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THE MINERALOGY AND SIZE OF AIRBORNE CHRYSOTILE AND ROCK FRAGMENTS: RAMIFICATIONS OF USING THE NIOSH 7400 METHOD

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The length and width of chrysotile and rock fragments that were collected on nine air-monitoring filters in the mine and plant of the Lowell asbestos mine in Vermont have been measured by transmission electron microscopy (TEM). Selective area electron diffraction (SAED) and energy dispersive x-ray analysis (EDS) were used to identify particles longer than 5 µm with a length-to-width aspect ratio of at least 3:1 (federal fiber). All federal fibers were found to be chrysotile or serpentinite rock fragment; no tremolite or other amphiboles were detected. Magnifications of 400× and 19 000× were used on five filters in an attempt to compare the size distributions of the federal fibers likely to be measured by using phase contrast optical microscopy (PCM) at 400× to those measured by TEM at higher magnification. The data from the mine show that (1) the size distribution of chrysotile determined at 19 000× differs substantially from that determined at 400× but the size distribution of rock fragment is nearly independent of the magnification; and (2) at 400×, 34% of the federal fibers were chrysotile, 39% were serpentinite rock fragment, and 27% were composite particles, not fibers. At 19 000×, the proportion of chrysotile increased to 77%, reflecting the increased visibility of chrysotile at high magnification. The proportions of chrysotile, serpentinite rock, and composite particles are such that if an air filter were analyzed at 400×, and 1.0 f/cc were determined to be the exposure, 0.3 f/cc would be chrysotile, 0.4 f/cc would be serpentinite rock, and 0.3 f/cc would be composite particles. If TEM were used at high magnification, the total federal fiber burden would rise to 1.6 f/cc with 1.2 f/cc chrysotile and 0.4 f/cc rock fragment. These results suggest that the proportion of federal fibers obtained by the standard PCM method that are actually asbestos may be lower in the chrysotile asbestos mining environment than that obtained in the commercial asbestos handling environments that were used in government quantitative risk assessments. The Occupational Safety and Health Administration excluded epidemiologic studies of asbestos miners and millers from its quantitative risk assessment because evidence showed the risk to be lower than in other industrial environments because of fiber size. Likewise, the use of the PCM National Institute for Occupational Safety and Health 7400 method, which was developed from data taken in commercial asbestos handling as an "index" of exposure, may not be valid in mining environments. TEM analysis of air filters may be necessary to assess chrysotile exposure adequately in mining environments.

Within any given industrial setting, there exists a positive correlation between the incidence of the asbestos-related diseases and the level of exposure to asbestos as established by phase contrast optical microscopy (PCM) and the membrane filter method.[1] However, it is generally not possible to predict closely the risk of disease within one industry by comparing exposures of its workers to exposures and disease incidence from a different industrial environment. For example, the incidence of asbestos-related diseases among Canadian chrysotile miners is less than would be predicted from the experience of textile workers or asbestos insulation workers.[2–5] Also, there is no elevated incidence of mesothelioma among anthophyllite asbestos miners of Paakila, Finland, although high incidence would be predicted based on the experience of crocidolite miners in Australia and South Africa.[6–8]

These observations lead to several possible hypotheses to explain the discrepancies. First, it may be that there are significant differences in the biological activity among the different asbestos minerals. For example, chrysotile fibers appear to dissolve or in some other way be removed from the body but crocidolite fibers do not, and because of this, the long-term effects of exposure to chrysotile may be quite different from the same level of exposure to crocidolite.[9] In fact, because of the behavior of chrysotile fibers in vivo, tremolite-asbestos, which in some cases has been identified as a contaminant in chrysotile, has received much attention as the possible etiologic agent for the diseases that are observed in those "solely" exposed to chrysotile.[10–13] Second, there may be significant differences in the size and shape of the respirable mineral particles making up the dust clouds in different industrial settings; differences that are simply not reflected in the PCM exposure measurements.[14]

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For example, because the average width of amosite fiber is greater than the average width of crocidolite fiber, more of the amosite fiber will be visible by PCM. In this case, therefore, more of the total airborne amosite fiber will be counted to assess exposure than of the total airborne crocidolite fiber when the total airborne fiber concentrations of the two are actually the same. Third, there may be differences in the aerodynamic characteristics among the asbestos minerals, such as chrysotile's propensity to curl compared to the relatively straight amosite fiber, that affect the fate of an inhaled fiber in the body. Although the relative importance of these variables is not well known, the explanation for the observed differences in risk for the same PCM-measured exposure is likely that both fiber type and fiber size play a role, and it is important that both of these variables be examined more closely.

