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ASBESTOS IN POTABLE WATER

by

William H. Hallenbeck
Carolyn S. Hesse
Edwin H. Chen
Kusum Patel-Mandlik
Arthur H. Wolff

School of Public Health
University of Illinois at the Medical Center
Box 6998, Chicago, Illinois 60680

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UNIVERSITY OF ILLINOIS
WATER RESOURCES CENTER
2535 Hydrosystems Laboratory
Urbana, Illinois 61801

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ABSTRACT

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Several published reports indicate that asbestos is found in public drinking water supplies. In order to determine the health effects of ingested asbestos, relevant animal and human studies were reviewed. From these studies, it was not possible to predict potential health hazards associated with ingestion of the levels of asbestos found in drinking water. Detailed examinations of water samples were carried out by electron microscopy in order to determine the precision of analysis for waterborne asbestos. Asbestos-cement pipes, used to transmit drinking water, were studied under field conditions. No significant release of asbestos was observed. Preliminary work suggests that asbestos is present in Lake Michigan. An investigation of possible sources of asbestos into Lake Michigan indicated that wet deposition of asbestos may be a principal pathway of asbestos contamination of Lake Michigan. A feasibility study was carried out to determine if asbestos is present in rainwater collected in the Chicago area. Chrysotile asbestos was found in Chicago rainwater at a level of 10^5 to 10^6 fibers per liter. This finding demonstrates that precipitation scavenges airborne chrysotile asbestos which may result in the contamination of surface waters.

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Preface

Asbestos is a generic term for a number of fibrous silicate minerals such as chrysotile, amosite, crocidolite, anthophyllite, tremolite, actinolite, and grunerite. Because asbestos does not burn or conduct electricity and can be woven into cloth, it has come to have over 3000 uses and consequent widespread distribution in the environment.

Asbestos has been well-documented as a carcinogen of the respiratory system in studies of asbestos workers. There is evidence which suggests that a large proportion of inhaled asbestos is subsequently swallowed. This transfer of asbestos is attributed to the respiratory clearance mechanism. Indeed, epidemiologic studies of asbestos workers have shown elevated rates of digestive system cancer as well as respiratory system cancer.

Recent reports indicate that the general population is ingesting asbestos in drinking water. Several studies at the University of Illinois Medical Center have been undertaken to define sources and the extent of asbestos contamination of Illinois water supplies. The analysis of water samples for asbestos was found to be a relatively new area of research. Detailed examinations of water samples were carried out by electron microscopy in order to determine the precision of analysis for asbestos.

One possible source of asbestos into drinking water may be asbestos-cement pipes which are used to transmit drinking water. Asbestos-cement pipes of various ages, lengths, and diameters were studied for possible release of chrysotile asbestos under field conditions. These pipes are presently in use in northeastern Illinois.

Preliminary work suggests that asbestos is present in Lake Michigan. Literature was reviewed and pertinent agencies were consulted in order to

determine possible sources of asbestos into Lake Michigan. The following topics were investigated: industrial effluents and emissions, natural deposits, intra- and inter-lake currents, and dredging operations. The results of this investigation indicated that wet deposition of asbestos into Lake Michigan may be a principal pathway of asbestos contamination of Lake Michigan. Hence, a feasibility study was carried out to determine if asbestos is present in rainwater collected in the Chicago area.

Overall, our studies indicate the need for considerable additional research on the sources and health effects of ingested asbestos.

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TABLE OF CONTENTS

Section I

A Review of the Health Effects of Ingested Asbestos*

W. H. Hallenbeck and C. S. Hesse

Abstract

Occupational studies indicate that a human health hazard may exist for ingested asbestos since the death rates due to digestive system cancers are elevated in asbestos workers. This finding may be related to the swallowing of asbestos that was inhaled and cleared from the respiratory system via the respiratory clearance mechanism. Published animal ingestion experiments have serious shortcomings in their design and execution which make their interpretation very difficult. Animal ingestion and human autopsy studies suggest that asbestos fibers may penetrate the digestive tract and migrate to other locations in the body.

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Introduction

Asbestos is a generic term for a number of fibrous silicate minerals such as chrysotile, amosite, grunerite, tremolite, crocidolite, anthophyllite, and actinolite.[1] There are over 3,000 industrial uses of asbestos,[2] and many reports indicate that asbestos has become an ubiquitous environmental contaminant. It has been found in air, drugs, beverages, food, and water supplies.[2-14,52-62] Since asbestos is being ingested by the general population and has been strongly implicated as an industrial carcinogen, a question has arisen as to the health effects of ingested asbestos. In order to answer this question we have examined relevant animal and human studies.

Occupational Studies

Evans, et al.[15] have demonstrated in rats killed immediately after exposure that up to 48% of an inhaled asbestos dose is swallowed. This transfer of material is attributed to the respiratory clearance mechanism. If the human respiratory clearance mechanism for asbestos is similar in efficiency to that of the rat, workers exposed to airborne asbestos also swallow a substantial dose.[16] Epidemiology studies have shown an increase in the rates of digestive system cancer as well as respiratory system cancer in asbestos workers.[17-23] We are concerned with those cancers which may be related to asbestos ingestion, i.e. digestive system cancers.

Selikoff and his co-workers[17,18] have done several epidemiology studies on the health effects of asbestos in workers. Overall, Selikoff[17] has found the incidence of gastrointestinal (GI) cancer in asbestos workers to be two to three times the level in the general population. This effect is more evident after 20 years or more from first exposure. In the following three occupational studies, it was found that cancer of the esophagus, stomach, colon or rectum caused from 5 to 10% of the total deaths.

One group studied by Selikoff, _et al._[17] was composed of 623 persons who were members of the New York - New Jersey branches of the Insulation Workers Union as of January 1, 1943. The study was based on their mortality experience through 1971. Cancer of the digestive system caused 41 deaths where 13 were expected. In a second investigation by Selikoff _et al._,[17] 877 workers, who were employed in an amosite asbestos plant between 1941 and 1945, were traced through 1971. Twenty-six deaths were due to cancer of the stomach, colon, and rectum, where 13 were expected. In a third study Selikoff, et al.[17] examined the causes of death among 17,800 asbestos insulation workers in the U.S. and Canada between January 1, 1967, and December 31, 1971. Fifty-five deaths were attributed to gastrointestinal cancer where 27 were expected. Selikoff[18] also compared cancer rates in asbestos workers who smoked cigarettes to those who did not. He found that cigarette smoking and inhalation of asbestos were synergistic for bronchogenic carcinoma but not for cancer of the colon or rectum.

In England, Newhouse[20] studied the mortality of workers engaged in the fabrication of asbestos products from crocidolite, amosite, and chrysotile. The male cohort consisted of 2,640 men who were employed since 1931 and followed until 1970; the female cohort of 922 started employment between 1936 and 1942 and were traced through 1968. In general, a significant excess of cancer deaths did not start appearing until 25 years after initial employment. The men experienced a significant excess of deaths from gastrointestinal cancer: 31 observed versus 20 expected (P*less than 0.05).[20] However, there was a less significant excess of deaths due to gastrointestinal cancer among the women: 11 observed deaths with 7.3 expected (P = 0.12).[20,21] The mortality rates of the general population of England and Wales were used for comparison.

*Probability

Enterline[22] compared the death rates of 2,833 white males who had been workers in the asbestos products industry to the rates of 6,424 who had worked in cotton textile plants for any period of time between 1948 and 1951. The number of deaths in each group was observed through June 30, 1963. The asbestos workers experienced 21 deaths due to digestive system cancer where 13.9 were expected. The cotton workers experienced 24 deaths due to digestive system cancer where 29.8 were expected. The general population was used as the reference.

The dose-response relationship between occupational exposure to asbestos and risk of digestive system cancer is very poorly defined.[19,23] Any attempt at this time to construct a quantitative dose-response relationship entails making tenuous assumptions concerning such variables as:

1. The concentration of asbestos to which a worker was exposed over his working lifetime
2. The fraction of inhaled asbestos which was subsequently swallowed
3. Co-variables such as smoking, diet
4. The distribution of fiber lengths swallowed

The last point is especially important since occupational exposures contain a broad range of fiber lengths, whereas environmental exposures (water, air) usually consist of only the shorter fibers. Thus it becomes very important to determine if fiber length is a factor in the causation of digestive system cancer.

Human Autopsy Studies

Evidence indicating that asbestos fibers can penetrate and migrate in humans is suggestive but inconclusive. Borow, et al.[44] performed autopsies on asbestos workers and found unspecified fibers in the parietal pleura, gastric and intestinal mucosa, spleen and liver. Pooley[45] has found unspecified types of fibers in the GI wall of asbestos workers. In one asbestos worker, Leicher[46] showed by using X-ray diffraction that asbestos was present in a peritoneal tumor. Godwin and Jagatic[47] found asbestos particles in the peritoneal mesotheliomas of two former asbestos workers. One of these persons was exposed for only six weeks. They also found asbestos in the intestinal mucosa and spleen of a third person with peritoneal mesothelioma. Hourihane[48] has reported finding asbestos fibers, which were mostly amphiboles, in tumor tissue by using phase contrast microscopy. Out of 36 cases of mesothelioma, most of which had asbestosis, he found fibers in 7 pleural (from 7 cases) and 7 peritoneal (from 7 cases) tumors. He also found fibers in the tumors of 4 of 19 cases with no known exposure to asbestos.

No conclusions can be drawn from these human autopsy studies because little or no mention was made concerning control techniques for contamination of the possibility that asbestos fibers can be found throughout normal tissue as well as diseased tissue.

Animal Studies

Published animal feeding studies have not definitively demonstrated that asbestos ingestion is either harmful or safe. Tumors have been produced in animals following the injection of asbestos fibers into the pleural and peritoneal cavities[25-31] but have not been demonstrated where ingestion was the only route of administration.[16,32-42] However, there is some evidence that ingested asbestos fibers may penetrate digestive system tissue and migrate to other locations in the body.[16,32,33,35.]

