

A Review of Scientific Literature Examining the Mining History, Geology, Mineralogy, and Amphibole Asbestos Health Effects of the Rainy Creek Igneous Complex, Libby, Montana, USA

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This article reviews the past 90 yr of scientific research directed on multiple aspects of the unique geology and environmental health issues surrounding the vermiculite deposit found at Libby, MT. Hydrothermal alteration and extensive weathering of the ultramafic units resulted in the formation of a rich deposit of vermiculite that was mined for 67 yr and used in numerous consumer products in its expanded form. Later intrusions of alkaline units caused hydrothermal alteration of the pyroxenes, resulting in formation of amphiboles. Some of these amphiboles occur in the asbestiform habit and have been associated with pulmonary disease in former miners and mill workers. Identification of these amphibole asbestos minerals has received little attention in the past, but recent work shows that the majority of the amphibole mineral species present may not be any of the amphibole species currently regulated by government agencies. Epidemiological studies on former miners have, nevertheless, shown that the amphibole asbestos from the Rainy Creek igneous complex is harmful; also, a recent study by the Agency for Toxic Substances and Disease Registry shows that residents of Libby who had not been employed in the vermiculite mining or milling operations also appear to have developed asbestos-related pulmonary diseases at a higher rate than the general public elsewhere. Since November 1999, the U.S. Environmental Protection Agency has been involved in the cleanup of asbestos-contaminated sites in and around Libby associated with the mining and processing of vermiculite.

The first examination of the Rainy Creek igneous complex (RCC) was during gold exploration in the late 19th century. Pardee and Larsen (1929) began their work in the area exploring the quartz veins in 1911. These early explorations, particularly by E. N. Alley, in which exfoliation of vermiculite was observed in the roof of audits, led to the discovery and large-scale mining of the vermiculite deposits in the area of Rainy Creek (Pardee & Larsen, 1929). (See Table 1 for a timeline of important events.) During the 1920s, the Zonolite Company began mining the deposit and developed uses for exfoliated (expanded) vermiculite. W. R. Grace Corporation purchased the mine from the Zonolite Company in 1963 and continued producing expanded vermiculite for products such as Zonolite[®] insulation and Monokote[®]

fireproofing, bulking agents, absorbents, and soil amendments. They increased production, and eventually the mine at Libby was the largest source of vermiculite worldwide. Along with the mine at Libby (Figure 1A–C), W. R. Grace also operated an export facility and local expansion facilities (until 1990), and continues to be a leading producer of vermiculite products. The mine at Libby ceased operation in 1990.

The vermiculite ore from the RCC is contaminated with varying amounts of amphibole asbestos (Figures 1D–F), which formed as a result of hydrothermal alteration of pyroxene minerals present in the units of the RCC. The morphology of these samples is characterized in Brown and Gunter (2002), and the crystal chemistry and crystal structure are presented in Gunter et al. (2003) and Bandli et al. (2003). The U.S. Geological Survey has conducted other research, and results have been presented at several international conferences and in several papers (Clark et al., 2003; Meeker et al., 2003; Van Gosen et al., 2005).

Several epidemiological studies have documented the toxicity of the amphibole asbestos minerals present in the RCC.

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TABLE 1
Timeline of events significant to vermiculite mining operations at Libby, MT

Year	Event
1919	E. N. Alley observes exfoliation of vermiculite in roof of mine audit
1923	Commercial mining of vermiculite begins on Vermiculite Mountain by E. N. Alley
1939	E. N. Alley's zonolite business becomes the Universal Zonolite Insulation Company
1944	First dust control equipment installed
1948	Universal Zonolite Insulation Company changes name to Zonolite Company
1954	First "wet" mill installed at Libby mine
1956	State of Montana conducts a study to examine the working conditions at the Zonolite Company facilities in Libby
1959	State of Montana conducts a follow-up study of the 1956 study and finds dust levels are lower, but asbestos content of dust collected in the vermiculite mill is determined to be 27%
1963	W. R. Grace purchases Zonolite Company
1964	W. R. Grace begins x-ray testing of employees
1970	Occupational Safety and Health Act creates Occupational Safety and Health Administration (OSHA)
1972	First federal regulations limiting exposure of workers to asbestos are enacted by OSHA (5 fibers/cm ³)
1973	Clean Air Act enacted placing limits on amounts of asbestos industries can release into the environment
1974	"Dry" milling of vermiculite ore discontinued
1977	W. R. Grace initiates policy of not hiring individuals who smoke cigarettes
1977	Federal Mine Safety and Health Act enacted to create safer working environment for miners
1986	W. R. Grace receives permission to expand vermiculite mine to 1004 acres (this is the largest area the mine will cover)
1990	September: Mining operations at Vermiculite Mountain end
1991	Reclamation at mine site begins
1994	W. R. Grace sells Vermiculite Mountain mine site to Kootenai Development Company
1997	Reclamation bond released on 900 acres of Vermiculite Mountain mine
1999	November, <i>Seattle Post-Intelligencer</i> publishes a series of articles about the high incidence of asbestos related lung disease among Libby, MT, residents
1999	November, U.S. EPA begins investigating asbestos contamination in and around Libby
2000	W. R. Grace initiates medical program to provide medical coverage for Libby residents and buys back Vermiculite Mountain mine site from Kootenai Development Company
2001	Agency for Toxic Substances and Disease Control begins health screening program for current and past Libby residents
2002	Libby added to U.S. EPA's National Priorities List
2002– 2006	Cleanup of W. R. Grace properties in and around Libby, private residences, and businesses in Libby ongoing. To date (April 2006) 604 properties have been cleaned up and ~1200 properties remain to be cleaned (personal communication, Courtney Zamora, EPA Libby Information Center)

Note. Data obtained from Montana Department of Environmental Quality (2000) and W. R. Grace (2000).

However, the species of amphibole present has been mischaracterized as tremolite asbestos in these studies. Recent work by several researchers has shown that the most abundant amphibole minerals present are actually winchite and richterite. The amphibole asbestos minerals present in the RCC appear to have significant effects on humans. The incidence of asbestosis, mesothelioma, and lung cancer is high in former mine workers, particularly those employed in the early unregulated workplace. The Agency for Toxic Substances and Disease Registry (ATSDR, 2001; Peipens et al., 2003) presented data that showed that a significant number of individuals who lived in Libby, but did not work in the mining or processing of vermiculite, show symptoms of diseases related to asbestos exposure.

MINING HISTORY

Mining and processing of vermiculite from the RCC continued uninterrupted from 1923 to 1990 (Table 1). E. N. Alley was the first individual to exploit the RCC vermiculite deposit in 1923. The Universal Zonolite Insulation Company and the Vermiculite and Asbestos Company were the first commercial ventures of the vermiculite deposits at Libby. In 1948, these two companies merged to become the Zonolite Company. In 1963 W. R. Grace Co. purchased the Zonolite Company and continued to mine the vermiculite deposit until 1990. W. R. Grace is the current owner of the mine property.

