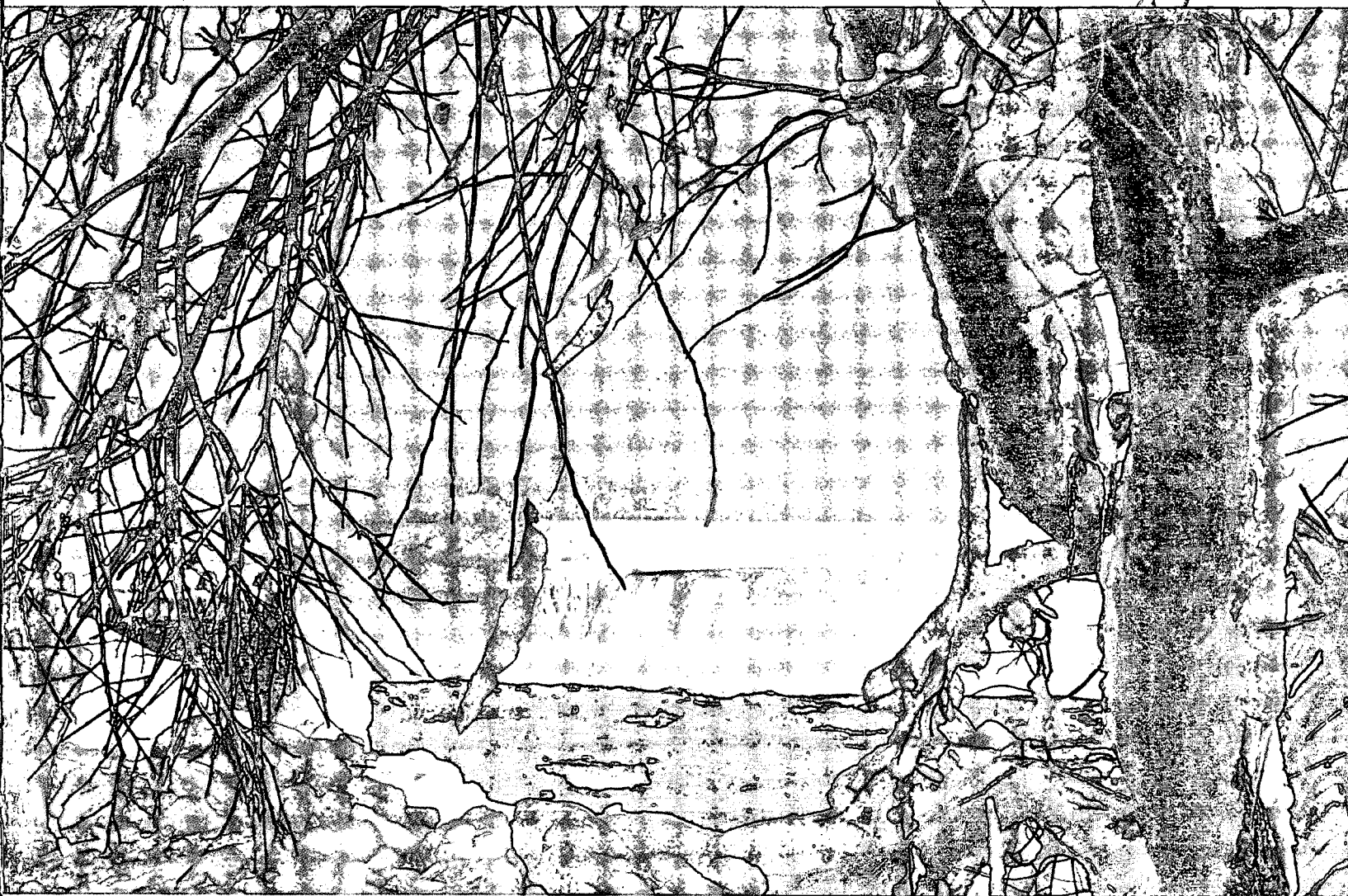




# Asbestiform Fibre Levels in Lakes Superior and Huron

R.W. Durham and T. Pang



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**SCIENTIFIC SERIES NO. 67**

*(Résumé en français)*

INLAND WATERS DIRECTORATE,  
CANADA CENTRE FOR INLAND WATERS,  
BURLINGTON, ONTARIO, 1976.



