

Airborne Fiber Concentration and Size Distribution of Mineral Fibers in Area with Serpentinite Outcrops in Aichi Prefecture, Japan

Kiyoshi SAKAI^{1*}, Naomi HISANAGA², Norihiko KOHYAMA²,
Eiji SHIBATA³ and Yasuhiro TAKEUCHI⁴

¹Nagoya City Public Health Research Institute, 1–11, Hagiya-cho, Mizuho-ku, Nagoya 467-8615, Japan

²National Institute of Industrial Health, Ministry of Labour, 6–21–1, Nagao, Tama-ku, Kawasaki 214-8585, Japan

³Department of Medical Technology, Nagoya University School of Health Science, 1–1–20, Daiko-minami, Higashi-ku, Nagoya 461-8673, Japan

⁴Department of Occupational and Environmental Health, Nagoya University Graduate School of Medicine, 65, Tsurumai-cho, Showa-ku, Nagoya 466-8550, Japan

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Abstract: Airborne fiber concentrations and size distributions of both asbestos and nonasbestos fibers were determined at property boundaries of 4 serpentinite quarries producing crushed stone (quarry property boundary), 10 sites within 10 km of the area with serpentinite outcrops (serpentinite area), and 2 sites in a reference area. The asbestos was identified in 7 rock and 3 soil samples collected in the serpentinite area. The geometric means of airborne concentrations of asbestos and nonasbestos fibers longer than 0.2 μm in length were: 384 and 447 fibers/liter (f/L) in the quarry property boundary (in operation), 12 and 124 f/L in the quarry property boundary (in closed), 5 and 103 fibers/liter in the serpentinite area, and less than 2 and 59 fibers/liter in the reference area, respectively. There was a significant difference in airborne concentrations of both asbestos and nonasbestos fibers among the areas. Tremolite/actinolite were found with chrysotile in the stones. Airborne concentrations of tremolite/actinolite were higher than those of chrysotile at the quarry property boundary and in the serpentinite area. Tremolite/actinolite were also found in the soils. There was no significant difference among the areas in the arithmetic means of fiber size distribution of both asbestos and nonasbestos fibers.

Key words: Asbestos, Nonasbestos fiber, Serpentinite, Fiber concentration, Fiber size distribution, Ambient air

Introduction

Serpentinite is primarily used for crushed stones and raw materials in fertilizer, construction, and the iron and steel industries. In Japan 1.8, 2.1 and 2.3 million tons of serpentinite were produced as crushed stone in 1980, 1985 and 1990, respectively¹⁾. Crushed serpentinite has often been produced at some open-pit quarries located in and around areas with

serpentinite outcrops. Serpentinite sometimes contains tremolite and anthophyllite as well as chrysotile^{2–5)}. There are some reports on air pollution by asbestos from natural sources in the United States. Airborne chrysotile concentrations in the vicinity of roads paved with asbestos-containing serpentine rock were about 1,000 times greater than those found in urban areas³⁾. Airborne chrysotile concentrations found in a recreational area located in a serpentine massive were found to be 100,000 to 10,000,000 times greater than those in urban areas⁴⁾. In Japan airborne

*To whom correspondence should be addressed.

asbestos concentrations in the vicinity of a serpentinite quarry producing crushed stone were higher than those in reference areas⁶. Asbestos was disseminated from asbestos mines and mills rather extensively⁷⁻¹⁰. The possibility was raised of air pollution by asbestos dispersed from serpentinite rock by natural means, such as weathering and landslides, or by human activities, such as quarrying, road building, construction and tilling the soil¹¹. On the other hand, animal data suggested that long and thin mineral fibers produced a higher incidence of mesothelioma than short and thick ones¹².

The objectives of this study are as follows: (1) to determine airborne fiber concentrations and size distributions of both

asbestos and nonasbestos fibers in and around the areas with serpentinite outcrops, (2) to identify the type of asbestos in both stone and soil samples collected in the area surveyed in this study.