The processes involved in mining and milling the vermiculite did not change significantly over the life span of the mine (Table 1). Initially, vermiculite ore was removed from

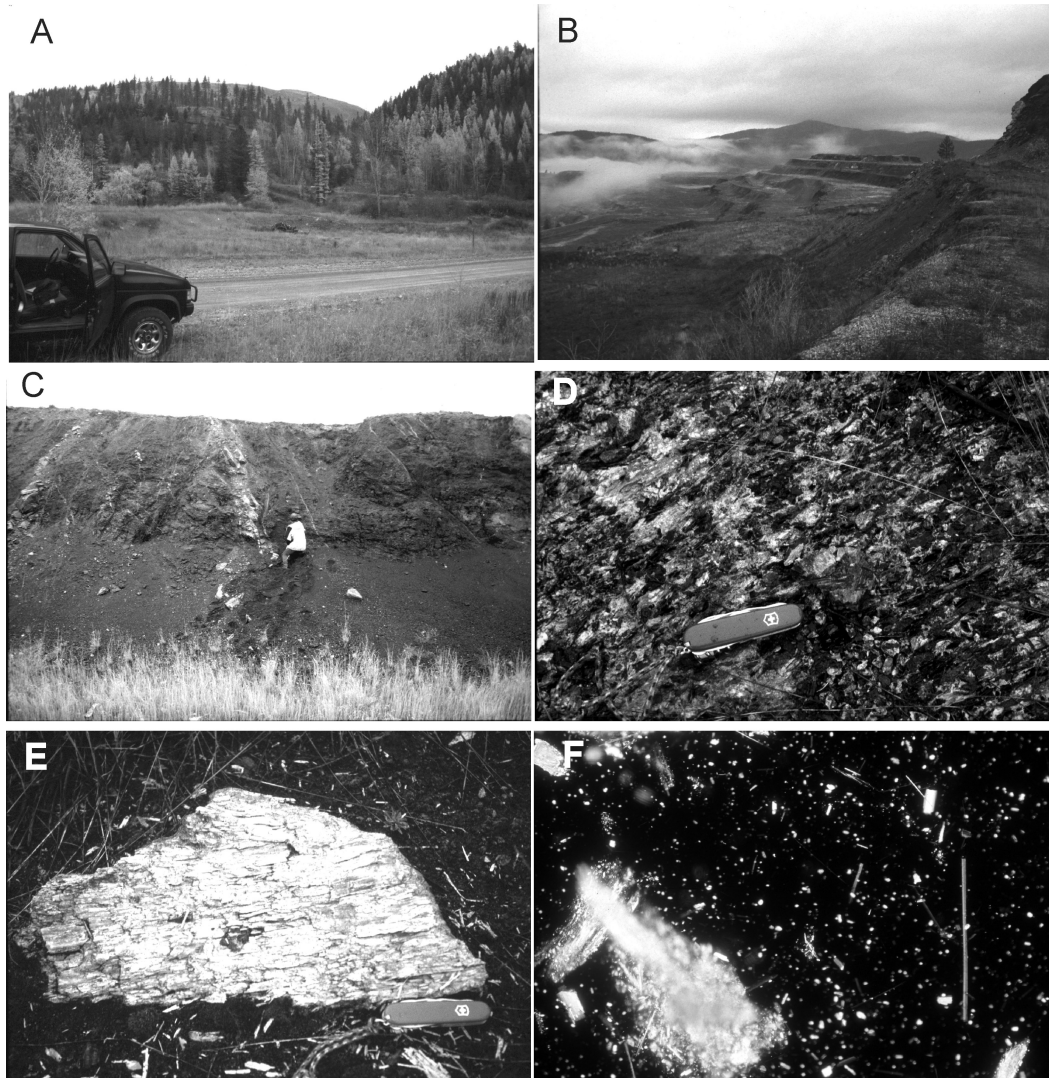


FIG. 1. (A) View from Rainy Creek Road east toward Vermiculite Mountain. (B) View from Vermiculite Mountain toward Libby with mine benches visible in the middle of the photo (benches approximately 7 m high). (C) View of mine bench showing amphibole asbestos vein. (D) Photograph of biotite pyroxenite. Light-colored grains are amphibole, medium-gray grains are pyroxenes, and dark-gray grains are biotite/vermiculite (knife for scale). (E) Photograph of boulder composed entirely of amphibole (knife for scale). (F) Photomicrograph of material from Vermiculite Mountain mine. High-aspect-ratio amphibole fragment (inclined extinction) visible on right side and large amphibole asbestos fiber bundle visible in lower left corner.

underground workings, but eventually surface mining methods (Figures 1B and C) were employed. The ore was generally very weathered and could be removed without blasting, but blasting was occasionally necessary. The mine was a large open pit that eventually covered several hundred acres (Figures 1A and B, and Figure 2). Ore was loaded onto trucks and hauled to a transfer point on the west end of the mine (U.S. EPA, 2001), where it was screened to remove the coarse fractions, and the remaining ore was transferred by conveyor to the concentrating/loading facility on the Kootenai River at the mouth of Rainy Creek (Figure 3) (Boettcher, 1963). In the mill, the vermiculite was concentrated

through a dry beneficiation process until 1954, when a wet beneficiation process was developed. Both processing methods were used until 1974, when the dry process was discontinued. Next, the concentrate was screened into five grades based on particle size. A portion of the vermiculite concentrate was then sent to an exfoliating and export plant in Libby. However, the majority of the vermiculite concentrate was transferred across the Kootenai River by conveyor for shipment by rail to expansion facilities across the United States (U.S. EPA, 2001).

Due to the low density of expanded vermiculite, most vermiculite from Libby was shipped as unexpanded concentrate



FIG. 2. Aerial photograph of former vermiculite mine. Photo is approximately 3 km across with north at top of image.

to expander plants nationwide. At the expanding facilities the vermiculite was heated in kilns to approximately 1100°C for a few seconds (Bassett, 1959). This rapid heating caused the water present in the vermiculite structure to vaporize into steam, which forced the layers of the sheet silicate apart and created the useable product (Figure 4). Expanded vermiculite is used in numerous consumer products. W. R. Grace marketed the majority of the expanded vermiculite originating from its Libby mine as Zonolite[®] insulation, but expanded vermiculite from Libby was also used as absorbents, fireproofing materials, industrial fillers, packaging material, and soil amendments. The mining and processing operations at Libby were very dusty by nature, and owners of the mine and various regulatory agencies worked to reduce the levels of dust to which workers were exposed. Regulations regarding acceptable limits of the amount of airborne asbestos fiber to which workers can be exposed are listed in Table 2, and these limits have decreased over time.

GEOLOGY

The RCC is an alkaline-ultramafic igneous complex in Lincoln County, MT, 7 miles northeast of Libby, and is geographically known as Vermiculite Mountain (Figure 5). The RCC lies in the basin of Rainy Creek and is much less resistant to erosion than the surrounding Belt series metamorphic rocks. The contact between the ultramafic and metamorphic units is topographically expressed in a significant increase in slope in the metamorphic units. There is also a significant decrease in the density of coniferous vegetation growing in soils over the ultramafic units (Boettcher, 1963). The rocks of the complex, where not exposed by mining, are covered by till (Larsen & Pardee, 1929). The geology of the RCC has been studied by several individuals—Goranson (1927), Pardee and Larsen (1929), Larsen and Pardee (1929), Kriegel (1940), Bassett (1959), and Boettcher (1963, 1966a, 1966b, 1967)—and is currently being studied by the U.S. Geological Survey. Boettcher provides the

