sample results.

Reported by activity performed, task associated with activity, individual and mean TEM concentrations.

Test Activity	Test Activity Task	Number Of Samples	[a]Number Of Detects (TEM)	TEM Conc. < 5 µm (structures/cm^2)	TEM Conc. > 5 µm (structures/ cm^2)	TEM Conc. Total Structures (structures/ cm^2)
Fire Line		8	4			
	Brush Clearer			896	ND	896
	Chainsaw Operator			ND	ND	ND
				ND	ND	ND
	Pulaski Operator			1,344	1,792	3,135
				448	448	896
	Comby Operator			896	ND	896
				ND	ND	ND
	Data Recorder			ND	ND	ND
	Mean Concentrations			448	280	728
Trail Maintenance		5	3			
	Brush Clearer			299	ND	299
				299	ND	299
				ND	ND	ND
	Chainsaw Operator			1,792	ND	1,792
	Data Recorder			ND	ND	ND
	Mean Concentrations			478	ND	478
Tree Measurement	Tree Measurer	5	4			
				ND	ND	ND
				ND	448	448
				448	ND	448
				ND	448	448
				448	448	896
	Mean Concentrations			179	269	448
[b]**Walking**	Walking	5	1			
				ND	ND	ND
				ND	ND	ND
				ND	ND	ND
				896	ND	896
				ND	ND	ND
	Mean Concentrations			179	ND	179

[a] ND = Non Detect, TEM analytical sensitivity = (448 – 896 s/cm^2)[b] one walking activity wipe sample (not reported) revealed chrysotile asbestos, not amphibole asbestos.

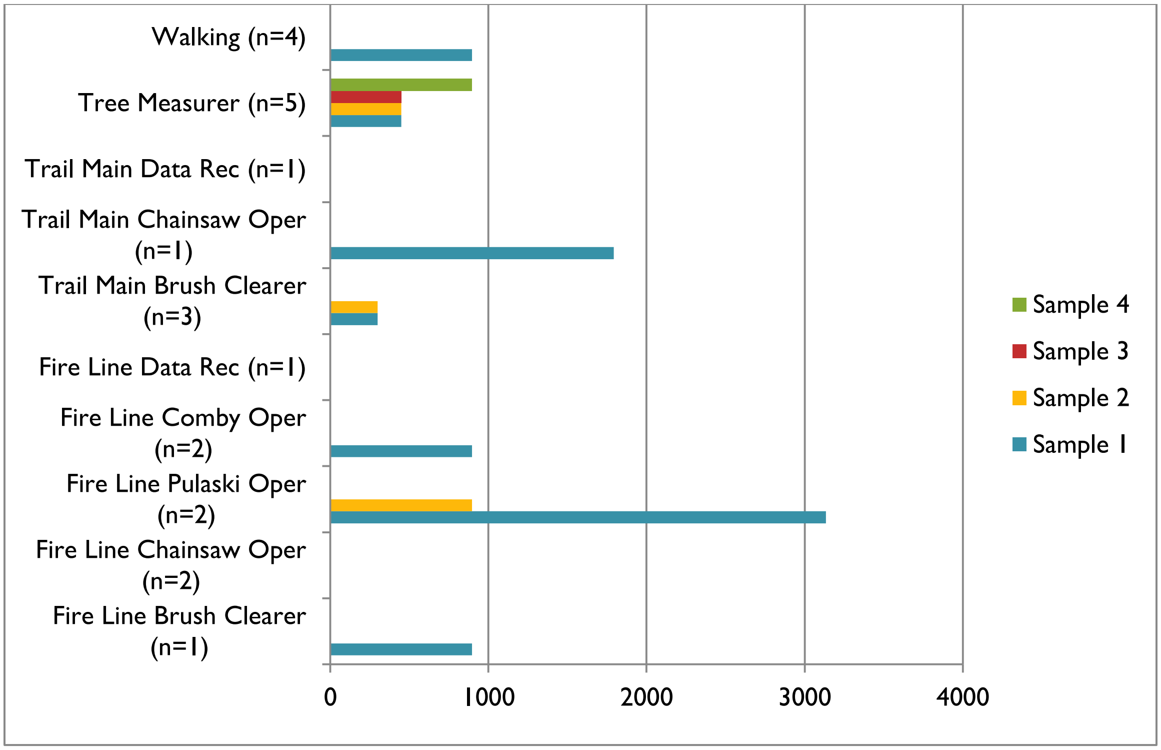

Figure 5. Forest Service occupational study Tyvek® clothing wipe sampling results presented as total asbestos (s/cm^2), TEM analytical sensitivity 448-896 s/cm^2

The tasks that revealed wipe sample concentrations greater than the AS for the fire line construction activity were brush clearer (1 of 1 sample), comby tool operator (1 of 2 samples) and Pulaski tool operator (2 of 2 samples). Four of the five tree measurement activity samples revealed concentrations greater than the AS. Two of three trail maintenance brush clearer and one of one trail maintenance chainsaw operator samples were greater than the AS, while one of five walking samples were greater than the AS. As noted with one PBZ walking sample, one of the walking activity wipe samples (not reported) revealed chrysotile asbestos. As noted previously, chrysotile asbestos is not part of the amphibole family. However, (Perkins and Harvey, 1993) also noted fibers and bundles with split ends resembling “commercial grade” asbestos as present but not common in the Rainy Creek Complex near Libby, Montana (Meeker et al., 2003).

The pre and post travel vehicle wipes collected for the FS 4872 vehicle driving activity simulations revealed concentrations below the AS for both the November and July trials. The pre travel vehicle wipe collected for the FS 401 vehicle driving activity simulation was also reported below the AS, while the post travel wipe sample revealed one amphibole fiber resulting in a concentration of 17,917 s/cm^2. The amphibole fiber detected was less than 5 µm long.

Post activity chainsaw bar wipe sample results are presented in Table 7. LA was detected on the chainsaw bar after all of the simulation activities. In terms of structure counts reported by the laboratory, 12 of 15 fibers were less than 5 µm long (not shown).

A scanning electron microscope spectra (not shown) for all wipe samples (clothing, equipment, vehicle) with detectable amphibole asbestos revealed measurable amounts of sodium and potassium in 73% of the samples.

Table 7. Post Forest Service occupational assessment chainsaw bar wipe sample results reported as TEM concentration of amphibole asbestos < 5 microns long, > 5 microns long and total structures per square centimeter.

Activity Performed	TEM (s/cm^2) < 5 µm	TEM (s/cm^2) > 5 µm	TEM (s/cm^2) Total Asbestos
Fire Line Construction	8,600	3,225	11,825
Trail Maintenance	2,688	ND[a]	2,688
Fire Line Construction	896	ND[a]	896

[a] ND = Non Detect, TEM analytical sensitivity = (896 s/cm^2)

5.2.4. Conclusions

Results from the FS activity simulations conducted within this study indicate that an exposure to LA may exist when work is performed in the Kootenai National Forest near the former vermiculite mine. Bark samples collected in the area where activity simulations were conducted revealed amphibole contamination ranging from 37 thousand to 15 million s per cm^2 of bark surface area. The lowest bark amphibole concentrations were observed in the Alexander Test Site, an area that was re-planted after a timber harvest in the early 1990s as a research plot for Tamarack trees. It is worth noting that trees in this location were planted after the vermiculite mine ceased operations. Contamination of these trees may indicate more recent dispersion of amphibole fibers. The highest bark amphibole concentrations were observed in aged Douglas Fir trees on the Rainy Divide Trail.

In terms of inhalation exposure potential associated with the FS tasks evaluated, fire line construction and tree measurement activities yielded detectable TEM PBZ concentrations. Detectable LA concentrations were not observed with trail maintenance and walking activities. Of the fire line activity tasks evaluated, the Pulaski tool operator and the brush clearer yielded the highest PBZ concentrations. PBZ concentrations for these fire line activity tasks revealed detectable LA concentrations for two separate trials conducted in two separate geographical areas. Five PBZ samples were collected for the tree maintenance activity. Of these, three samples revealed detectable LA. These three samples were also collected in two separate trials conducted in two separate geographical areas. In terms of individual fiber counts, fifty-seven percent of PBZ asbestos structures were < 5 µm long. This is consistent with other research performed regarding amphibole asbestos in tree bark (Hart et al., 2007; Ward et al., 2006).

It is worth noting that the operation of the Pulaski tool employed in fire line construction involves clearing vegetation to mineral grade soil. Therefore, it is unclear whether LA exposure associated with this task is derived from vegetation or soil sources.

In addition to the airborne exposure potential associated with FS activities, there is a potential for clothing and equipment contamination. Composite wipe samples collected from the investigators' forearm, shin, and chest revealed detectable amphibole asbestos in fifty-two percent of the samples collected. Clothing contamination was observed in samples from each of the four activities evaluated; fire line construction, tree measurement, trail maintenance, and walking. In addition, the wipe samples collected from the chainsaw bar after each trial (n = 3), revealed amphibole contamination ranging from 896 to 11,825 s/cm^2. Clothing and equipment contamination may serve as a secondary source of exposure to FS personnel. Cross contamination of vehicle cabs, vehicle boxes, equipment storage areas, equipment maintenance areas, and offices may occur as a result of clothing and equipment contamination.

Although the objective of this study was to assess the potential exposure associated with FS occupational activities, the potential for public exposure to LA cannot be ignored. Libby and the surrounding area are known for clean water, beautiful scenery, and recreational activities such as hiking, hunting boating and skiing. As noted earlier, the simulation areas are accessible to the general public. The frequency of recreational use by the general public was not evaluated in this study; however, hunters were observed near the simulation site during the bark collection phase of this study. In an effort to inform the public about the amphibole contamination in the Kootenai National

Forest, FS management have published a brochure that outlines safeguards to minimize dust generation and transport of fibers on clothing.

The forested areas near the simulation sites were historically used for timber harvests as observed by numerous clear-cut plots. In the past, FS personnel visited the Alexander Test plot and areas accessible via roadways 4872 and 401 on a weekly basis. Since the awareness of amphibole contamination in tree bark, FS travel in this area has been reduced. In addition, fire fighting in this area is currently performed from the air only.

This research was funded as a small project/pilot study in order to assess potential FS exposure to LA. A limited number of samples were collected within a relatively small geographical area. Future research is planned to assess FS exposure potentials with the activities evaluated in this study throughout a range of meteorological conditions (i.e., different seasons) as well as other activities (i.e., fire fighting), in expanded radii from the former vermiculite mine. In addition, vehicle cabs, offices and equipment storage and maintenance facilities should be evaluated for potential LA contamination.

5.3. Research Aim 3: The Impact of Weatherizing Homes with Vermiculite Insulation

5.3.1. Introduction

Weatherization practices are recommended for homes to reduce the amount of energy needed for heating and cooling (USEPA 2012d). High energy costs substantially impact low-income families who are the least equipped to improve their home’s energy efficiency (AEP 2013). Funding sources for low-income energy assistance, including weatherization, include the Department of Energy’s Weatherization Assistance Program, the Department of Health and Human Service’s (DHHS) Low Income Home Energy

Assistance Program (LIHEAP) and charitable programs associated with local utilities. In Montana, weatherization funds are directed to local Human Resource Development Councils who implement weatherization improvements in qualifying homes. An estimated 1,500 to 2,000 qualified homes are weatherized each year throughout the state.

Unfortunately, weatherization services are denied to approximately 200 high-energy-burden LIHEAP-recipient households annually due to the presence of asbestos containing materials (ACM) in their homes, either as loose-fill insulation in attics, in pipe or duct insulation, or in certain wall, ceiling and siding materials. Due to potential health and safety hazards to residents and weatherization workers, the U.S. Department of Energy weatherization rules prohibit weatherization agencies from weatherizing homes with vermiculite attic insulation (VAI) containing asbestos or other ACM that are friable or brittle and could potentially become airborne.

The Montana DPHHS Intergovernmental Human Services Bureau was the recipient of a LIEAP Residential Energy Assistance Challenge Program grant. The grant funded the Asbestos Safe Weatherization Demonstration Project. This section 5.3 of this thesis presents the results from 37 homes weatherized through the Asbestos Safe Weatherization Demonstration Project, in which the primary goal was to develop and test procedures that would allow for the safe and effective weatherization of low-income homes with asbestos.

Expanded vermiculite was used in numerous commercial products throughout the 20th century. Vermiculite is a mineral that expands when subjected to heat, forming pockets of air that made the material marketable as an insulation product and soil conditioner. The largest size or grade 1 was used as a loose-fill insulation for attic and wall cavities in dwellings and buildings, referred to here as vermiculite attic insulation (VAI). Expanded vermiculite was also used in refrigerators and furnaces (Keigel, 1940), in potting soils and other horticultural products (Potter, 1997, USEPA, 2000), as a carrier for various chemicals, including herbicides, insecticides, and fertilizers and as an aggregate for insulation plasters, and in the manufacture of fire-resistant, insulating wall-boards and acoustic tile. Very fine sizes were reported to be used as extenders in gold and bronze paints and inks due to their silver to gold colors that were produced upon expansion (Keigel, 1940).

The precise number of U.S. homes insolated with Zonolite brand VAI from the Libby mine is unknown (Gunter et al. 2005; USEPA 2011b; Zalac 2003); however, vermiculite was widely distributed via processing plants throughout the country and may be present in millions of homes nationwide (USEPA 2011b). The fact that a very large estimated number of homes were insulated with Libby vermiculite represents an ongoing potential public health hazard. 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 EPA has made such a determination under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA). As a component of the cleanup efforts under the Superfund program, EPA has removed VAI from hundreds of Lincoln county homes (USEPA 2012b). This cleanup effort has not extended into other Montana counties and currently there are no assistance programs for homeowners living in structures containing VAI outside of the Libby Superfund site.

In addition to vermiculite insulation, many homes (especially those constructed from 1930 to 1970) contain serpentine asbestos in commercial products, as asbestos has been found to be present in 3000-4000 commercial products (Dodson and Hammer 2006). Examples of products used in home construction that may contain asbestos include thermal insulation, floor tiles, roofing tiles or shingles, gaskets, ceiling texture materials, and siding (Dodson and Hammer 2006).

This research was divided into two phases. The objective of Phase I of this research described in Spear et al. (2012) was to confirm the presence of VAI and/or other ACM in homes via bulk sampling and to assess the potential for living space contamination associated with these sources. Baseline data from the Phase I study were used to develop sampling strategies, personal protective equipment (PPE) selections, and exposure control strategies for Phase II of the research reported in this manuscript. The aim of Phase II was to determine the impact of weatherization activities in asbestos laden homes on potential living space contamination and weatherization worker exposure, with the ultimate goal of developing asbestos safe weatherization protocols.