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## Abstract

Asbestiform fibre concentrations from less than 0.1 million to 87.3 million per litre were measured in the open waters of Lake Superior during 1973. The most commonly occurring mineral fibre was chrysotile, although fibres of the amphibole mineral cummingtonite were found in the western arm of Lake Superior near the Reserve Mining tailings dumping site at Silver Bay, Minnesota. The results suggest that the zone of influence of this dumping of asbestiform fibres does not extend far out into the lake and that it decreases from June through November. Rivers flowing into Lake Superior which were analyzed all had low chrysotile fibre concentrations below 8 million per litre, as did samples taken in spring 1974 from Lake Huron and Georgian Bay.

## Résumé

Au cours de 1973, on a mesuré des teneurs aussi faibles que 0.1 million à 87.3 millions de fibres amiantacées par litre dans les eaux libres du lac Supérieur. La fibre minérale la plus courante était le chrysotile bien qu'on ait trouvé des fibres de cummingtonite de type amphibole dans le bras ouest du lac, près des décharges de résidus de la *Reserve Mining* de Silver Bay au Minnesota. D'après les résultats de cette étude, la zone d'influence de ces décharges ne s'étendrait pas loin dans le lac et elle diminuerait de juin à novembre. Les échantillons prélevés dans des tributaires du lac Supérieur présentaient tous de faibles teneurs en chrysotile (moins de 8 millions de fibres par litre); de même que ceux prélevés au printemps de 1974 dans le lac Huron et la baie Georgienne.

# Asbestiform Fibre Levels in Lakes Superior and Huron

R.W. Durham and T. Pang\*

## INTRODUCTION

In December 1972 International Joint Commission public hearings on pollution of the Upper Great Lakes from land use activities were held in Duluth. Reference was made at that time to the public health hazard arising from dumping of taconite tailings containing asbestos minerals into Lake Superior by Reserve Mining at Silver Bay, Minnesota. The production of cancer in humans by inhalation of asbestiform fibres is well documented, but the correlation between stomach cancer and ingestion of such fibres has only recently been suggested by Merliss (1). A study of stomach cancer mortalities in 24 countries showed Japan to have the highest rate, seven times higher than for the United States. Merliss relates this high incidence to the fact that rice bought by the Japanese consumer is treated with talc containing fibrous asbestos minerals. A study of cancer mortality over the period 1950-69 in the city of Duluth, Minnesota, however, did not show any effect of the entry of asbestiform fibres into the drinking water supply starting in 1955 (2).

As Lake Superior is used as a drinking water source for many Canadian communities, Environment Canada started an investigation early in 1973 to determine whether or not transboundary movement of asbestiform fibres from Reserve Mining's operation occurred. Samples of water were taken from many stations in Lake Superior during five regular monitoring cruises of the lake by vessels from Canada Centre for Inland Waters (CCIW) during 1973 and subsequently from Lake Huron during the 1974 field season. An analytical method to determine fibre concentration and size distribution was developed, using a transmission electron microscope. The method is described, along with some initial results, in a previous publication (3).

The first step in the analysis was to concentrate the fibres from 250 ml of sample by ultracentrifugation for 2 hr at 20,000 rpm (about 30,000 g). The fibres were resuspended ultrasonically in one millilitre of fibre-free distilled water. Because of the low concentrations of particulates in Lake Superior and Lake Huron waters no interference with the fibre counting from clumps of detrital material occurred, so no further processing was required.

One microlitre of this suspension was transferred to a 3-mm carbon-coated grid and dried under a heat lamp before insertion into the Siemens 101 transmission electron microscope. The grid was then scrutinized at a low magnification to determine the sample coverage, usually about 100 grid squares of a 200-mesh grid. The magnification was then increased to 20,000 and all the fibres counted in the 100 grid square openings. The ratio of total area to open area of a grid square was measured on each grid in order to calculate the total fibres in the 1- $\mu$ l aliquot from the number counted in the grid square openings. This ratio averaged 2.4 for the batch of grids used in these analyses.

A calibrated eye-piece was used to determine the size distribution of the fibres. The shortest fibre length recorded was 0.05  $\mu$ m.