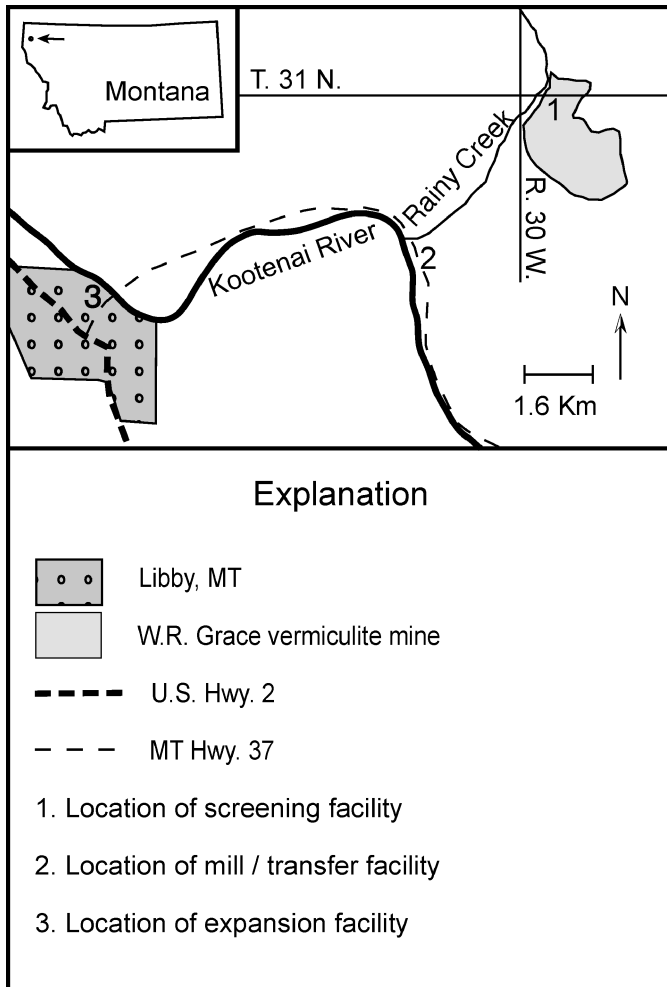


FIG. 3. Map of Libby, Montana and former vermiculite mine. Adapted from U.S. EPA (2001).

most detailed and most recently published geologic and mineralogical information on the RCC in its entirety.

The rocks of this igneous complex formed by intrusion into argillite, limestone, and dolostone of the Precambrian Belt series (Wallace Formation) (Figure 5). The structure present in the Belt series of north-northwest-trending folds is unrelated to the intrusion of the RCC. The magma intruded into the axis of a slightly southeasterly plunging syncline (Figure 5). The rocks of the RCC consist of biotitite, biotite pyroxenite, magnetite pyroxenite, syenite, trachyte, phonolite, and granite (Boettcher, 1967). Workers prior to Boettcher (1967) collectively describe the biotite pyroxenite and magnetite pyroxenite as pyroxenite. The main body of the complex is a stock composed predominantly of biotite pyroxenite, magnetite pyroxenite, and biotitite. A large, irregularly shaped body of syenite crosscuts the pyroxenites (Figure 5). All of these units are crosscut by trachyte and phonolite dikes, which are, in turn, cut by granitic dikes (Boettcher, 1963).

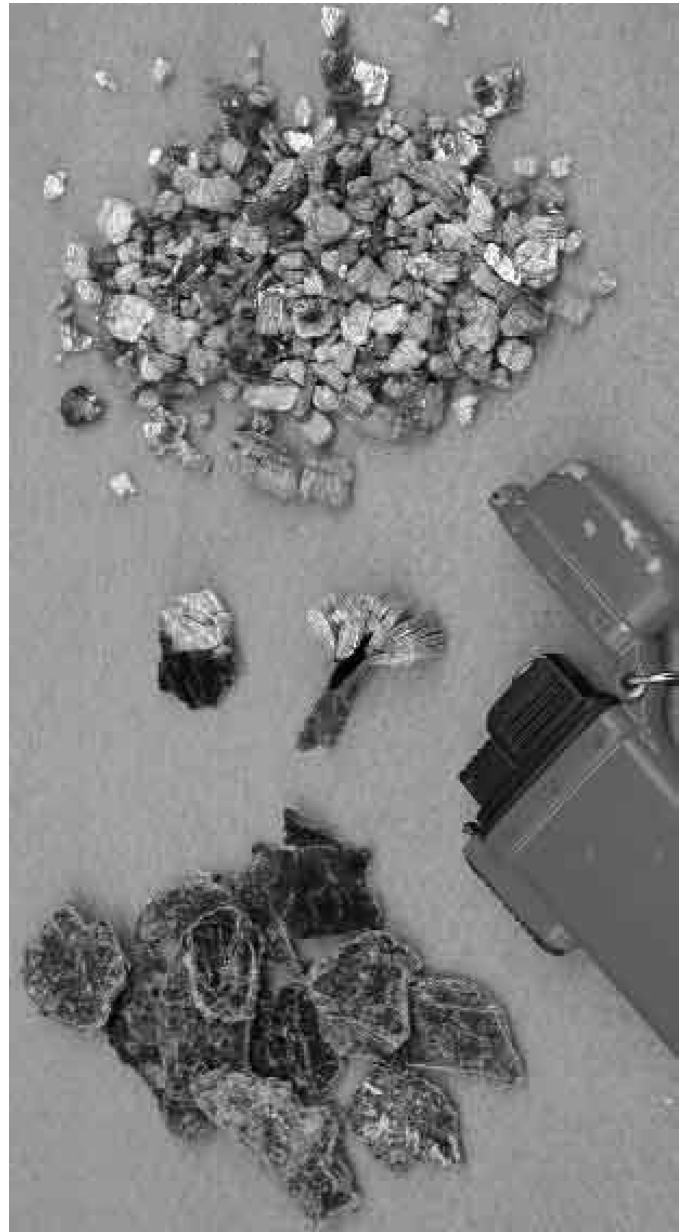


FIG. 4. Photograph showing raw (unexpanded) vermiculite on bottom, partly expanded vermiculite at center, and completely expanded (exfoliated) vermiculite on top (cigarette lighter for scale).

Ultramafic Units

Biotitite. The central and topographically highest unit of the complex is a coarse-grained biotitite and comprises approximately 5% of the intrusion. The biotitite is composed almost entirely of anhedral books of biotite that are generally larger than 10 cm and show no preferred orientation (Boettcher, 1966a). The biotitite was thought by Boettcher to have formed near the roof of the magma chamber in the presence of higher concentrations of alkali metals, metal sulfides, and other volatiles relative to the

TABLE 2
Regulations for occupational exposure and environmental releases of mineral dust

Year	Regulation	Exposure limit
1946	ACGIH	5 mppcf ^a
1968	ACGIH	12 fibers/cm ³
1972	OSHA	5 fibers/cm ³ 8 h TWA ^b
1973	Clean Air Act	Sets no specific release levels, but mandates practices for handling asbestos-containing materials
1976	OSHA	2 fibers/cm ³ 8-h TWA
1977	Mine Act	2 fibers/cm ³ 8-h TWA
1986	OSHA	0.2 fibers/cm ³ 8-h TWA
1992	OSHA	0.1 fibers/cm ³ 8-h TWA, and deregulates nonasbestiform amphiboles

Note. 1946 and 1968: Recommendations made by the American Conference of Governmental Industrial Hygienists. The exposure levels recommended at these times were not enforced by any regulatory agency.