In view of the long latent period of asbestos carcinogenesis, the length of time allowed for study is important. Most of the published feeding experiments have been conducted for relatively short periods of time.[16,32-36,40]

Some feeding studies have not specified the size of test or control groups used, and some apparently used no controls. Where controls were used, none of the feeding experiments to data has used large enough numbers of test and control animals to observe a statistically significant incidence of cancer of the digestive system.[16,32-40] In asbestos workers the death rate due to cancer of the digestive system ranges up to 10%.[17] If this 10% rate were extrapolated to animal studies, a minimum of 50 test and 50 control animals would be needed to observe a statistically significant difference between groups at $P = 0.05$.[43]

The length distribution of asbestos fibers used in a feeding experiment should be characterized since it appears that the fibrous property of asbestos, rather than the chemical composition or presence of trace metals, determines its carcinogenicity.[26,27,24,49-51] Most of the published feeding experiments have not specified the distribution of fiber lengths.[16,32-37,40]

Some papers do not indicate which tissues were examined for asbestos or what type of microscopy was used. In a given weight of asbestos, a large proportion of the fibers can only be observed with an electron microscope. Those studies purporting to demonstrate the absence of fibers in tissues based on light microscopic examination are of doubtful significance.[32,34,35,37-39]

Sometimes adequate measures to prevent contamination of tissues have not been taken. Fiber contamination can come from the environment during tissue preparation or from the gut lumen. Some of the contamination problems encountered include unfiltered tap water used to rinse tissues, tissues weighed on an asbestos contaminated balance, and cross-contamination of tissues.[32,34,35] Proper use of controls would detect problems related to contamination.

Samples for examination are prepared from either tissue digests or tissue sections. In comparison to tissue section preparations, tissue digest preparations increase the probability of finding low levels of fibers because a greater mass of tissue is used. Therefore, ingestion studies which demonstrate the absence of fiber penetration based only on tissue section preparations are of doubtful significance.[32,34-37]

Summary

Asbestos is an ubiquitous low level environmental contaminant.[2-14,52-62] Hence, a question has arisen as to the health effects of ingested asbestos.

Studies of asbestos workers indicate that exposure to high levels of airborne asbestos increases the risk of cancer of the digestive system.[17-23] This finding may be related to the swallowing of asbestos that was inhaled and cleared from the respiratory system via the respiratory clearance mechanism.[15] The dose-response relationship between occupational exposure to asbestos and risk of digestive system cancer is very poorly defined.[19,23] Therefore it is difficult to evaluate the potential health hazard of low levels of ingested asbestos. Studies involving autopsies of asbestos workers are generally inconclusive but do suggest that asbestos may penetrate digestive system tissue.

Studies involving the feeding of asbestos to animals[16,32-42] are also generally inconclusive mainly because insufficient numbers of test and control animals were used. In addition, the duration of most of the studies was shorter than the animals' usual life span. However, there is some evidence that asbestos may penetrate digestive system tissue and migrate to other locations in the body.[16,32,33,35]

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Section II

Precision of Analysis for Waterborne Chrysotile Asbestos by Transmission Electron Microscopy*

W. H. Hallenbeck, E. H. Chen,
K. Patel-Mandlik, A. H. Wolff

Abstract

Detailed examinations of water samples were carried out to determine the precision of analysis for chrysotile asbestos by transmission electron microscopy (TEM). Since the frequency distribution of counts fit a Poisson distribution, several statistical inferences were made, including (1) an estimate of precision and (2) a model for determining the probability of observing chrysotile as a function of its concentration in water and magnitude of area scanned by TEM.

*This section was accepted for publication in the Bulletin of Environmental Contamination and Toxicology.

Introduction

Asbestos is a generic term for a number of fibrous silicate minerals such as chrysotile, amosite, grunerite, tremolite, crocidolite, anthophyllite, and actinolite. In 1970 chrysotile accounted for about 95% of the 750,000 short tons of asbestos used commercially in the U.S.[1,2] There are many reports which indicate that asbestos is an ubiquitous environmental contaminant. It has been found in ambient air,[3-5] indoor air,[5] parenteral drugs,[6] beverages,[7-9] food,[10] cosmetics,[11] the vicinity of asbestos manufacturing operations,[12-14] and water supplies.[7,15-19] Our knowledge of the health effects of asbestiform minerals is derived mainly from studies of occupational exposure. There is reason to expect that a large fraction of inhaled asbestos is swallowed and thereby constitutes an ingestion exposure.[23] This ingestion exposure may be causally related to the excess of digestive system cancers in several occupational studies.[20-22]

Our interest in the health significance of environmental asbestos has led to our examination of electron microscopic (EM) methods for the detection of waterborne chrysotile. The concentration of asbestos in water is generally reported in terms of fibers or fibrils per liter. A fiber is a reducible unit of asbestos which is capable of further longitudinal splitting into fibrils. Henceforth, the term "unit" will be used to refer to either a fibril or fiber.

Published reports of the asbestos content of various media generally do not contain an estimate of precision.[3-19] We suggest that it is very important to define the precision of EM methodology since only a very small percentage of a collected sample is actually examined. For example, only about 0.01% of a water sample is actually examined by EM. This paper describes the method used and results obtained in our effort to determine the precision of the analysis for waterborne chrysotile by transmission electron microscopy (TEM).

Experimental Procedure

The methods and statistics developed here apply to quantitation of chrysotile asbestos in drinking water. Each of two drinking water samples of groundwater origin was analyzed for its content of chrysotile asbestos using the following procedure.

1. A sample was mixed by multiple inversions.
2. 200 ml of water were filtered through a Millipore membrane filter of 0.45μm pore size and 47mm diameter within 24 hours of collection.
3. The filter was placed in a petri dish and allowed to dry in a vacuum dessicator.
4. A randomly selected 3.05mm diameter punch from each filter was placed on a 200 mesh copper TEM grid of equal diameter. The grid substrate was carbon coated 0.25% Formvar.
5. Acetone was filtered through a 0.1μm Nucleopore membrane filter and used to charge the boiler of a condensation washer. The grid-punch combination was placed in the condensation washer for two hours with an acetone reflux rate of approximately one drop/second.

6. Ten grid squares were randomly selected and examined by TEM at 26,280x and 100kv. Chrysotile asbestos was confirmed by both morphology and selected area electron diffraction (SAED) pattern. In general, there are three types of information needed for the complete identification of asbestos: morphological information obtained by high magnification TEM, crystallographic information obtained by SAED patterns, and microchemical information obtained by energy dispersive spectrometry. There is good agreement among investigators that the morphology and SAED pattern of chrysotile are sufficiently distinct to permit visual identification by TEM.[7,14,17,24-26]
7. Steps 4-6 were repeated for nine more punches from the same membrane filter. Thus 10 punches were randomly selected from each membrane filter.

Results

For each of the two drinking water samples 10 grid squares from each of 10 grids were scanned for their content of chrysotile units. Hence the basic unit of observation is a single grid square. Table 1 presents the frequency distributions of counts. Selected descriptive statistics and estimation values are presented in Table 2.

Discussion

The variance test was performed to test the Poisson goodness-of-fit for the distribution of chrysotile units per grid square.[27] The results given in item 4 of Table 2 show a good fit for both water samples. Thus, we may state that the number of chrysotile units per grid square follows a Poisson distribution.

The number of chrysotile units is usually presented in terms of number per liter of water. The conversion from average units per grid square to units per liter is as follows.

Area of one grid square = 8.236×10^{-5} cm^2
Effective filter area = 13.847 cm^2
Volume of water filtered = 200 ml
Units/liter = [(Average units/grid square)/(area of one grid square)] x (effective filter area) x (dilution factor) = $(8.41 \times 10^5)\bar{X}$

Since we recognize that the number of chrysotile units per grid square follows a Poisson distribution, confidence intervals can readily be obtained.[28] The 95% confidence intervals are given in item 5 of Table 2. It is interesting to note that confidence intervals can be set up even if $\bar{X} = 0$. For example, the 95% confidence limits for counting 100 grid squares with $\bar{X} = 0$ are 0 and 3.10×10^4 units per liter.

When the distribution of the number of chrysotile units per grid square is understood, statistical tests can be carried out to test the hypothesis that two water samples have the same number of chrysotile units. After the statistical test was applied to the two water samples, there was not enough evidence to say that the two water samples contained different levels of chrysotile units per liter $(p>0.10)$.[28]

Usually 10 to 20 grid squares are scanned in routine analyses. If the number of chrysotile units per grid square follows a Poisson distribution with the expected value λ, we will be able to predict how many zero counts will be encountered. Given in Table 3 are the Poisson probabilities of the number of chrysotile units per grid square for four values of λ. The best estimate of λ is the sample mean $\bar{X}$. If we know the approximate value of λ for a given water sample, then the Poisson probability of finding a zero count in a single grid square, P_o, can be obtained as in Table 3. The probability that N grid squares will yield a zero count is simply P_o^N. Table 4 presents P_o^N for several values of λ and N. It is obvious that counting only 10 grid squares for $\lambda \leq 0.08$ would result in a zero count more than 45% of the time; the conventional counting of 20 grid squares would result in a zero count more than 20% of the time. For $0.06 \leq \lambda \leq 0.07$, 50 grid squares should be counted to reduce the probability of a zero count to less than 5%. In order to exclude the possibility that counts were made on background contamination, six blank grids were prepared by washing pure Millipore membrane filter punches onto grids. A total of 45 grid squares was scanned and no chrysotile units were confirmed. If $0.06 \leq \lambda \leq 0.07$, there is only a 4-7% probability of observing no units in 45 grid squares. Thus it is unlikely that background contamination can account for the chrysotile units confirmed in the two groundwater samples.

Increasing the number of grid squares to be scanned will inevitably increase cost and time. One can solve this problem by filtering large volumes of water. Another possible solution to the problem is based on the property that the sum of k Poisson variates with $\lambda_1, \lambda_2, \ldots, \lambda_k$ is a Poisson variate with an expected value of $\lambda_1 + \lambda_2 + \ldots + \lambda_k$.[29] For a given water sample, all λ's are of identical value. As an illustration of this point, the data obtained by scanning 100 grid squares were converted to units per grid data (see Table 5). The Poisson distribution of chrysotile units per grid square can be generalized to the Poisson distribution of chrysotile units per grid.[30] It also follows that it is theoretically possible to stack several filter punches vertically on a single grid and still maintain a Poisson distribution of grid square and grid counts. This procedure may obviate the need to filter large volumes of water.

