



## ENVIRONMENTAL SAMPLING AND ANALYSIS OF PARTICULATE MATTER: ASBESTOS

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### Introduction

There is, at present, no practical absolute method for the routine measurement of exposure to airborne asbestos fibers. The technical difficulties of the electron microscope for dust counting make its routine use unfeasible; when it is used, the results are heavily biased in favor of very small fibers.

Each of the methods considered measures something different and, therefore, they do not correlate well. Ratios between results by different methods should be used for qualitative prediction only.

Each method is strongly influenced by one attribute of the dust cloud. The biologically optimum method cannot be selected unless it is known which attribute is biologically most appropriate.

The preferred index of asbestos exposure is fibers longer than 5 microns counted on membrane filters at 430X phase contrast (PC). The method of counting is convenient and practical, and fibers > 5 microns constitute a direct index of asbestos fiber exposure.

At the New York Academy of Sciences Conference on the Biological Effects of Asbestos in 1965, Gilson<sup>(1)</sup> noted the proliferation of dust sampling methods and appealed for international comparability of dust measurement data to be related to biological data.

There could be as many as 100 indices of exposure to the asbestos in a dust cloud. In a United States Public Health Service (USPHS) epidemiological study of the asbestos products industry, the choice was narrowed down to the count and weight methods shown in Table 1. Based on the results of analysis of almost 10,000 samples, this paper reviews previously published relationships between these methods in order to:

- (1) exhibit the effect of product and operation,
- (2) reveal the bias of the methods, and
- (3) evaluate the usefulness of the indices derived. (2)

Table 1  
Selected Measures of Fiber Exposure

| Count                                    | Weight                             |
|------------------------------------------|------------------------------------|
| Total dust in impinger at 100X           | Total dust                         |
| Total fiber on MF at 430X PC             | Respirable dust                    |
| Fibers > 5 microns on MF at 430X PC      | Chrysotile by magnesium            |
| Total fiber on MF by electron microscope | Respirable chrysotile by magnesium |

### Impinger Relationships

The early epidemiologic data on asbestos were based on the impinger method and, until the beginning of the present study, most sampling in the United States usually was done by this method, since only in this way could the exposure be evaluated against the threshold limit value (TLV) of 5 million particles per cubic foot (MPPCF).<sup>(3, 4)</sup> Consequently, it is necessary to develop ratios between present methods of measurement and impinger counts performed in the traditional manner so that the results that would have been obtained years ago had present methods been used might be predicted.

In the early days of counting dust particles obtained in impinger samples in asbestos textile factories, few fibers were seen, and as a result all particles were counted to give an estimate of overall dustiness. The lack of visible fibers in these samples was due in part to the inefficiency of the impinger in the collection of particles of such small aerodynamic size and in part to the low resolving power of the 10X objective used. As a result of recent studies of the asbestos industry, techniques were evolved for detecting many of the airborne fibers previously missed.<sup>(5, 6)</sup> Membrane filters with a pore size of 0.8 micron are known to have surface collection efficiencies close to 100% for particles down to a few hundredths of a micron. Airborne dust samples collected on these filters may be viewed by high resolution transmitted light microscopy if the filter can be rendered transparent or "cleared." Acetone, glycerol triacetate, immersion oil, and other materials have been used to clear membrane filters. However, the fiber being sought frequently has an index of refraction very near the index of the cleared filter. Thus, under bright field illumination, the fiber may be invisible. Phase contrast illumination, which utilizes the interference patterns produced by the combination of two out-of-phase columns of light, is capable of revealing objects imbedded in media with an index of refraction only slightly different from that of the object. The sensitivity of this method to index differences necessitates the use of glycerol triacetate or a diethyl oxalate-dimethyl phthalate mixture to clear the filter perfectly. Clearance of the filter with either of the liquids alone may cause displacement of the particles by too rapid solution of the filter. To prevent this, the particles are either fixed with perspex in chloroform before clearing with glycerol triacetate (British practice) or pure membrane filter material is added to the diethyl oxalate-dimethyl phthalate mixture before it is used to clear a filter to slow the rate of filter solution (USPHS method). With these techniques, fibers whose thickness is only a few tenths of a micron can be seen.