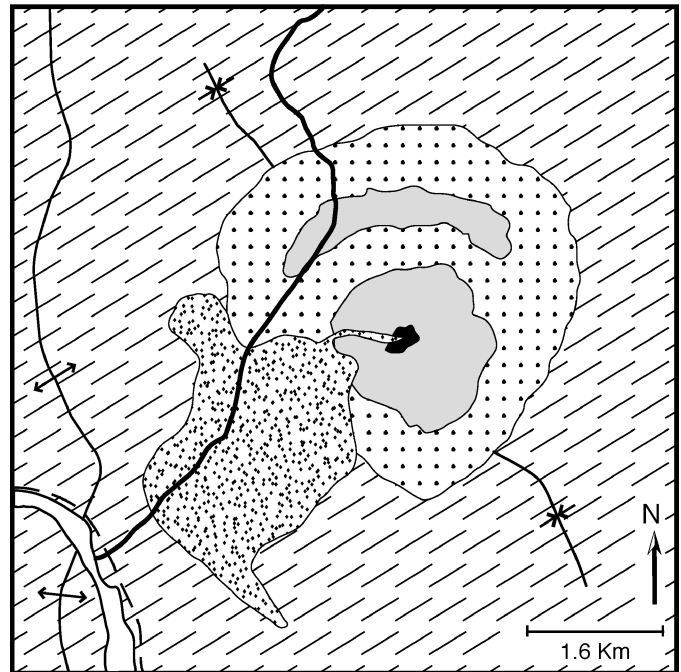
^amppcf, millions of particles per cubic foot.

^bTWA, time-weighted average.

surrounding pyroxenites. Larsen and Pardee (1929) mentioned a "biotite rock," but it does not appear to be the biotite unit described by Boettcher (1967). The Larsen and Pardee (1929) "biotite rock" was described as being almost entirely altered to vermiculite, whereas the biotite described by Boettcher is composed of unaltered biotite with only small amounts of vermiculite. Bassett (1959) does not describe this unit in any detail.

Feldspar occurs as wedges between books of biotite and make up less than 10% of the rock. Small amounts (<2%) of pyrite and calcite are also present as secondary alteration products. Calcite is evenly distributed throughout the biotite as a secondary alteration product of the biotite (Boettcher, 1966a). The contact between the biotite and the biotite pyroxenite is gradational over 3 m. The contact zone is also expressed in a compositional change, where feldspar content decreases to zero while diopside increases and vermiculite content increases significantly (Boettcher, 1966a).

Biotite Pyroxenite. The biotite pyroxenite completely surrounds and has a gradational contact with the inner biotite (Figure 5). The biotite pyroxenite makes up approximately 20% of the intrusion (Boettcher, 1967). In hand sample, its color is dark green and it is very weathered and friable (Figure 1D). The biotite pyroxenite ranges in grain size from fine-grained (<1 mm) to coarse-grained (>10 cm) and is composed of variable amounts of clinopyroxene (diopside), biotite, vermiculite, and hydrobiotite. This unit was the source of all the mineable vermiculite, and vermiculite content varies significantly, but on average is 25 wt% of this rock (Boettcher, 1966a). Unaltered biotite can be found locally within the biotite pyroxenite. Boettcher (1966a) did not observe any chemical zoning but did describe



Explanation

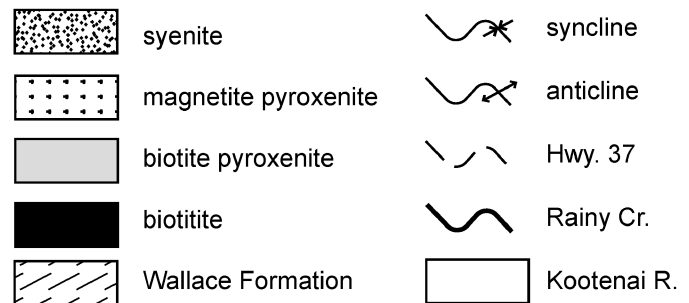


FIG. 5. Geologic map of the RCC. Adapted from Boettcher (1966a).

a textural gradient. Bassett (1959) observed areas where pyroxene crystals were horizontally oriented but were crosscut by veins of fine-grained pyroxenite where the pyroxene was oriented vertically. This would indicate some sort of vertical flow of the magma prior to complete crystallization, according to Bassett (1959). Apparently, this feature does not occur over large areas of pyroxene-bearing units, and Boettcher (1966a) contradicts Bassett (1959) by noting that most of the pyroxene crystals do not show this preferred orientation. The largest grains of diopside occur nearest to the contact with the biotite. Larsen and Pardee (1929) did not observe any type of textural gradient and felt the grain size distribution was more or less random; however, at the time of their study, outcrops of the biotite pyroxenite were limited (i.e., more of this rock was exposed by later mining). Fluorapatite is the most common accessory mineral and occurs as interstitial euhedral crystals and as small crystals within the diopside crystals (Boettcher, 1966a). The biotite

pyroxenite and biotitite appear to be comagmatic (Boettcher, 1967). Several dikes of magnetite pyroxenite have intruded into the biotite pyroxenite, indicating a discontinuity in the intrusion of the ultramafic portion of the complex.

Magnetite Pyroxenite. The magnetite pyroxenite has a uniform grain size (0.7–3 mm) and is composed of diopside, magnetite, and apatite, with andradite, titanite, and biotite or vermiculite as accessory minerals. It constitutes approximately 40% of the intrusion (Boettcher, 1966a). The orientation of the magnetite pyroxenite relative to the two inner units is like that of a ring dike (Figure 5). The magnetite pyroxenite completely surrounds the inner units and the contact dips slightly outward from the center of the complex in all directions (Boettcher, 1966a). Diopside and apatite crystals are aligned and dip out from the center at various angles. The magnetite pyroxenite also forms numerous small dikes that crosscut the biotite pyroxenite. The emplacement of the magnetite pyroxenite is thought to have occurred as an intrusion into a zone of weakness that formed between the Wallace Formation and the biotite pyroxenite (Boettcher, 1967). The distinguishing feature of the magnetite pyroxenite in thin section is the “island in sea” texture of the pyroxene and interstitial magnetite. The diopside present in both of the pyroxenites is aluminum deficient as a result of the early fractionation of the biotitite from the crystallizing magma. Larsen and Pardee (1929) stated that observable amounts of magnetite occur only in pyroxenites that contained less than 20% biotite. Larsen and Pardee (1929) also noted the presence of garnet in pyroxene-rich areas of the stock. Bassett (1959) noted the increase of magnetite and apatite content in the pyroxenite near its contact with the Wallace Formation.

Alkaline Units

The remainder of the complex (approximately 35%) is composed of various alkaline rocks: syenite, nepheline syenite, trachyte, phonolite, alkaline pegmatite, and alkaline granites. The largest alkaline unit is an irregularly shaped body of variably altered syenite that is located in the southwest portion of the complex (Figure 5) and transects the earlier ultramafic units. This syenite has been altered, as evidenced by the replacement of nepheline by muscovite (Boettcher, 1966a). Syenite also occurs as dikes of varying width and is probably genetically related to the alkaline pegmatite dikes (Boettcher, 1966a). These dikes crosscut all of the ultramafic units. The smaller syenite dikes exhibit some compositional and textural variability that could be attributed to multiple intrusions of syenite magma (Larsen & Pardee, 1929). Intrusion of these dikes into the pyroxenite units caused significant wall rock alteration, resulting in the amphibolitization of pyroxene minerals. However, where these dikes occur in the biotitite, little alteration of the biotite is observed (Boettcher, 1966a). Dikes of trachyte, phonolite, and alkaline granite crosscut both these syenite and alkaline pegmatite dikes. The trachyte and phonolite dikes are interesting in that no wall-rock alteration resulted from their intrusion. This feature, along with other petrologic characteristics described by

Boettcher (1966a), indicates that these dikes penetrated very near, if not at, the surface.