Conclusions

It is important to report the precision of an observed count of asbestos units. Equally important is an estimate of the probability of observing asbestos units as a function of concentration and area scanned. A model for specifying precision and probability has been described for chrysotile units in the 10^4 - 10^5 units per liter range. Further research is needed: (1) to determine whether the Poisson distribution also holds for higher concentrations of chrysotile, and (2) to verify that the distributions of grid square and grid counts follow a Poisson distribution for stacked punches.

TABLE 1

Frequency Distributions of Chrysotile Units by Grid Square

Count of Chrysotile Units/grid square	Number of Grid Squares Sample A	 Sample B
0	95	93
1	4	7
2	1	0
TOTAL	100	100

TABLE 2

Selected Descriptive Statistics and Estimation Values

	Sample A	Sample B
(1) Number of grid squares scanned, N	100	100
(2) Standard deviation, s (units/grid square)	0.28	0.26
(3) Variance, s^2	0.08	0.07
(4) Variance test $x^2 = (N-1)\ s^2 / \bar{x}$	127	93
p-value	0.06	0.70
(5) Point estimates and confidence interval		
(a) Mean, $\bar{x}$ (units/grid square)	0.06	0.07
(b) $8.41 \times 10^5\ \bar{x}$ (units/liter)	5.05×10^4	5.89×10^4
95% confidence interval		
Lower limit (units/liter)	1.85×10^4	2.36×10^4
Upper limit (units/liter)	1.10×10^5	1.21×10^5

TABLE 3

Poisson Probabilities as a Function of λ and Count/Grid Square

Count of Chrysotile Units/Grid Square	λ			
	0.05	0.06	0.07	0.08
0	0.951	0.942	0.932	0.923
1	0.048	0.057	0.065	0.074
≥2	0.001	0.001	0.003	0.003

TABLE 4

Probability of a Zero Count as a Function cf λand Number of Grid Squares Scanned

N	λ			
	0.05	0.06	0.07	0.08
10	0.61	0.55	0.49	0.45
20	0.37	0.30	0.24	0.20
30	0.22	0.17	0.12	0.09
40	0.13	0.09	0.06	0.04
45	0.10	0.07	0.04	0.03
50	0.08	0.05	0.03	0.02

TABLE 5

Frequency Distributions and Means
of Chrysotile Units by Grid

Count of Chrysotile Units/Grid		Number of Grids	
		Sample A	Sample B
0		6	4
1		3	5
2		0	1
3		1	0
	TOTAL	10	10
Mean (units/grid)		0.6	0.7

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Section III

Is Chrysotile Asbestos Released From Asbestos-Cement Pipe Into Drinking Water?*

William H. Hallenbeck
Edwin H. Chen, Ph.D.
Carolyn S. Hesse
Kusum Patel-Mandlik, Ph.D.
Arthur H. Wolff, D.V.M.

Abstract

Fifteen asbestos-cement pipes of various ages, lengths, and diameters were studied for possible release of chrysotile asbestos under field conditions. The pipes were located in northeast Illinois. Paired water samples were taken from water before and after it flowed through the pipes. The water samples were analyzed for chrysotile asbestos by transmission electron microscopy. Chrysotile was identified on the basis of morphology and electron diffraction pattern. Under the conditions and limitations of this study, no significant release of chrysotile from asbestos-cement pipe was observed.

*This section has been forwarded for publication in the _Journal of the American Water Works Association._

Introduction

More than 200,000 miles of asbestos-cement water distribution pipe have been placed into service in the United States since its introduction in 1930. Over 30% of all the water distribution pipe currently being sold in the U.S. is made of asbestos-cement. Its principal attributes are corrosion resistance, light weight, and low cost.[1]

Asbestos is a generic term for a number of fibrous silicate minerals. "Chrysotile" refers to the serpentine ($Mg_6Si_4O_{10}(OH)_8$) variety of asbestos.[2] Approximately 80% of the asbestos used to fabricate asbestos-cement pipe is chrysotile asbestos.[3]

A question has arisen concerning the possible release of asbestos fibers from asbestos-cement pipe into drinking water. Occupational epidemiology studies indicate that a human health hazard may exist for ingested asbestos since death rates due to cancer of the digestive system are elevated in asbestos workers.[4] This finding may be related to the swallowing of asbestos that was inhaled and cleared from the respiratory system via respiratory clearance mechanisms.[5]

The Johns-Manville Corporation has studied two asbestos-cement pipes in two different municipal water systems in order to determine if chrysotile was released. Two pipes of unspecified ages were studied over a period of about one year. Paired water samples were taken two to three times a month from water before and after it flowed through the pipes. It was concluded that both pipes released chrysotile on the order of nanograms per liter at the 10% level of significance.[3]

Since the effect of pipe age on the release of chrysotile was unknown, our objective was to study a broad range of pipe ages for possible release of chrysotile asbestos. A primary determinant of our experimental design was related to the time needed to analyze a water sample for chrysotile by transmission electron microscopy (TEM). Approximately six man-hours are required to analyze a water sample for chrysotile by the method described below. Hence the study was designed to obtain a manageable number of water samples associated with a variety of pipe ages.

Method

Sample collection. Asbestos-cement pipes of various ages were identified in fifteen public water systems in northeast Illinois. Two water samples (200 ml each) were taken for each asbestos-cement pipe studied; one was taken before and the other was taken after each length of pipe. These paired samples were taken within fifteen minutes of each other at existing water taps, e.g., homes, public buildings, gas stations, fire hydrants. Samples were collected from February through May of 1975. The preparation of glass sample bottles consisted of hot detergent washing, rinsing with distilled water (pre-filtered through a 0.1μm Millipore membrane filter), and rinsing with specimen water prior to collection. The following water system parameters were obtained: pipe age, length, and diameter; total dissolved solids; total hardness; alkalinity; calcium; and pH.

Sample preparation. The paired water samples were prepared and analyzed under nearly identical conditions in order that the before sample could serve as a reference for the after sample. Throughout sample preparation and TEM analysis, the analyst was not aware of the before/after identity of a water sample. Before/after samples were prepared and analyzed in random order. Water samples were mixed by multiple inversions of the glass sampling bottles and filtered through a 0.45μm Millipore membrane filter (47mm diameter) within 24 hours of collection. The filter was placed in a plastic petri dish with the lid slightly ajar and dried in a vacuum desiccator. A randomly selected punch (3mm diameter) from each filter was placed, sample side down, on a 200 mesh TEM grid (3.05mm diameter). The grid substrate was either carbon or 0.25% Formvar coated with carbon. The grid and punch were washed in an acetone condensation washer for two hours at a reflux rate of about one drop per second. The acetone was filtered through a 0.1μm Nucleopore or 0.2μm Fluoropore membrane filter prior to use.

Sample analysis. In the early stages of our study, the viewing screen of our TEM was not designed for obtaining length and diameter measurements. Therefore many of the earlier samples were scanned for fiber count data only. Ten grid squares were randomly selected and scanned at 100kv and from 22,000 to 26,000 magnification. After the viewing screen had been modified, an additional punch was randomly selected from each of the previously prepared filter papers. Ten grid squares were randomly selected and scanned to obtain fiber count, length, and diameter data.

Since several minerals have morphological characteristics similar to chrysotile,[6] fibers were identified as chrysotile by both morphology and selected area electron diffraction (SAED) pattern. There is good agreement among investigators that the morphology and SAED pattern of chrysotile are sufficiently distinct to permit visual identification by TEM.[7-11]

Length and diameter data were used to compute the mass of a fiber. "Fiber" designates either a fibril or bundle of fibrils. A fibril was assumed to be cylindrical[12,13] and to have a density of 2.5 picograms (pg)/μm^3.[2] Hence, pg/fibril = $2.5 \times \pi \times L \times R^2$, where L = length in µm and R = radius in µm. The mass of a bundle of fibrils was calculated by summing the masses of its individual fibrils.

Detection limits for fiber counts.

Groundwater: One fiber per ten grid squares corresponds to 8.02×10^4 fibers per liter. One fiber per 20 grid squares corresponds to 4.10×10^4 fibers per liter.

Lake Michigan water: One fiber per ten grid squares corresponds to 5.84×10^4 fibers per liter. One fiber per 20 grid squares corresponds to 2.92×10^4 fibers per liter. (The difference between groundwater and Lake Michigan detection limits arises from the slightly different effective diameters of the filtration funnels used).

Results and Discussion

Table 1 contains the age, length, and diameter of each asbestos-cement pipe and the saturation indices of the various water systems. Saturation indices (I) were calculated according to the method of Nordell.[14] When $I = 0$, the water is in equilibrium with respect to $CaCO_3$; when I is positive, the water is oversaturated and will deposit $CaCO_3$; when I is negative, the water is undersaturated and will dissolve $CaCO_3$. Most of the water systems had $I \geq 0$, which implies that pipes in these systems may have had a layer of $CaCO_3$ on their interior walls. A few systems had $I < 0$, which implies that the water may have been in direct contact with asbestos-cement.

Tables 2 and 3 contain, for each of the water samples, the number of grid squares scanned by TEM, the average number of chrysotile fibers per grid square, the average mass per grid square, and standard deviations. The average number of fibers per grid square was significantly correlated with the average mass per grid square. Rank correlations[15] for groundwater and Lake Michigan systems were 0.651 ($P < 0.05$) and 0.393 ($P < 0.05$), respectively. This means that, in general, mass increased as fiber count increased.

The data in Tables 2 and 3 can be analyzed in an overall sense by applying the sign test[15] to all fifteen pairs of before/after samples. On the basis of average fiber counts, eight "after" samples showed an increase in chrysotile, six showed a decrease, and one pair was tied. The observation of eight increases was not significant (P = 0.395). On the basis of average mass data, there were seven increases and eight decreases. The occurrence of seven increases was also not significant (P = 0.500).