Larger fibers (over 1 micron thick) may sometimes be identified by dispersion staining.<sup>(7)</sup> If a transparent particle is immersed in a liquid with a steeper dispersion curve, that is, large variation in index of refraction with wavelength, and if the index of the particle matches the index of the liquid at some wavelength in the visible range, a dispersion color will be produced. When the objective lens is equipped with a central stop in the back focal plane of the lens, the primary image will be eliminated and only the light deviated by the dispersion will be seen. The color of the particle thus observed will depend on the wavelength of light at which the index of the particle matches the index of the liquid. A wide variety of high dispersion liquids covering the index of refraction range from 1.350 to 2.11 are available. With the proper liquid, transparent isotropic particles, such as glass fibers, may be easily identified. For anisotropic minerals, such as asbestos, a mixture of colors is produced and identification becomes more difficult. The addition of a polarizer results in the production of the single dispersion color corresponding to the index of refraction of the fiber along the axis observed. By rotating the polarizer and noting the "highest" (most red) color observed, the various indices along different axis may be measured and the particle identity determined. In this manner asbestos may be differentiated from other minerals although determination of the type of asbestos is more difficult due to a variation in the index of refraction with composition. By using an objective with a centered stop (centered screening or dark field dispersion), isotropic particles of about 1 micron may be identified.

The ratios shown in Table 2 were developed from simultaneous pairs of impinger-membrane filter samples and are expressed as the fiber counts equivalent to 1 mppcf by impinger. Equivalence means the fiber concentration that would be found, on the average, when the impinger samples were giving, on the average, a concentration of 1 mppcf in the same product area. The product areas are: textile for asbestos spun and woven products, friction for brake linings and clutch facings, and asbestos cement pipe.

Table 2  
Impinger/Membrane Filter Count Ratios  
(fibers/ml for 1 mppcf)

| Product  | Total            | Fibers           |           | Number of Pairs |
|----------|------------------|------------------|-----------|-----------------|
|          |                  | > 5 $\mu$        | >10 $\mu$ |                 |
| Textile  | 10.3             | 5.9              | 2.7       | 500             |
| Friction | 3.7              | 2.2              | 1.0       | 200             |
| Pipe     | 4.8 <sup>a</sup> | 1.9 <sup>a</sup> | 0.7       | 100             |

<sup>a</sup> Correlation coefficient not significant at 95%.

In all cases the correlation coefficients from the large numbers of pairs of simultaneous samples are not over 0.6, as would be expected from pairs of measurements of a real environment, each having its own intrinsic error and not necessarily measuring the same thing. The impinger results are counts of all visible particles (predominantly grains with generally less than 20% fiber). These few fibers seen in the impinger sample are a small fraction of the fibers counted on the membrane filters, since the membrane filter is a more efficient collector of fibers and the magnification/illumination used results in improved visibility. In the case of the impinger versus total and > 5-micron fiber ratios in the pipe plants, the correlation coefficient was not significantly different from zero at the 95% confidence level, and the hypothesis that the measures of exposure are unrelated cannot be rejected. In this case a usable prediction ratio will probably need to be based on ratios calculated for subsets within pipe manufacturing operations.

#### Size and Visibility

Table 3 is broken down by operation within product area. Despite the fact that the textile operations and the three mixing operations are all cases of secondary dust dispersion (that is, the small dust particles already exist in the bulk material and are being shaken loose) whereas grinding and finishing are primary dispersion (that is, the dust is being produced and dispersed simultaneously by mechanical abrasion), the proportions are quite consistent. Although mode of dispersion undoubtedly has an effect on the proportion of large particles, this effect apparently has a negligible influence on the number distribution of particles in the size range of biological significance. Similarly, the variation in bulk asbestos quality, from the long fiber used in textile to the short fiber used in pipe, has no apparent effect on the count proportions of fibers by length.

Vorwald, (8) in his early animal studies of the toxicity of asbestos, suggested that the long fibers are more toxic than the short, and this possibility has been raised again by the results of recent investigations. (9) While this possibility undoubtedly influenced researchers in England and in the United States to count fibers longer than 5 microns, the present continued use of this index is based on the reduction of visibility variability, referred to above, as well as on the biological implications.