MINERALOGY

Two major processes have significantly influenced the mineralogy of the RCC: magmatic differentiation and hydrothermal alteration. The biotitite and the biotite pyroxenite are believed to have been the first units to crystallize from the original ultramafic magma (Boettcher, 1967). The early crystallization of large amounts of biotite preferentially differentiated aluminum from the melt. This early separation of biotite from the melt was facilitated by a high p_{H_2O} (Boettcher, 1967). Boettcher concluded that the biotitite, biotite pyroxenite, and magnetite pyroxenite are comagmatic. However, the magnetite pyroxenite is not linearly related to the other two units. Later, syenite, trachyte, phonolite, and pegmatites intruded the previous units from a much more felsic magma and resulted in the alteration of diopside to amphiboles and biotite to vermiculite and hydrobiotite (Boettcher, 1967).

Presently, the two main minerals of interest are vermiculite and amphibole asbestos. The RCC contains a very large reserve of easily mineable vermiculite. Vermiculite, as discussed earlier, is a very useful industrial mineral. However, the health effects associated with the amphibole asbestos minerals present in the pyroxenite units makes mining and milling of the vermiculite from this deposit a health hazard. Previous workers have examined the incidence of asbestos-related disease associated with the amphibole minerals present in the RCC, and their work is examined in a later section. Persistent classification of the amphibole asbestos from the RCC as tremolite has been shown to be inaccurate. Proper mineralogical classification of amphibole minerals requires precise chemical analysis (Leake et al., 1997), and the classification of amphiboles from the RCC is discussed in a later section.

Biotite, Vermiculite, and Hydrobiotite

The biotitite unit is almost entirely composed of biotite, while biotite comprises roughly 40% of the unaltered biotite pyroxenite and slightly less of the magnetite pyroxenite. The alteration of the biotite in the biotite pyroxenite resulted in the formation of the vermiculite and hydrobiotite. The vermiculite is a result of weathering of biotite. Bassett (1959) and Boettcher (1966b) explored the chemical conditions necessary for the conversion of biotite to vermiculite. The hydrobiotite and amphibole asbestos are the product of much higher temperature hydrothermal processes (Boettcher, 1966b). Boettcher (1966b) proposed two separate phases of alteration: high-temperature hydrothermal alteration and lower temperature weathering. Bassett (1959) proposed that vermiculite and hydrobiotite formed as a result of varying degrees of supergene alteration.

The optical properties of the biotite from the biotitite and biotite pyroxenite are within the range of most ordinary biotites and exhibit a light brown to greenish brown pleochroism (Table 3). The biotite from the RCC has an average $d_{(001)}$ value of 10.1

TABLE 3
Optical properties of minerals present in the RCC

Mineral	α	β	γ	2V (°)
Biotite ^a	1.569(3)	1.609(1) ^h	1.609(1) ^h	11.0
Biotite ^b	1.570 ⁱ	1.610 ^{h,i}	1.610 ^{h,i}	5.0
Hydrobiotite ^a	—	1.575(1) ^h	1.575(1) ^h	13.0
Hydrobiotite ^b	1.560 ⁱ	—	1.595 ⁱ	5.0
Vermiculite ^a	1.519(3)	1.545(1)	1.545(1) ^h	9.0
Vermiculite ^b	1.530 ⁱ	—	1.555 ⁱ	5.0
Pyroxene from biotite	1.678(1)	1.684(1)	1.702(1)	124.0
Pyroxenite ^a	—	—	—	—
Pyroxene from magnetite	1.692(1)	1.698(1)	1.717(1)	122.0
Pyroxenite ^a	—	—	—	—
Diopside ^c	1.685 ⁱ	1.699 ⁱ	1.719 ⁱ	79.8 ^j
Aegirine ^d	1.742(3)	1.768(3)	1.787(3)	81.0 ^j
Winchite ^e	1.621(1)	1.631(1)	1.637(1)	75.1 ^j
Winchite ^e	1.618(1)	1.628(1)	1.634(1)	75.1 ^j
Winchite ^f	1.6265(5)	1.6349(6)	1.6381(10)	58(1)
Winchite ^f	1.6251(4)	1.6345(5)	1.6389(8)	64(2)
Winchite ^f	1.6221(14)	1.6316(6)	1.6387(5)	73(2)
Winchite ^f	1.6214(4)	1.6340(5)	1.6397(5)	63(1)
Winchite ^f	1.6238(4)	1.6329(4)	1.6392(7)	63(1)
Winchite ^f	1.6228(11)	1.6305(5)	1.6388(12)	66(1)
Winchite ^f	1.6243(9)	1.6323(5)	1.6417(4)	67(1)
Winchite ^f	1.6253(49)	1.6305(3)	1.6337(12)	80(1)
Winchite ^f	1.6177(6)	1.6351(5)	1.6369(6)	67(1)
Winchite ^f	1.6226(10)	1.6342(7)	1.6391(6)	60(1)
Winchite ^f	1.6257(5)	1.6370(6)	1.6407(4)	59(1)
Tremolite ^g	1.599–1.612	1.615–1.628	1.627–1.638	95.3–114.5 ^j

Note. α , β , and γ are unitless. For discussion and definitions of optical properties see Bloss (1999).

^aBoettcher (1966a).

^bBassett (1959).

^cLarsen and Pardee (1929).

^dGoranson (1927).

^eWylie and Verkouteren (2000).

^fBandli et al. (2003).

^gVerkouteren and Wylie (2000).

^h β and γ cannot be equal with an observed 2V; however, precision in the measurement of β and γ does not allow for their values to be distinguished.

ⁱNo reported error.

^jCalculated 2V values.

Å (Boettcher, 1966b). The vermiculite of the RCC was shown by Boettcher (1966b) to have an upper stability limit of 350°C based on hydrothermal studies. The chemistry of vermiculite indicates that it was the result of leaching of biotite by groundwater. A lower content of alkali metals and higher amount of Fe³⁺ than that of biotite indicates a low-temperature leaching process altered the biotite to vermiculite (1, 2, and 3 in Table 4). The hydrobiotite was shown in the same study to have an upper stability limit of as high as 480°C. The hydrobiotite has a 1:1 stacking sequence of vermiculite and biotite that is not inherited from the biotite. This, along with the lack of a direct chemical relationship between hydrobiotite (4 in Table 4) and biotite (hy-

drobiotite has significantly less K, Al, and Si and more Fe³⁺ than the biotite), indicates that a much higher temperature hydrothermal alteration process worked to alter the original biotite (Boettcher, 1966b).