## SAMPLING AND ANALYSIS

The general locations of the sample stations in Lakes Superior and Huron are shown in Figures 1 and 2. Water samples were taken generally at 1 m below the surface, but at some stations they were also taken near the bottom and at the thermocline. Other samples, defined as integrated, were taken while lowering the sample container from 15 to 50 m. The samples were stored in 25-l polyethylene containers until analyzed either at CCIW, Ontario Research Foundation (ORF) or McMaster University. The analytical methods used by the other two laboratories have been described in the literature (4,5) and differ mainly in the techniques used to concentrate the fibres.

Although the majority of fibres from the lakes observed in the electron microscope were identified as chrysotile by their characteristic hollow-tube appearance (Fig. 3), fibres of another asbestiform mineral were present in samples from the western arm of Lake Superior. These were identified as cummingtonite by comparing the electron diffraction pattern (Figs. 4a and 4b) of such a fibre with that of a cummingtonite fibre from the taconite tailings of Reserve Mining (3).

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The detection limit for samples analyzed at CCIW is defined as that concentration which would produce a number of fibres on the electron microscope grid equal to that found due to background. The highest background count found on a carbon-coated grid before evaporation of the processed sample aliquot was 10 fibres per 100 grid openings, the average area covered by the evaporated sample aliquot. This number corresponds to a concentration of 0.1 million fibres per litre in a 250-ml sample. For each analysis the number of fibres on the grid before sample evaporation was measured and subtracted from the total.

## RESULTS AND DISCUSSION

Tables 1,2,3 and 4 give the values obtained for the concentrations of fibres and the dates of the cruises during which the samples were taken. The range of values from

0.1 million to 87.3 million fibres per litre is comparable with other published measurements of fibre levels in surface waters. Cunningham and Pontefract (6) found asbestiform fibre concentrations varying from 2 million to 173 million per litre in a survey of tap water in several cities and towns in Ontario and Quebec. Kay (7) reported from 0.14 million to 25.3 million fibres per litre in samples from the drinking water supplies of a large number of Ontario cities and towns. The samples were analyzed by Ontario Research Foundation. The type of asbestiform fibre found in both these studies was chrysotile. Cook, Glass and Tucker (8), however, reported that analyses of the Duluth water supply during 1973 gave values of from 1 million to 30 million fibres per litre of amphibole asbestos. The predominant amphibole mineral present was cummingtonite originating from the operations of Reserve Mining at Silver Bay.

Table 1. Asbestos fibre concentrations in Lake Superior (millions of fibres per litre)

Station	Depth	Cruise				
		73-03-103	73-03-104	73-03-106	73-03-107	73-03-108
2	1 m		1.9	4.1	10.9	1.0
	bot			4.6		1.6
12	int		1.6			
22	int		3.0			
25	int		2.7			
31	1 m		BDL	0.6		1.0
34	int		1.3			
36	int		3.4			
39	int		82.0			
43	1 m		1.5	1.4	7.5	0.3
	bot		BDL	1.7	1.4	0.1
53	1 m			2.2		2.5
	bot			3.6		3.6
62	int		2.8			
68	1 m			5.8		3.5
	bot			3.3		3.0

1 m = Sample from 1 m below surface

bot = Sample from 1 m above lake bottom

int = Integrated sample from 15 to 50 m

BDL = Below detection limit of  $0.1 \times 10^6$  fibres per litre

Cruise	Date 1973
73-03-103	June 15-28
73-03-104	July 26-Aug. 3
73-03-106	Sept. 4-18
73-03-107	Oct. 9-29
73-03-108	Nov. 18-Dec. 3

Table 1. (Cont.)