This study was undertaken in order to examine the mineralogy and size distribution of the airborne particles at the Lowell chrysotile mine in Vermont. The Lowell mine is located in the upper Missisquoi Valley just south of the international boundary with the Canadian province of Quebec. Chrysotile has been produced from this mine and nearby pits for more than 100 yr. More than 80% of the asbestos mined in the United States has come from this mining district, and the Lowell mine is the largest and most productive mine in the area. It is located in the same geologic terrane as the larger asbestos deposits in Quebec.[15] The geology of the Lowell mine and vicinity is described in detail by Cady et al.[16]

Specifically, the authors were interested in determining what minerals would be included in a PCM fiber count, how the specific minerals differ in particle size and shape, and how the use of transmission electron microscopy (TEM) affects determination of fiber exposure. The authors also wanted to determine if tremolite was an important constituent of airborne dust in the mine because it is widely assumed that tremolite is ubiquitous in chrysotile deposits.[10-13,17-19] Estimates of tremolite asbestos contamination in chrysotile mines range as high as 1.5%.[20]

EXPERIMENTAL MATERIALS AND METHODS

Samples

Nine air filters from the Lowell mine were provided by the United States Mine Safety and Health Administration (MSHA). No information was provided on the airflow or duration of collection. Six of the filters contained particles collected in the mine, two of the filters were from the mill, and one was from the bagging room. The filters were collected in 1987. The samples were prepared for TEM analysis by AMA Laboratories in Laurel, Md., according to the procedures specified by the Environmental Protection Agency[21] for air filter analysis in school buildings. Two 200-mesh copper grids were prepared from each filter.

High Magnification

Randomly chosen grid openings were scanned at a magnification of 19 000×. All particles longer than 5 μm with an aspect ratio of 3:1 or greater (federal fibers) were identified and their length and width determined directly from the screen by comparison to a calibrated scale. The precision of this method of measurement is estimated at 10%.[22] Identification was based on selective area electron diffraction (SAED) pattern, qualitative chemical analysis by energy dispersive x-ray analysis (EDS), and morphology. For a federal fiber to be positively identified as chrysotile, it had to have the appropriate chemical composition and possess either an obvious tubular structure or a characteristic diffraction pattern. Fiber analysis continued until at least 50 federal fibers from each filter were measured and identified.

Low Magnification

Randomly chosen grid openings were first scanned at a magnification of 400×. All particles that met the federal fiber size criteria, as determined by comparison to a calibrated scale on the screen, were designated. A diagram of the grid opening with the position of these federal fibers was made. Each designated federal fiber was then examined at a magnification of 19 000×. The federal fibers were identified as chrysotile or "other" based on the diffraction pattern and tubular morphology and, if necessary, EDS. For federal fibers exhibiting diffraction patterns that could be amphibole, EDS would have been used to identify the amphibole tentatively. However, no amphibole particles were found. The length and width of all chrysotile and other federal fibers were measured in the same manner described above.

RESULTS

Mineralogy

In this study, a total of 517 federal fibers were measured and identified. A total of 63% were found to be chrysotile and 37% were identified as other. For the 256 federal fibers examined at high magnification, all those identified as "other" had a Si:Mg ratio and a diffraction pattern (when obtainable) consistent with the serpentinite minerals antigorite or lizardite. MSHA, by using an automated imaging system, had previously studied these filters and identified 477 federal fibers on the basis of qualitative chemical composition and, in part, diffraction patterns.[23] MSHA identified a large portion of the federal fibers as antigorite/lizardite and found small amounts of a number of other minerals, including magnetite, plagioclase feldspar, chlorite, quartz, enstatite, olivine, diopside, and calcite. MSHA did not identify tremolite or any other amphibole.

When the federal fibers designated at low magnification were examined at 19 000×, a large portion were not fibers at all but were composed of a linear array of smaller particles. These particles were sometimes both chrysotile and serpentinite rock fragments, and they sometimes were only serpentinite rock fragments. A total of 29% of the elongated particles from the mine were found to be such "composite particles." Of the elongated particles from the mill sample, 10% were also composite particles. Although there is a significant difference between the number of composite particles found in

TABLE I. Distribution of Particles at 400× Magnification

	Chrysotile		Other Minerals		Composite Particles	
Filter	(No.)	(%)	(No.)	(%)	(No.)	(%)
Mine 3	20	30	27	40	20	30
Mine 4	14	29	21	44	13	27
Mine 6	18	31	17	29	23	40
Mine 7	12	20	36	61	11	19
Mine 9	36	49	15	20	23	31
Mine total	100	33	116	38	90	29
Mill 5	22	44	23	48	5	10
Total mine and mill	122	34	139	39	95	27

the mill (one filter) and in the mine, it is impossible to tell whether this is caused by real differences in the nature of the airborne dust cloud in these two environments or to a difference in filter loading (see Table I). Because "composite particles" were not fibers, they are not used in comparing the size distributions at 400× and 19 000×.