Bassett (1959) provided an alternate hypothesis for the formation of the vermiculite in the RCC. Bassett concluded that continued supergene alteration of augite resulted in the formation of vermiculite. Hydrothermal alteration of augite by fluids rich in K and Si resulted in the formation of biotite. Supergene alteration of this biotite increased Ca and Mg and decreased the K content of the biotite and resulted in the formation of hydrobiotite. Further supergene alteration of the hydrobiotite resulted

TABLE 4
Reported chemical analysis of various minerals from the RCC

	1	2	3	4	5	6	7	8	9	10
SiO ₂	40.60	38.63	35.57	35.60	53.94	51.59	56.60	56.10	56.70	51.91
Al ₂ O ₃	14.10	13.08	11.47	11.85	0.76	1.86	0.50	0.40	0.71	0.38
TiO ₂	—	1.55	1.06	1.13	0.26	0.61	—	—	0.28	0.91
Cr ₂ O ₃	—	0.23	0.18	0.03	0.20	—	—	—	0.06	—
Fe ₂ O ₃	2.30	2.50	7.49	10.28	1.13	2.86	—	—	4.71	21.79
FeO	1.70	8.75	0.34	0.81	1.91	3.26	6.00	4.20	0.87	1.48
MnO	—	0.14	0.06	0.08	0.07	0.18	0.10	0.30	0.07	0.58
MgO	27.00	19.94	22.57	20.17	16.93	14.67	20.20	21.00	21.95	3.08
CaO	—	0.18	0.73	1.44	24.55	24.35	8.30	8.80	6.15	5.53
Na ₂ O	1.20	0.26	0.00	0.16	0.17	0.29	3.20	3.40	5.15	10.46
K ₂ O	8.20	10.00	0.96	3.17	—	0.05	0.70	0.80	1.80	0.22
SrO	—	0.005	0.01	0.005	0.04	0.14	—	—	0.02	—
BaO	—	0.45	0.10	0.17	0.01	—	—	—	—	—
P ₂ O ₅	—	0.06	0.06	0.07	—	—	—	—	—	—
V ₂ O ₃	—	—	—	—	—	—	—	—	—	3.98
F	1.50	0.30	—	0.21	—	—	—	—	1.30	—
S	—	—	—	—	—	—	—	—	—	0.13
NiO	—	0.02	0.02	0.00	—	0.00	—	—	0.23	—

Note. All values wt% column headings: 1, biotite from Larsen and Pardee (1929); 2, biotite RCB-12 from Boettcher (1966a); 3, vermiculite RCSa-59 from Boettcher (1966a); 4, hydrobiotite RCSa-36 from Boettcher (1966a); 5, pyroxenite from biotite pyroxenite (P-100) from Boettcher (1966a); 6, pyroxenite from magnetite pyroxenite (P-103) from Boettcher (1966a); 7, amphibole 1 from Wylie and Verkouteren (2000); 8, amphibole 2 from Wylie and Verkouteren (2000); 9, richterite from Larsen (1942); 10, aegirine from Pardee and Larsen (1929).

in formation of vermiculite. Bassett (1959) stated that separate areas of unaltered augite and unaltered biotite were evidence for two stages of alteration, hydrothermal and supergene.

The vermiculite, hydrobiotite, and biotite appear similar at the mine; however, upon heating, the vermiculite will expand while the biotite will not (Figure 4). This property of vermiculite is a diagnostic property and the trait that makes it a commercially valuable mineral. Vermiculite will expand to over 20 times its original volume, which gives it high thermal insulation and increased liquid absorption properties. Bassett (1959) mentioned that miners used subtle color differences as an ad hoc method to distinguish areas in the mine richer in vermiculite than biotite or hydrobiotite, as opposed to testing by expanding vermiculite grains.

Recent work by Gunter et al. (2005) provides more chemical analysis of the vermiculite from the Rainy Creek Complex. The data collected were compared to data collected from other known deposits of vermiculite that have been mined and used in industrial products. The results show that vermiculite from this deposit is significantly different from other vermiculite deposits. By determining the Ba, Cr, and V content of a bulk vermiculite sample, it is possible to distinguish vermiculite originating from the Libby deposit from vermiculite originating other deposits.

Pyroxenes

The pyroxenes present in the pyroxenite units are predominantly light green, non-pleochroic diopside (Boettcher, 1966a)

or augite (Bassett, 1959). Pyroxene accounts for over half of the mineral content in the pyroxenite units. In hand sample, the diopside has perfect (100) parting and is emerald green in the biotite pyroxenite and darker green in the magnetite pyroxenite (Boettcher, 1966a). The optical properties of several pyroxenes from the RCC are shown in Table 3. The refractive index values are slightly higher for the diopside that occurs in the magnetite pyroxenite than those that occur in the biotite pyroxenite. The iron content of the diopside (5 and 6 in Table 4) is elevated in later crystallizing units and especially when diopside is found in association with magnetite.

The RCC also contains aegirine that has been examined by Goranson (1927) and Pardee and Larsen (1929). The aegirine is of interest because of its increased vanadium content (10 in Table 4). It occurs as black acicular crystals up to 2.5 cm in length that project from the walls of veins or as radiating nodules embedded in other minerals of the pegmatites cutting the pyroxenites and biotite. The pegmatites in the biotite generally contain greater amounts of aegirine than those in the pyroxenite units (Boettcher, 1963).

Amphiboles

Amphibolitization of the pyroxenes in the biotite pyroxenite produced nearly all the amphiboles present in these rocks. Amphibole asbestos occurs in the gangue of the Libby vermiculite

deposits and has been viewed as an accessory mineral by previous workers and was thus not studied in significant detail. Until recently, there has been some confusion as to the classification of the asbestos minerals at Libby. Pardee and Larsen (1929) named the amphibole asbestos minerals tremolite but stated there were "considerable" amounts of Na and Fe present in their samples. The U.S. EPA and its contractors mischaracterized these minerals as tremolite (U.S. EPA, 2000). The transmission electron microscopy–energy-dispersive spectroscopy (TEM-EDS) data presented in the U.S. EPA study (U.S. EPA, 2000) of vermiculite garden products shows that the samples from Libby vermiculite contain significant amounts of Na and K. This would mean these amphibole minerals could not be tremolite. However, the inaccurate name tremolite or actinolite persists in U.S. EPA literature and in the popular press. The optical properties of the amphibole mineral winchite from the RCC are listed in Table 3 and have properties very similar to tremolite.

The precise classification of the amphiboles from the RCC has received little attention until very recently. The amphiboles present in the RCC have been called tremolite-actinolite (Larsen & Pardee, 1929), richterite (Larsen, 1942), tremolite-actinolite (Bassett, 1959), tremolite (Boettcher, 1963), richterite (soda tremolite) (Deer et al., 1963), and winchite (Wylie & Verkouteren, 2000; Gunter et al., 2003; Bandli et al., 2003; Meeker et al., 2003). With the exception of Larsen (1942), no chemical analysis was available to correctly identify the amphibole minerals from the Rainy Creek Complex until Wylie and Verkouteren (2000) (7, 8, and 9 in Table 4). This led to several researchers reexamining the amphibole mineralogy of this deposit. Recent work has shown the composition of amphibole

minerals found in the Rainy Creek Complex to consist of calcic and sodic–calcic amphiboles that lie in composition ranges that include tremolite, winchite, richterite, and magnesioriebeckite (Meeker et al., 2003). Chemical formulas calculated from recent electron microprobe analyses of amphiboles from the Rainy Creek Complex are presented in Table 5.

Other work has been performed on these amphibole minerals in order to characterize the morphology of the amphibole particles. Bandli and Gunter (2002) present morphological features observed with scanning electron microscopy (SEM). The observations of large bundles of fibers show how particles are flattened on the (100) crystallographic direction. Brown and Gunter (2002) used polarized light microscopy to determine aspect ratios (length:width, length:thickness, width:thickness) of a population of amphibole particles from the Rainy Creek Complex. Similar measurements were made on National Institute of Science and Technology (NIST) reference tremolite asbestos and a nonasbestos tremolite sample. This is significant because there may be a correlation between the crystal form of a particle [cleavage along the (110) crystallographic plane or twinning along the (100) crystallographic plane] and effects on human health (Zoltai, 1981; Dorling & Zussman, 1987). There also may be a correlation between increased disease potential and other dimensional ratios that have yet to be examined (Davis et al., 1991).