The ratio of the number of fibers seen at 970X PC to those seen at 430X PC on the same filter is shown in Table 4. These results are based on a relatively small number of samples, and the confidence limits may be quite wide. They do, however, indicate that the increase in visibility is roughly the same for fibers longer than 5 microns as for total fibers. Thus, the critical factor in fiber resolution at this level of magnification is fiber diameter.

Table 3  
Fiber Size Distribution

| Operation                       | 430X Phase Contrast<br>% of Total Fibers |                         |
|---------------------------------|------------------------------------------|-------------------------|
|                                 | Longer<br>than 5 $\mu$                   | Longer<br>than 10 $\mu$ |
| Textile                         |                                          |                         |
| Fiber preparation and carding   | 50                                       | 25                      |
| Spinning, twisting, and weaving | 61                                       | 38                      |
| Friction                        |                                          |                         |
| Mixing                          | 68                                       | 30                      |
| Grinding, cutting, and drilling | 63                                       | 31                      |
| Pipe                            |                                          |                         |
| Mixing                          | 57                                       | 28                      |
| Finishing                       | 58                                       | 27                      |
| Insulation                      |                                          |                         |
| Mixing                          | 55                                       | 27                      |
| Finishing                       | 50                                       | 29                      |

Table 4  
Ratio of 970X to 430X Fiber Concentrations  
(phase contrast)

| Operation                       | Fibers |          |
|---------------------------------|--------|----------|
|                                 | Total  | >5 $\mu$ |
| Textile                         |        |          |
| Fiber preparation and carding   | 2.0    | 2.1      |
| Spinning, twisting, and weaving | 1.8    | 1.9      |
| Friction                        |        |          |
| Mixing                          | 1.1    | 1.1      |
| Grinding, cutting, and drilling | 1.0    | 1.1      |
| Pipe                            |        |          |
| Mixing                          | 1.2    | 1.2      |
| Finishing                       | 1.6    | 1.8      |
| Insulation                      |        |          |
| Mixing                          | 1.0    | 1.1      |
| Finishing                       | 1.8    | 2.0      |

As shown in Table 4, 430X PC may count as few as half of the fibers >5-microns visible at 970X PC. Even total fiber counts at 430X PC, which are about twice as high as over—5-micron 430X PC counts (Table 3), still include less than 10% of the true number of fibers in the air.

Since all light microscope methods can measure only a small part of the whole airborne fiber cloud, it is important to consider the use of electron microscope counts as an exposure index. Certain disadvantages, such as preparation time, equipment cost, and lack of general availability, are obvious. Several other problems should also be considered. Samples of a particle density appropriate for light microscope counting are too sparse for electron microscopy unless a very large number of fields are counted. Special techniques are required for random field selection to avoid counter bias. Most important is the occasional presence of large numbers of very small fibers (about 0.1-micron thick by 1.0-micron long). Since these small fibers often occur in groups, it is possible that they are airborne as clumps (of respirable size) rather than as single fibers.

A number of electron microscope preparations of samples previously counted by optical microscopy were made. The counts of longer fibers on electron micrographs did not appear to be greater than those obtained by optical microscopy. The technical difficulties involved in the preparation and counting of asbestos samples make quantitative conclusions impossible from the data obtained, but it appeared that the major difference between electron microscope counts and optical counts was in those fibers shorter than 1-micron. These small fibers, when they are present, dominate the electron microscope count. Even if the other technical difficulties mentioned could be overcome, what would result, from an electron microscope count method, would be fiber concentrations strongly biased by these very small fibers. Present evidence does not indicate that they should be given such a dominant role.<sup>(2)</sup>

#### Weight Relationships

It is possible to count asbestos fibers on one-half of a membrane filter at 430X PC and to analyze the other half for magnesium. In this way, the direct count/weight ratios shown in Table 5 were obtained. Several difficulties with this method are readily apparent. Only chrysotile asbestos is determined; thus, the crocidolite used in some types of asbestos cement pressure pipes is not included in the weight of asbestos. Further, all other sources of magnesium, especially the nonfibrous parent rock associated with the chrysotile asbestos, interfere directly.