5.3.2. Previous Studies

While substantial literature exists regarding occupational asbestos exposure, limited information is available concerning asbestos exposure in residential settings (Ewing et al. 2010). The majority of studies associated with residential living space asbestos contamination have focused on exposure and related disease among household members of occupationally exposed workers (Anderson et al. 1979; Epler et al. 1980; Kilburn et al. 1985; Sider et al. 1987; NIOSH 1995; Whitehouse 2004; Miller 2005; Perez et al. 2008) or residential exposure in areas near asbestos-related industries or naturally occurring asbestos deposits (Pan et al. 2005; Reid et al. 2007; Kumaqai et al. 2010; Adgate et al. 2011).

Cowen (1997) discussed contractor asbestos exposures from a building demolition which contained VAI . The majority of bulk VAI samples collected prior to demolition revealed less than 0.1% asbestos, with detectable concentrations ranging from 0.1% to 5-10% actinolite and/or tremolite. The initial demolition work was conducted without dust suppression and air monitoring revealed asbestos concentrations ranging from 13 to 172 structures per mL (s/mL) by transmission electron microscopy (TEM).

A study (USEPA, 2003A) was conducted to estimate asbestos exposures from vermiculite insulation in containment structures and occupied and unoccupied Vermont homes with asbestos concentrations in bulk VAI samples ranging from non-detect to <0.1% by TEM. The implications of this study were that routine disturbances of vermiculite insulation by homeowners can result in asbestos exposure via inhalation of airborne fibers.

Activity-based air and surface sampling was conducted primarily in the attic spaces of three homes to evaluate amphibole asbestos exposures during specific activities in attics containing VAI (Ewing et al. 2010). Personal and area air sampling revealed significant concentrations of airborne amphibole asbestos above background concentrations when VAI was disturbed with the highest personal and area concentrations observed when VAI was moved aside with a dry sweeping method.

The above referenced studies provided initial insight into potential exposures associated with demolition of structures containing VAI and the potential for exposure associated with activities that may be performed primarily in the attic of homes with

VAI. However, the impact of weatherizing homes with VAI and/or other ACM on potential living space contamination, has not been evaluated.

5.3.3. Weatherization Measures

Typical weatherization measures may be generally divided into structure assessment or mitigation activities. Assessment measures include a general safety evaluation of the structure, an assessment of building tightness, an evaluation of combustion appliance efficiencies and emissions (furnaces, water heaters, etc.), an appraisal of current home insulation properties and estimate of insulation requirements, and a calculation of potential moisture loads. After the home has been thoroughly assessed, weatherization mitigation measures are prescribed. These include air sealing measures (sealing bypasses), insulation improvements (installing insulation in attic, crawl space, basement, and/or side walls), door and window improvements (installing weather stripping, thresholds, and/or sweeps or replacing doors; re-glazing, glass replacement, sash replacement and/or locks), indoor moisture reduction measures (adding bathroom, drier, kitchen and/or attic venting) and combustion appliance improvements (repairing or replacing furnaces, water heaters, etc).

Weatherization activities are generally not considered to cause indoor air problems by adding new airborne pollutants; however, many weatherization practices such as installing storm windows, weather stripping, caulking, and insulation, reduce the amount of outdoor air infiltrating into a home; thereby, increasing the concentrations of indoor air pollutants from sources inside the home (USEPA 2012d). Indoor air quality issues such as excessive moisture, mold growth and radon accumulation can be the result of over tightening of homes (Manuel 2011).

5.3.4. Precautions Taken

Since the homes considered for this study were known to have asbestos sources, several substantial precautionary modifications to weatherization activities were prescribed. These modifications include the following:

In the Phase I component of this project (Spear et al. 2012), researchers conducted a visual inspection of each home and collected bulk samples of VAI and/or other suspect sources of ACM when found. When the presence of asbestos was confirmed in VAI and/or other bulk sources of ACM via independent laboratory analyses, baseline air and surface sampling was performed to assess potential living space contamination. Air and surface concentrations of 0.01 f/mL (70 s/mm2) (confirmed by TEM analysis) and 10,000 structures per square centimeter (s/cm2), respectively, were adopted for this project as values, that if exceeded, required the home to be cleaned by a state licensed asbestos abatement contractor and cleared via air sampling prior to the home being considered for the Phase II component of this research.

The home occupant(s) were instructed to remain out of the home until the home was cleared via sample results. A stipend check was issued to the home occupant(s) to minimize the economic hardship associated with this requirement.

Prior to conducting any building tightness assessments, all identified potential pathways to VAI (holes or cracks in ceiling) were sealed.

To minimize the potential of drawing asbestos fibers into the living space from VAI and or other ACM, the blower test used to assess building tightness was performed under positive pressure only, not under negative pressure.

Interior attic entry ways in homes with VAI were sealed. Exterior attic entrances were constructed (if necessary) and additional fiberglass insulation was blown into the attic from this exterior access only. No VAI was removed from the homes.

No drilling or exterior wall insulation blow-in was conducted on homes with asbestos siding. If a home with asbestos siding required additional wall insulation, cellulose insulation was blown-in from internal walls within a 6 mil plastic containment structure.

All weatherization workers participating in this project first completed Asbestos Worker training. In addition, all weatherization crew supervisors completed Asbestos Inspector and Asbestos Project Contractor/Supervisor training.

All personnel on site (weatherization workers, supervisors, and researchers) received medical clearance to wear respiratory protection prior to the inception of the research. Level C personal protective equipment was worn by all personnel when conducting or when in the vicinity of weatherization measures that may disturb asbestos sources (blowing attic or wall insulation, installing bathroom ventilation fan, etc).

Living space high volume air and weatherization worker personal breathing zone (PBZ) air sampling was conducted continuously throughout the weatherization processes. In addition, at the conclusion of all weatherization activities in the home, surface wipe sampling was conducted. If any of the living space high volume air or surface wipe samples exceeded 0.01 f/mL (70 s/mm^2) (confirmed by TEM analysis) or 10,000 s/cm^2, respectively, the home was cleaned by a state LLAC and cleared via air sampling prior to permitting the occupant(s) to re-enter the home.

An additional precaution, unrelated to VAI or ACM in homes, was the practice of lead-safe weatherization protocols in all homes constructed prior to 1978 (DOE 2002; DOE 2008).

5.3.5. Methods

The objective of the sampling in this Phase II segment of the project was to evaluate the impact of weatherization to the interior living spaces of the home and to evaluate potential occupational exposures to asbestos associated with weatherizing homes with VAI or other ACM. These objectives were evaluated via high-volume air sampling, surface sampling and personal breathing zone (PBZ) sampling.

5.3.5.1. High-Volume Air Sampling

High-volume air sampling was conducted in homes during each weatherization measure using Gast Model 1532 High Flow Vacuum Pumps. A minimum of five high volume air pumps were used simultaneously and positioned throughout the living spaces of each home. Pumps were pre and post calibrated at 9.5 – 9.9 liters per minute (L/min) with a Bios Defender 510 dry cal primary flow calibrator. Each high volume air sampler was placed so that it encountered normal air circulation. Asbestos sampling cassettes fitted with 0.8 µm 25 mm mixed cellulose ester membrane (MCE) filters were positioned five to six feet above the ground at a 45 degree downward angle. Although the high-volume sampling was conducted during each weatherization measure, in order to achieve the minimum sample volume of 1200 liters, the average high-volume sample duration of 2.5 hours commonly encompassed more than one weatherization task. At the conclusion of sampling, the cassettes were removed, capped and sent to an independent laboratory. The laboratory used was accredited by the American Industrial Hygiene Association (AIHA), the National Voluntary Laboratory Accreditation Program (NVLAP), and the New York State Department of Health Environmental Laboratory Approval Program. The air samples were analyzed for asbestos per National Institute for Occupational Safety and Health's Manual of Analytical Methods (NMAM) 7400, Asbestos and Other Fibers by PCM (NIOSH, 1994a). Samples that revealed PCM concentrations greater than 0.01 f/mL were further analyzed by EPA's Asbestos Hazard Emergency Response Act (AHERA), Airborne Asbestos by TEM (USEPA, 1987). In the event that none of the area high volume air samples revealed PCM concentrations greater than 0.01 f/mL, the two highest PCM samples from each daily sample set were selected for TEM analysis. The results of the PCM samples are reported as fibers per milliliter of air sampled (f/mL). The results of the TEM samples are reported as structures per milliliter of air sampled (s/mL).

5.3.5.2. Personal Breathing Zone Sampling

The potential occupational asbestos exposure to weatherization workers associated with weatherization measures in the homes was assessed by PBZ sampling. A minimum of two workers performed weatherization measures in each home during each sample period. PBZ samples were collected with asbestos sampling cassettes containing 25 mm 0.8 µm pore size MCE filters positioned in the breathing zone of each worker.

SKC Aircheck 224 sampling pumps were calibrated pre and post each sample period with a Bios Defender 520 primary flow meter at an average flow rate of 3.0 Liters per min (L/min).

PBZ samples were analyzed by the independent laboratory for fibers per NMAM 7400. Samples that revealed PCM concentrations greater than 0.1 f/cc (OSHA's 8-hour time weighted average Permissible Exposure Limit) were further analyzed by EPA's Asbestos Hazard Emergency Response Act (AHERA), Airborne Asbestos by TEM (USEPA, 1987).

5.3.5.3. Surface Sampling

Surface wipe samples provide an indication of the potential of all weatherization measures conducted to contribute to living space contamination. Immediately prior to any weatherization measures in each home, a 20 x 20 cm piece of 0.3 ml plastic was secured to a minimum of five horizontal surfaces (interior window sills, dressers, table tops, etc.) with painters tape. A 10 x 10 cm disposable manila template was then secured on each plastic base. These plastic templates were placed in approximately the same area as the baseline surface samples collected prior to weatherization in the Phase I assessment. At the conclusion of all weatherization tasks, including cleanup of debris and dust, and removal of protective barriers, surface wipes were collected from these template locations. SKC Ghost Wipes pre-moistened with deionized water were used to collect samples from the template areas using the American Society for Testing and Materials (ASTM) D 6480-05 procedures, Wipe Sampling for Settled Asbestos (ASTM 2006). Surface wipe samples were analyzed by TEM by the independent laboratory. The results of the surface samples are reported as asbestos structures per square centimeter of surface area sampled (s/cm^2). Ten percent field blanks were submitted for the high volume air, PBZ air, and surface wipe samples.

5.3.5.4. Baseline and Clearance Concentrations Adopted for this Project

As discussed in the Section 4.3.1. of this manuscript, air and surface concentrations of 0.01 f/mL (70 s/mm^2) (confirmed by TEM analysis) and 10,000 structures per square centimeter (s/cm2), respectively, were adopted for this project as values, that if exceeded, required the home to be cleaned by a state licensed asbestos abatement contractor (LAAC) and cleared via air sampling prior to the home occupants being cleared to re-enter the home. The air concentration of 0.01 f/mL (70 s/mm^2) represents the Montana state asbestos abatement project clearance concentration (Montana Asbestos Work Practices and Procedures Manual 2005).

Little scientific research has been performed to quantify “background” surface levels typically seen in homes. The 10,000 s/cm^2 surface contamination level established for this project was not intended to be a fine line between safe and unsafe. According to USEPA (2003b), “establishing action levels based upon indoor dust levels is not straightforward. There are two primary reasons for this:

Unlike air samples, there are no established regulatory or health-based standards to guide the determinations of acceptable concentrations of asbestos in indoor dust.

The relationship between the concentration of asbestos in dust and the resultant concentration in air (the medium that actually determines human exposure and risk) is highly variable. This is because the relationship depends on a long list of different factors, most important of which is the nature and frequency of dust disturbance. This means that it is difficult to calculate a value in dust that corresponds to an acceptable level in air, and it is even harder to try to select a level in dust based on site-specific measurements." (USEPA 2003b). A review of available literature indicates that a surface may be considered "clean" when the asbestos concentration is below 1,000 s/cm2. A surface would be considered contaminated when the asbestos concentration is greater than 100,000 s/cm^2 (Millette and Hays 1994). Based on existing scientific literature, an acceptable background level for surface samples of 10,000 s/cm^2 was adopted for this research.

5.3.6. Results

5.3.6.1. Phase 1 Background Results Summary

As discussed previously, the information presented in this manuscript is considered Phase II of a larger research project. Thirty-seven homes containing asbestos sources were weatherized through this Phase II research. Originally, 46 homes were evaluated in the Phase I assessment reported in Spear et al. (2012). Nine homes were not weatherized for the following reasons; five of the homeowners or renters did not meet economic qualifications, one home could not be feasibly cleaned prior to weatherization, two homeowners requested to be dropped from the study, one homeowner passed away prior to scheduling weatherization.

Bulk sampling results obtained in Phase I of this research (Spear et al., 2012) for the 37 homes that were weatherized during Phase II are presented in Table 8. VAI was present in 32 of the 37 homes and one of the homes without VAI contained vermiculite insulation in two walls. All bulk samples of VAI analyzed contained asbestos. Twenty-six samples of bulk ACM were collected in these homes. Seventeen of these samples contained greater than one percent asbestos. The majority of positive bulk ACM samples were collected in basement areas and were chrysotile-based thermal system insulation (TSI) materials. Seven homes contained both VAI and other ACM, while four homes contained only ACM other than VAI.

Homes that revealed any living space air or surface sample from the Phase I assessment above the air and/or surface clearance concentrations adopted for this project were cleaned and cleared (via air sampling) by a LAAC prior to participation in Phase II weatherization. Twenty one homes required cleaning prior to Phase II weatherization. With the exception of one home, all of the homes requiring cleaning prior to Phase II weatherization were due to surface concentrations of chrysotile exceeding the background concentration of 10,000 s/cm^2 adopted for this project. Amphibole asbestos, associated with VAI, was detected in the living space of 26% of the homes assessed in Phase 1, while chrysotile asbestos, associated with other ACMs, was detected in the living space of 98% of the homes.