Since the current OSHA and U.S. EPA regulations do not regulate all amphibole asbestos minerals, it is crucial to understand the precise definition of the mineralogy of any asbestos containing material. The health effects associated with exposure to the amphibole asbestos from this location have been documented, and this particular asbestos mineral is harmful to

TABLE 5
Chemical formulas for amphiboles collected from the Rainy Creek Complex and pure end-member formulas

Amphibole name	Chemical formula
Winchite ^b	$(K_{0.13}Na_{0.25})(Na_{0.63}Ca_{1.27})(Mn_{0.01}Fe_{0.72}^{2+}Mg_{4.28}Al_{0.09})Si_{8.04}O_{22}(OH,F,Cl)_2$
Winchite ^b	$(K_{0.15}Na_{0.33})(Na_{0.61}Ca_{1.34})(Mn_{0.03}Fe_{0.50}^{2+}Mg_{4.45}Al_{0.07})Si_{8.01}O_{22}(OH,F,Cl)_2$
Winchite ^c	$(K_{0.19}Na_{0.21})(Na_{0.95}Ca_{0.97}Mn_{0.01}Fe_{0.07}^{2+})(Fe_{0.22}^{2+}Fe_{0.53}^{3+}Mg_{4.21}Ti_{0.02}Al_{0.02})(Al_{0.02}Si_{7.98})O_{22}(OH,F,Cl)_2$
Winchite ^c	$(K_{0.13}Na_{0.16})(Na_{0.61}Ca_{1.39})(Mn_{0.03}Fe_{0.21}^{2+}Fe_{0.48}^{3+}Mg_{4.28})(Al_{0.02}Si_{7.94})O_{22}(OH,F,Cl)_2$
Winchite ^c	$(K_{0.13})(Na_{0.97}Ca_{0.84}Mn_{0.01}Fe_{0.03}^{2+})(Fe_{0.22}^{2+}Fe_{0.66}^{3+}Mg_{4.06}Ti_{0.03}Al_{0.03})(Si_{8.10})O_{22}(OH,F,Cl)_2$
Winchite ^d	$(K_{0.11}Na_{0.21})(Na_{0.69}Ca_{1.09}Mn_{0.01}Fe_{0.21}^{2+})(Fe_{0.10}^{2+}Fe_{0.34}^{3+}Mg_{4.53}Ti_{0.02}Al_{0.01})(Al_{0.01}Si_{7.99})O_{22}(OH,F,Cl)_2$
Tremolite ^d	$(K_{0.13}Na_{0.25})(Na_{0.46}Ca_{1.54}Mn_{0.01})(Mn_{0.01}Fe_{0.10}^{2+}Fe_{0.04}^{3+}Mg_{4.60}Ti_{0.01}Al_{0.02})(Al_{0.01}Si_{7.99})O_{22}(OH,F,Cl)_2$
Richterite ^d	$(K_{0.22}Na_{0.44})(Na_{0.78}Ca_{1.12}Mn_{0.01}Fe_{0.09}^{2+})(Fe_{0.41}^{2+}Fe_{0.09}^{3+}Mg_{4.47}Ti_{0.02}Al_{0.02})(Al_{0.03}Si_{7.97})O_{22}(OH,F,Cl)_2$
Magnesioriebeckite ^d	$(K_{0.15}Na_{0.20})(Na_{1.59}Ca_{0.33}Mn_{0.01}Fe_{0.07}^{2+})(Fe_{0.11}^{2+}Fe_{1.12}^{3+}Mg_{3.69}Ti_{0.06}Al_{0.02})(Al_{0.02}Si_{7.98})O_{22}(OH,F,Cl)_2$
^a Tremolite ^e	$Ca_2Mg_5Si_8O_{22}(OH)_2$
^a Richterite ^e	$Na(CaNa)Mg_5Si_8O_{22}(OH)_2$
^a Winchite ^e	$(CaNa)Mg_4(Al,Fe^{3+})Si_8O_{22}(OH)_2$
^a Magnesioriebeckite ^e	$Na_2(Mg_3Fe_2^{3+})Si_8O_{22}(OH)_2$

^aEnd-member chemical formulas.

^bWylie and Verkouteren (2000).

^cBandli et al. (2003).

^dMeeker et al. (2003).

^eLeake et al. (1997).

humans. This would suggest that current regulations regarding amphibole asbestos should be revised to include all amphibole asbestos minerals. Many of the optical properties of the winchite asbestos varieties are similar to those of tremolite asbestos, but winchite asbestos is not regulated. This is problematic, as numerous cases of asbestos-related lung disease have been associated with the Libby vermiculite mining operation. The threat that amphibole asbestos poses to workers of the Zonolite Company and W. R. Grace was studied by McDonald et al. (1986a, 1986b, 1988, 2004), Amandus and Wheeler (1987), and Amandus et al. (1987a, 1987b). Regardless of the mineral species or regulations, it is clear that the amphibole asbestos mineral present at Libby should be regulated in order to prevent unnecessary risk to public health.

HEALTH EFFECTS

Health effects observed in Libby workers have been typical of other groups exposed to amphibole asbestos. The amphibole asbestos occurs as a contaminant in the vermiculite ore that was mined there. It is important to note that no adverse health effects have been observed from exposure to vermiculite alone (Ross et al., 1993). Lockey et al. (1984) examined a group of vermiculite workers who were exposed to "tremolite-asbestos" in the vermiculite ore, and determined that occupational exposure to asbestos-contaminated vermiculite could cause pleural changes. Other epidemiological studies showed substantially increased risks of lung cancer, malignant mesothelioma, and pleural changes (McDonald et al., 1988; Amandus et al., 1987b). McDonald et al. (2004) is a continuation of the studies of this cohort of vermiculite miners and adds to the knowledge of the health of miners exposed to vermiculite mined at Libby.

The studies of McDonald et al. (1986a, 1986b, 1988), Amandus and Wheeler (1987), and Amandus et al. (1987a, 1987b) were performed in parallel and studied the health of men who were involved with mining and processing the vermiculite from the RCC. W. R. Grace funded the McDonald et al. (1986a, 1986b, 1988) studies, and the National Institute for Occupational Safety and Health (NIOSH) funded the Amandus and Wheeler (1987) and Amandus et al. (1987a, 1987b) studies. Both studies estimated the amounts of airborne asbestos to which workers were exposed and calculated standard mortality ratios (SMRs) for various diseases. These studies provide the definitive evidence that the dust produced from the mining and processing of vermiculite from the RCC is harmful to humans.