Table II gives the distributions of chrysotile and federal fibers identified as "other" according to sample location and the magnification. At low magnification, more than half of the federal fibers were found to be a mineral other than chrysotile. When the filters were first examined at high magnification, most of the federal fibers were found to be chrysotile and the proportion of other federal fibers was significantly less than at low magnification. The difference between the proportion of airborne chrysotile in the mine and the proportion of airborne chrysotile in the mill or bagging room is statistically insignificant (5% level of significance).

TABLE II. Mineralogical Distribution of Single Particles at 400× and 19 000× Magnification

	Chrysotile		Other Minerals	
Filter	(No.)	(%)	(No.)	(%)
400×				
Mine 3	20	43	27	57
Mine 4	14	40	21	60
Mine 6	18	51	17	49
Mine 7	12	25	36	75
Mine 9	36	71	15	29
Mine total	100	46	116	54
Mill 5	22	49	23	51
19 000×				
Mine 8	38	70	16	30
Mine 6	46	92	4	8
Mine 4	35	69	16	31
Mine total	119	77	36	23
Mill 1	44	88	6	12
Bagging room 10	43	84	8	16
Plant total	87	86	14	14

Size Distributions

The frequency distributions of log length, log width, and log aspect ratio of federal fibers collected from the mine at 400× and 19 000× magnification that are not chrysotile are given in Figure 1a–f. Log values are used to approximate normal distributions. The similarity of these distributions indicates that there is little evidence for systematic bias in the characterization of these federal fibers introduced by changing magnifications over this range. The ranges, modal classes, and means are similar. However, only the means of log length from the low and high magnifications are statistically indistinguishable (t-test). Mean log width and mean log aspect ratio are not identical even though they are close in magnitude.

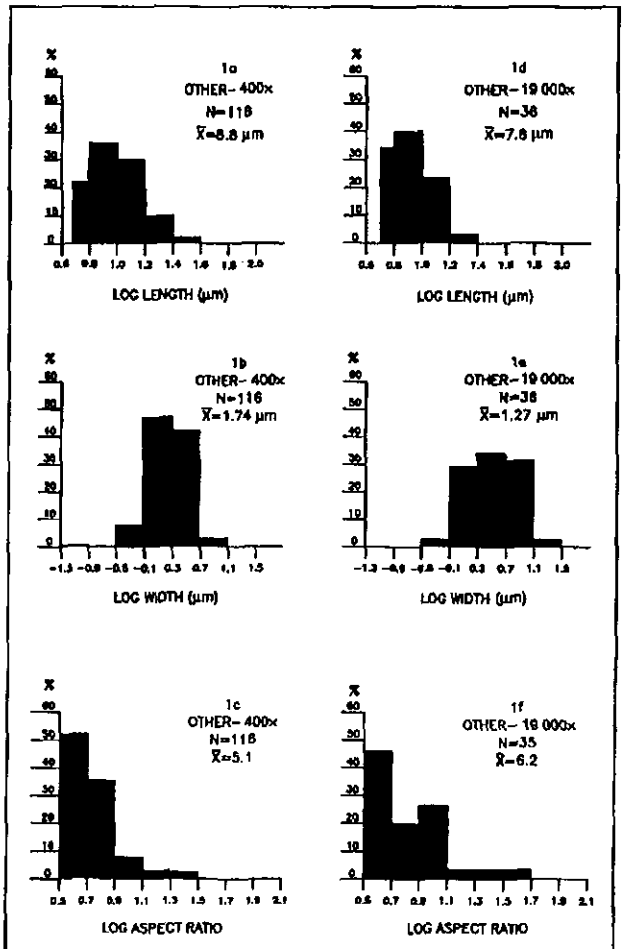

FIGURE 1. Nonasbestos particle dimension distributions

The frequency distributions of log length, log width, and log aspect ratio of chrysotile collected from the mine at 400× and 19 000× are given in Figure 2a–f. Changing magnification has had a significant impact on the distributions of these variables. Thin, short, high-aspect-ratio fibers dominate the populations analyzed at high magnification.