Table 8. Vermiculite attic insulation (VAI) and bulk ACM results for the 37 homes weatherized

Home	Vermiculite attic insulation (VAI) containing asbestos present	Number of bulk ACM samples collected	Number of bulk ACM samples containing asbestos (n)	Location & Analytical Results (%) from other ACM analyzed by PCM
1	PRESENT	0	0	N/A[1]
2	PRESENT	0	0	N/A
3	NO	5	3	Basement & boiler room TSI: 30-60% Chrysotile
4	NO	3	3	Basement TSI: 40-70% Chrysotile Outside Siding: 10-20% Chrysotile
5	NO	0	0	N/A
6	PRESENT	3	1	Basement TSI: 31-40% Chrysotile
7	PRESENT	3	2	Basement TSI: 40-50% Chrysotile Basement Furnace: 40-50% Chrysotile
8	PRESENT	0	0	N/A
9	NO[2]	0	0	N/A
10	PRESENT	0	0	N/A
11	PRESENT	2	0	ND
12	PRESENT	0	0	N/A
13	PRESENT	0	0	N/A
14	PRESENT	1	0	ND[3]
15	PRESENT	1	1	Basement TSI: 30-40% Chrysotile
16	PRESENT	0	0	N/A
17	PRESENT	1	1	Basement TSI: 60-70% Chrysotile
18	PRESENT	0	0	N/A
19	PRESENT	0	0	N/A
20	PRESENT	0	0	N/A
21	PRESENT	0	0	N/A
22	PRESENT	0	0	N/A
23	PRESENT	0	0	N/A
24	PRESENT	2	2	Basement TSI: 40-50% Chrysotile Outside Siding: 10-20% Chrysotile
25	PRESENT	0	0	N/A
26	PRESENT	0	0	N/A
27	PRESENT	1	1	N/A
28	PRESENT	1	1	Basement TSI: 60-70% Chrysotile
29	PRESENT	0	0	N/A
30	PRESENT	0	0	N/A
31	PRESENT	1	1	Basement TSI: 60-70% Chrysotile
32	PRESENT	0	0	N/A
33	PRESENT	0	0	N/A
34	PRESENT	0	0	N/A
35	PRESENT	0	0	N/A
36	PRESENT	1	1	Basement TSI: 40-50% Chrysotile
37	PRESENT	0	0	N/A

[1] N/A= no samples analyzed; [2] vermiculite attic insulation was not present; however, vermiculite was identified in wall cavities; [3] ND= none detected

5.3.6.2. Phase II High Volume Air Sampling Results

Airborne asbestos was detected in high-volume air samples in 28 of the 37 homes weatherized. Of these, chrysotile asbestos was detected in the air in 26 homes, and LA (including actinolite-tremolite), was detected in 14 homes. Eleven homes had both LA and chrysotile detected in the air during weatherization activities. LA was detected alone, without the presence of chrysotile, in air samples in three homes. Amosite asbestos, probably associated with TSI, was detected in three air samples from two different homes. Figure 3 shows the asbestos fiber counts by type and fiber length of asbestos detected in high-volume air samples during weatherization activities. The most common type of asbestos detected in high-volume air samples was chrysotile, and the majority of fibers detected were less than 5 um in length.

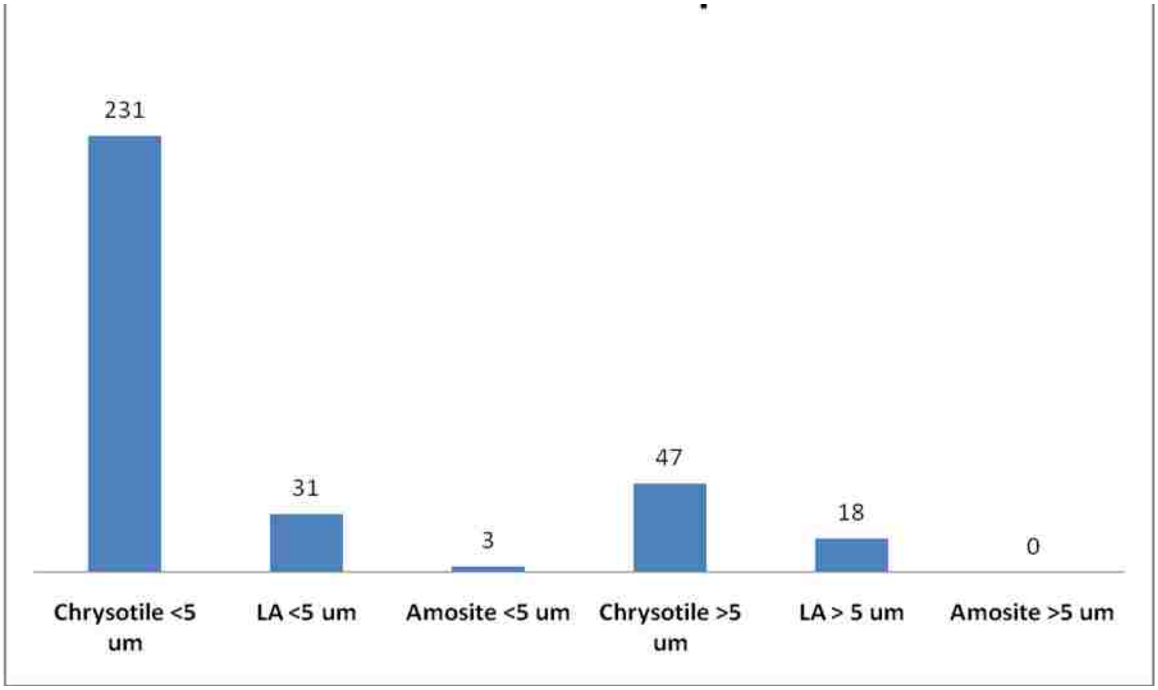

Figure 6. Weatherization high volume air sampling results: asbestos fiber counts by type and length of asbestos

Summary statistics for the high-volume air sample results are shown below in Table 9. Six hundred eighty-eight high-volume air samples (excluding field blanks) collected in the 37 homes during weatherization measures were analyzed by PCM. The mean PCM concentration for these samples was 0.053 fibers/cubic centimeter (f/cc), with a standard deviation (SD) of 0.0157 and a maximum concentration of 10.22 f/cc. Samples with PCM concentrations greater than the clearance concentration of 0.01 f/cc, were further analyzed by TEM. If none of the samples from a single-day sample set exceeded this value, the two highest PCM samples were selected for TEM analysis.

Table 9. Weatherization high-volume air sample summary results
Mean PCM (f/mL), mean TEM for asbestos structures less than 5 µm long (s/mL), mean TEM for asbestos structures greater than 5 µm long (s/mL), and mean TEM total asbestos structures (s/mL)

Sample Type	Number of Samples	Mean (s/mL)	SD[1]	Maximum
PCM f/ mL	6	0.05	0.01	10
TEM s/mL < 5 um	5	0.00	0.02	0.44
TEM s/mL ≥ 5 um	5	0.00	0.00	0.11
TEM Total s/mL	5	0.00	0.00	0.56

[1] Standard Deviation

Five hundred-nine (74%) of the PCM samples were analyzed by TEM. Of these, 107 (21%) samples revealed detectable levels of asbestos. The mean TEM concentration for asbestos fibers < 5 um was 0.0048 s/mL and the mean TEM concentration for asbestos fibers > 5 um was 0.0011 s/mL. The mean TEM concentration for total asbestos structures (< 5 um and > 5 um) was 0.0059. In direct comparison with the high-volume air clearance concentration of > 0.01 s/cc adopted for this project, 14 (2.8% of total) high-volume air samples for asbestos fibers > 5 um long exceeded this clearance concentration. Considering both short (< 5 um) and long (> 5 um) asbestos fibers, 69 (13.6%) of the total high-volume air samples exceeded this clearance concentration of > 0.01 s/mL.

Airborne asbestos was detected in 28 (76%) of the homes weatherized. Thirty four samples were greater than or equal to the air clearance level of 0.01 s/mL for short (< 5 um in length) structures and 11 samples were greater than or equal to the clearance level for long (> 5 um in length) asbestos structures. These samples were collected in seven separate homes. These homes required cleaning by a LAAC and clearance via air sampling prior to the residents re-occupying.

The weatherization measures performed during this project were categorized into six main sections: home auditing/initial blower door, sealing penetrations into attic, adding attic/kneewall insulation, adding wall insulation, final blower door and miscellaneous activities (window/door replacement, adding bathroom fan, installing basement batting, adding attic vents and cleanup). During the performance of the weatherization measures by the HRC crew, detailed field notes of the work were kept by the researchers. Based on the field notes, the six sections of weatherization measures were further divided into 16 different activities: audit, initial blower door, sealing penetrations in attic, attic blow-in, drilling holes in exterior walls, drilling holes in interior walls, exterior wall blow-in, interior wall blow-in, final blower door, HEPA vacuuming, basement batting installation, attic batting installation, door work, window work, and miscellaneous activities which included adding bathroom fan, adding attic vents or mudding access holes.

Due to the need to run the high-volume pumps for at least two hours to meet the minimum required air volume sample (1200 liters), several weatherization activities often were performed during the collection of individual high-volume air samples. Therefore, the results from the high-volume samples may be influenced by more than one weatherization activity. A one way analysis of variance (ANOVA) assessment was used to evaluate the 16 specific weatherization measures with airborne asbestos concentrations measured. The results seem to be suggestive that certain weatherization activities have a larger impact on asbestos concentrations than others. It would appear from this analysis that when interior holes are drilled for blowing wall insulation, and when insulation is blown into the walls and attic through access points inside the home, high volume air sample asbestos concentrations are elevated. Additionally, the category identified as miscellaneous also revealed elevated concentrations. However, a review of this analysis revealed that because of the complex interactions of the multiple variables, it is not possible to state statistically that one variable had a larger impact than another on the sample results collected. One important factor that cannot truly be addressed statistically is the home itself. Each home that was selected for weatherization had a different age, interior volume, condition of building materials, and variety of building materials used. It is necessary therefore, to consider each home on a separate basis when assigning risk factors to individual weatherization measures.

5.3.6.3. Phase II Personal Breathing Zone Sample Results

Personal breathing zone (PBZ) samples were collected to evaluate the potential asbestos inhalation hazard to weatherization workers associated with this research and to ensure that the appropriate personal protective equipment (PPE) was selected. PBZ sampling was conducted for the work periods and was not task specific. The mean sample duration for PBZ samples was six hours. Excluding field blanks, 246 samples were collected from the PBZ of four different weatherization crew members and analyzed by PCM. PBZ samples were collected from the crew leader, two crew leader assistants, furnace technician, and supervisors. Two hundred twelve (86%) of the PCM samples were analyzed by TEM-AHERA. Summary sample time weighted statistics for the PBZ sampling results are presented in Table 10. The majority of PBZ samples revealed greater than five non-asbestos fibers (cellulose, organic, etc.) when analyzed by TEM. Therefore, TEM concentrations for asbestos structures greater than 5 µm long may provide the most accurate exposure estimate for comparison with the 0.1 f/mL occupational exposure limit. Due to the large variability of sample volumes and analytical sensitivities, mean concentrations were calculated by using a value of zero for non-detect concentrations. In terms of TEM mean concentrations, this method may reflect an uncertain estimate of true mean and actual risks may be higher or lower (USEPA 2008c).

Table 10. Weatherization PBZ summary results
Mean PCM (f/mL), mean TEM for asbestos structures less than 5 µm long (s/mL), mean TEM for asbestos structures greater than 5 µm long (s/mL), and mean TEM total asbestos structures (s/mL)

Sample Type	Number of Samples (n=)	Mean	SD[1]	Maximum
PCM (f/ mL)	246	1.12	2.61	19.55
TEM s/mL < 5 um	212	0.29	1.76	18.60
TEM s/mL ≥ 5 um	212	0.08	0.46	5.37
TEM Total s/mL	212	0.37	2.14	21.49

[1] Standard Deviation

While the highest PBZ appeared to be associated with the crew leader and crew leader assistants, an ANOVA assessment of exposures measured for specific job tasks revealed no significant differences (alpha = 0.05) in PCM or TEM PBZ concentrations between crew members. Asbestos was detected in breathing zone samples collected at 24 separate (65%) homes. Fifty-two PBZ samples (64%) revealed chrysotile asbestos, 23 samples (28%) revealed LA and 6 samples (7%) revealed actinolite-tremolite. In terms of PBZ samples with TEM concentrations for asbestos structures greater than 5 µm exceeding the 0.1 f/mL occupational exposure limit, 12 samples (46%) revealed chrysotile asbestos, 12 samples (46%) revealed LA and 2 samples (8%) revealed actinolite-tremolite.

Initially, weatherization crew members wore half mask air purifying respirators with P100 filters for all weatherization tasks conducted near asbestos sources. However, during the course of this project, the highest LA exposures were observed on days where workers entered attics containing VAI to blow in additional fiberglass insulation. Therefore, the prescribed personal protective equipment was modified to include full face air purifying respirators for attic insulation blow in to ensure that respiratory protection maximum use concentrations were not exceeded.

5.3.6.4. Post Weatherization Surface Sampling Results

Excluding field blanks, a total of 218 surface samples were collected in numerous rooms at the conclusion of weatherization work in each home and analyzed by TEM. Summary statistics for the surface sampling results are shown in Table 11. One hundred eighty four (84%) of these surface samples revealed asbestos concentrations less than the analytical sensitivity (AS). Thirty-two surface samples revealed asbestos concentrations greater than the AS but less than the surface background level of 10,000 s/cm^2. Seventy-eight percent of these samples revealed chrysotile asbestos, while 22% revealed LA or actinolite-tremolite. Two surface samples revealed asbestos concentrations greater than the surface background level of 10,000 structures/cm^2. One sample revealed chrysotile and LA asbestos, while the other sample revealed chrysotile. These homes required cleaning by a LAAC and clearance via air sampling prior to the residents re-occupying. One home also required cleaning due to high volume air sampling concentrations exceeding the clearance level, while another home revealed surface asbestos concentrations above the clearance level without any high volume air samples exceeding clearance levels.

Table 11. Weatherization surface wipe sampling summary results
Mean and maximum TEM concentrations (structures per cm^2) for asbestos structures less than 5 µm long and greater than 5 µm and total asbestos structures

Sample Type	Number of Samples	Mean (s/cm^2)	SD[1]	Maximum (s/ cm^2)
TEM s/cm^2 < 5 um	216	471	2624	34,127
TEM TEM s/cm^2 > 5 um	216	78.8	385.1	3,413
TEM Total TEM s/cm^2	216	534	2911	37,540

[1] Standard Deviation

5.3.7. Discussion

The asbestos minerals identified in high volume air, PBZ air and surface samples in this study, listed in order of decreasing abundance by asbestos structure counts were chrysotile, LA, actinolite/tremolite and amosite. While there is substantial evidence supporting the hypothesis that chrysotile is present in numerous historic building materials (TSI, flooring, siding, etc.), and LA is present in VAI derived from the former mine near Libby, Montana, the source(s) of actinolite/tremolite and amosite are not as clear.

Recent mineralogical studies describing the amphibole asbestos minerals from Vermiculite Mountain have defined their composition as primarily winchite and richterite (Wylie and Verkouteren 2000; Gunter et al. 2003) with a decreased presence of tremolite and magnesioriebecktite (Meeker et al. 2003). Historically, amphibole minerals in Libby vermiculite were inaccurately described as tremolite or tremolite-actinolite (Larsen and Pardee 1929; Bassett 1959; Boettcher 1963) and this mischaracterization has persisted in literature (Bandli and Gunter 2006). The very similar optical properties of winchite and tremolite are hypothesized by Bandli and Gunter (2006) as an explanation for the historical misclassification.