Materials and Methods

Study area description

Figure 1 shows the location of the areas where serpentinite crops out in eastern area of Aichi Prefecture, Japan¹³. The area is surrounded by forested mountains on its three sides. There were no human-created sources such as the large-

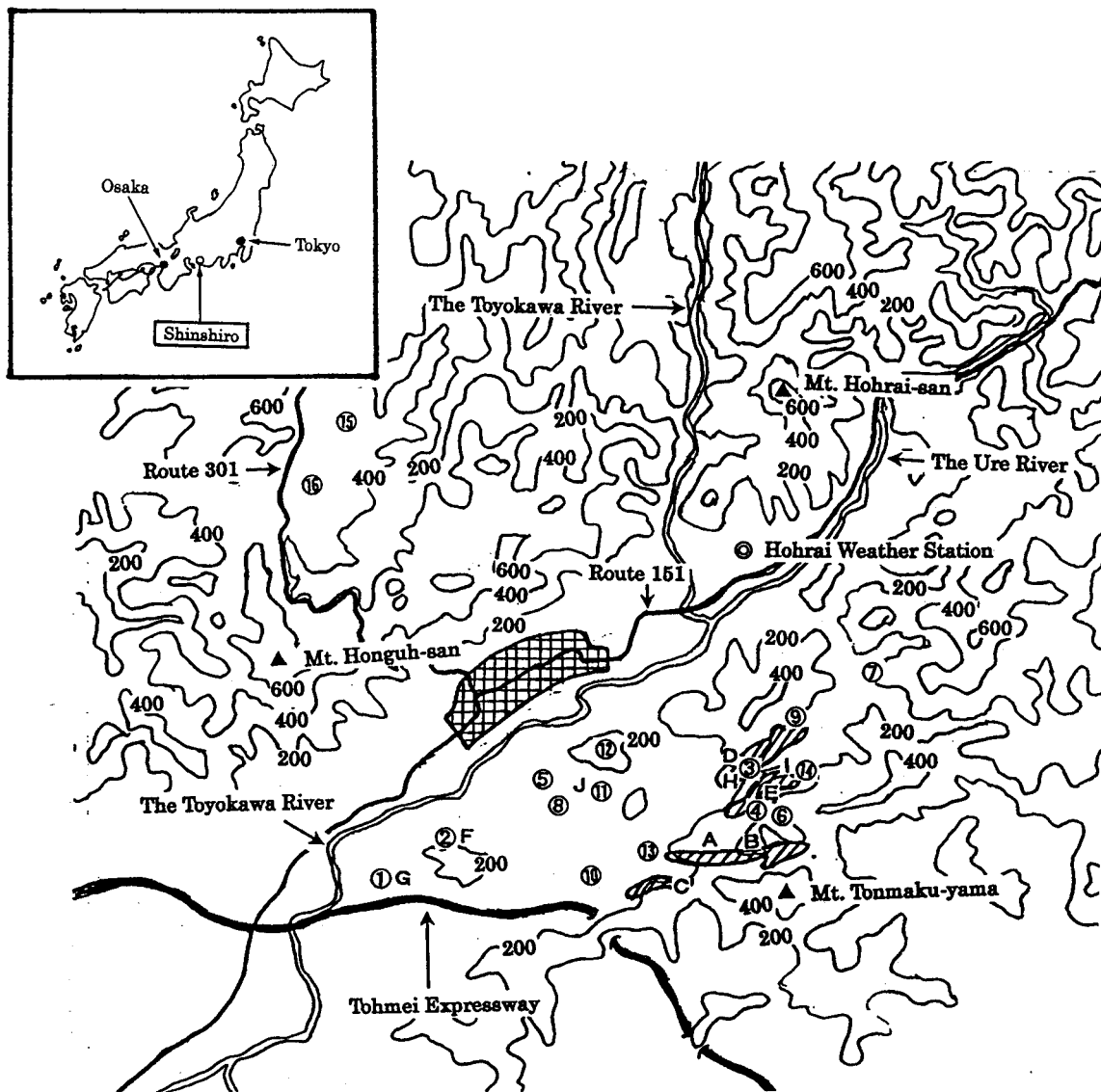


Fig. 1. Sampling sites of ambient airs and bulk samples in Shinshiro area.

~ : for ambient air, A~J: for bulk sample. —200—: Contour line in meter. ▨: Urban area of Shinshiro City. ▩: Serpentinite area (Inomata M, 1978).

scale manufacture and use of asbestos products within this area, but were some open-pit quarries producing crushed stone and areas sticking out the serpentinite ground due to road buildings and landslides. The downtown part of Shinshiro City (population: about 36,000) is located at a distance of about 5 km northwest of an area with serpentinite outcrops. The reference area was Tsukude Village located at a distance of about 20 km northwest of an area with serpentinite outcrops.