A comparison of the size distributions of serpentinite rock fragment and chrysotile shows that these two populations are distinct, whether examined at low or high magnification. However, there is significant overlap at both low and high magnification in all three dimensional parameters, and it is not possible to discriminate effectively between these populations by introducing a simple dimensional parameter such as an aspect ratio of 20:1 or a width of 1 μm as may be appropriate in other environments and has been suggested elsewhere.[24–26]

DISCUSSION

Estimates of the minimum width of a fiber that can be seen by phase contrast microscopy at 400× range from 0.1 to 0.5 μm.[27]

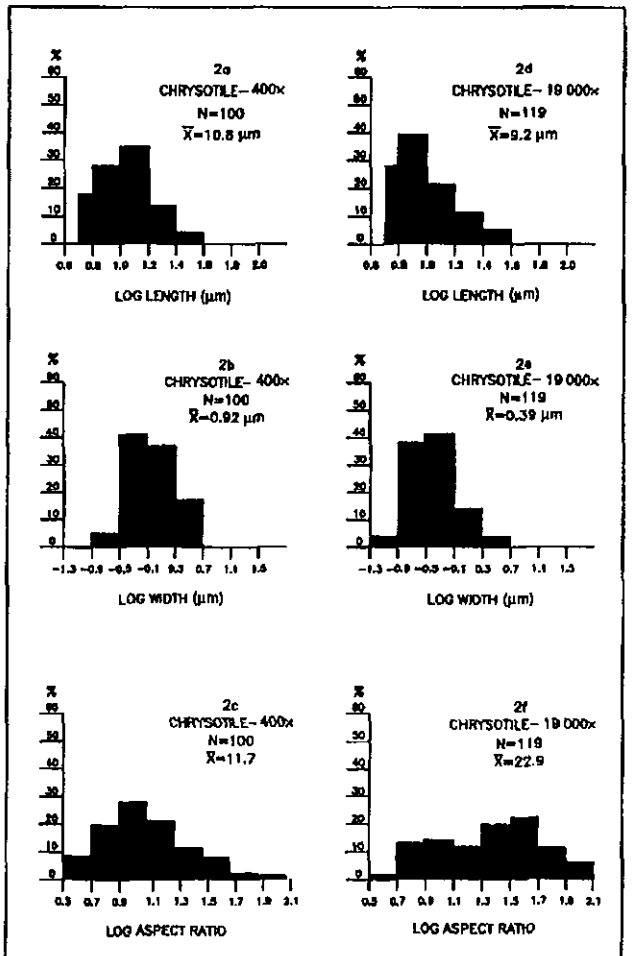

FIGURE 2. Chrysotile particle dimension distributions

Whether the observations made at 400× by TEM duplicate what would be seen optically by phase contrast microscopy is difficult to evaluate. Visibility is not only a function of the resolution of the system, it is also a function of the magnification, the difference in the index of refraction of the fiber and the substrate, and the visual acuity of the observer.

A comparison of width distributions of chrysotile at 400× (Figure 2b) and at 19 000× (Figure 2e) shows abundant chrysotile fibers with widths less than 0.4 μm present on the air filters. It is also evident that most of these fibers are not visible at 400×.

Figure 3 shows the aspect ratio frequency distribution of chrysotile fibers with widths greater than 0.4 μm and those that are visible at 400×, scaled so that the frequency of the modal class at 400× is equivalent to the frequency at 19 000×. The similarity in the distributions between the chrysotile greater than 0.4 μm and that visible at 400× reinforces the conclusion that the visibility of chrysotile at 400× on the TEM is comparable to the visibility of chrysotile by phase contrast microscopy.

Accurate assessment of the airborne fiber content in occupational settings where epidemiologic studies have shown a correlation between exposure to airborne fiber and incidence of asbestos-related diseases is essential for risk assessment and forms the basis for establishing acceptable exposure limits in occupational settings. If federal fibers other than asbestos are included in unequal proportions in different occupational settings, the reliability of the occupational exposure standard for all environments is suspect. Very few data are available on what proportion of a fiber count, as established by the membrane filter method, is composed of federal fibers that are *not* asbestos.[14] This study clearly indicates that for this mining environment, the proportion is large (54%); it would be inappropriate to use exposure data from this mine to assess risk in another environment where the proportion of asbestos is different.