Although tremolite has been described in literature as a minor (6%) component of the amphibole minerals from Vermiculite Mountain (Meeker et al. 2003), it is also considered a mineral component (contaminant) of Canadian chrysotile, a major source for commercially produced asbestos products (Dodson and Hammer 2006). Therefore, the source, ACM or VAI, of actinolite-tremolite in homes within this study cannot be confirmed.

Like chrysotile, amosite was used in commercial applications in the United States (Dodson and Hammer 2006). Therefore, amosite sources in homes within this study are assumed to be ACM.

The information presented in this article was derived from Phase II of a larger research project. In the Phase I baseline assessment of homes (prior to any weatherization work) (Spear et al. 2012), detectable levels of asbestos were found in 15 samples (9.5% of high volume air samples analyzed by TEM). These samples were collected in 11 separate homes. One of these samples exceeded the clearance concentration of 0.01 s/mL. Twenty-two of the 37 homes that were weatherized required cleaning and clearance by a LAAC prior to the Phase II weatherization work. With the exception of the one Phase I air sample exceeding the clearance concentration, all of these homes required cleaning by a LAAC because of elevated surface concentrations of chrysotile asbestos.

The air and wipe sampling results from the Phase II weatherization assessment are quite different from the Phase I data. In the Phase II weatherization assessment, detectable levels of asbestos were found in 106 samples (21% of the high volume air samples analyzed by TEM). These samples were collected in 28 separate homes. Forty-five (9%) of these samples exceeded the clearance concentration of 0.01 s/mL and were collected in seven separate homes. In addition to the elevated air concentrations of asbestos revealed in the Phase II weatherization sampling, the type of asbestos identified also differed. Of the seven homes that exceeded the high volume air clearance concentration, two were due to serpentine (chrysotile), two due to amphibole, and 3 due to both chrysotile and amphibole air concentrations. These Phase II high volume air sampling results correlate well with the types of asbestos identified through PBZ sampling.

The results presented in this research in comparison with results found in the Phase I baseline assessment are similar to those described by Ewing et al. (2010) and USEPA (2003) in that disturbances of VAI increased the likelihood of asbestos exposure via inhalation of airborne fibers. In addition to amphibole asbestos associated with VAI, weatherization activities also increased the potential for inhalation of chrysotile asbestos associated with ACM.

Phase I baseline sampling revealed chrysotile surface contamination above the background asbestos surface concentration of 10,000 s/cm2 in 21 homes. These 21 homes along with one additional home were cleaned and cleared via air sampling by a licensed asbestos abatement contractor prior to participating in the Phase II weatherization. Surface asbestos concentrations provide some indication of prior airborne asbestos dispersal in the home, but it is not possible to determine when or how this may have occurred. It is important to note that 56% of homes with detectable concentrations of chrysotile asbestos in Phase I surface samples contained no sources of ACM identified through visual inspection and bulk sampling. We hypothesize that historical construction materials containing ACM were not accounted for; therefore ACMs were underestimated in the homes. For example, homes may have contained external asbestos siding, flooring, etc., that were now covered by newer materials.

Whereas the Phase I baseline sampling revealed primarily surface asbestos contamination, surface asbestos concentrations exceeding the background surface concentration of 10,000 s/cm^2 were revealed in only two homes through the Phase II weatherization sampling. One of these was due to chrysotile and the other due to chrysotile and Libby amphibole. There was no correlation between the Phase II high volume air and surface sampling data. This may be due to an insufficient time allowed for dispersed asbestos fibers to settle out of the air onto surfaces. Therefore, high volume air sampling, not surface sampling, is recommended for future weatherization clearance indices.

Our study had some limitations. The 37 homes that were weatherized in this study were previously identified as containing VAI and/or ACM. Therefore, only asbestos-positive homes were considered for this project. In addition, home occupants were required to demonstrate low-income eligibility in order to participate in this study, resulting in economic bias. Additionally, all of the homes considered for this study were in Montana (Figure 2). Due to the geographical proximity of these homes to the former Libby, MT Zonolite Mine, there is a high likelihood that vermiculite in Montana homes was derived from the Libby Mine. However, because the Libby Zonolite mine supplied up to 80% of the world's vermiculite and since vermiculite processing facilities were located throughout the United States, this limitation may be insignificant. As noted previously, only the asbestos content in suspect ACM, identified through visual inspection, was quantified; therefore, the historical presence of ACM in homes may be underestimated.

In addition to the limitations described above, in order to achieve the minimum sample volume required for high volume air sampling methodologies, there were several occasions where more than one weatherization activity was performed during the collection of the high volume air samples. The high volume air sampling results where, therefore, influenced by more than one weatherization activity. This approach presented a challenge in terms of identifying weatherization measures that are most likely to disperse asbestos fibers in the home. Future research should be performed in a controlled setting with high volume air and PBZ sampling dedicated to single weatherization activities.

While additional fiberglass attic insulation was blown in to the attic spaces of all the homes in this study, VAI was not removed prior to this activity. The options available for homeowners outside of Lincoln County Montana with VAI in their homes are limited. These options include leaving the VAI in place or abating (removing) the VAI and replacing with other insulation sources, i.e., fiberglass insulation. Removal of VAI or other ACM sources in the home should only be performed by a licensed asbestos abatement contractor. Based on the results of this study as well as Ewing et al. (2010), Cowen (2007) and USEPA (2003a); there is an increased risk of asbestos exposure when VAI and/or ACM are disturbed. Future research is recommended to evaluate the impact of VAI abatement (removal) practices on potential living space contamination.

5.3.8. Conclusion

These data revealed that performing weatherization measures in homes containing asbestos has the potential to disturb asbestos-containing materials and disperse asbestos fibers into the air. This presents a risk to weatherization workers and home occupants of inhaling asbestos. Of the 37 homes weatherized in this study, six homes were found to have asbestos contamination beyond established air clearance levels, one home had surface dust contamination beyond established surface dust clearance levels and one home had both air and surface dust contamination beyond established clearance levels. All of these homes were cleaned by a licensed asbestos abatement contractor by HEPA vacuuming and wet wiping surfaces. Samples collected after cleaning were found to be within established background levels and the homeowners were allowed to reoccupy the residences.

The majority (79% and 80% respectively) of high-volume air and personal breathing zone (PBZ) air samples from this study did not reveal detectable airborne concentrations of asbestos. However enough test samples did reveal detectable concentrations that careful consideration should be given when performing weatherization work in homes with asbestos. Airborne asbestos was detected in 76% of the homes of the weatherized. Significantly, airborne asbestos was detected during numerous weatherization measures; suggesting that weatherization practices as a whole, not single weatherization activities, may contribute to the disturbance and dispersal of asbestos fibers into the air.

In summary, future weatherization practices in homes containing asbestos should include control measures similar to those described in the Precautions Taken section of this manuscript. These control measures include high volume air clearance sampling, containment practices and worker protection. Although both living space high volume air and surface sampling were employed in this research, asbestos was detected in air samples more frequently than wipe samples. This may be due to the time required for asbestos fibers to settle onto surfaces once they are liberated. Living space high volume air sampling is recommended as a clearance factor for future research.

6. Research Aims Conclusion

The primary objective of this research was to evaluate the potential for LA exposure associated with tree bark and VAI sources. While LA has most likely accumulated on tree bark throughout the 70 plus years of vermiculite mining, this reservoir for LA has only recently been discovered. Libby vermiculite was used in homes as VAI and wall insulation for several decades in the 20th century and is most likely to remain in the majority of homes. With exception of Libby, MT, limited research has been performed to assess the impact of VAI to home occupants or to individuals that may disturb insulation through residential related activities. Additionally, outside of Libby, MT, there are limited public assistance programs for homeowners living in homes with VAI.

The activities performed in Aims 1 – 3 revealed the potential to disperse LA into the breathing zone. As discussed in section 3.1, epidemiology studies considering former mine and mill workers exposed to LA have demonstrated clear and significant increases in ARD, including asbestosis, lung cancer, and mesothelioma. While several engineering modifications were made in the Zonolite Mountain facility, resulting in decreased exposures over time, concentrations exceeding 400 x's the current occupational exposure limit of 0.1 f/mL have been reported for several job classifications (Amandus et al., 1987). These exposures are substantially higher than those revealed in Aims 1 -3. In addition, the duration of exposure experienced by former mine and mill workers is likely to be considerably higher than public firewood harvesters, USDA FS workers that spend a portion of their work week in the forested area near the former vermiculite mine, or weatherization workers that may encounter a home with VAI. Individual susceptibility is also a factor in assessing the likelihood of disease.

Compounding this issue, it is important to note that ARDs have been observed in Libby area residents with no direct occupational exposures and that the most common health outcome, pleural changes, has been demonstrated in Libby residents and the Marysville, OH cohort with low lifetime cumulative fiber exposure levels (Peipins, 2003; Rohs et al., 2008).

The prevalence of pleural abnormalities increases substantially with increasing exposure pathways according to community health screenings (Peipins et al., 2003). Unless an aggressive control option is implemented, LA tree bark contamination in the forested areas near the former vermiculite is likely to remain for decades. In addition, VAI is likely to remain in homes throughout the United States. Identifying pathways of exposure associated with these sources and prescribing methods to minimize exposure is a critical step to minimizing the risk of ARD or pleural changes. Conclusions specific to the 3 research Aims are presented in the following sections 6.1 and 6.2.

6.1. Kootenai National Forest Activity Based Exposure Assessments

The firewood harvest simulations conducted as Aim 1 revealed a strong potential for LA exposure associated with all harvesting activities in the Kootenai National Forest and support the Aim 1 hypothesis that there is a potential for airborne LA exposure and clothing contamination associated with this activity. This conclusion is based on the fact that LA was detected in all breathing zone and clothing wipe samples.

Tree bark, soil and duff measurements surrounding the former vermiculite mine suggest a concentration gradient, with the highest LA concentrations reported in closest proximity to the mine (USEPA, 2008a). As illustrated in Figure 1, the Aim 1 firewood harvest simulations were conducted over one mile from the former mine. It is anticipated that firewood harvests conducted closer to the mine may reveal higher breathing zone and clothing contamination concentrations, while those conducted further from the mine may reveal lower concentrations than what was observed with this research.

There were no differences in LA concentrations measured in the breathing zone of the chainsaw operator, operator assistant, and wood stacker, implying that the plume was equally distributed among the tasks. Therefore, the individuals stacking wood are likely to have similar exposures to the chainsaw operator. Other potential variables that may influence breathing zone and clothing LA concentrations are the season of the year when firewood is harvested and the condition of the trees harvested. Wipe samples from the firewood harvest trial conducted in July revealed 6 times higher LA concentrations than those collected during the October trials. This may be due to increased precipitation on vegetation in the fall compared to summer months or due to condition of trees harvested. Dead trees with signs of decay or rot generated more visible particulate matter than those with minimal decay.

At the time of the proposed Aim 2 research, the USEPA bark, soil and duff sampling program had not been concluded. Aim 2 research extended into the Kootenai National Forest outside of the USEPA restricted zone established at the time of this research. We postulated that LA concentrations in tree reservoirs would decrease as the distance from the source (the former vermiculite mine) decreased. Preliminary bark sampling, reported in Hart et al., (2009) and section 5.2 of this document, confirmed this hypothesis.

While the Aim 2 research confirmed the potential for airborne LA exposure and clothing contamination associated with USDA FS employee activities, thereby, supporting the Aim 2 hypothesis, the exposure potential was lower than what was revealed in the Aim 1 firewood harvest study. Personal breathing zone sampling revealed detectable LA in 25% of the samples analyzed by TEM versus 100% in the firewood harvest study. We postulate that this is most likely associated with reduced, but still substantial (37,000 – to 15,000,000 structures/cm^2 compared to 2.5 -110 million structures/cm^2) concentrations of LA from tree bark (Hart et al., 2009; Ward et al., 2006) as well as soil and duff sources. In addition to the concentration of LA in bark, the type of activity performed was an important variable. Activities that are most likely to result in breathing zone exposures are those that disturb vegetation as well as soil. The highest inhalation exposures were observed during fireline construction where both vegetation and soil is disturbed and during tree measurement activities where close contact to tree bark is necessary in order to measure tree diameters. Simply walking through the forest did not reveal detectable levels of LA in the breathing zone. However, all activities revealed the potential for clothing contamination. This contamination may serve as a secondary source of LA exposure to co-workers or family members (NIOSH, 1995).

In the firewood harvest and USDA FS occupational assessment, the majority (> 60 %) of LA fibers detected in air, Tyvek® clothing wipe and equipment wipe samples were < 5 µm in length. This is consistent with ambient air monitoring trends reported for Libby (ATSDR, 2003). It is hypothesized that LA was dispersed from the 70 plus years of mining at vermiculite mountain and intercepted, impacted or diffused on tree bark surfaces. The LA fiber characteristics described in Aims 1 and 2 as well as Ward et al. (2006) may provide insight into the fiber characteristics associated with historic dispersions. Future exposure assessments should consider concentrations of fibers less than and greater than 5 µm to provide the most accurate exposure estimates.

Personal breathing zone sampling conducted in Aims 1 and 2 were compared to occupational exposure limits due to the lack of any relevant non-occupational exposure limit at the time of the studies. Considering the OSHA, NIOSH, and ACGIH time weighted average exposure limit of 0.1 f/mL for an 8 hour exposure duration, the analytical sensitivities for the PBZ PCM and TEM analyses were 0.009 – 0.19 f/mL and 0.012 – 0.037, respectively. As discussed in section 2.8 of this document, at the time of this thesis, a LA reference concentration 2×10^{-5} f/mL has been proposed by the USEPA. This RfC is considerably lower than the analytical sensitivities reported for Aims 1 and 2.

Therefore, it is speculated that a portion of the samples reported as non-detect for the Aims 1 and 2 studies may have revealed measureable concentrations if a lower analytical sensitivity were applied, such as an analytical sensitivity applicable to the proposed RfC. If data were applied directly from the firewood harvest and occupational study in Aims 1 and 2 to assess potential non-cancer health risks associated with these activities, there is a potential error of substantially under estimating the risks.

As a result of this research and additional activity-based exposure assessments sponsored by the USEPA, several Superfund and USDA FS work practice modifications have been made. These include extending the OU3 boundary from the original location in close proximity to the former mine to the Lake Koocanusa boundary (Figure1), modifying USDA FS work practices to minimize tree and soil disturbance including extending the boundary by which fires are fought by air only, enhancing USDA FS personal protective equipment policies, and conducting bark sampling prior to timber sales in areas within a few miles of the former mine.

It is likely that the LA contamination on tree surfaces surrounding the former vermiculite mine will remain for decades. Therefore, best management practices should include avoiding activities that are most likely to disturb trees and soil, conducting activities in the area during months with substantial precipitation, and identifying alternative sites for firewood harvesting.