Station	Depth	Cruise				
		73-03-103	73-03-104	73-03-106	73-03-107	73-03-108
73	1 m		BDL	3.7	24.8	1.1
	bot			7.4		2.8
80	int		1.5			
84	1 m			45.0		0.7
	bot			1.5		1.3
	int		1.5			
89	1 m		2.2	23.3	0.6	0.8
92	int		1.7			
102	1 m			19.0		1.5
	bot			2.9		2.3
	int		5.2			
115	int		1.9			
127	1 m		BDL	3.0		1.2
	bot		0.1	2.3		2.3
	int		0.6		11.0	
133	int		0.9			
138	1 m		1.5	12.6	12.4	0.6
	bot		0.8	0.3	1.3	1.6
	int		2.5			
142	1 m		BDL	5.4	1.4	2.2
	bot			1.9		1.6
	int	15.5				
149	int		3.1			
152	1 m			14.0		1.1
	bot			4.0		0.9
157	int		1.8			
169	int		5.0			
173	1 m			21.0		1.0
	bot			3.9		1.6
180	int		17.0	3.6		
189	1 m			6.9		1.7
	bot			2.0		1.9
	int		3.4			
201	int			1.0		
203	1 m		9.5	0.8	0.3	0.1
	bot					0.4
	int	87.3	3.1		2.9	
204	int			0.9		
207	int		8.0			



The sample taken during the first cruise, 73-03-103 at station 203, about 8 km offshore from Silver Bay showed a high fibre level with a preponderance of cummingtonite fibres. The samples from subsequent cruises however, had much lower fibre concentrations and the cummingtonite contribution became progressively less until it disappeared in cruises 73-03-107 and 73-03-108. This effect could have been caused by an offshore wind at the time of the first cruise, moving the Reserve Mining tailings farther out into the lake. Another possibility is that a hesitation in the strong southwesterly inshore current during early spring (9) allowed the tailings to diffuse out farther. Although cummingtonite fibres large enough to be identified by electron diffraction generally occurred only in the western arm of Lake Superior, samples from a few stations elsewhere in the lake occasionally were found to contain fibres that morphologically were not identifiable as chrysotile. These fibres, which were classified as amphiboles, always represented a small fraction of the fibres counted on the electron microscope grid. For example, the cruise 73-03-104 1-m sample at station 2 was found to contain 19% of amphibole fibres, but none were found in samples from the next three cruises. These amphibole fibres are more likely to be of local origin than from the taconite mining operations in Minnesota.

Transmissometer measurements during cruise 73-03-106 showed a band of high opacity coincident with the thermocline, indicating a sharp density difference at this depth. Density currents from the taconite tailings sinking to the bottom of the epilimnion and then spreading along the thermocline might have caused fibres to be trapped in this band. Samples were taken therefore from within the band at stations near the tailings dumping site as well as at stations well removed from it. The results given in Table 2 clearly show that no major concentration of fibres occurred in the band. In fact, for station 127 the thermocline result was lower than those for surface and bottom samples. At station 203, the only other station where another depth sample was taken, the thermocline value was higher than that for the surface sample. Generally, there appears to be no correlation between concentration and depth at which the samples were taken.

Table 2. Fibre concentrations of water samples from Lake Superior taken near the thermocline on cruise 73-03-106

Station	23	175	181	203	207	127
Depth(m)	25	22	20	13	30	25
Millions of fibres per litre	5.0	1.5	0.3	4.6	1.1	0.7

In some cases the surface values were higher than bottom values, and in other cases the converse was true.

The concentrations of asbestiform fibres in Lake Huron and Georgian Bay were surveyed during the spring of 1974. The samples were taken during three cruises from 11 stations and the results are shown in Table 3. All the fibres measured were chrysotile, and the concentrations ranged from below the detection limit to 4.9 million fibres per litre. These correspond to the lower portion of the range of values found in Lake Superior. The fibre size distributions for stations in the two lakes given in Figure 5 show a marked similarity, with 95% of the fibres being less than 1  $\mu\text{m}$  in each case.

Table 3. Concentration of asbestos fibres in Lake Huron and Georgian Bay

Cruise	Station	Depth(m)	Fibre concentration in millions per litre
74-05-101	10	1	0.5
74-05-101	33	1	4.9
74-02-101	2	1	0.6
74-02-101	4	1	0.3
74-02-101	9	1	BDL
74-02-101	21	1	0.7
74-02-101	22	1	0.5
74-02-101	29	1	0.9
74-02-101	36	1	0.3
74-02-102	38	1	BDL
74-02-102	56	1	0.6

BDL = Below detection limit of  $0.1 \times 10^6$  fibres per litre.

Cruise	Date
74-02-101	April 23-27
74-05-101	April 27-May 1
74-02-102	May 13-18

Analyses of water samples taken near the mouths of tributaries of Lake Superior gave low values as shown by Table 4. The fibres observed were chrysotile, so that these rivers, at least, were not a source of amphibole fibres in Lake Superior.