Since the individual characteristics of the fifteen asbestos-cement pipes were different, we also carried out statistical tests on each before/after sample pair. The individual pairs of samples were analyzed first in terms of fiber count data. In order to apply a parametric test, the distribution of fiber counts per grid square must be known. A previous study has shown that when the concentration of chrysotile is low, fiber counts per grid square follow a Poisson distribution for a given water supply.[16] Corroborating this earlier finding, it can be seen in Table 2 that as the average number of fibers per grid square increased the variance increased (rank correlation = 0.930, P < 0.01). This was a strong indication that, in general, fiber counts per grid square followed the Poisson distribution. Therefore, to test the significance of increase of fiber count from before to after for each pair of samples, a parametric test was carried out which was based on the comparison of two Poisson distributions.[17] The results expressed in terms of P-Values are given in Table 4. Only the Bradley Road system had a significant increase in fiber count (P = 0.01).

Individual members of before/after pairs were tested for goodness of fit of the Poisson distribution.[18] In several cases, the before or after or both member(s) of a pair may not follow the Poisson distribution at the 5% significance level (see footnote of Table 4), and therefore the nonparametric Mann-Whitney test[15] was used to test the significance of increase of fiber count from before to after for each pair of samples. These results are given in Table 4 and are in agreement with those of the Poisson test.

The distribution of mass data was not studied and therefore the nonparametric Mann-Whitney test was used to test the significance of increase of mass from before to after for each pair of samples. The results are given in Table 4 and reveal no significant increases.

It may be hypothesized that fibers which are already present in the water may break down in transit and thus create the appearance of chrysotile release, in terms of fiber count, from asbestos-cement pipe. Some evidence was found to support this hypothesis from the following analysis of Lake Michigan systems. All the fiber lengths from the before samples were compared to all the fiber lengths in the after samples by using the Mann-Whitney test. The lengths of the after fibers were found to be somewhat shorter (P = 0.0630) than those of the before samples. The same analysis of groundwater systems did not reveal a significant difference between before and after fiber lengths (P = 0.382). Therefore an increase was considered significant only when both fiber count and mass data demonstrated a significant increase. Referring to Table 4, no sample pairs satisfied these two criteria at the 5% level of significance.

It is possible that chrysotile may be released from pipes at levels below our detection limit or at levels which are obscured by laboratory contamination or chrysotile already present in the water. Laboratory contamination may result from the presence of chrysotile in the air, membrane filters, or grid coatings. Since an insufficient number of control samples was used to determine the level of laboratory contamination, no inferences regarding the absolute level of chrysotile in drinking water can be made from the data in Tables 2 and 3.

In conclusion, fifteen asbestos-cement pipes of various ages, lengths, and diameters were studied for possible release of chrysotile asbestos. Under the conditions and limitations of this study, no significant release of chrysotile from asbestos-cement pipe was observed.

Table 1

Characteristics of Asbestos-Cement Field Pipe and Saturation Indices

Water Systems	Pipe Age (years)	Length (feet)	Diameter (inches)	Saturation Index
Groundwater Systems				
Westmont	0.5	200	12	-0.9
Lisle	1	3,000	8	0.4
Hoffman Estates	18	3,700	8	0.5
Rolling Meadows	20	4,400	12	0.1
York Center	27	1,100	6	0.6
Lake Michigan Systems				
Bannockburn	1	1,300	10	0.2
Bradley Road	14	1,000	6	0.2
Zion-Benton	18	10,000	8	0
Waukegan	19	600	6	---
Zion	26	1,150	6	0
Midlothian	27	1,500	6	0
Blue Island	30	3,248	8	0
Brookfield	35	40,000	20	0
Glenview	37	1,500	6	-0.7
Highland Park	40	275	6	-0.6

Table 2

Number of Grid Squares Scanned, Average Number of Fibers Per Grid Square and Standard Deviation (in Parenthesis)

	BEFORE PIPE		AFTER PIPE	
Water Systems	Number of Grid Squares Scanned	Average Number of Fibers* Per Grid Square	Number of Grid Squares Scanned	Average Number of Fibers* Per Grid Square
Groundwater Systems				
Westmont	20	0.15 (0.49)	20	0.10 (0.31)
Lisle	10	0.10 (0.32)	10	0.60 (1.90)
Hoffman Estates	20	0.25 (0.72)	20	0 ----
Rolling Meadows	20	0.40 (0.60)	20	0 ----
York Center	20	0.05 (0.22)	20	0.25 (0.72)
Lake Michigan Systems				
Bannockburn	20	0.10 (0.31)	20	0.30 (0.57)
Bradley Road	20	0.10 (0.31)	20	0.65 (0.88)
Zion-Benton	10	0.10 (0.32)	10	0.30 (0.67)
Waukegan	20	0.20 (0.70)	20	0.10 (0.31)
Zion	10	0.10 (0.32)	10	0.10 (0.21)
Midlothian	20	0.40 (1.35)	20	0.45 (0.94)
Blue Island	20	1.15 (1.57)	20	0.95 (1.23)
Brookfield	10	0.70 (0.82)	10	0.30 (0.48)
Glenview	20	0.05 (0.22)	20	0.25 (0.64)
Highland Park	20	0.15 (0.37)	20	0.20 (0.52)

* Fiber designates either a fibril or bundle of fibrils.

Table 3

Number of Grid Squares Scanned, Average Mass (10^{-15} grams) Per Grid Square and Standard Deviation (in Parenthesis)

Water Systems	BEFORE PIPE Number of Grid Squares Scanned	BEFORE PIPE Average Mass per Grid Square	AFTER PIPE Number of Grid Squares Scanned	AFTER PIPE Average Mass per Grid Square
Groundwater Systems				
Westmont	20	0.15 (0.51)	20	0.92 (3.80)
Lisle	10	0.13 (0.41)	10	0.44 (1.40)
Hoffman Estates	20	0.28 (2.80)	20	0 ---
Rolling Meadows	20	1.06 (2.54)	20	0 ---
York Center	20	0.03 (0.12)	20	0.27 (0.79)
Lake Michigan Systems				
Bannockburn	10	0.53 (1.68)	10	3.78(11.22)
Bradley Road	10	5.97(15.21)	10	0.41 (1.16)
Zion-Benton	10	0.41 (1.28)	10	1.25 (3.57)
Waukegan	20	17.21(62.15)	20	4.71(20.84)
Zion	10	0.28 (0.89)	10	0.34 (1.06)
Midlothian	10	1.27 (4.02)	10	1.05 (2.72)
Blue Island	10	29.90(46.78)	10	6.63(14.69)
Brookfield	10	0.92 (1.21)	10	0.28 (0.47)
Glenview	10	0.39 (1.24)	10	0.82 (2.60)
Highland Park	10	0.24 (0.76)	10	0.10 (0.26)

Table 4

P-Values for the Test of Hypothesis that Chrysotile was Released from Selected Asbestos-Cement Pipes

Water Systems	P-VALUES Fiber Count Data Mann-Whitney Test	 Poisson Test	 Mass Data Mann-Whitney Test
Groundwater Systems			
Westmont	0.51	>0.50	0.34
Lisle	0.47	0.10†	0.47
Hoffman Estates	0.79	>0.50†	0.79
Rolling Meadows	0.97	>0.50	0.97
York Center	0.28	>0.10†	0.27
Lake Michigan Systems			
Bannockburn	0.37	>0.10	0.14
Bradley Road	0.02*	0.01*	0.54
Zion-Benton	0.33	>0.10	0.34
Waukegan	0.51	>0.50†	0.51
Zion	0.51	0.50	0.47
Midlothian	0.22	>0.10†	0.25
Blue Island	0.70	>0.50†	0.92
Brookfield	0.83	>0.50	0.86
Glenview	0.28	>0.10	0.47
Highland Park	0.48	>0.10	0.37

* Significant

† The before or after or both member(s) of a pair may not follow the Poisson distribution.

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Section IV

Potential Sources of Asbestos into Lake Michigan

C. S. Hesse and W. H. Hallenbeck

Abstract

Since preliminary work suggests that asbestos is present in Lake Michigan, numerous agencies were contacted and literature reviewed in order to determine possible sources of asbestos into Lake Michigan. It was learned that one industrial operation formerly discharged process wastewater, which probably contained asbestos, into Lake Michigan. Another industrial operation discharged wastewater which contained asbestos into a Lake Michigan tributary. No industries are known to discharge asbestos at the present time. A natural deposit of chrysotile asbestos exists in Wisconsin, and amphibole asbestos deposits exist in the Upper Peninsula of Michigan. There is no evidence which indicates that these natural sources of asbestos are being eroded. Currents in Lake Michigan and harbor dredging activities with subsequent open water disposal were investigated as methods by which asbestos could be transported and dispersed throughout Lake Michigan. Although it has been hypothesized that water may flow from Lake Superior into Lake Michigan, it seems unlikely that asbestos fibers from Lake Superior could be found in detectable levels in Lake Michigan. A list of industries with NESHAP permits to use asbestos was compiled as it was felt that wet and dry deposition of asbestos fibers could be a major source of asbestos into Lake Michigan.

Introduction

Recent preliminary work has suggested that asbestos fibers may be present in raw and finished water taken from Lake Michigan.[1-6] Possible sources of asbestos into Lake Michigan were investigated by contacting numerous agencies and individuals. The following modes of pollution were investigated: air and water pollution, erosion of natural deposits of asbestos, and mechanisms by which asbestos could be dispersed, i.e., dredging operations and lake currents.

I. Investigation of Industries Which May Discharge Asbestos in Wastewater

By feeding relevant Standard Industrial Code[7] numbers into the U. S. EPA STORET computer system, an attempt was made to compile a list of industries which discharge asbestos-contaminated wastewater into the Lake Michigan drainage basin.[8] STORET contains information regarding industries with National Pollutant Discharge Elimination System (NPDES) permits. When NPDES permits were checked at the U.S. EPA Region V office in Chicago, it was learned that federal regulations do not require specification of asbestos discharges on NPDES permits.[9,10] Therefore, the water pollution control agencies in Illinois, Indiana, Michigan, and Wisconsin were contacted for information regarding industries that might be discharging asbestos into the Lake Michigan basin. The results of this effort follow.