6.2. Weatherizing Homes with Vermiculite Insulation

The majority (79% and 80% respectively) of high volume and PBZ air samples from the Aim 3 weatherization study presented in section 5.3 of this thesis did not reveal detectable concentrations of airborne asbestos. However, the high volume air samples that did reveal detectable asbestos concentrations were collected in 28 (76%) of the 37 homes assessed in this study. Therefore, these results support the hypothesis that weatherization activities in homes with VAI and/or other ACM would result in detectable concentrations of asbestos in airborne samples and surface dust samples.

While the focus of this study was weatherization of homes, similar results may occur when vermiculite or other asbestos containing materials are disturbed through home renovation, energy efficiency blower door testing, home new construction additions, etc. Control measures, such as containment strategies, living space high volume clearance sampling, and worker protection are recommended for future weatherization activities or any activity with the potential to disturb asbestos sources in the home.

The focus of this study was weatherizing homes with VAI, however, because of the age of homes evaluated in this study (majority constructed prior to 1960), commercial based chrysotile asbestos was anticipated and included in the air and surface analyses. Bulk samples of suspect ACM (chrysotile sources) and VAI were collected and confirmed through PLM analyses in Phase I. Of the 37 homes that were weatherized, 33 (89%) contained VAI, 11 (30%) contained other ACMs and 7 (19%) contained both VAI and ACM. Although the majority homes contained vermiculite sources, the majority (65%) of the asbestos structures detected in living space high volume air samples were chrysotile and 35% were amphiboles. Therefore, careful consideration should be given to minimize disturbance of potential chrysotile sources in homes as well as vermiculite amphibole sources.

Homes in this study with VAI that revealed obvious pathways to living spaces such as holes or cracks in the ceiling were repaired (sealed) prior to any weatherization activities. Similar measures are recommended in any home containing VAI. While the structural features of the home are important in terms of minimizing LA dispersal from VAI sources, occupant activities, such as storing materials in the attic or renovation work, may also contribute to living space contamination. Homeowners should take necessary precautions to minimize potential pathways into living spaces. These include sealing any obvious holes or crack in the ceiling, sealing interior attic hatches, and avoiding entrance into the attic. If attic entrance is critical for the safety of the structure, appropriate containment practices should be employed to minimize dispersion into the living space and personal protective equipment such as Tyvek® clothing and respiratory protection should be worn.

As discussed in the Introduction of this dissertation, the precise number of homes containing VAI is unknown. While over 1,000 residential and commercial properties have been cleaned through Superfund activities in Lincoln County, no formal action has been initiated concerning the numerous properties with VAI outside of Lincoln County. The exposure potential associated with VAI, especially associated activities that may disturb this insulation, warrant attention from a U.S. public health standpoint.

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7. Appendix A. Research Aims 1 and 2 refereed publications

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Evaluation of Asbestos Exposures during Firewood-Harvesting Simulations in Libby, MT, USA—Preliminary Data

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Research was conducted in order to assess potential exposure to asbestos while harvesting firewood from amphibole-contaminated trees near Libby, MT, USA. Three firewood-harvesting simulations took place in the summer and fall of 2006 in the Kootenai Forest inside the US Environmental Protection Agency (EPA) restricted zone surrounding the former W.R. Grace vermiculite mine. Another simulation was conducted near Missoula, MT, USA, which served as the control. The work practices following each simulation were consistent throughout each trial. Personal breathing zone (PBZ) asbestos concentrations were measured by phase contrast microscopy (PCM) and transmission electron microscopy (TEM). Surface wipe samples of personal protective clothing were measured by TEM. The mean (n = 12) PBZ PCM sample time-weighted average (TWA) concentration was 0.29 fibers per milliliter, standard deviation (SD = 0.54). A substantial portion (more than five fibers per sample) of non-asbestos fibers (cellulose) was reported on all PBZ samples (excluding field blanks) when analyzed by TEM. The mean (n = 12) PBZ TEM sample TWA concentration for amphibole fibers <5-μm long was 0.15 fibers per milliliter (SD = 0.21) and the mean (n = 12) PBZ TEM concentration for amphibole fibers >5-μm long was 0.07 fibers per milliliter (SD = 0.08). Substantial amphibole fiber concentrations were revealed on Tyvek® clothing wipe samples. The mean concentration (n = 12) was 29 826 fibers per square centimeter (SD = 37 555), with 91% (27 192 fibers per square centimeter) comprised fibers <5-μm long. There were no significant differences in PBZ and wipe sample concentrations among the tasks performed by four investigators. Each of these three simulations were consistent in demonstrating that amphibole fibers are released from tree reservoirs during firewood-harvesting activities in asbestos-contaminated areas and that the potential for exposure exists during such activities.

Keywords: amphibole; asbestos; electron microscopy; fibers; Libby; Montana; tree bark

INTRODUCTION

For 70 years, a mining operation located 7 miles northeast of Libby, MT, USA, may have supplied 80% of the world's vermiculite (USEPA, 2007). In the early 1920s, Dr Edward Alley founded the Zonolite Company in Libby. Soon after, the mine and processing facility at Vermiculite Mountain (also known as Zonolite Mountain) was developed. W.R. Grace purchased the site in 1963 and continued operation of the mine until 1990.

Vermiculite was mined and processed primarily for use as building insulation and as a soil conditioner. However, in the case of the Libby ore, it proved to be a hazardous resource. Throughout the deposit, veins of vermiculite ore were contaminated with a toxic form of naturally occurring fibrous, asbestiform amphibole (Pardee and Larsen, 1929). Today, many areas surrounding the abandoned surface mine and decommissioned processing facilities are contaminated with amphibole fibers as well as are many of the homes within the Libby area.

Former Libby mine and mill workers exposed to amphibole fibers have a high incidence of pleural plagues, asbestosis, lung cancer and mesothelioma

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(McDonald *et al.*, 1986; Amandus and Wheeler, 1987; Amandus *et al.*, 1987; Dearwent *et al.*, 2000; Peipins, 2003). The relationship between mesothelioma and asbestos exposure has been adequately explored, with at least 70% of mesothelioma cases reported in direct correlation to asbestos exposure (Hammond *et al.*, 1965; McDonald and McDonald, 1977, 1980; McDonald *et al.*, 1986; NCI, 2005). Furthermore, asbestosis mortality in the Libby area was found to be 40–80 times higher than the expected; and lung cancer was found to be 20–30% higher than the expected (ATSDR, 2003).

It has recently been discovered that tree bark samples collected within the town of Libby, within the EPA-restricted mine area and along the railroad corridor west of town also contain varying levels of amphibole contamination (Ward *et al.*, 2006). Analyses to date have yielded substantial amphibole fiber concentrations ranging from 41 to 530 million fibers per gram of bark, while a bark sample collected ∼11 kilometers west of town along the railroad line had concentrations of 19 million fibers per gram. A conversion of these mass-based concentrations to areal concentrations (to reflect surface area contamination) revealed concentrations in excess of 100 million amphibole fibers per square centimeter.

In addition to vermiculite mining, much of the economy in Libby has historically been supported by the harvesting and processing of timber. Western Montana logging companies own ∼315 000 acres of land surrounding the Libby mine that could potentially be harvested. Because firewood is the cheapest source of fuel in the Libby area, it is the most common source of residential heating during the cold Libby winters. There are an estimated 1300 wood stoves in use in Libby, with at least some of the firewood being harvested within the Libby valley and surrounding forests.

Previous results from tree bark sampling suggest a potential for asbestos exposure to those who harvest or disturb contaminated wood within the Libby area (Ward *et al.*, 2006). Despite the reliance on local timber resources in Libby, currently no definitive efforts exist to evaluate the potential for asbestos exposure during the common practice of harvesting firewood for residential home heating. The research within this study presents preliminary data that evaluate the potential of amphibole exposure associated with firewood harvesting within a known asbestos-contaminated area. Research trials were conducted inside the Libby EPA-restricted zone where amphibole contamination in tree bark was previously demonstrated (Ward *et al.*, 2006).

METHODS

During the summer and fall of 2006, three separate firewood-harvesting simulations were conducted on US Forest Service property in an area of the Kootenai Forest inside the EPA-restricted zone surrounding the former W.R. Grace vermiculite mine. These trials were conducted ∼30 to 35 m off of Rainy Creek road ∼1.5 km up Rainy Creek road from Highway 37 (Fig. 1). Another simulation was conducted near Missoula, MT, USA (∼4 h southeast of Libby) to serve as a control.

All the investigators participating in this study completed a 40-h Occupational Safety and Health Administration (OSHA) Hazardous Waste Operations and Emergency Response course or demonstrated competency via education and/or professional certifications (industrial hygiene PhD, certified industrial hygienist). In addition, investigators participated in a training/planning session developed specifically for the harvest simulations. A site safety and health plan was also written and submitted to the Libby EPA supervisor for approval. All investigators obtained medical clearance to wear negative pressure respiratory protection and passed quantitative fit tests within the past year.

Trees selected for the harvesting simulation at each site consisted of three to four standing dead and three to four downed trees. The location of each simulation site was identified and recorded using a Garmen Etrex 12 channel global positioning system. Tree species were identified and documented. Prior to harvesting, a minimum of one 200-gm bark sample was collected from two sides of each tree ∼1.2 m from the base. Additional bark samples were collected from randomly selected harvested trees at 1.2 m intervals from the base to the treetop. The bark was collected by prying off sections with a small pry bar and placing them in labeled plastic bags. The bags were then sealed and labeled and the pry bar was cleaned with a wet wipe after each collection. The bark samples were preserved for later analysis by transmission electron microscopy (TEM).

New Poulan® model 3416 gas chain saws were used for each research simulation trial. The chain saw was replaced prior to each trial in order to avoid cross-contamination and to ensure that the condition of the chain (sharpness) remained consistent. The harvesting simulation process at each site consisted of downing the tree, removing tree branches and sawing the log into 30-cm-long blocks. The blocks were then gathered and stacked in a pile ∼20 m away. Four to five investigators participated in each simulation trial, with the work practices employed by each investigator remaining consistent throughout each of the trials. One investigator operated a chain saw, while a second investigator assisted the chain saw operator by clearing debris, moving downed trees and holding downed trees steady while being sawed. Two investigators gathered the wood blocks and stacked them into piles. An additional investigator was present for trials 2 and 3 and served as a data recorder.

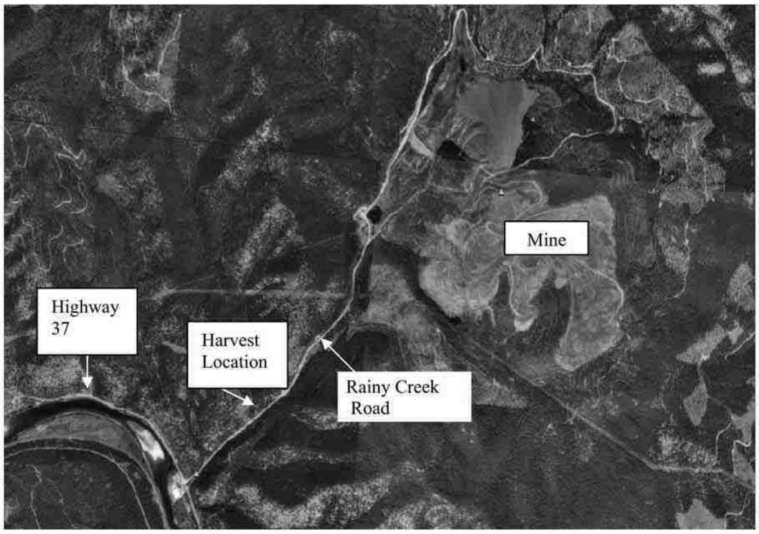

Fig. 1. Location of the 2006 firewood-harvesting simulations conducted off Rainy Creek road, near the former vermiculite site in the EPA-restricted zone near Libby, MT, USA. The distance from Highway 37 to the Harvest Location was ~1.5 km.

Personal breathing zone (PBZ) samples were collected during the trials using non-conductive three-piece asbestos sampling cassettes. The cassettes contained 25 mm, 0.8 μm pore size mixed cellulose ester membrane filters. SKC Aircheck 224 sampling pumps were calibrated before and after each trial with a Gilian® Gilibrator 2 primary flow meter at a flow rate of 4 l min^{-1}. Throughout each trial, each investigator wore a sampling pump with the asbestos cassette placed in the breathing zone. PBZ samples were analyzed for fibers per National Institute for Occupational Safety and Health's Manual of Analytical Method 7400, Asbestos and Other Fibers by phase contrast microscopy (PCM) (NIOSH, 1994) and for asbestos per EPA's Asbestos Hazard Emergency Response Act's (AHERA), Airborne Asbestos by TEM (USEPA, 1987). AHERA requires selected area electron diffraction and energy dispersive X-ray analysis to determine mineral type and elemental composition (asbestos types). AHERA analysis was enhanced by recording individual fiber dimensions rather than classifying them into two size categories. Fibers classified as 'actinolite/tremolite' also included the winchite/richterite fibers characterized by Meeker *et al.* (2003).

All air samples were analyzed by DataChem Laboratories (Cincinnati, OH, USA), a laboratory accredited by the American Industrial Hygiene Association (PCM), the National Voluntary Laboratory Accreditation Program (TEM) and the New York State Department of Health Environmental Laboratory Approval Program (PCM and TEM). PBZ samples submitted included 10% field blanks.

In addition to PBZ sampling, surface wipe sampling of the outer layer of Tyvek® clothing was conducted at the conclusion of each trial. The wipe sampling protocol followed the American Society for Testing and Materials (ASTM) D 6480-05 procedures, Wipe Sampling for Settled Asbestos (ASTM, 2006). Wipes were collected with SKC Ghost Wipes pre-moistened with deionized water. A 10 by 10 cm SKC disposable manila paper template was used for each wipe. A wipe sample was gathered on each investigator's chest and upper thigh. The site of the chest wipe sample and thigh sample (right/left) was randomly selected. The two wipe samples collected for each investigator were submitted for analysis as a composite sample. In addition to the post-harvest wipes collected, pre-harvest wipes, inner layer Tyvek wipes and 10% field blanks were analyzed. The wipe samples were analyzed for asbestos per ASTM's D 6480-05 Method, TEM Asbestos Analysis (ASTM, 2006) by DataChem.

The average duration of each firewood harvest simulation was 89 min, with 45–50 min dedicated to the harvest simulations and the remaining time associated with bark collection. The harvest duration was limited to minimize the potential to overload the sample media.