McDonald et al. (1986a, 1986b, 1988) showed that there was a significantly higher incidence of lung cancer and nonmalignant respiratory disease in workers of the Libby vermiculite mine. The study examined the exposure levels and health histories of 406 men employed for at least 1 yr before 1963. It was determined that the SMR for lung cancer (SMR = 2.45) and nonmalignant respiratory disease (SMR = 2.55) were significantly higher for this cohort than for the White male population of the United States (McDonald et al., 1986a). The exposure levels were variable and dependent on workstation activity (Table 6)

TABLE 6
Fiber exposure estimates (fibers/ml)

Workstation	Year						
	Pre-1950	1955	1960	1965	1970	1975	1980
Dry mill ^a	101.5	101.5	101.5	22.1	22.1	—	—
Dry mill ^b	168.4	168.4	168.4	33.2	33.2	—	—
Wet mill ^a	—	—	—	—	3.9	1.5	0.8
Wet mill ^b	—	—	—	—	3.2	2.0	0.8
Drilling ^a	—	12.5	12.5	12.5	5.2	5.2	0.8
Drilling ^b	23.0	23.0	23.0	23.0	9.2	0.6	0.6
Concentrate loading ^a	24.0	15.0	15.0	9.0	9.0	4.8	0.2
Concentrate loading ^b	82.5	27.7	10.7	10.7	3.2	0.2	0.2
Skip area ^a	—	68.8	68.8	15.0	15.0	2.0	0.6
Skip area ^b	88.3	88.3	88.3	17.4	17.4	0.6	0.6
River dock ^a	—	—	12.0	12.0	12.0	12.0	0.7
River dock ^b	116.9	42.5	17.0	17.0	17.0	5.1	0.5

^aMcDonald et al. (1986a).

^bAmandus et al. (1987a).

(McDonald et al., 1986b). It was shown that after W. R. Grace acquired the mine, dust levels decreased significantly. It was also shown that for each fiber-year of exposure there was a 1% increase in the probability of a worker developing lung cancer (McDonald et al., 1988).

Amandus and Wheeler (1987) and Amandus et al. (1987a, 1987b) replicated the studies of McDonald et al. (1986a, 1986b, 1988). These studies examined the exposure levels and health histories of 575 men who had been hired before 1970 and were employed for at least 1 yr. Exposure estimates (Amandus et al., 1987a) were determined using previous measurements and workstation activities. Amandus et al. (1987a) estimated the exposure rates for workers at the Libby vermiculite operations and determined that from the onset of mining to the mid 1980s there was a significant decrease in the levels of airborne asbestos (Table 6). These data were used to determine individual cumulative fiber exposure (fiber-years). The estimates show that, in general, exposure was highly variable depending on a worker's workstation activity and that dust levels had significantly decreased for all work activities after 1963, when W. R. Grace acquired the mine. SMR for lung cancer (SMR = 2.44) and SMR for nonmalignant respiratory diseases (SMR = 2.42) were found to be significantly higher than that of the general White male population in the United States. The increase in the risk of developing lung cancer was determined to be 0.6% for each fiber-year of exposure (Amandus & Wheeler, 1987).

These two studies show that the asbestos minerals present in the Libby vermiculite ore posed a significant health risk to workers who were exposed at high levels. However, these studies incorrectly refer to the amphibole asbestos mineral present as tremolite. It was noted in Nolan et al. (1991) that amphibole

asbestos samples from Libby that had been previously identified as tremolite were not in fact tremolite; Nolan et al. used energy-dispersive spectroscopy (SEM-EDS) to examine the chemical composition of multiple samples and found that the chemistry of samples from Libby was not consistent with that of tremolite. Recent mineralogical work on the amphibole minerals from Libby has shown that winchite is most likely the predominant species of amphibole asbestos present (Wylie & Verkouteren, 2000; Gunter et al., 2003; Bandli et al., 2003; Meeker et al., 2003).

Both McDonald et al. (1986a) and Amandus et al. (1987a) showed that workers employed in the unregulated workplace (before 1970) were exposed to significantly higher levels of amphibole asbestos than those employed later. Table 6 presents estimated amphibole asbestos dust levels at various workstations during the life of the mine. Dust levels were not measured in all years, so dust levels were assumed to remain constant until the next measurement was made. The data show several significant changes in the amounts of dust to which the workers at Libby were exposed. Dust levels were extremely high in the dry mill before W. R. Grace acquired the operation in 1963, and dust levels were reduced by approximately 75% by 1965. With the elimination of the dry mill in 1974, the largest source of airborne amphibole asbestos fibers was removed, and with the introduction of federal regulations in 1972 (Table 2), the fiber exposure for workers was reduced further. The McDonald et al. (1986a) estimates are consistently lower than those of Amandus et al. (1987a), but both show the consistent trend of decreasing fiber exposure with time.

Preliminary results of a recent Agency for Toxic Substances and Disease Registry study (ATSDR, 2001; Peipens et al., 2003) of Libby residents reveal that 18% of the population has pleural abnormalities; 2% had no direct exposure to asbestos, and 5% of those who had no direct exposure (0.1% of the study group) have lung abnormalities consistent with asbestos exposure. This may mean that the asbestos present at Libby is hazardous even at very low exposure levels. Peipens et al. (2003) determined that the greater the number of exposure pathways a person was exposed to, the higher is the likelihood that lung abnormalities would be observed in chest x-rays. It was also determined that the most significant risk factors were age, employment at the vermiculite mine, living with an employee of the mine, and being male. However, there is very little information about how much asbestos residents of Libby were actually exposed to. The major concern is that environmental exposure to the amphibole asbestos from Libby is harmful. Browne and Wagner (2001) found that there are no reliable data to support claims that increased environmental exposure to amphibole asbestos leads to an increased incidence of mesothelioma, but their data do not include the recent, ongoing studies in Libby. Peipens et al. (2003) imply that there is a higher risk for individuals living in Libby prior to 1990 developing pleural abnormalities as a result of environmental exposures only. However, the significance of their data is being debated within the epidemiological community (see *Environmental Health Perspectives*, February 2004, Corre-

spondence). Pfau et al. (2005) showed an increased autoimmune response to individuals exposed to asbestos in Libby when compared to those not exposed. This result shows that a relationship may exist between certain autoimmune responses and asbestos exposure even at very low exposure levels.

SUMMARY

The RCC alkaline-ultramafic igneous intrusion was mined for 67 yr for vermiculite, which has numerous industrial applications. It still contains significant amounts of vermiculite ore. However, the geologic processes that created the vermiculite also worked to create amphibole asbestos, which proved harmful to those exposed to it at elevated levels for extended periods of time. As pressure from the regulatory agencies and residents of Libby to remove asbestos contamination from the vermiculite mining and milling operations in Libby and elsewhere around the United States continues, more information about exactly what species of amphibole minerals will be required. The classification of the amphibole asbestos present in vermiculite products that originated from the Libby mine has been clouded in confusion. For the past 6 yr, the U.S. EPA and the media have continued to call these amphibole minerals tremolite, when indeed they are not. Correct classification of these asbestiform amphiboles will require a change in the regulations to protect human health. Ironically and interestingly, many of the health risks of tremolite have been based on the misconception that the amphibole asbestos found at Libby was tremolite. We now know this to be incorrect.

Since November 1999, the U.S. EPA has been actively involved in abatement of asbestos contamination resulting from the vermiculite mining and milling operations at Libby. The main focus of the U.S. EPA's cleanup effort has been on the Export Plant at the mouth of Rainy Creek, but several other sites in Libby, including the Libby High School, Burlington Northern and Santa Fe Railway yard, and multiple residential areas, are being considered for asbestos cleanup projects. Currently, the Libby area is listed as a Superfund site and cleanup activities are ongoing. Considerable sums of money have already been spent cleaning up the former export plant and several other locations in Libby where vermiculite was used for various purposes. In order to clean up all asbestos contamination in Libby, considerably more funds will be required. A final decision by the U.S. EPA as to how it will deal with asbestos contamination at Libby will be made in the near future. It will also be necessary to decide how to handle the vermiculite insulation that was used in approximately 1 million homes across the United States (Gunter et al., 2005) and to examine the levels of asbestos contamination that occurred at the numerous vermiculite expansion facilities that were operated nationwide.

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