The weather and average (maximum) velocity of the wind at Hohrai weather station (elevation: 81 m, Fig. 1) located near an area with serpentinite outcrops were as follows; March 6, 1996: fine, 1.5 (4.0) m/s, April 22, 1996: fine, 1.8 (4.0) m/s, April 27, 1996: fine, 2.1 (5.0) m/s, May 23, 1996: fine, 2.0 (4.0) m/s.

Sampling and preparation

Airborne samples: Fig. 1 shows the sites surveyed. Table 1 gives a short description of each sampling site. These sites were classified as in a property boundary of serpentinite quarries producing crushed stone (quarry property boundary), an area within 10 km from an area with serpentinite outcrops (serpentinite area), and a reference area. In 4 open-pit serpentinite quarries, two quarries were in operation and others were closed. Each air sample was collected on membrane filter (Millipore AA, 47 mm in diameter) that was mounted in an open-faced filter holder. The filter holders

were positioned about 1 meter above the ground. The air sampling was done for 2 hours at a flowrate of 10 L/min. The direct transfer method¹⁴⁾ was employed to prepare samples for transmission electron microscopy.

Bulk samples: The sites where we collected the bulk samples are shown in Fig. 1. The stone samples were collected at 5 serpentinite quarries producing crushed stone, one cliff and one roadside. The soil samples were collected from the soil surface at two fields and beside a field. Stones and soil were crushed into powder with an agate mortar. The powders suspended in distilled water were filtered through Nuclepore® filters (pore size: 0.2 µm). These filters were transferred into TEM mesh samples by using chloroform to dissolve the filters.

Analytical methods

The prepared air samples were scanned at a magnification of 20,000 by means of analytical transmission electron microscopy (ATEM) (H-800, HITACHI). Only particles with approximately parallel sides and an aspect ratio (length/diameter) of more than 3 were considered to be fibers. For each sample, either a minimum of 100 fibers or 50 grid openings were counted. Asbestos was morphologically (e.g., the hollow tube structure for chrysotile) identified with the help of an energy-dispersive X-ray (EDX) analyzer (7000Q, KEVEX). The X-ray diffraction (XRD) method was used for identification of the minerals, especially for tremolite/

Table 1. Description of the different types of sampling sites of ambient air

Site No.	Type of area	Name of place	Location situation	Date
1	Quarry property boundary ^{a)}	Teruyama	Southern border of serpentinite quarry producing crushed stone (in operation)	96. 4.27
2	(in operation)	Yanai	Eastern border of serpentinite quarry producing crushed stone (in operation)	96. 4.27
3	Quarry property boundary ^{a)}	Yoshikawa	Southern border of serpentinite quarry producing crushed stone (in closed)	96. 3. 6
4	(in closed)	Tsugeno	Southern border of serpentinite quarry producing crushed stone (in closed)	96. 3. 6
5	Serpentinite area ^{b)}	Kuroda	Vicinity of intersection (motor traffic volume: very few)	96. 4.22
6		Tsugeno	Roadside of key street (about 50 m from site No. 4, moter traffic volume: few)	96. 3. 6
7		Shimoyosida	Parking lot of kindergarden (asphalt pavement)	96. 4.27
8		Kuroda	Roadside of farm road (about 80 m from site No. 5)	96. 4.22
9		Takenowa	Opening in a grove	96. 4.27
10		Nakauri	Precinct of a shrine	96. 5.23
11		Kuroda	Opening on the south of a assembly hall	96. 4.22
12		Kazekiriyama	The roadside of unpaved woodland path (moter traffic volume: none)	96. 4.22
13		Nakauri	Precinct of a shrine	96. 5.23
14		Tugeno	Vicinity of a intersection (moter traffic volume: very few)	96. 3. 6
15	Reference area ^{c)}	Tsukude	Shrine ground surrounding with trees	96. 5.23
16		Tsukude	Roadside in rice field	96. 5.23

a) Property boundary of a serpentinite quarry producing crushed stone. b) Area within 10 km of an area with serpentinite outcrops. c) Area far from more than 10 km of an area with serpentinite outcrops.

actinolite, in bulk samples. Tremolite and actinolite cannot be clearly distinguished by qualitative EDX analysis as they chemically resemble each other, and were combined into one group as tremolite/actinolite in calculating in this study. The type of nonasbestos fibers was not identified. The type of nonasbestos fibers was classified with the combination of components of which contents were more than 30%. The fiber which showed no elemental peaks or very small peaks was classified as a biological one¹⁵⁾. These fibers were not counted.