A. Illinois

Presently, the State of Illinois has regulations specifically covering asbestos discharges. Illinois regulations stipulate that water containing asbestos shall not be discharged to sewers or bodies of water in Illinois without first applying the best available treatment.[11] The asbestos-containing sludge material from a manufacturing plant is to be transported to a sanitary landfill in leak-proof containers and buried.[11]

The Illinois Environmental Protection Agency was contacted for information regarding asbestos-containing effluents in Illinois. At the present time, no industries are known to discharge asbestos in industrial wastewaters to surface waters in Illinois.[12] However, before 1968 the Johns-Manville Products Corporation at Waukegan discharged process water to Lake Michigan after passing the water through its settling pond system.[12,13] The discharged process water probably contained asbestos. Currently, most of the process water from this plant is recycled for reuse.[12,13] Nonrecycled process water (containing organic waste) and sewer wastewater are sent to Waukegan's sewage treatment plant.[13]

B. Indiana

Information was requested from the Indiana State Board of Health on industrial effluents that might contain asbestos and the regulations which govern such effluents. No information on industrial sources of asbestos in effluents was received. From the water quality regulations received,[14-23] it appears that the State of Indiana does not have specific regulations governing asbestos in effluents.

C. Michigan

A list of three chloralkali plants which might have been sources of asbestos to surface waters in Michigan was obtained from the Michigan Department of Natural Resources.[24,25] Only one of these plants, Hooker Chemical in Montague, is in the Lake Michigan drainage basin. The asbestos could originate from the backwashing of chlorine cells which contain asbestos.[24] Before installing a system to remove asbestos (around April or May 1976),[24,25] the plant was reportedly discharging measurable amounts of asbestos into surface waters in Michigan.[25] Michigan's present policy is to discourage discharging of asbestos to surface waters.[25]

D. Wisconsin

The Wisconsin Department of Natural Resources was contacted for information regarding point sources of asbestos effluents in Wisconsin. Presently, the State of Wisconsin does not require the reporting of asbestos in industrial effluents.[26] However, the state is considering adding asbestos to the list of substances for which reporting is required.[26] At the present time no industries in Wisconsin that are located in the Lake Michigan drainage basin are known to discharge asbestos-contaminated wastewaters into surface waters.[26]

II. Investigation of Possible Industrial Sources of Asbestos Air Emissions

It is known that particulate matter as well as other pollutants can be transferred from the atmosphere to bodies of water. This can occur by rainout as condensation nuclei, rainfall washout, or dry deposition (settling out) from the atmosphere.[27-32] Therefore, point sources of asbestos air emissions in Wisconsin, Indiana, Illinois, and Michigan were investigated as possible sources of asbestos into Lake Michigan. Building construction, demolition and repair, landfills, and nonpoint sources which could be sources of emissions [33-35] were not investigated. Air emissions occurring in more distant states were not investigated either.

The federal regulation of no visible emissions of asbestos or use of control equipment applies to operations using asbestos.[11,36,37] Even when an industry is in compliance, quantities of microscopic asbestos may still be escaping to the ambient air.[38,39]

Johns-Manville at Waukegan, Illinois, is located on the shore of Lake Michigan. Asbestos emissions from this plant and its adjacent waste disposal site have been studied.[38,39] Even though efforts are being made to stabilize the waste disposal site, measurable amounts of asbestos have been found downwind from the site.[38] Analysis of asbestos in air streams before and after baghouse collectors has shown that the mass collection efficiency was greater than 99%. However, 10^5 fibers/m^3 (lengths > 1.5μm) and 10^8 fibers/m^3 (lengths < 1.5μm) were found downstream from the baghouse.[39] Therefore, even though the best available control technology is being used at this plant, it is a likely source of airborne asbestos to Lake Michigan.

A list of asbestos-users[40,41] subject to the National Emissions Standards for Hazardous Air Pollutants (NESHAP) was provided by the U.S. EPA. This information was validated and updated to 1976 by the respective air pollution agencies in each of the four states: Division of Air Pollution Control, Illinois Environmental Protection Agency; Air Pollution Control Division, State of Indiana Board of Health; Air Quality Division, State of Michigan Department of Natural Resources; and Bureau of Air and Solid Waste Management, State of Wisconsin Department of Natural Resources. Table 1 is the updated list (to 1976) of industries with NESHAP permits to use asbestos.

III. Investigation of Possible Natural Sources of Asbestos

The locations of natural deposits of asbestos and the possibility of erosion of these deposits were investigated as possible sources of asbestos into Lake Michigan.[25,46-57] Our investigation revealed the following:

A. Deposits of amphibole asbestos occur in the Upper Peninsula of Michigan within the drainage basin of Lake Michigan.[46-48]

1. Amphibole asbestos is associated with some of the iron deposits in the Marquette and Menominee Iron Ore Districts in Michigan.[46] Cummingtonite-actinolite asbestos often occurs in rocks which are rich

in iron ore.[49] Monitoring of mine runoff in streams near the Groveland, Republic, Empire and Humbolt iron ore mines did not reveal detectable levels of asbestos in the water.[25] At least one of these, the Groveland Mine, is known to have amphiboles present in the ore.[46]

2. Amphibolite, a rock composed largely of amphibole, is being mined in Dickinson County, near Randville, Michigan.[46,47]

B. There are no reports of any amphiboles contained in any of the rocks being mined in Wisconsin.[46]

C.- A natural deposit of short fiber chrysotile asbestos occurs locally in Marinette County, Wisconsin. This deposit, which is not being worked commercially, is found "along fractures in small knobs of serpentine."[46]

No other evidence of geological deposits of asbestos was found during this investigation. Kramer at McMaster University is presently studying the natural erosion of asbestos mineral fibers in the Great Lakes area.[50] He has not found any evidence to expect that natural erosion of chrysotile asbestos occurs in detectable amounts.

IV. Investigation of Currents as a Means of Dispersing Asbestos in Lake Michigan

Water currents in Lake Michigan and through the Straits of Mackinac were investigated in order to determine how asbestos might disperse throughout Lake Michigan and between lakes.

The Great Lakes Environmental Research Laboratory (GLERL) of the National Oceanic and Atmospheric Administration collected data on current flow through the Straits of Mackinac between May 21 and November 27, 1973, by placing current meters at various depths across the width of the Straits.[58-60] It was found that water currents through the Straits of Mackinac are complex.[58,59] In general, the net flow of water during the May-November period was from Lake

Michigan to Lake Huron 60% of the time.[60] When the water layers* were considered separately, the net flow of the epilimnetic water was generally eastward through the Straits before, during, and after thermal stratification.[58-60] Before and after stratification, the hypolimnetic water (below about 20 feet) has a net flow into Lake Huron.[58,59] However, during periods of stratification (July through September), the hypolimnetic water generally flows westward into Lake Michigan. It has been hypothesized that the hypolimnetic water flowing into Lake Michigan is coming from Lake Superior.[59] If this hypothesis is true, it may be possible for water containing asbestos (from Reserve Mining dumping activities at Silver Bay, Minnesota) to flow into Lake Michigan. Even if Lake Superior water does flow into Lake Michigan, it appears to be unlikely that Lake Superior could be a significant source of asbestos into Lake Michigan as Lake Michigan has a long flushing time (69 years** to 137 years†),[59] and less than 8% of the total volume of water entering Lake Michigan comes through the Straits.[59] Therefore, it would take a long time for an appreciable quantity of asbestos fibers from Lake Superior to accumulate throughout Lake Michigan. In addition, the preliminary data on asbestos fibers in Lake Michigan[1-3,6] identified chrysotile to be much more common than amphiboles, whereas Reserve Mining's tailings are contaminated with amphiboles.[62-65]

Models of currents of Lake Michigan exist along with some data on currents.[66-74] The existing models do not adequately predict the characteristics of Lake Michigan currents.[73,74] More testing of these models is in progress.[73,74]

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* Between July and September, the water column in the Straits is density stratified into layers: the hypolimnion (bottom) during summer stratification is a region of cold water; the thermocline is a region where water temperature decreases rapidly as depth increases; and the epilimnion (surface water) has a uniform temperature due to mixing by wave action and wind. In summer, the epilimnion is the warmest layer.[61]

** Based on current flow rates determined from the GLERL study on currents through the Straits of Mackinac.

† Based on calculations using the mean annual net flow through the Straits.

Preliminary data indicates that near shore currents (usually within 8-10 km) flow parallel to the shore of Lake Michigan. Such a coastal stream could carry any pollutants from a near shore outfall some distance along the shore before being dispersed throughout the lake. Additional work is in progress to assess the effect of the near shore currents on pollutant dispersal.[74]

V. Investigation of Dredging Activities as a Possible Mechanism of Asbestos Dispersal in Lake Michigan

The Army Corps of Engineers was contacted for information regarding dredging activities (See Table 2).[75,76] Disposal of dredged material may transport asbestos in the sediments of harbors and navigational channels to open water in Lake Michigan. Asbestos deposition in sediments may result from industrial discharges, erosion of natural sources, or deposition from air.

Dredged material that is designated as polluted by the Army Corps of Engineers is placed within confined disposal areas.[75] Only the dredged material that the Army Corps of Engineers considers to be "clean" is dumped at open water sites.[75] All the disposal sites are located within the coastal stream, which could carry suspended material along the shore for some distance.

Two of the dredged harbors in the Lake Michigan Basin are of particular interest. The dredged material from White Lake Harbor, Michigan, is classified as "clean" by the Army Corps. However, it is possible that dredgings from this harbor contain asbestos from the wastewater discharges of the Hooker Chemical Company. The dredgings from the Menominee River and Harbor were disposed of in open water before 1969. (The site is now inactive.) In the unlikely event that erosion of natural sources of asbestos occurs in Northern Wisconsin and the Upper Peninsula of Michigan, the sediments of the Menominee River and Harbor may contain asbestos.