Table 4. Asbestos fibre concentrations in Lake Superior tributaries

River	Sample location	Date sampled	Millions of fibres per litre
Black	Highway 17	5/2/74	7.1
Pic	Highway 17	5/2/74	0.2
Michipicoten	Highway 17	6/2/74	0.2
Kaministiquia	Highway 17	4/2/74	0.1
Nipigon	Highway 17	6/2/74	0.1
Pine	Highway 61	4/2/74	BDL

BDL = Below detection limit of  $0.1 \times 10^6$  fibres per litre

Table 5. Interlaboratory comparison of fibre analyses in water samples from Lake Superior (millions of fibres per litre)

Cruise	Station	Canada Centre for Inland Waters†	Ontario Research Foundation†	McMaster University*
73-03-103	142	15.5	7.0	—
73-03-103	203	87.3	28.3	—
73-03-104	43	1.4	0.6	—
73-03-104	73	BDL	1.5	—
73-03-104	89	2.2	0.4	—
73-03-104	138	1.4	0.8	—
73-03-104	142	BDL	1.4	—
73-03-106	2	1.4	0.7	4.1
73-03-106	73	3.6	0.6	3.7
73-03-106	142	1.1	8.0	5.4
73-03-108	2	1.1	0.9	1.0
73-03-108	142	0.8	0.9	2.2

\*Duplicate sample

†Same sample

BDL = Below detection limit of  $0.1 \times 10^6$  fibres per litre

A direct comparison of analytical methods between the Canada Centre for Inland Waters and the Ontario Research Foundation is possible, as both laboratories analyzed a set of 12 samples from Lake Superior. The results are not consistent at low values of fibre concentration (Table 5). At higher values, such as occurred in cruise 73-03-103, the CCIW results are clearly higher than ORF by about a factor of three. The larger number of fibres deposited on the electron microscope grid from the higher concentration samples reduces the statistical error in counting the fibres, which would account for the more consistent results from these samples.

Results of five analyses by McMaster University can be compared with results of similar samples analyzed by CCIW and ORF. The samples analyzed by McMaster were duplicate samples taken during cruises 73-03-106 and 73-03-108 at the same stations and immediately after those analyzed at CCIW and ORF. The results obtained by McMaster are not too different from those obtained by the other laboratories for similar samples, although a slight trend to higher results is apparent. There are insufficient data to establish this trend precisely, however.

## ACKNOWLEDGMENTS

The authors appreciate the help of M.R. Thompson with the analyses at CCIW. Dr. E. Chatfield and Dr. J.R. Kramer were responsible for the analyses carried out at Ontario Research Foundation and McMaster University respectively.