RESULTS AND DISCUSSION

Multiple tree bark samples were collected from standing dead or fallen trees selected for harvesting during both the control harvest in Missoula and the firewood-harvesting simulations conducted within the Libby restricted zone. The samples were collected from common coniferous tree types [lodgepole pine (*Pinus contorta*), ponderosa pine (*Pinus ponderosa*), larch (*Larix occidentalis*) and Douglas fir (*Pseudotsuga menziesii*)] found within the area and are representative of the types of trees typically burned during residential home heating in western Montana. Amphibole fibers were not detected in bark samples collected from Missoula, MT, USA. Eight bark samples analyzed to date from the Libby EPA-restricted zone (collected in the same area where the firewood-harvesting simulations were conducted) revealed substantial amphibole fiber concentrations ranging from 7 to 97 million fibers per square centimeter of bark surface area. These concentrations are consistent with amphibole contamination in tree bark previously reported by Ward *et al.* (2006). Fiber dimension analyses of the bark samples revealed that the majority of the asbestos fibers detected were <5 microns in length. Results from the bark samples collected in these trials showed that all identified fibers were typical of the Libby vermiculite amphibole contaminants, with typical elemental composition of Si > Mg > Ca > Fe > Na > K (Meeker *et al.*, 2003). There were no length-based differences in the elemental composition of fibers.

PBZ samples collected during the firewood-harvesting trials were analyzed for asbestos by both PCM and AHERA TEM. Fibers were observed on all samples analyzed by PCM, excluding field blanks. The PCM fiber concentrations from the control (Missoula) trial ranged from 0.01 to 0.02 fibers per milliliter. The National Institute for Occupational Safety and Health (NIOSH) PCM method cannot identify fiber types (Dodson and Hammar, 2006), but AHERA TEM analysis revealed fibers in the control samples to be organic, non-asbestos (cellulose), with no asbestos concentrations above the AHERA TEM analytical sensitivity (AS) of (0.009–0.01 structures per milliliter).

Table 1 presents PBZ air sampling results, including the mean PBZ asbestos concentrations (measured by PCM and AHERA TEM, respectively) and the standard deviation (SD) from the three harvest trials per task (chain saw operator, operator assistant and wood stackers 1 and 2). While the PBZ sample from the chain saw operator's assistant revealed the highest mean total asbestos concentration (column 5, Table 1), overall no significant differences were observed in PBZ asbestos concentrations between tasks.

Differences were observed in the concentration of shorter fibers (<5 μm long) compared to the concentration of longer fibers (>5 μm long) ($P = 0.055$) for PBZ air samples. The mean concentration of asbestos fibers <5 μm long for all samples gathered from the Libby EPA-restricted zone trials was 0.15 fibers per milliliter, SD = 0.21, while mean concentration of asbestos fibers >5 μm long for all samples gathered from the Libby EPA-restricted zone trials was 0.07 fibers per milliliter SD = 0.08 (row 6, Table 1). Three of 12 analyses for fibers >5 μm long from the Libby EPA-restricted zone trials revealed concentrations that were less than the AS of 0.0148, 0.0145 and 0.0148 fibers per milliliter, respectively. In order to perform statistical analysis on concentrations that were less than the AS, a value equal to the AS divided by the square root of 2 was used (Hornung and Reed, 1990).

In terms of fiber counts reported by the laboratory (not shown), 69% of the fibers collected on PBZ samples were <5 μm long. This is consistent with ambient air sampling trends reported for Libby (ATSDR, 2003).

Due to the lack of public exposure limits for asbestos applicable to this situation, PBZ concentrations were compared with occupational exposure limits. The current occupational 8-h time-weighted average (TWA) exposure limit for asbestos is 0.1 fiber per milliter for fibers >5 μm in length, with an aspect ratio (length:width) ≥3:1, as determined by PCM (OSHA, ACGIH, 2001). The NIOSH recommended exposure limit for asbestos is identical except that it is based on a 10-h TWA (NIOSH). In addition to

Table 1. Mean PBZ air sampling results reported in fibers per milliliter (f/ml) and SDs from three firewood harvest simulation trials conducted in the EPA-restricted zone near Libby, MT, USA

Task performed	Mean PCM sample TWA (f/ml)	Mean TEM sample TWA (f/ml) <5 μm	Mean TEM sample TWA (f/ml) >5 μm	Mean TEM sample TWA (f/ml) total asbestos
Chain saw operator, $n = 3$	0.72 (1.06)	0.07 (0.03)	0.04 (0.03)	0.11 (0.06)[a]
Operator assistant, $n = 3$	0.26 (0.32)	0.26 (0.37)	0.14 (0.14)	0.40 (0.51)[a]
Wood stacker 1, $n = 3$	0.07 (0.06)	0.09 (0.12)	0.04 (0.05)[b]	0.13 (0.17)
Wood stacker 2, $n = 3$	0.12 (0.10)	0.19 (0.24)	0.05 (0.07)[c]	0.24 (0.31)
Total mean for all tasks, $n = 12$	0.29 (0.54)	0.15 (0.21)	0.07 (0.08)	0.22 (0.29)

Results are reported by task performed (chain saw operator, operator assistant and wood stackers 1 and 2).

[a] One of three samples had loose material on the filter and was prepared using an indirect preparation method.

[b] Two samples were less than the AS of 0.0145 and 0.0148 structures per milliliter, respectively. AS divided by the square root of 2 was used to calculate mean concentration.

[c] One sample was less than the AS of 0.0148 structures per milliliter. AS divided by the square root of 2 was used to calculate mean concentration.

the TWA permissible exposure limit, OSHA has defined an excursion limit of 1.0 fiber per milliliter averaged over a sampling period of 30 min.

For individual PBZ harvest trial samples for fibers >5 μm (not shown in Table 1), two of three samples from both the chain saw operator and the operator's assistant exceeded the OSHA exposure limit of 0.1 fiber per milliliter, assuming an 8-h exposure duration, while one of three PBZ samples from both of the wood stackers exceeded the OSHA exposure limit assuming an 8-h exposure duration when analyzed by PCM.

A substantial portion of cellulose (from sawdust) fibers was expected in PCM analyses. AHERA TEM analyses were performed to describe the fiber population. In terms of fiber counts reported by the laboratory (not shown in Table 1), more than five non-asbestos fibers (organic, gypsum) were identified on all PBZ AHERA TEM samples. AHERA TEM analyses for the concentration of asbestos fibers >5 μm revealed that 3 of 12 samples exceeded the OSHA PEL, assuming an 8-h exposure duration (not shown in Table 1). These samples were collected on the chain saw operator's assistant and wood stackers 1 and 2 during the firewood harvest trial 3.

The current regulatory methods of counting fibers based on fiber length and aspect ratio may not adequately describe the risk of asbestos-related health effects. Fiber size, shape and composition contribute collectively to health risks in ways that are currently being evaluated (ATSDR, 2003). Although we compared concentrations of asbestos >5 μm to occupational exposure limits, the concentrations of fibers <5 μm may contribute to health risks.

Surface wipe sampling of the outer layer of Tyvek clothing was conducted at the conclusion of each trial. These wipe samples were analyzed for asbestos fibers by TEM with results presented in Table 2. All the field blank, inner layer Tyvek and pre-harvest outer layer Tyvek wipe samples showed no asbestos contamination and were below the AS (878 structures per square centimeter) for the D 6480-05 TEM method. There was a striking difference between the sizes of the asbestos fibers (length) measured from the suits following the firewood-harvesting simulations, with significant concentrations of the shorter fibers (<5 μm) found when compared to longer fibers (>5 μm in length) ($P = 0.038$). The mean concentration of asbestos fibers <5 μm long for all Libby restricted zone post-harvest wipe samples was 27 192 fibers per square centimeter, SD = 36 749. The mean concentration of fibers >5 μm in length for all Libby restricted zone post-harvest wipe samples (and for each job description) was more consistent compared to the smaller fibers detected, with 2634 fibers per square centimeter (SD = 1983) measured. One of 12 wipe sample analyses for fibers >5 μm long revealed concentrations that were less than the AS of 5270 fibers per square centimeter. In order to perform statistical analysis on concentrations that were less than the AS, a value equal to AS divided by the square root of 2 was used (Hornung and Reed, 1990).

Wipe samples collected from the chain saw operator and the chain saw operator's assistant after harvest trial 1 showed concentrations of asbestos fibers <5 μm long at least six times the asbestos wipe concentrations measured from the two wood stackers

Table 2. TEM wipe sampling results from three firewood harvest simulation trials conducted in the Libby EPA-restricted zone near Libby, MT, USA

Task performed	Harvest trial	TEM (f/cm^2) <5 μm	TEM (f/cm^2) >5 μm	TEM (f/cm^2) total asbestos
Chain saw operator	1	100 123	3726[a]	103 849
	2	4830	878	5708
	3	15 848	4953	20 801
Operator assistant	1	108 905	3513	112 418
	2	5709	439	6148
	3	14 134	2827	16 961
Wood stacker 1	1	16 863	2108	18 971
	2	5709	439	6148
	3	14 135	3392	17 527
Wood stacker 2	1	6324	2108	8432
	2	6587	439	7026
	3	27 140	6785	33 925
Total mean for all tasks, $n = 12$		27 192 (36 749)	2634 (1983)	29 826 (37 555)

Results are reported by task performed (chain saw operator, operator assistant and wood stackers 1 and 2). f/cm^2 = fibers per centimeter square.

[a] One sample was less than the AS of 5270 structures per square centimeter. AS divided by the square root of 2 was used to calculate mean concentration.

(column 3, Table 2). However, this same trend was not observed for harvest trials 2 and 3. There were no statistically significant differences observed in wipe asbestos concentrations between the four investigators.

CONCLUSION

Results from the firewood-harvesting simulations conducted within this study indicate that amphibole fibers can become liberated from trees when harvesting firewood in asbestos-contaminated areas. Bark samples collected in the same area where the firewood-harvesting simulations were conducted revealed substantial amphibole fiber concentrations ranging from 7 to 97 million fibers per square centimeter of bark surface area. The majority of the PBZ samples collected during the EPA-restricted zone harvest simulations showed concentrations above analytical sensitivities (21 of 24 samples), while PBZ samples collected during a control harvest simulation did not detect asbestos fibers above TEM analytical sensitivities. A higher concentration of shorter fibers (<5 µm) was observed on the PBZ air samples compared to longer fibers (>5 µm), and the task performed by each investigator was not a factor in their PBZ exposures. The wood stackers had PBZ exposures comparable to the investigators much closer to the source; i.e. the chain saw operator and the chain saw operator's assistant. The lack of difference in exposure between the investigators indicates that the plume was not narrowly localized.

In addition to the airborne exposure potential associated with harvesting amphibole-contaminated trees, there is also a strong potential for clothing contamination. Wipe samples collected from the investigators' chest and thigh revealed asbestos fiber contamination above the AS in 23 of 24 samples. Clothing contamination may serve as a secondary source of exposure to those that harvest amphibole-contaminated wood. In addition, family members, etc., not directly exposed to asbestos during firewood harvests, may be exposed while laundering contaminated clothing. As noted with PBZ samples, there were no significant differences in wipe sample concentrations between the four investigators. And, consistent with the PBZ samples, a higher concentration of fibers <5 µm was observed on the wipe samples compared to longer fibers (>5 µm).

The authors recognize that the firewood-harvesting simulations presented in this study represent near worst-case scenarios. The study was conducted on US Forest Service land within the EPA-restricted zone. This area is currently secured and not available to the public for firewood harvesting. However, areas within the Libby EPA-restricted zone have historically been utilized for public firewood harvesting and commercial logging. Amphibole contamination in tree bark has been demonstrated in areas near Libby that are outside of the Libby EPA-restricted zone. The results of this study suggest that similar exposure potentials may exist to members of the public when harvesting firewood or to commercial loggers working in the Libby area. Further studies are needed to address the degree of amphibole contamination in tree species outside of the Libby EPA-restricted zone and the related risk to members of the public as well as occupational exposure groups.

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Research Article

An Evaluation of Potential Occupational Exposure to Asbestiform Amphiboles near a Former Vermiculite Mine

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Amphibole asbestos (AA) has been detected on the surface of tree bark in forests neighboring an abandoned vermiculite mine near Libby, Montana. In the present study, simulations were performed to assess potential AA exposure associated with United States Department of Agriculture Forest Service (FS) occupational activities. Bark samples were collected prior, and personal breathing zone (PBZ) and Tyvek clothing wipe samples were collected during and immediately after trials that simulated FS activities. Transmission electron microscopy (TEM) analyses revealed AA bark concentrations up to 15 million structures per square centimeter (s/cm^2). AA was detected in 25% of the PBZ TEM samples. AA was detected on wipe samples collected from all activities evaluated. This research demonstrates the potential for airborne exposure and transport of AA in the Kootenai National Forest. These findings are especially relevant to those that work in the area and to the general public who may conduct recreational activities.

1. Introduction

Libby, Montana (population ~2700, 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. While the Libby vermiculite had useful insulating and soil conditioning properties, ore from the mine (in operation from the 1920s–1990) was contaminated with fibrous and nonasbestiform amphiboles in veins throughout the deposit [1]. Approximately 30–40% of the amphiboles are asbestiform and include winchite, richterite, tremolite, and magnesioriebeckite; differing in their relative proportions of cations (Mg, Ca, Fe, Na, K) [2–7].

Over 70 years of mining amphibole-contaminated vermiculite has led to amphibole asbestos (AA) contamination in areas surrounding the abandoned mine and in other areas throughout the town. Libby was added to the Environmental Protection Agency's (EPA) National Priorities List in October 2002. In 2005, researchers discovered that trees surrounding the former vermiculite mine served as reservoirs for AA [8]. Transmission electron microscopy (TEM) analysis of bark samples from trees near the vermiculite mine yielded amphibole fiber concentrations in excess of 100 million amphibole structures per square centimeter of bark surface (s/cm^2). Contamination has also been identified in trees near transportation corridors where vermiculite was transported from Libby to processing facilities around the country [8].

In 2006, research was conducted to assess potential exposure to AA associated with harvesting firewood within the EPA-restricted zone [9]. Personal breathing zone (PBZ) and Tyvek clothing wipe samples revealed that AA was liberated from tree bark during harvesting tasks and that a potential exists for direct inhalation exposure and clothing contamination.

In September, 2007, EPA and W.R. Grace entered into an agreement to determine the nature and extent of contamination and any threat to the public health, welfare, and the environment caused by the release or threatened release of hazardous substances, pollutants, or contaminants at or from the former mine site. In 2007/2008 EPA contractors collected bark samples from forested areas surrounding the former mine site and found AA bark contamination ranging from less than the limit of detection (LOD) to 20 million s/cm^2. AA contamination on tree bark extends several kilometers (km) from the mine site outside of the EPA restricted zone [10].