Conclusions

Our investigation has located two asbestos- (mainly chrysotile)[77,78] using industries that discharged process wastewaters into surface waters in the Lake Michigan drainage basin (see map, p.IV-17). Before 1968, the Johns-Manville Corporation in Waukegan, Illinois, discharged wastewater into Lake Michigan.[12,13] Even though this wastewater had been passed through an "extensive settling pond system,"[13] asbestos was probably still present in the effluent. Before May of 1976, Hooker Chemical Company in Montague, Michigan, discharged water with measurable amounts of asbestos into White Lake,[25] which is in the Lake Michigan drainage basin. No industries are known to discharge asbestos-containing effluents into Lake Michigan or the Lake Michigan drainage basin at the present time.

One chrysotile deposit and several amphibole deposits (generally associated with iron ore deposits) exist in the Lake Michigan drainage basin[46-48] (see map, p.IV-17). Limited monitoring of iron mine runoff for asbestos has not revealed detectable amounts of asbestos.[25] Currently work on the erosion of natural deposits of asbestos in the Great Lakes basin is in progress at McMaster University.[50] The investigators do not expect that erosion of chrysotile asbestos occurs in detectable amounts.[50]

Investigation of water currents in Lake Michigan revealed that a current exists which runs parallel to the shore. Work is being done to determine what effect this shoreline stream actually has on pollution dispersal.[72] A current flow from Lake Superior into Lake Michigan has been hypothesized.[59] However, even if this current does exist, it is unlikely that Lake Superior could be a significant source of asbestos into Lake Michigan since Lake Michigan has a long flushing time (69-137 years),[59] and a relatively small amount of the water entering Lake Michigan comes through the Straits of Mackinac.

In addition, amphibole is the more common type of asbestos in Lake Superior,[62,65] while chrysotile is more commonly reported in the preliminary data on Lake Michigan.[1-3,6]

Dredging activities of the Army Corps of Engineers may transport asbestos fibers in sediments of harbors and navigational channels to open water in Lake Michigan. Asbestos could have settled into the sediments from natural and/or anthropogenic sources. Two Army Corps projects are of particular interest: the Menominee Harbor project which may have produced dredgings contaminated from possible erosion of natural deposits and the White Lake Harbor project which may have produced dredgings contaminated from industrial effluents. The open water disposal site of the former project is classified as inactive, while the site of the latter project is still in active use.

There are numerous industries in the four states surrounding Lake Michigan that have NESHAP permits to use asbestos. Quantities of microscopic asbestos may be emitted from these operations. Even when the best available technology is used to control asbestos emissions, numerous small fibers can be found downwind from the control device.[39,78] In addition to point sources covered by NESHAP, there are numerous other sources of airborne asbestos fibers, e.g., building construction, demolition, and repair; the wearing of automotive brake linings and clutch facings; and the weathering, wearing away, and alteration of numerous asbestos-containing products.[33-35] The numerous sources not covered by NESHAP permits may be contributing as much or more asbestos to the air than the industries covered by NESHAP.

Airborne asbestos fibers could be deposited by wet and/or dry deposition into Lake Michigan directly or into the Lake Michigan drainage basin. At least half the water entering Lake Michigan is from precipitation[59,32] largely because Lake Michigan occupies one third of its drainage basin.[32] We have done some preliminary work to determine if asbestos could be found in rainwater. This research is discussed in section V, "Chrysotile Asbestos in Rainwater."

Recommendations for Further Study

The following areas are recommended for further research to determine the presence and sources of asbestos in Lake Michigan.

1. Water taken downstream from the Hooker Chemical Company as this company used to discharge asbestos-containing wastewater prior to May of 1976

2. Lake Michigan water near the mouth of the Menominee River, whose sub-basin contains natural deposits of chrysotile and amphibole asbestos

3. Lake Michigan water and wet and dry deposition over Lake Michigan near the Johns-Manville Products Corporation in Waukegan, Illinois

4. Wet and dry deposition over Lake Michigan, especially near urban areas

5. Storm water runoff from urban areas as it may contain asbestos from brake lining wear, building demolition and construction, etc.

6. Hypolimnetic water in the Straits of Mackinac since some Lake Superior water (which may contain amphibole asbestos) may enter Lake Michigan

TABLE 1

Industries in Illinois, Indiana, Michigan, and Wisconsin Which Have NESHAP Permits to Use Asbestos (1976)

Illinois[42]

The Flintkote Company	Chicago Heights
Abex Corporation	Chicago
Armstrong Cork Company	Kankakee
Dana Corporation	Chicago
Caterpillar Tractor Company	Morton
Caterpillar Tractor Company	Mapleton
Caterpillar Tractor Company	Aurora
Caterpillar Tractor Company	Decatur
Johns-Manville Products Corporation	Waukegan
Babbitrite Company	Peoria
Borg Warner, Spring Division	Bellwood
Pure Asphalt Company	Chicago
Rockford Clutch	Rockford
United States Gypsum Company	Chicago
Coleman Cable	River Grove
Grundy Industries	Joliet
Olin Corporation	Marion
Raybestos Manhattan	Elmhurst
Resinoid Engineering Corporation	Skokie
Claud S. Gordon Company	Richmond
Daubert Chemical Company	Chicago
Victor Gasket Division	Robinson
GAF Corporation	Joliet

Illinois (Continued)

Crane Packing Company	Morton Grove
Western Electric Company	Chicago
Borg and Beck	Chicago

Indiana[43]

Bendix	South Bend
Firestone Industrial Products	Noblesville
Gatke Corporation	Warsaw
H.K. Porter	Huntington
Inmont	Huntington
National Friction Products	Logansport
The Proko Company	Cambridge City
Raybestos Manhattan	Crawfordsville
Rostone Corporation	Lafayette
Standard Industrial Products	Evansville
Union Carbide	Indianapolis
World Bestos	New Castle
Brake Supply Company	Evansville
Parr, Inc.*	Elkhart
Atlantic Richfield	East Chicago
Owens Corning	Valparaiso

* Not in compliance. Indiana is the only state for which we have compliance information.

Michigan[44]

Allied Materials	Detroit
Allied Materials	Warren
Amtech	Ferndale

Table 1 continued

Michigan (Continued)	
Auto Specialties	Hartford
Auto Specialties	St. Joseph
BASF Wyandotte	Wyandotte
Bear Paint Company	Grand Rapids
Bendix, Hydraulics Division	St. Joseph
Brown-Morse Company	Muskegan Heights
Chevy-Saginaw, GMC	Saginaw
Chrysler Corporation	Trenton
Dow Chemical	Midland
E.H. Sheldon	Muskegan
Guardsman Chemicals	Grand Rapids
Hooker Chemical	Montague
Kelsey-Hayes	Jackson
Mercury Paint Company	Detroit
Mortell	Warren
Pennwalt	Wyandotte
U.S. Gypsum	River Rouge
U.S. Gypsum	Detroit

Wisconsin[45]

Filter Material, Inc.	Waupaca
Universal Refractories	Oak Creek
W.R. Grace and Company	Milwaukee
Weyerhaeuser Company	Marshfield
Wisconsin Gasket & Manufacturing Co.	Milwaukee
U.S. Gypsum Company	Walworth

TABLE 2

Dredging Operations under the Jurisdiction of the Detroit and Chicago Districts of the Army Corps of Engineers

Name of Project	Classification of Dredged Material: Polluted[b]	Clean	Type of Open Water Disposal for Clean Dredged Material[a]: Near Shore[c]	Deep (miles from harbor)
Detroit District				
Muskegon Harbor, Mich.		x	x	1
Grand Haven Harbor, Mich.	x	x	x	1
Holland Harbor, Mich.	x[d]	x	x	1
Saugatuck Harbor, Mich.		x	x	1.5
White Lake Harbor, Mich.		x	x	1
Pentwater Harbor, Mich.		x	x	1.5
Ludington Harbor, Mich.		x	x	1.5
Manistee Harbor, Mich.		x	x	1.5
Portage Lake Harbor, Mich.		x	x	2
Arcadia Harbor, Mich.		x	x	1
Frankfort Harbor, Mich.	x[d]	x	x	1
Leland Harbor, Mich.		x	x	1
Charlevoix Harbor, Mich.		x	x	1
St. Joseph Harbor, Mich.	x	x	x	1
South Haven Harbor, Mich.	x[d]	x	x	1.5

TABLE 2 (Continued)

Chicago District			
Algoma Harbor, Mich.		x	2
Big Suamico Harbor, Wis.		x	1.5
Chicago Area		x	10[h]
Green Bay Harbor, Wis.		x	1[g],8
Kenosha Harbor, Wis.		x	3[j]
Kewaunee Harbor, Wis.		x	2.5[j]
Kipling Channel, Wis.		x	1
Manitowoc Harbor, Wis.	x	x	2.5[j]
Menominee Harbor, Mich.		x	2[i]
Michigan City Harbor, Ind.		x	2.5
Milwaukee Harbor, Wis.		x	5[i]
Oconto Harbor, Wis		--	1[e]
Pensaukee Harbor, Wis.		x	1.5[g]
Port Washington Harbor, Wis.		x	2
Racine Harbor, Wis.		x	2.5[h]
Sheboygan Harbor, Wis.		x	1.5[j]
Sturgeon Bay Canal, Wis.		x	1.5[f], 2.5
Two Rivers Harbor, Wis.		x	2
Waukegan Harbor, Ill.		x	2.5

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[a] Unless otherwise specified, the following types of disposal are still active.

[b] Polluted dredgings are placed in confined areas.

[c] Near-shore disposal areas are along the 18-foot lake contour and/or the shoreline of Lake Michigan.

[d] Polluted areas will not be dredged until a confined disposal area is obtained.