The detection limit was about 2 fibers/liter (f/L) when the sampled air volume was 1200 L. The length and diameter of each fiber were measured by ATEM at two magnifications, i.e. 2,000~20,000 and 5,000~50,000, respectively. The shortest fiber counted by ATEM was 0.2 μm in length. The mean number of fibers counted per sample was 61.1 (27~112) fibers.

Thirty fibers from each bulk sample were identified by ATEM. When at least one fiber of a given type was seen in a sample, the sample was classified as “positive”.

Statistical methods

The mean fiber concentrations were shown as geometric means. Where no fibers of a given type were seen, the concentration of the fiber type was set at one half the detection limit to allow geometric mean calculations. The means of fiber size dimensions (length, diameter, aspect ratio) of the airborne fibers are shown as geometric ones. The mean fiber size dimension by area is shown as the arithmetic one. Fiber concentrations and size dimensions of airborne samples

among the areas were compared by means of Kruskal-Wallis test.

Results

Types of mineral fibers in bulk samples

Table 2 lists the collecting sites and fiber types of both asbestos and nonasbestos fibers found in the bulk samples examined. Out of 7 stone samples, tremolite/actinolite was found in all samples, and chrysotile was found in 2 samples. Nonasbestos fibers were considered to be antigorite, chlorite, mica and others by their EDX spectra as listed in Table 2. Fiber mainly composed of S and Ca (fibrous calcium sulfates) was not found in any of the stone samples.

In all soil samples, tremolite/actinolite was found, whereas chrysotile was not found. Fibrous aluminum silicates were found in all the soil samples, whereas fibrous calcium sulfates were not found. Fibrous magnesium silicates were found in one sample. Amosite, crocidolite and anthophyllite were not found in any of the bulk samples.

Airborne fiber concentration of asbestos and nonasbestos fibers

Table 3 shows airborne asbestos concentrations at the quarry property boundaries, in the serpentinite areas and the reference areas. In 4 air samples obtained at the quarry property boundaries, tremolite/actinolite was found in all of the samples, whereas chrysotile was found in only 2 air samples. In 10 air samples obtained in the serpentinite areas, tremolite/actinolite was found in 7 air samples and

Table 2. Asbestos and nonasbestos fibers found from bulk stone and soil samples

Type of sample		Stone							Soil		
Site		A	B	C	D	E	F	G	H	I	J
Name of place		Fukutsu	Fukutsu	Kamewari	Yoshikawa	Tsugeno	Yanai	Teruyama	Yoshikawa	Tsugeno	Kuroda
Situation		Cliff with outcrop of serpentinite	unpaved road	serpentinite quarry	serpentinite quarry	serpentinite quarry	serpentinite quarry	serpentinite quarry	field	field	the side of field
Asbestos	Chrysotile	-	-	-	-	+	+	-	-	-	-
	Tremolite/actinolite	+	+	+	+	+	+	+	+	+	+
Nonasbestos fibers (Combination of main components)	Mg+Si	+	-	+	+	+	+	+	+	-	-
	Mg+Al+Si+Fe	-	+	-	-	-	-	-	-	-	-
	Al+Si+K+Fe	-	-	-	-	-	-	-	+	+	+
	Al+Si	-	-	-	+	+	-	-	+	+	+
	Ti	+	-	-	-	-	-	-	-	-	-

considered to be antigorite, considered to be chlorite, considered to be mica minerals, considered to be kaoline minerals, considered to be rutile.
+: detected, -: not detected.

chrysotile in 4 air samples. No asbestos was found in 2 air samples in the reference area. Asbestos such as amosite, crocidolite and anthophyllite were not found in any of the air samples.

The geometric mean of airborne fiber concentrations for asbestos longer than $0.2 \mu\text{m}$ was 384 f/L at the quarry property boundary (in operation), 12 f/L at the quarry property boundary (in closed), 5 f/L at the serpentinite area and less than 2 f/L at the reference area. There was a significant difference among the areas in airborne asbestos concentrations ($p=0.034$).