Occupational exposure to AA is associated with significant increases in asbestosis, lung cancer, and pleural cancer compared to the rest of the U.S. population [11]. High incidences of asbestos-related disease have been reported in former mine and mill workers [12–14]. While asbestos-related disease in the general Libby population has also been reported, risk associated with lower level exposures has not been as clearly defined. 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 6668 participants [15]. Although the focus of the [15] study was to describe lung abnormalities in the general Libby population, significant factors for predicting pleural abnormalities included occupational pathways [16]. Additional occupational and nonoccupational mesothelioma cases have been identified since the end of the last follow-up [17–19], and current mortality figures indicate one new case per year in Lincoln County, Montana. For the last five-year period for which figures are available (2000–2004), there were five mesothelioma deaths (two female) in Lincoln County, making it the third-highest county in the USA in terms of age-adjusted death rate per million population at 56.1 [20].

Much of the land surrounding the former vermiculite mine is owned by the United States Department of Agriculture (USDA) and private logging companies. USDA Forest Service (FS) personnel frequently travel on roadways and trails in the Kootenai forest. To date, there have been no occupational exposure assessments of FS employees pertaining to AA. The purpose of this research was to evaluate the potential for occupational AA exposure as a result of FS activities in the Kootenai National Forest. The potential for AA exposure was evaluated through the analysis of PBZ samples and Tyvek clothing wipe samples collected during and immediately after trials that simulate FS tasks.

2. Material and Methods

2.1. Preliminary Work. Preliminary work for this research was conducted in the fall of 2007. Investigators met with FS personnel and discussed tasks typically performed (and roadways and trail systems most commonly used) in areas within an 8 km radius of the former vermiculite mine. In addition, prevailing wind data via a Windrose were obtained [21].

Tree bark samples were also collected during this time to determine if AA contamination was present in areas frequented by FS personnel near the former vermiculite mine, but outside of the EPA's restricted zone, and within prevailing wind locations from the mine. Bark samples were collected from several tree species: Tamarack (Larix laricina), Douglas fir (Pseudotsuga menziesii), and Ponderosa pine (Pinus ponderosa) employing [8] methods. The location of each tree sampled was identified and recorded using a Garmen Etrex 12 channel global positioning system (GPS). A minimum of one 200 gram bark sample was collected from two sides of each tree approximately 1.2 m from the base. The bark was collected by prying off sections with a small pry bar and placing them in labeled plastic bags. The bags were then sealed and the pry bar was cleaned with a wet wipe after each collection. The bark samples were preserved for later analysis by TEM.

The activities selected for evaluation included driving on roadways, walking through forested areas, performing tree measurement activities, performing trail maintenance, and constructing a fire line. Tree measurement, trail maintenance, and fire line construction activities were demonstrated by FS personnel in an area with no known AA contamination. Tree measurement tasks are typically performed by at least two foresters in a plot of 10–12 trees. Tree diameter is measured with diameter tape. Tree height is then measured by securing loggers tape to the tree surface approximately 1.2 m from the ground and walking while unrolling the tape 9–15 m away from the tree. A clinometer is then used to indirectly measure tree height. Along with tree diameter and tree height, tree measurement activities usually include visually evaluating all the trees in the plot for disease.

Fire line construction is performed by a minimum of four foresters. The objective of the fire line is to construct a 1-2 m fuel break with combustible materials cleared to a mineral soil base. The type of fire line constructed, flat scrape or cup trench, is dependent on the slope grade. The first task performed in fire line construction is removal of trees and brush. This is performed by a chainsaw operator and a brush clearer. A Pulaski tool, a comby (combination) tool, and/or a Rogue hoe are then used to clear vegetation approximately 30–35 cm to mineral soil.

Trail maintenance activities are similar to fire line construction in that a chainsaw operator and brush clearer remove vegetation growth from the trail; however, the trail is not cleared to mineral grade soil. Trail maintenance also involves a wider corridor 2-3 m, and trees are limbed with the chainsaw to a height of 2.4 m to allow for transportation by horseback.

FS personnel do not currently employ the use of personal protective equipment (PPE) beyond level D when performing field tasks in the Kootenai forest. Therefore, the tasks typically conducted by FS personnel were simulated by the research team. In an effort to minimize risks associated with the task simulations, FS personnel provided training on vehicle safety procedures, emergency radio communication, procedures for minimizing hunting related risks, and procedures for wild animal encounters. The investigators were also issued a radio for emergency communication. The investigators were suited in level C PPE while performing task simulations. This PPE consisted of hooded Tyvek coveralls, neoprene gloves, Tyvek booties, a half mask air purifying respirator with P100 filters, work boots, hard hat, and orange reflective vests (during hunting season only). All investigators obtained medical clearance to wear negative pressure respiratory protection and passed

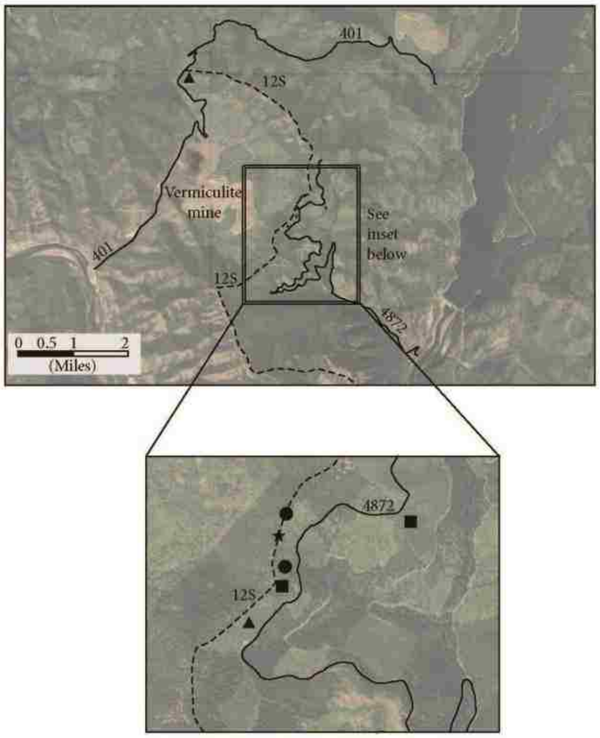

Figure 1: Map illustrating the location of Forest Service activity task simulations in relation to the former vermiculite mine. United States Department of Agriculture Forest Service Roadways 4872 and 401 and Rainy Divide Trail/Loop 12S are identified. Simulation activities are identified via the following symbols: fire line construction: triangle; tree measurement: square; walking: circle; and trail maintenance: star.

quantitative fit tests within the past year. This project was approved by the University of Montana's Institutional Review Board for the Use of Human Subjects in Research.

The PPE selected for this research presented a potential heat stress risk to the investigators. This risk was minimized by conducting the task simulations in the early morning and evening hours. In addition, task durations associated with the most physical simulation, fire line construction, were minimized and adequate fluid intake and work breaks were emphasized.

2.2. Research Methods. Simulations were performed in July of 2008. The meteorological conditions during the sampling period included temperatures from 15.8 to 25.5 °C, 20%–24% humidity, and wind speeds from 8–18 km per hour. Morning dew condensation on vegetation was observed during early morning trials, but no measured precipitation was reported.

Two simulation trials each were performed for the following tasks: (1) driving on FS roads, (2) walking through forested areas, (3) tree measurement, and (4) fire line construction activities. In addition, one trail maintenance activity was performed. One driving simulation was also conducted in November of 2007, when preliminary data collection necessitated roadway driving. All of the simulations were conducted on FS land north and east of the former mine and EPA-restricted zone (Figure 1).

Potential AA exposure was assessed via PBZ sampling and Tyvek clothing wipe sampling for all tasks with the exception of roadway driving. The roadways selected for the roadway driving task include FS Roads 4872 and 401 (Figure 1). Prior to driving up these roadways from paved access ways, a 10 × 10 cm disposable Manila template was secured to the rear vehicle bumper with duct tape. The template was then wiped three times with SKC Ghost wipes premoistened with deionized water. These wipes were then discarded and a 4th wipe was used to gather a pretravel vehicle wipe. The wipe sampling protocol followed the American Society for Testing and Materials (ASTM) D 6480-05 procedures, Wipe Sampling for Settled Asbestos [22]. This 4th wipe was placed in a labeled plastic bag and sealed. The vehicle was then driven to the terminal destination (Figure 1) and parked while the investigators got out of the vehicle and performed other task simulations. Other task simulations were conducted at least eight meters from the vehicle. Investigators then returned to the vehicle and drove down the roadways to the same location where the pretravel vehicle wipe was collected. A posttravel vehicle wipe sample, employing the methods described above, was then collected and placed in a labeled plastic bag and sealed. The wipe samples were analyzed for asbestos per ASTM's D 6480-05 Method, TEM Asbestos Analysis [22] by ALS Laboratories (Cincinnati, OH), a laboratory 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). Wipe samples submitted included ten percent field blanks.

The total distances driven for the FS Roadway 4872 and 401 activities were 25 and 21 km, respectively. The average vehicle speed was 16–24 km per hour. Other vehicle traffic, ahead of the test vehicle, was noted for the November roadway driving assessment, and no other vehicle traffic was observed during the remaining roadway driving activities.

PBZ samples were collected during the walking, tree measurement, fire line construction, and trail maintenance simulation trials using conductive three piece asbestos sampling cassettes. The cassettes contained 25 mm 0.8 micron (μm) pore size mixed cellulose ester membrane filters. SKC Aircheck 224 sampling pumps were calibrated before and after each trial with a Bios Defender 520 primary flow meter at an average flow rate of three liters per minute (L/min). Throughout each trial, each investigator wore a sampling pump with the asbestos cassette placed in the breathing zone. PBZ samples were analyzed for fibers per National Institute for Occupational Safety and Health's Manual of Analytical Method (NMAM) 7400, Asbestos and Other Fibers by phase contrast microscopy (PCM) [23], and for asbestos per EPA's Asbestos Hazard Emergency Response Act's (AHERA), Airborne Asbestos by TEM [24]. AHERA requires selected area electron diffraction and energy dispersive X-ray analysis to determine mineral type and elemental composition (asbestos types). Fibers classified as "actinolite/tremolite" also included the winchite/richterite fibers characterized by Meeker et al. [6]. Asbestos structures greater than 0.5 μm long with an aspect ratio (length : width) greater than or equal to 5 : 1 are recorded in the AHERA analysis. Data were reported as the concentration of asbestos structures less than ($<$) 5 μm long and the concentration of asbestos structures greater than or equal to ($\geq$) 5 μm long. All air samples were analyzed by ALS Laboratories. PBZ samples submitted included ten percent field blanks.

In addition to PBZ sampling, surface wipe sampling of the outer layer of Tyvek clothing was conducted at the conclusion of each walking, tree measurement, fire line construction, and trail maintenance simulation trial. The wipe sampling protocol followed the American Society for Testing and Materials (ASTMs) D 6480-05 procedures, Wipe Sampling for Settled Asbestos [22]. Wipes were collected with SKC Ghost wipes premoistened with deionized water. A 10 by 10 cm SKC disposable Manila paper template was used for each wipe. A wipe sample was gathered on each investigator's chest, forearm, and shin. The site of the chest, forearm, and shin sample (right/left) was randomly selected. The three wipe samples collected for each investigator were submitted for analysis as a composite sample. In addition to the postsimulation trial wipes collected, presimulation trial wipes and ten percent field blanks were collected and analyzed. The wipe samples were analyzed for asbestos per ASTM's D 6480-05 Method, TEM Asbestos Analysis [22] by ALS.

The average duration of each activity simulation was 66 minutes. The fire line construction activities were conducted for 31–42 minutes and the remaining task durations were 70–90 minutes. The fire line activity was shorter in duration simply because of the physical nature of the task. An effort was made to minimize potential overloading of the PBZ filters and, as described above, a shorter duration was selected for the fire line construction activities to minimize potential heat stress hazards to the investigators.

FS personnel loaned the research team equipment in order to perform task simulations. The tools included a new Stihl Model MS361 chainsaw, Pulaski tool, comby tool, diameter tape, clinometer, and forester tape. These tools were wiped with wet wipes prior to and immediately after each simulation trial. At the conclusion of the fire line construction and trail maintenance trials, and prior to equipment cleaning, one wipe sample was collected on the chainsaw bar. The wipe samples were collected using the methods described above and placed in labeled bags and sealed. The wipe samples were analyzed for asbestos per ASTM's D 6480-05 Method, TEM Asbestos Analysis [22] by ALS.

A minimum of two investigators conducted the walking simulation trials. The tree measurement simulation trials were conducted by three investigators; two investigators conducted tree diameter and height measurements, while the third investigator served as the data recorder. Fire line construction simulation trails were conducted with five investigators; one investigator each served as a chainsaw operator, brush clearer, Pulaski tool operator, comby tool operator, and data recorder. Five investigators conducted the trail maintenance simulation trials; one served as a chainsaw operator, three served as brush clearers, and one was the data recorder.

All simulation activities were performed within a 4.8 km radius of the former vermiculite mine. Fire line construction simulations were conducted near the Rainy Divide stock trail head (12S) and in a forested area northwest of FS roadway 4872. Tree measurement simulation activities were performed in the Alexander Test Site and in a forested area northwest of FS roadway 4872. Trail maintenance simulation activities were performed on the Rainy Divide Trail (12S). Walking activities were performed in the forested area northwest of FS roadway 4872 and Rainy Divide Trail (12S). The location of each simulation trial in relation to the former vermiculite mine is illustrated in Figure 1. The area selected for the majority of the simulations, near roadway 4872, is accessible by vehicle travel for approximately 8 km up roadway 4872 from the paved roadway (228) (Figure 1). Past this point, the roadway is currently restricted to general

Table 1: Tree bark sample results—Forest Service land northeast of the former vermiculite mine.

Location $n = (8)$	Tree Species	Amphibole structures/cm^2
Alexander Test Site	Tamarack	36,898
Alexander Test Site	Tamarack	158,583
Alexander Test Site	Tamarack	112,336
Rainy Divide Trail 12S	Ponderosa pine	568,137
Rainy Divide Trail 12S	Douglas Fir	12,356,979
Rainy Divide Trail 12S	Douglas Fir	15,383,941
Rainy Divide Trail 12S	Douglas Fir	13,377,926
Bark sample collected in Missoula, MT. Serves as control sample	Ponderosa pine	[a]ND

[a] ND = Nondetect.

public vehicle traffic but may be accessed by nonmechanized means or FS vehicles. The Rainy Divide stock trail head (12S) is available for general public and FS travel from the northern section of roadway 401 (Figure 1).

3. Results

3.1. Tree Bark Sampling Results. Seven bark samples collected from trees northeast of the former vermiculite mine showed substantial AA contamination, ranging from 37 thousand to 15 million structures/cm^2 of bark surface area (Table 1). These concentrations are consistent with AA contamination in tree bark previously reported by Ward et al. [8]. Fiber dimension analyses of the bark samples revealed that the majority of the asbestos fibers detected were $<5\,\mu$m long. Fibers exhibited mineral characteristics consistent with Libby amphiboles. Amphibole fibers were not detected in bark sample collected from the Missoula, MT tree (control).