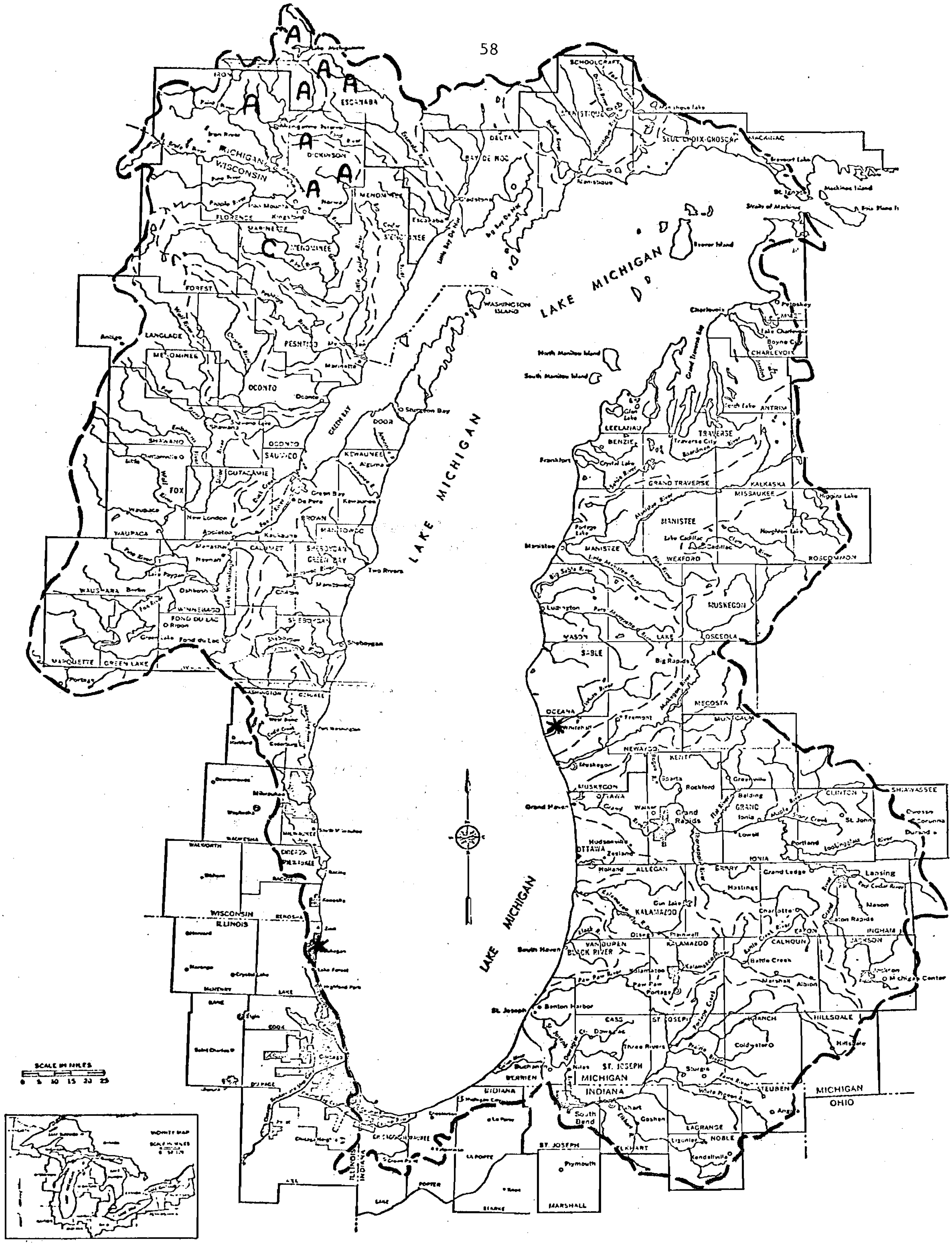
[e-j] Inactive sites last used in 1948,[e] 1965,[f] 1966,[g] 1968,[h] 1969,[i] 1970.[j]

Map of the Lake Michigan
Drainage Basin[8]

Legend*

Boundary of the drainage basin	— —
General locations of amphibole asbestos deposits	A
General location of chrysotile asbestos deposits	C
Asbestos industries that used to discharge process wastewater	*

*Note: All locations are approximate.


LAKE MICHIGAN
LAKE MICHIGAN
LAKE MICHIGAN
LAKE MICHIGAN
GREEN BAY
WASHINGTON ISLAND
Beaver Island
North Manitou Island
South Manitou Island
Mackinac Island
Straits of Mackinac
St. Ignace
MICHIGAN
WISCONSIN
ILLINOIS
INDIANA
OHIO
SCHOOLCRAFT
MANISTIQUE
SEUL CHOIX-GROSCAP
MACKINAC
DELTA
BAY DE NOC
ESCANABA
IRON
DICKINSON
MENOMINEE
FLORENCE
MARINETTE
FOREST
LANGLADE
PESHTIGO
OCONTO
DOOR
KEWAUNEE
SHAWANO
OUTAGAMIE
FOX
WAUPACA
BROWN
MANITOWOC
CALUMET
SHEBOYGAN
WAUSHARA
WINNEBAGO
FOND DU LAC
MARQUETTE
GREEN LAKE
WASHINGTON
OZAUKEE
MILWAUKEE
WAUKESHA
RACINE
WALWORTH
KENOSHA
McHENRY
LAKE
KANE
COOK
DU PAGE
LAKE
PORTER
LA PORTE
ST. JOSEPH
MARSHALL
ELKHART
LAGRANGE
NOBLE
STEUBEN
BERRIEN
CASS
ST. JOSEPH
BRANCH
HILLSDALE
VAN BUREN
BLACK RIVER
KALAMAZOO
CALHOUN
JACKSON
ALLEGAN
BARRY
EATON
INGHAM
OTTAWA
IONIA
CLINTON
SHIAWASSEE
GRAND
MUSKEGON
KENT
NEWAYGO
MONTCALM
OCEANA
MECOSTA
SABLE
MASON
LAKE
OSCEOLA
MUSKEGON
MANISTEE
WEXFORD
MISSAUKEE
ROSCOMMON
GRAND TRAVERSE
KALKASKA
BENZIE
LEELANAU
TRAVERSE
ANTRIM
CHARLEVOIX
Manistique
Escanaba
Gladstone
Marinette
Oconto
Sturgeon Bay
Green Bay
De Pere
Kewaunee
Two Rivers
Manitowoc
Appleton
New London
Neenah
Oshkosh
Ripon
Fond du Lac
Sheboygan
Port Washington
Milwaukee
Racine
Kenosha
Waukegan
Lake Forest
Highland Park
Chicago
Elgin
Gary
La Porte
South Bend
Elkhart
Goshen
Plymouth
Kendallville
Angola
Niles
Sturgis
Coldwater
Hillsdale
Three Rivers
Buchanan
St. Joseph
Benton Harbor
South Haven
Paw Paw
Kalamazoo
Portage
Battle Creek
Marshall
Albion
Jackson
Michigan Center
Lansing
Mason
Eaton Rapids
Charlotte
Hastings
Grand Ledge
Holland
Zeeland
Hudsonville
Grand Haven
Grand Rapids
Walker
Lowell
Portland
Ionia
Belding
St. Johns
Owosso
Corunna
Durand
Rockford
Sparta
Greenville
Muskegon
Whitehall
Fremont
Big Rapids
Ludington
Manistee
Cadillac
Lake Cadillac
Houghton Lake
Higgins Lake
Frankfort
Crystal Lake
Traverse City
Torch Lake
Boyne City
Lake Charlevoix
Charlevoix
Petoskey
Antigo
Portage
SCALE IN MILES
0 5 10 15 20 25
VICINITY MAP
SCALE IN MILES

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Section V

Chrysotile Asbestos in Rainwater*

C. S. Hesse and W. H. Hallenbeck

Abstract

A feasibility study was undertaken in order to determine if Chicago area rainwater contains chrysotile asbestos. Rainwater samples were collected at one Chicago and two suburban Chicago locations. The Chicago location was near a busy intersection and building construction. The two suburban stations were located in residential areas. All three sites were within four blocks of a major expressway. None of the suburban rainwater samples contained levels of chrysotile which were significantly above laboratory contamination levels. All the Chicago rainwater samples contained significant levels of chrysotile. This finding demonstrates a mechanism by which asbestos can be transferred from air to surface water.

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Introduction

Occupational exposure to asbestos has been well documented as a health hazard.[1-10] However, at the present time it is very difficult to evaluate the potential health hazards associated with environmental exposure to asbestos at the levels found in ambient air or drinking water.[1,2] The term "asbestos" refers to several fibrous minerals composed of impure magnesium silicates such as chrysotile, amosite, crocidolite, anthophyllite, termolite, actinolite, and cummingtonite.[11,12] Since asbestos does not burn or conduct electricity and can be woven into cloth, it has come to have over 3,000 uses[13] and consequent widespread distribution in the environment.

Recent preliminary work has suggested that asbestos is present in raw and finished drinking water supplied by Lake Michigan.[14-19] Since about half the water entering Lake Michigan is from precipitation,[20,21] it has been recommended that wet deposition into Lake Michigan, especially near urban areas, should be examined for asbestos (see Section IV).

Collecting samples over Lake Michigan is a complicated and expensive procedure. Therefore, a preliminary study was conducted to determine if chrysotile asbestos could be found in rainwater at more accessible, land-based locations. Such a study would demonstrate a mechanism by which airborne asbestos could be transferred to surface waters.

Method

Sample collection. Rainwater samples were collected at one Chicago and two suburban Chicago locations. The Chicago samples were collected on a horizontal roof-top which was approximately 30 feet above street level and near a busy intersection and building construction. The suburban samples were collected in residential neighborhoods which had no building construction and less traffic than the Chicago site.

The suburban sites were either 25 feet above ground on an open stairway landing or two feet above ground on a cement block fence. All three sites were within four blocks of a major expressway.

Plastic buckets with 363 cm^2 openings were used to collect the rainwater samples. All labware which came into contact with the samples was washed with detergent and warm water, rinsed with water which had been prefiltered through a 0.1 micrometer (μm) pore-size Millipore membrane filter, and covered and stored until needed.

An attempt was made to collect a rainwater sample as soon as possible after a period of rain started. The time lag between when a period of continuous rain started and when collection commenced varied from less than five minutes to approximately 30 minutes. The lag time for intermittent rainfalls varied from four to 7½ hours.