Table 5  
Count/Weight Ratios

| <u>Product</u> | <u>Type of<br/>Fiber Count</u> | <u>Geometric<br/>Mean<br/>(fibers per <math>\mu\text{g}</math>)</u> | <u>Geometric<br/>Standard<br/>Deviation</u> |
|----------------|--------------------------------|---------------------------------------------------------------------|---------------------------------------------|
| Textile        | Total                          | 14,500                                                              | 2.5                                         |
|                | > 5 $\mu$                      | 6,700                                                               | 3.3                                         |
|                | > 10 $\mu$                     | 3,400                                                               | 3.5                                         |
| Friction       | Total                          | 26,300                                                              | 3.4                                         |
|                | > 5 $\mu$                      | 13,900                                                              | 3.6                                         |
|                | > 10 $\mu$                     | 5,300                                                               | 4.4                                         |
| Pipe           | Total                          | 46,500                                                              | 2.8                                         |
|                | > 5 $\mu$                      | 22,500                                                              | 2.9                                         |
|                | > 10 $\mu$                     | 8,300                                                               | 3.4                                         |

Although count data did not exhibit any consistent shift in fiber length distribution with product, operation, or mode of dispersion, the weight of asbestos, calculated from the magnesium content, gave decreasing fiber weight in this order: textile, friction, pipe (Table 5). When the count/weight ratios were used to convert the tentative TLV of 12 fibers (> 5  $\mu$  length) per milliliter to the equivalent gross weight concentrations of asbestos (based on magnesium determinations), the gross weight concentrations were 2 mg/m<sup>3</sup> for textile, 1 mg/m<sup>3</sup> for friction, and 0.5 mg/m<sup>3</sup> for pipe. A single value for the weight TLV for all product areas would be equivalent to a fourfold difference in fiber threshold between textile and pipe plants.

## Discussion

Biologically active fibers have long been an important public health problem. As early as 1900, asbestos fibers were identified as the causative agent in a chronic pneumoconiosis (asbestosis) occurring among asbestos textile workers. Later investigators noted an excess of cancer occurring among workers exposed to asbestos fibers. A relationship between the known excess of cancer in urban environments and asbestos has been suggested by the presence of "asbestos" or "ferruginous" bodies in the lungs of a high proportion of urban dwellers. (Editor's Note: In this regard, see review by Caensler, E.A. and Addington, W.W., "Asbestos or Ferruginous Bodies," New Engl. J. Med. 280: 488-492 (February 27, 1969).)

Fibrous glass has been given considerable attention as a possible hazard of the general type of asbestos, although current manufacturing practices produce few fibers capable of penetrating into the pulmonary air spaces. A trend toward the production of smaller glass fibers and the emergence of small, respirable, graphite, silicon carbide, and other man made fibers suggests that the problem of the health effects of fibers will increase. These effects are of concern, not only to the industrial population, but also to urban dwellers who may breathe fibers from many sources; occupants of buildings with fibrous glass-lined ducts and others who may unknowingly become exposed. (Editor's Note: A review of bioeffects information on both of these types of fibrous dusts appears in the Proceedings of the Fibrous Dust Seminar, November 22, 1968, Medical Series Bulletin No. 16-70, Industrial Hygiene Foundation, Pittsburgh, Pa.)

To protect the health of the population and avoid penalizing a valuable economic material which is causing no harm, it is important to learn as much as possible about fibers in materials, in the air, and in the body. Fibers occurring in the environment must be detected and identified so they can be traced to their source. Quantitative estimates of fiber concentration in industrial and community air are needed to establish any relationship between disease patterns and to determine the effectiveness of control measures. The trace chemical constituents of these fibers are of importance in uncovering the mechanisms of the diseases resulting from exposure. (10)

## International Comparability

At the present time the goal of international comparability has been partly achieved by the adoption of the concentration in numbers of fibers longer than 5 microns counted on membrane filter samples at 430X magnification with phase-contrast (PC) illumination as the standard method in both Britain and the United States. (6, 11)

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