Table 3 shows fiber concentrations for nonasbestos fibers in the air samples. Fibrous aluminum silicate and fibrous calcium sulfate were found in all of the air samples. Fibrous magnesium silicate was found in all of the air samples taken at the quarry property boundary, but was found in only 4 out of 10 air samples obtained in the serpentinite areas and one of 2 air samples in the reference areas. The geometric

mean of airborne fiber concentrations for nonasbestos fibers was 447 f/L at the quarry property boundary (in operation), 124 f/L at the quarry property boundary (in closed), 103 f/L at the serpentinite area and 59 f/L at the reference area. There was a significant difference in airborne nonasbestos fiber concentrations among the areas ($p=0.036$). Although there was no significant difference in airborne concentrations of both fibrous magnesium silicate and fibrous aluminum silicate among the areas ($p=0.055$), the geometric means of fiber concentrations of these nonasbestos fibers were highest in the air samples obtained at the quarry property boundary (in operation).

The mean (range) percentage of asbestos in total mineral fibers was 47 (42–51) % at the quarry property boundary (in operation), 9 (8–10) % at the quarry property boundary (in closed), 9 (0–26) % in the serpentinite areas and 0% in the reference areas.

Table 3. Fiber concentrations of asbestos and nonasbestos fibers in ambient air

Site No	Type of area	Fiber concentration (fibers/liter)							
		Asbestos*			Nonasbestos fiber (combination of main components)*				
		Total	Chrysotile	TR/AC ¹⁾	Total	Al + Si	S + Ca	Mg + Si	Others
1	Quarry property boundary ^{a)}	740	<13	740	714	390	52	143	130
2	(in operation)	199	<5	199	280	104	19	57	100
	Geometric mean	384	<9	384	447	202	31	90	114
3	Quarry property boundary ^{a)}	14	2	12	153	33	45	17	59
4	(in closed)	11	8	2	101	21	54	5	21
	Geometric mean	12	4	5	124	26	49	9	35
5	Serpentinite area ^{b)}	33	2	30	96	40	23	<2	33
6		16	2	15	97	19	19	2	58
7		10	<3	10	78	31	13	3	31
8		5	<3	5	207	24	32	5	145
9		5	2	2	136	52	19	<2	66
10		5	2	2	92	19	40	<2	33
11		4	<2	4	60	9	40	<2	11
12		<2	<2	<2	106	17	64	2	24
13		<2	<2	<2	167	40	42	<2	85
14		<2	<2	<2	68	7	28	<2	33
	Geometric mean	5	2	4	103	22	29	2	41
15	Reference area ^{c)}	<2	<2	<2	64	17	9	7	31
16		<2	<2	<2	55	18	22	<2	16
		Geometric mean	<2	<2	<2	59	17	14	3
	p-value (Kruskal-Wallis test)	0.034	ND ²⁾	0.051	0.036	0.10	0.18	0.055	0.13

*: All fibers longer than $0.2 \mu\text{m}$. 1) Tremolite and actinolite (amosite, crocidolite and anthophyllite were not found). 2) Not done, because the detection limits in quarries (in operation) were far from those in other sites. a) Property boundary of a serpentinite quarry producing crushed stone. b) Area within 10 km of an area with serpentinite outcrops. c) Area far from more than 10 km of an area with serpentinite outcrops.

Airborne fiber size distribution of asbestos and nonasbestos fibers

Table 4 summarizes the geometric means of length, diameter and aspect ratio of asbestos in each area. There was no significant difference among the areas in the arithmetic means of asbestos size distributions.

Table 5 summarizes the geometric means of length, diameter and aspect ratio of nonasbestos fibers in each sample at the quarry property boundary, in the serpentinite area and in the reference area. There was no significant difference among the areas in fiber size distributions of nonasbestos fibers.

The arithmetic mean length of asbestos in all of the samples (2.8 μm) was significantly longer than that of nonasbestos fibers (1.2 μm) ($p < 0.01$). The aspect ratio of asbestos in all air samples (9.5) was significantly greater than that of nonasbestos fibers (5.7) ($p < 0.01$). The arithmetic mean length of chrysotile in all air samples (1.2 μm) was significantly shorter than that of tremolite/actinolite (3.2 μm) ($p < 0.01$). Consequently, the arithmetic mean diameter of chrysotile

in all air samples (0.07 μm) was significantly smaller than that of tremolite/actinolite (0.46 μm) ($p < 0.01$).

Fibers with a high probability of producing mesothelioma in animal experiments (length $> 8 \mu\text{m}$ and diameter $\leq 0.25 \mu\text{m}$)¹²⁾ were not found in any of the air samples. The percentage of fibers producing a relatively higher incidence of mesothelioma (length $> 8 \mu\text{m}$ and $0.25 \mu\text{m} < \text{diameter} \leq 1.5 \mu\text{m}$, or length $> 4 \mu\text{m}$ and diameter $\leq 1.5 \mu\text{m}$)¹²⁾ to total mineral fibers was 23 (15~30) % at the quarry property boundary (in operation), 5 (4~5) % at the quarry property boundary (in closed), 10 (3~17) % in the serpentinite area and 15 (14~15) % in the reference area.