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**Figures 1 to 5**

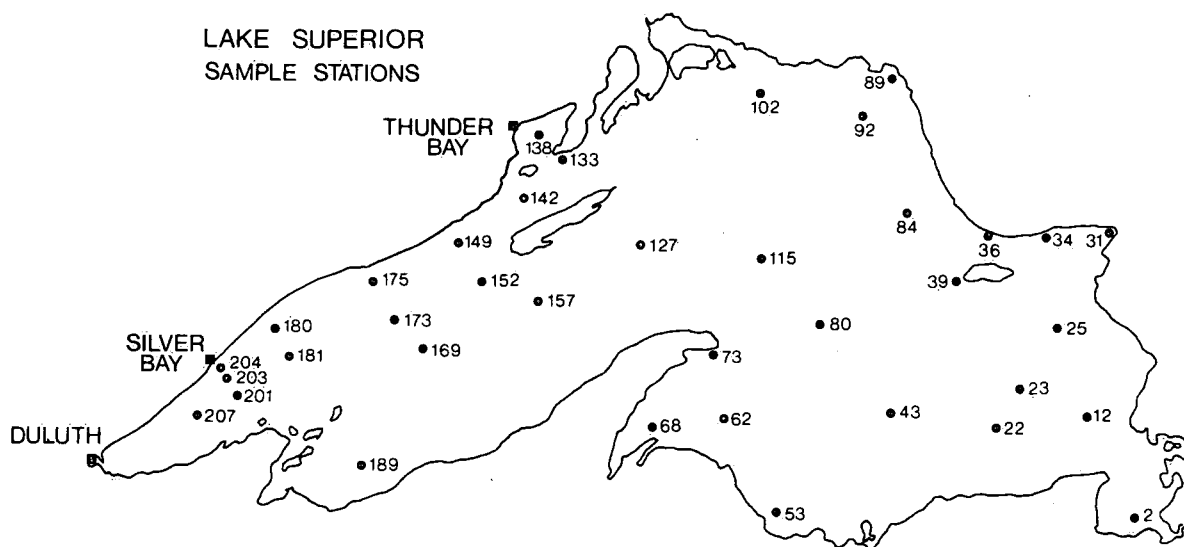


Figure 1. Locations of sample stations in Lake Superior.

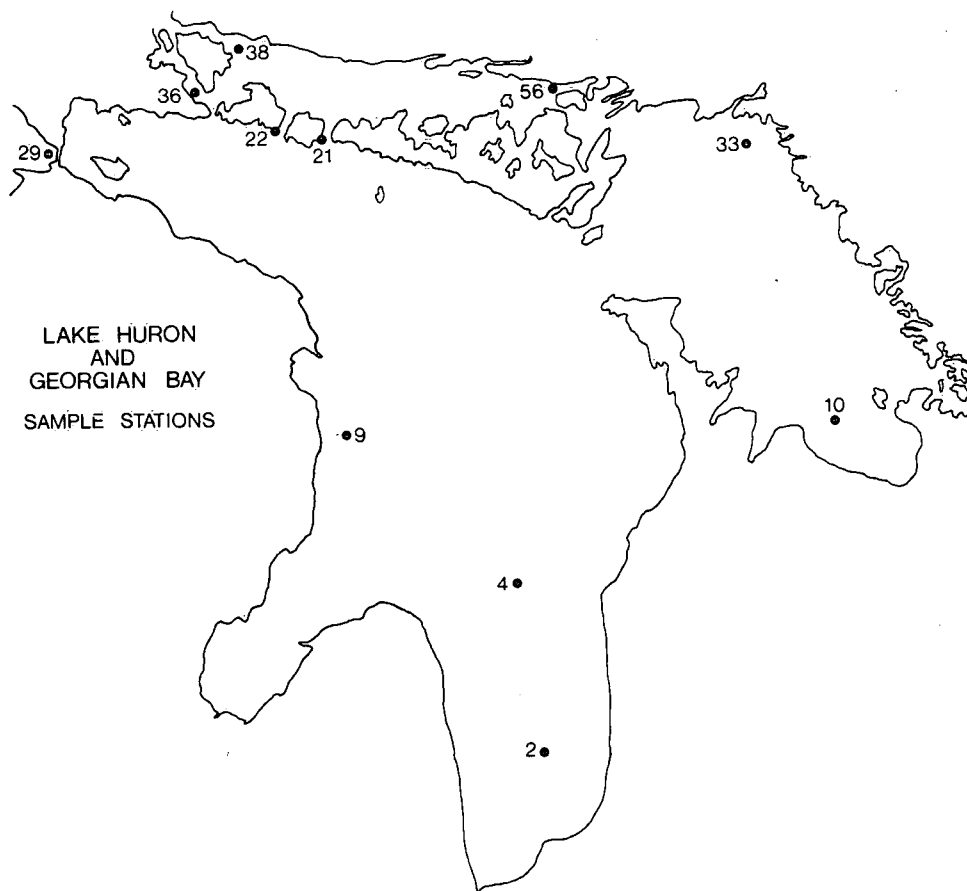


Figure 2. Locations of sample stations in Lake Huron and Georgian Bay.