Because the distributions of length, width, and aspect ratio of serpentinite rock fragments visible at 400× are so similar to the distributions visible at 19 000×, it is reasonable to assume that most serpentinite rock federal fibers that are visible at 19 000× would also be visible at 400×. Of the serpentinite rock federal fibers visible at 19 000×, 3% have widths less than 0.3 μm and none visible at 400× have widths this small, so the populations are clearly not identical. Nonetheless, the populations are similar enough that predictions based on the assumption that they are the same can be reasonably made. Accepting this assumption, the concentration in fibers per cubic centimeter of serpentinite rock federal fibers would be the same whether the filters were examined with low or high magnification. If an air filter from this mine were analyzed at 400× and the total fiber concentration were determined to be 1.0 f/cc, the proportion of serpentinite rock fragment, chrysotile, and composite particles would be 0.4, 0.3, and 0.3, respectively. If the same filter were examined at 19 000× and the concentration of serpentinite fragments would remain constant at 0.4 f/cc, the likely concentration of chrysotile would be 1.2 f/cc.

The proportions of chrysotile and rock fragment found on an air filter may vary greatly as a function of the environment. That the proportion of rock fragment will be considerably greater in a mining environment than in an industrial setting where a commercial asbestos product was applied is certainly reasonable; it may also vary from one mine to another. Not only will the amount of rock fragment meeting the dimensional criteria for a fiber as defined by the Occupational Safety and Health Administration (OSHA) vary from one mine to another, but the proportion of asbestos meeting these criteria may also vary. For example, the distribution of length and width of airborne chrysotile at the Lowell mine differs significantly from that at the chrysotile mines in Quebec as described by Gibbs and Hwang.[28] Therefore, these data cannot be applied directly to all other mining environments.

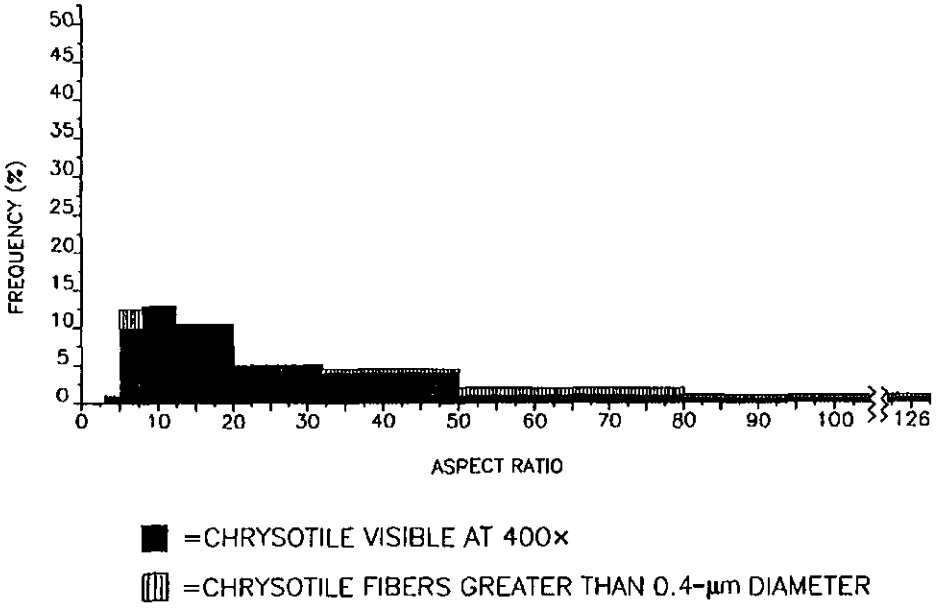

FIGURE 3. Chrysotile asbestos aspect ratio distributions

These results do suggest that the proportion of federal fibers obtained by the standard PCM method that are actually asbestos may be lower in the chrysotile asbestos mining environment than that obtained in the commercial asbestos handling environments (i.e., textile, insulation). The quantitative risk assessments were calculated from these commercial asbestos environments; ultimately, the levels of permitted exposure in regulatory standards were established. OSHA, in its rulemaking deliberations on asbestos, stated that "There is some evidence that risks in asbestos mining and milling operations are lower than other industrial operations due to differences in fiber size." For this reason, the epidemiologic data from mining and milling operations were not considered.[29] These data show that the use of the PCM National Institute for Occupational Safety and Health 7400 method, which was developed from data taken in commercial asbestos handling environments for the purpose of providing an "index" of actual asbestos exposure, may not be valid in the mining environments. The use of the PCM method in mining environments can present a significant problem in determining true asbestos exposure levels. This problem must be addressed by using more sophisticated means of analysis such as TEM.

The fact that neither tremolite or actinolite were found among the airborne federal fibers from the Lowell chrysotile mine points up the need to determine the abundance of tremolite-asbestos throughout the chrysotile mining industry. These data indicate that tremolite may not contaminate all chrysotile deposits at measurable levels. Before tremolite-asbestos can be assumed to be the active agent in all chrysotile exposure, more data on its abundance are needed.

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