3.2. PBZ Sampling Results. PBZ samples collected during the FS simulation activities were analyzed for asbestos by both PCM and by AHERA TEM. Table 2 presents individual sample and Mean PBZ air sampling results, reported for each simulation activity (fire line construction, tree measurement, trail maintenance, and walking) as well as by the task(s) associated with the activity. Mean concentrations were calculated by using a value of zero for nondetect concentrations. In terms of TEM mean concentrations, this method may reflect an uncertain estimate of true mean and actual risks may be higher or lower [25]. Fibers were observed on all samples analyzed by PCM, excluding field blanks.

The current occupational 8-hour time weighted average (TWA) exposure limit for asbestos is 0.1 fiber per mL for fibers $>5\,\mu$m long, with an aspect ratio greater than or equal to 3 : 1, as determined by PCM (OSHA, ACGIH, 2001). The National Institute for Occupational Safety and Health (NIOSH) recommended that exposure limit for asbestos is identical except that it is based on a 10-hour TWA (NIOSH). In addition to the TWA permissible exposure limit, OSHA has defined an excursion limit of 1.0 fiber per mL averaged over a sampling period of 30 minutes.

For individual PBZ FS simulation trial samples for fibers $>5\,\mu$m, 10 of 24 samples (forty-two percent) exceeded the OSHA exposure limit of 0.1 fiber per mL, assuming an eight-hour exposure duration, when analyzed by PCM. These 10 PBZ samples were all collected during the fire line construction simulation activity.

A substantial portion of cellulose (from forest vegetation) fibers was expected in PCM analyses; therefore, AHERA TEM analyses were performed to describe the fiber population. In terms of fiber counts reported by the laboratory (not shown in Table 2), one to five nonasbestos fibers (organic, gypsum) were identified on all PBZ AHERA TEM samples. Twenty-five percent of the PBZ samples revealed concentrations greater than the analytical sensitivity (AS) when analyzed by AHERA TEM. These samples were collected during the fire line construction and tree measurement simulation activities. AHERA TEM analyses for the concentration of asbestos fibers $>5\,\mu$m revealed that none of the samples collected exceeded the OSHA PEL, assuming an 8-hour exposure duration (not shown in Table 2).

Although the simulations for each task were conducted in two separate geographical areas (Figure 1), no differences in PBZ concentrations were observed for each simulation based on the area that the simulation activity was conducted (not shown in Table 2).

The tasks that revealed PBZ concentrations greater than the AS for the fire line construction activity were brush clearer (2 of 2 samples) and Pulaski tool operator (2 of 2 samples). Two of five tree maintenance activity samples revealed concentrations greater than the AS. One of the walking activity PBZ samples revealed chrysotile asbestos (not shown in Table 2). Chrysotile asbestos is not part of the amphibole family, and this PBZ sample contamination may have been derived from sources other than the vermiculite mine.

A review of the scanning electron microscope (SEM) energy dispersive X-ray spectroscopy (EDS) spectra (not shown) for PBZ samples with detectable amphibole asbestos revealed measurable amounts of sodium and potassium in 100% of the samples. Recent research has demonstrated that amphiboles originating from the vermiculite deposit contain sodium and potassium that can be observed in the SEM-EDS spectra [5].

Table 2: PBZ data reported by activity performed, task associated with activity, PCM and TEM individual, and mean sample concentrations.

Test activity	Test activity task	Number of samples	[a]Number of detects (PCM)	PCM conc. (fibers/mL)	[b]Number of detects (TEM)	TEM conc. <5 μm (structures/mL)	TEM conc. >5 μm (structures/mL)	TEM conc. total structures (structures/mL)
Fire Line		10	10		4			
	Brush Clearer			0.354		0.0277	0.0277	0.0544
				0.249		ND	0.0367	0.0367
	Chainsaw Operator			0.384		ND	ND	ND
				0.242		ND	ND	ND
	Pulaski Operator			0.438		0.0524	0.0262	0.0786
				0.410		0.0332	0.0664	0.0996
	Comby Operator			0.446		ND	ND	ND
				0.238		ND	ND	ND
	Data Recorder			0.220		ND	ND	ND
				0.117		ND	ND	ND
	Mean Concentrations			0.302		0.011	0.016	0.027
Trail Maintenance		5	5		0			
	Brush Clearer			0.059		ND	ND	ND
				0.045		ND	ND	ND
				0.024		ND	ND	ND
	Chainsaw Operator			0.063		ND	ND	ND
	Data Recorder			0.015		ND	ND	ND
	Mean Concentrations			0.041		ND	ND	ND
Tree Measurement	Tree Measurer	5	5		2			
				0.021		ND	ND	ND
				0.035		ND	0.0162	0.0162
				0.015		ND	ND	ND
				0.038		0.0134	ND	0.0134
				0.062		ND	ND	ND
	Mean Concentrations			0.034		0.003	0.003	0.006
[c]Walking	Walking	4	4		0			
				0.021		ND	ND	ND
				0.043		ND	ND	ND
				0.011		ND	ND	ND
				0.019		ND	ND	ND
	Mean Concentrations			0.024		ND	ND	ND

[a] PCM Analytical Limit of Detection : (0.009 – 0.19 f/mL). [b]ND : Non detect, TEM analytical sensitivity : (0.0123 – 0.0367 s/mL). [c]One walking activity PBZ sample (not reported) revealed chrysotile asbestos, not amphibole asbestos.

3.3. Wipe Sampling Results. Surface wipe sampling of the outer layer of Tyvek clothing was conducted at the conclusion of each activity simulation trial. These wipe samples were analyzed for asbestos fibers by TEM, with summary results presented in Table 3. All of the field blank and preactivity Tyvek wipe samples showed no asbestos contamination and were below the AS (448 structures per cm^2) for the D 6480-05 TEM method. Fifty-two percent of postactivity wipe samples revealed concentrations greater than the AS. While the concentrations of AA were associated with the fire line construction activity, AA was detected on wipe samples collected from all of the activities evaluated.

The tasks that revealed wipe sample concentrations greater than the AS for the fire line construction activity were brush clearer (1 of 1 sample), comby tool operator (1 of 2 samples), and Pulaski tool operator (2 of 2 samples). Four of the five tree measurement activity samples revealed concentrations greater than the AS. Two of three trail

Table 3: Clothing wipe sample data reported by activity performed, task associated with activity, individual, and mean TEM concentrations.

Test Activity	Test Activity Task	Number Of Samples	[a]Number Of Detects (TEM)	TEM Conc.<5 μm (structures/cm^2)	TEM Conc.>5 μm (structures/cm^2)	TEM Conc. Total Structures (structures/cm^2)
Fire Line		8	4			
	Brush Clearer			896	ND	896
	Chainsaw Operator			ND	ND	ND
				ND	ND	ND
	Pulaski Operator			1,344	1,792	3,135
				448	448	896
	Comby Operator			896	ND	896
				ND	ND	ND
	Data Recorder			ND	ND	ND
	Mean Concentrations			448	280	728
Trail Maintenance		5	3			
	Brush Clearer			299	ND	299
				299	ND	299
				ND	ND	ND
	Chainsaw Operator			1,792	ND	1,792
	Data Recorder			ND	ND	ND
	Mean Concentrations			478	ND	478
Tree Measurement	Tree Measurer	5	4			
				ND	ND	ND
				ND	448	448
				448	ND	448
				ND	448	448
				448	448	896
	Mean Concentrations			179	269	448
[b]Walking	Walking	5	1			
				ND	ND	ND
				ND	ND	ND
				ND	ND	ND
				896	ND	896
				ND	ND	ND
	Mean Concentrations			179	ND	179

[a] ND : NonDetect, TEM analytical sensitivity : (448–896 s/cm^2). [b]One walking activity wipe sample (not reported) revealed chrysotile asbestos, not amphibole asbestos.

maintenance brush clearer and one of one trail maintenance chainsaw operator samples was greater than the AS, while one of five walking samples were greater than the AS. As noted with one PBZ walking sample, one of the walking activity wipe samples (not reported) revealed chrysotile asbestos. As noted previously, chrysotile asbestos is not part of the amphibole family. However, fibers and bundles with split ends resembward ling commercial grade asbestos have been identified but are not common in the Rainy Creek Complex near Libby, Montana [6].

The pre- and post- travel vehicle wipes collected for the FS 4872 vehicle driving activity simulations revealed concentrations below the AS for both the November and July trials. The pretravel vehicle wipe collected for the FS 401 vehicle driving activity simulation was also reported below the AS, while the posttravel wipe sample revealed one amphibole fiber resulting in a concentration of 17 917 s/cm^2. The amphibole fiber detected was less than 5 μm long.

Postactivity chainsaw bar wipe sample results are presented in Table 4. AA was detected on the chainsaw bar after all of the simulation activities. In terms of structure counts reported by the laboratory, 12 of 15 fibers were less than 5 μm long (not shown).

A SEM-EDS spectra (not shown) for all wipe samples (clothing, equipment, vehicle) with detectable amphibole

TABLE 4: Postactivity TEM chainsaw bar wipe sample results reported as concentration of amphibole asbestos <5 microns long, >5 microns long, and total structures per square centimeter.

Activity Performed	TEM (s/cm^2) <5 μm	TEM (s/cm^2) >5 μm	TEM (s/cm^2) Total Asbestos
Fire Line Construction	8,600	3,225	11,825
Trail Maintenance	2,688	ND[a]	2,688
Fire Line Construction	896	ND[a]	896

[a] ND : NonDetect; TEM analytical sensitivity : (896 s/cm^2).

asbestos revealed measurable amounts of sodium and potassium in 73% of the samples.

4. Conclusions

Results from the FS activity simulations conducted within this study indicate that an exposure to AA may exist when work is performed in the Kootenai National Forest near the former vermiculite mine. Bark samples collected in the area where activity simulations were conducted revealed amphibole contamination ranging from 37 thousand to 15 million structures per cm^2 of bark surface area. The lowest bark amphibole concentrations were observed in the Alexander Test Site, an area that was replanted after a timber harvest in the early 1990s as a research plot for Tamarack trees. It is worth noting that trees in this location were planted after the vermiculite mine ceased operations. Contamination of these trees may indicate more recent dispersion of amphibole fibers. The highest bark amphibole concentrations were observed in aged Douglas Fir trees on the Rainy Divide Trail.

In terms of inhalation exposure potential associated with the FS tasks evaluated, fire line construction and tree measurement activities yielded detectable AS TEM PBZ concentrations. Detectable AA concentrations were not observed with trail maintenance and walking activities. Of the fire line activity tasks evaluated, the Pulaski tool operator and the brush clearer yielded the highest PBZ concentrations. PBZ concentrations for these fire line activity tasks revealed detectable AA concentrations for two separate trials conducted in two separate geographical areas. Five PBZ samples were collected for the tree maintenance activity. Of these, three samples revealed detectable AA. These three samples were also collected in two separate trials conducted in two separate geographical areas. In terms of individual fiber counts, fifty-seven percent of PBZ asbestos structures were <5 μm long. This is consistent with other research performed regarding amphibole asbestos in tree bark [8, 9].

It is worth noting that the operation of the Pulaski tool employed in fire line construction involves clearing vegetation to mineral grade soil. Therefore, it is unclear whether AA exposure associated with this task is derived from vegetation or soil sources.

In addition to the airborne exposure potential associated with FS activities, there is a potential for clothing and equipment contamination. Composite wipe samples collected from the investigators' forearm, shin, and chest revealed detectable amphibole asbestos in fifty-two percent of the samples collected. Clothing contamination was observed in samples from each of the four activities evaluated: fire line construction, tree measurement, trail maintenance, and walking. In addition, the wipe samples collected from the chainsaw bar after each trial ($n = 3$) revealed amphibole contamination ranging from 896 to 11 825 s/cm^2. Clothing and equipment contamination may serve as a secondary source of exposure to FS personnel. Cross contamination of vehicle cabs, vehicle boxes, equipment storage areas, equipment maintenance areas, and offices may occur as a result of clothing and equipment contamination.

Although the objective of this study was to assess the potential exposure associated with FS occupational activities, the potential for public exposure to AA cannot be ignored. Libby and the surrounding area are known for clean water, beautiful scenery, and recreational activities such as hiking, hunting boating, and skiing. As noted earlier, the simulation areas are accessible to the general public. The frequency of recreational use by the general public was not evaluated in this study; however, hunters were observed near the simulation site during the bark collection phase of this study. In an effort to inform the public about the amphibole contamination in the Kootenai National Forest, FS management has published a brochure that outlines safeguards to minimize dust generation and transport of fibers on clothing.

The forested areas near the simulation sites were historically used for timber harvests as observed by numerous clear-cut plots. In the past, FS personnel visited the Alexander Test plot and areas accessible via roadways 4872 and 401 on a weekly basis. Since the awareness of amphibole contamination in tree bark, FS travel in this area has been reduced. In addition, fire fighting in this area is currently performed from the air only.

This research was funded as a small project/pilot study in order to assess potential FS exposure to AA. A limited number of samples were collected within a relatively small geographical area. Future research is planned to assess FS exposure potentials with the activities evaluated in this study throughout a range of meteorological conditions (i.e., different seasons) as well as other activities (i.e., fire fighting), in expanded radii from the former vermiculite mine. In addition, vehicle cabs, offices, and equipment storage and maintenance facilities should be evaluated for potential AA contamination.

5. Competing Interests

One of the authors (TMS) has served as an expert witness for plaintiffs' attorneys in litigation involving asbestos exposure in Libby, MT.

Abbreviations

AHERA:	Asbestos Hazard Emergency Response Act
AIHA:	American Industrial Hygiene Association
AS:	Analytical Sensitivity
ASTM:	American Society for Testing and Materials
ATSDR:	Agency for Toxic Substances and Disease Registry
Ca:	Calcium
COBRE:	Center of Biomedical Research Excellence
EDS:	Energy Dispersive X-ray Spectroscopy
EPA:	Environmental Protection Agency
Fe:	Iron
FS:	Forest Service
GPS:	Global Positioning System
K:	Potassium
LOD:	Limit of Detection
Mg:	Magnesium
Na:	Sodium
NIH:	National Institute of Health
NIOSH:	National Institute for Occupational Safety and Health
NMAM:	NIOSH Manual of Analytical Methods
NVLAP:	National Voluntary Laboratory Accreditation Program
OSHA:	Occupational Safety and Health Administration
PBZ:	Personal Breathing Zone
PCM:	Phase Contrast Microscopy
PPE:	Personal Protective Equipment
SD:	Standard Deviation
SEM:	Scanning Electron Microscope
TEM:	Transmission Electron Microscopy
TWA:	Time Weighted Average
USDA:	United States Department of Agriculture.

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