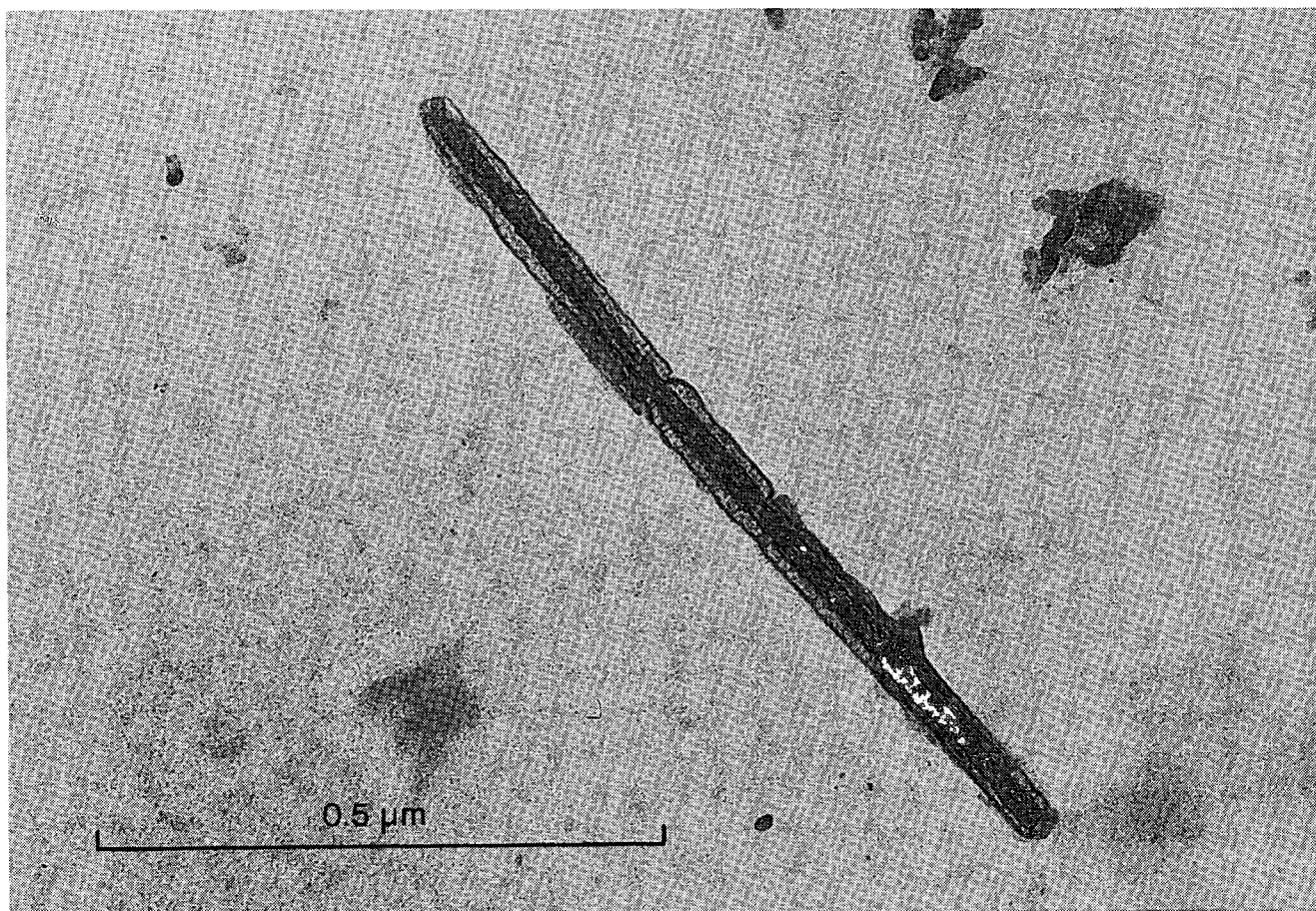


Figure 3. Electron micrograph of chrysotile fibre from Lake Superior.