Immediately before the rainwater samples were collected, the collection buckets were given two final rinses with 0.1 μm filtered water. The latter of the two rinses was stored in a one-half gallon plastic bottle and used as a control for contamination. Collection buckets were located where overhanging structures could not drip into them. As soon as a rainfall ended, the collected rainwater was transferred to a one-half gallon plastic bottle. Table 1 gives the characteristics of the individual rainwater samples collected.

<u>Sample preparation</u>. The control and rainwater samples were filtered (usually within three days of collection) through 47 mm Millipore membrane filters with a pore size of 0.45 μm. The filters containing the samples were placed into plastic petri dishes and dried in a vacuum desiccator. Either whole membrane filters or sections of an extremely dirty filter were ashed in an International Plasma Corporation IPC 1000 low temperature asher by a method similar to that of Bishop.[22] The radio-frequency energy was held at 25 watts for approximately 15 minutes and then increased to 75-80 watts for 30-35 minutes. Oxygen at a pressure

of one torr was used as the reaction gas. Samples were allowed to cool to room temperature inside the asher. Approximately five ml of 0.1 μm filtered water were added to the ash in each crystallization dish. The samples were sonicated 5-10 minutes in order to disperse any large clumps of debris, mixed with more filtered water, and refiltered through 0.45 μm Millipore membrane filters. The filters were stored in petri dishes and dried in the vacuum desiccator as before.

In order to prepare a sample for electron microscopy, a random 3 mm punch from the membrane filter was placed on a 0.25% Formvar and carbon coated, 200 mesh, copper transmission electron microscope grid. The membrane filter was dissolved in acetone vapors in a condensation washer with a reflux rate of approximately one drop per second. The acetone was prefiltered through a 0.2 μm Fluoropore filter.

Sample analysis. A Philips 300 transmission electron microscope was used to scan grids at 100 kilovolts and a magnification of approximately 23,000-24,000 times. Random numbers corresponding to the vernier settings of the specimen stage on the electron microscope were used to select the ten grid squares scanned for each sample. When a fiber was located, its morphology and selected area electron diffraction pattern were used to determine if it was chrysotile. Whenever a positive identification was made, its length and width were measured so that a mass estimate could be made.

The term "fiber" indicates a unit of asbestos which could be composed of a single fibril or a bundle of fibrils. The number of chrysotile asbestos fibers per liter of rainwater was calculated by the following equation:

$$X_f = 1000\ F\ (\bar{x} - \bar{y})/VA$$

where

X_f = concentration of asbestos in fibers per liter of rainwater

$\bar{x}$ = average number of fibers per grid square for rainwater

$\bar{y}$ = average number of fibers per grid square for the respective control

V = volume of rainwater filtered in ml

F = effective filter area = 9.62 cm^2

A = area of one grid square = 8.24×10^{-5} cm^2

1000 = conversion factor from ml to liter.

In order to estimate the mass of chrysotile in a liter of rainwater, an individual chrysotile fibril was assumed to be cylindrical[23] and have a density of 2.5 picograms (pg) per cubic micrometer.[1] The concentration of asbestos in rainwater was calculated by the following equation:

$$X_m = \frac{1000\ F\ \pi\ 2.5}{NAV}\left(\sum_{i=1}^{r} L_i R_i^{\,2} - \sum_{j=1}^{c} L_j R_j^{\,2}\right)$$

where

V, F, A, 1000 are the same as above

X_m = concentration of asbestos in picograms per liter of rainwater

2.5 = density of chrysotile in picograms per cubic micrometer

N = number of grid squares scanned = 10

r = total number of rainwater fibrils in N grid squares

c = total number of control fibrils in N grid squares

L = length of a fibril in micrometers

R = radius of a fibril in micrometers.

Results and Discussion

The goodness-of-fit test for a Poisson distribution was applied to fiber count data for each sample, all controls combined, and all rainwaters combined.[24-26] All these data were found to follow a Poisson distribution. Therefore, a test of the significance of difference between two Poisson variables was used in order to determine if the number of chrysotile fibers found in a given rainwater sample was significantly greater than the number of fibers found in its control.[24,27] The two members of a rainwater/control pair were prepared at the same time and, therefore, subjected to similar amounts of contamination. None of the rainwater samples collected at the suburban Chicago locations contained significantly greater numbers of chrysotile fibers than their respective controls (at $\alpha = 0.05$ for a one-tailed test). However, all the Chicago rainwater samples contained significantly higher numbers of chrysotile fibers than their respective controls. See Table 2 for p-values. Table 4 contains the concentrations of chrysotile in the Chicago rainwater samples in terms of fibers per liter adjusted for contamination.

The rank sum test[28] was applied to the mass data for each pair of samples. None of the rainwater samples collected at the suburban Chicago locations contained masses of chrysotile fibers significantly greater than their respective controls (at $\alpha = 0.05$ for a one-tailed test). All the Chicago rainwater samples contained significantly greater masses of chrysotile fibers than their respective controls. See Table 3 for p-values. Table 4 contains the concentrations of chrysotile in the Chicago rainwater samples in terms of mass per liter adjusted for contamination.

Referring to Table 4, it should be noted that the highest concentrations of asbestos are associated with the shortest lag times of collection. This relationship is consistent with findings for other pollutants.[29] The Chicago sample from August 6, 1976, contained the highest concentration of chrysotile asbestos and had no collection lag time. This sample contained a mixture of wet and dry deposition since collection commenced during a very brief shower and continued through several rainless hours. During subsequent intermittent rainfall, 100 ml of rain were collected. Therefore, the relatively high level of asbestos in this sample may reflect dry deposition and/or a high concentration of asbestos in the initial rainfall.

The amount of a pollutant washed out of the atmosphere is proportional to the airborne concentration of a pollutant.[30] Therefore, the large difference between the chrysotile content of Chicago and suburban Chicago rainwater may be related to a difference in airborne concentrations. Differential airborne concentrations could be attributed to differences in traffic flow and subsequent brake-lining wear, building construction and demolition, and manufacturing activities.

In conclusion, samples of rainwater collected in Chicago contained chrysotile asbestos ranging from 10^5 to 10^6 fibers per liter. This finding demonstrates that precipitation scavenges airborne chrysotile asbestos which may result in the contamination of surface waters.

Table 1. Characteristics of Rainwater Collection

Date collected	Volume collected (ml)	Sampling height (ft)	Collection lag time*	Collection time (hr)
<u>Chicago Samples</u>				
5-28-76	660	30	< 30 min	6
6-18-76	520(260)**	30	10 min	2
9-09-76	520	30	5 hrs	4
10-05-76	400	30	7½ hrs	2
8-06-76***	100(25)	30	0	17
<u>Suburban Chicago Samples</u>				
6-13-76	850	25	4 hrs	2
6-27-76	140(70)	25	20 min	1½
6-29-76	370 (185)	25	minutes	4
7-20-76	600 (150)	2	10 min	1
7-31-76	300	2	10 min	7½
9-19-76	120	2	< 5 min	2

*Approximate length of time from beginning of rain until collection commenced.

**The volume analyzed is given in parenthesis if it differed from the volume collected.

***This "rainwater" sample contained a mixture of wet and dry deposition as the bucket was in place before the rain started.

Table 2. Number of Chrysotile Asbestos Fibers Found in Rainwater and Control Samples

Date collected	Number of chrysotile fibers in ten grid squares Rain	 Control	P-values for test of significance of difference between two Poisson variables*
Chicago Samples			
5-28-76	10	0	0.005**
6-18-76	25	0	< 0.005**
9-09-76	12	2	0.01**
10-05-76	6	0	0.025**
8-06-76***	56	5	< 0.005**
Suburban Chicago Samples			
6-13-76	6	4	> 0.10
6-27-76	3	5	> 0.5
6-29-76	6	5	> 0.10
7-20-76	5	4	> 0.10
7-31-76	1	1	> 0.10
9-19-76	8	2	0.10

*One sided test (of the alternative hypothesis) that the level of chrysotile in the rainwater sample was greater than that in its control.

**Significant at $\alpha = 0.05$.

***This "rainwater" sample contained a mixture of wet and dry deposition as the bucket was in place before the rain started.

Table 3. Mass of Chrysotile Asbestos Fibers Found in Rainwater and Control Samples

Date collected	Mass of chrysotile found in ten grid squares (picograms)		P-values for test of significance of difference (Rank-Sum Test)*
	Rain	Control	
<u>Chicago Samples</u>			
5-28-76	0.145	0	0.032**
6-18-76	0.596	0	< 0.001**
9-09-76	0.565	0.052	0.022**
10-05-76	0.121	0	0.032**
8-06-76***	0.806	0.540	0.038**
<u>Suburban Chicago Samples</u>			
6-13-76	0.868	0.024	0.3
6-27-76	0.035	0.601	0.780
6-29-76	2.267	0.038	0.456
7-20-76	0.054	0.033	0.3
7-31-76	0.008	0.016	0.5
9-19-76	0.279	0.027	0.062

*One sided test (of the alternative hypothesis) that the level of chrysotile in the rainwater sample was greater than that in its control.

**Significant at $\alpha = 0.05$.

***This "rainwater" sample contained a mixture of wet and dry deposition as the bucket was in place before the rain started.

Table 4. Concentration of Chrysotile Asbestos in Chicago Rainwater Samples Adjusted for Contamination Found in Their Respective Controls*

Date sample collected	Collection lag time	Concentration in fibers per liter	95% confidence interval of concentration in fibers per liter[24,27]		Concentration in nanograms per liter
			lower	upper	
10-05-76	7½ hr	1.8×10^5	0.64×10^5	3.8×10^5	3.5
9-09-76	5 hr	2.2×10^5	1.1×10^5	4.1×10^5	12
5-28-76	30 min	1.8×10^5	0.85×10^5	3.3×10^5	2.6
6-18-76	10 min	1.1×10^6	0.73×10^6	1.7×10^6	27
8-06-76**	0 min	2.4×10^7	1.7×10^7	3.1×10^7	120

*Only those rainwater samples that differed significantly from their controls at $p \leq 0.05$ are given in this table.

**This "rainwater" sample contained a mixture of wet and dry deposition as the bucket was in place before the rain started.

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