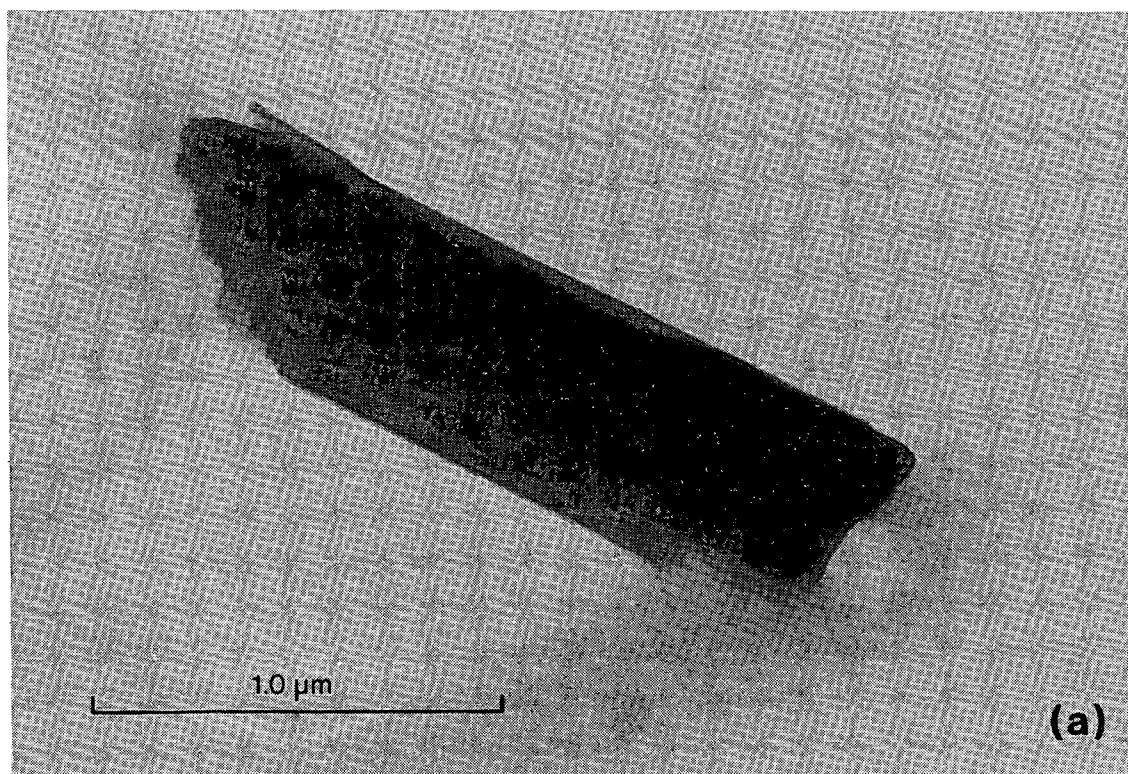


Figure 4(a). Electron micrograph of cummingtonite fibre from western arm of Lake Superior.



Figure 4(b). Electron diffraction pattern of cummingtonite fibre shown in Fig. 4(a).

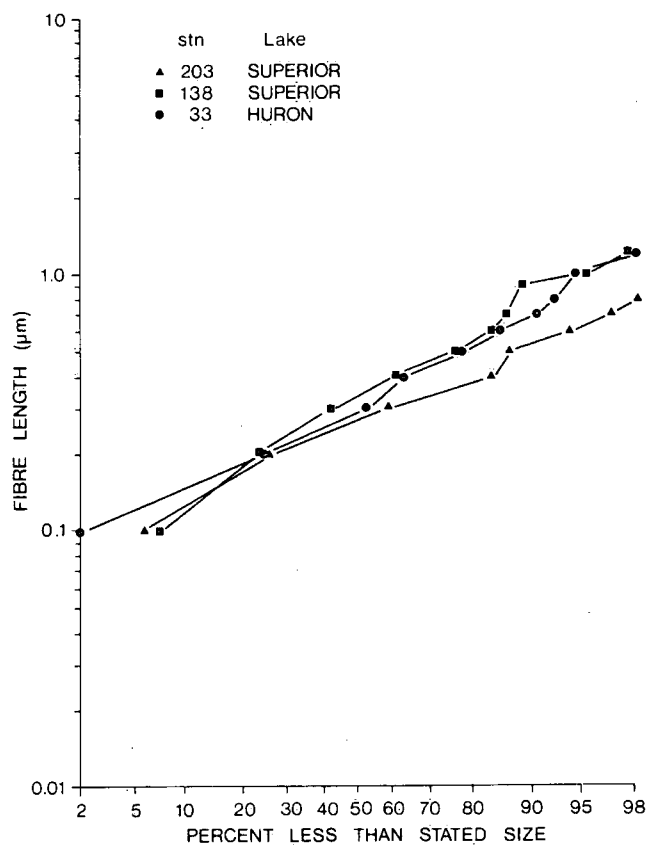


Figure 5. Size distribution of asbestiform fibres in Lakes Superior and Huron.

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