Discussion

Asbestos and nonasbestos fibers in bulk samples

Serpentine minerals are principal constituents of serpentinite. Serpentine minerals are chrysotile, antigorite and lizardite. Chrysotile, antigorite and lizardite all have

Table 4. Fiber size dimension of asbestos in ambient air

Site No	Type of area	Total asbestos			Chrysotile			Tremolite/actinolite		
		Length (μm)	Diameter (μm)	Aspect ratio	Length (μm)	Diameter (μm)	Aspect ratio	Length (μm)	Diameter (μm)	Aspect ratio
1	Quarry property boundary ^{a)}	3.1 (2.3)	0.43 (2.3)	7.2 (1.5)	—	—	—	3.1 (2.3)	0.43 (2.3)	7.2 (1.5)
2	(in operation)	2.6 (2.4)	0.35 (2.3)	7.3 (1.7)	—	—	—	2.6 (2.4)	0.35 (2.3)	7.3 (1.7)
	Arithmetic mean	2.82	0.389	7.3	—	—	—	2.82	0.389	7.3
3	Quarry property boundary ^{a)}	1.7 (2.1)	0.21 (2.7)	8.4 (1.9)	0.8	0.04	20.0	2.0 (1.1)	0.29 (1.9)	7.0 (1.7)
4	(in closed)	1.8 (2.1)	0.083 (2.6)	21.8 (1.8)	1.3 (1.6)	0.051 (1.2)	26 (1.6)	1.8 (2.0)	0.45 (1.2)	11.8 (1.5)
	Arithmetic mean	1.77	0.146	15.1	1.06	0.046	23.0	1.92	0.368	9.4
5	Serpentinite area ^{b)}	3.6 (2.5)	0.51 (2.9)	7.0 (1.6)	0.8	0.05	16.0	4.0 (2.4)	0.60 (2.4)	6.6 (1.5)
6		5.7 (2.1)	0.62 (2.4)	9.3 (1.5)	2.7	0.18	15.0	6.1 (2.1)	0.68 (2.3)	8.9 (1.5)
7		3.9 (1.3)	0.62 (1.2)	6.3 (1.3)	—	—	—	3.9 (1.3)	0.62 (1.2)	6.3 (1.3)
8		3.8 (1.3)	0.42 (1.3)	9.1 (1.6)	—	—	—	3.8 (1.3)	0.42 (1.3)	9.1 (1.6)
9		1.3 (2.3)	0.13 (5.1)	10.0 (2.2)	0.7	0.04	17.5	2.3	0.40	5.8
10		1.6 (3.1)	0.16 (6.8)	10.1 (2.2)	0.7	0.04	17.5	3.5	0.60	5.8
11		2.3 (1.8)	0.27 (1.8)	8.5 (1.0)	—	—	—	2.3 (1.8)	0.27 (1.8)	8.5 (1.0)
12		—	—	—	—	—	—	—	—	—
13	—	—	—	—	—	—	—	—	—	
14	—	—	—	—	—	—	—	—	—	
	Arithmetic mean	3.16	0.387	8.6	1.23	0.078	16.5	3.69	0.512	7.3
15	Reference area ^{c)}	—	—	—	—	—	—	—	—	—
16		—	—	—	—	—	—	—	—	—
	Arithmetic mean	—	—	—	—	—	—	—	—	—
	Total (arithmetic mean)	2.84	0.343	9.5	1.17	0.067	18.7	3.21	0.464	7.7

Geometric mean (geometric standard deviation). —: A fiber was not found. a) Property boundary of a serpentinite quarry producing crushed stone. b) Area within 10 km of an area with serpentinite outcrops. c) Area far from more than 10 km of an area with serpentinite outcrops.

similar chemical compositions of magnesium silicate. Chrysotile has a fibrous form of a hollow tube and lizardite is in irregular plate-like crystal form, so that fibrous magnesium silicate is considered to be antigonite. The stone samples collected in this study were of a yellowish green color, and were confirmed to be serpentinite by XRD analysis. Tremolite/actinolite was identified in the stone samples by XRD, and their coexistence with chrysotile was identified by the analytical electron microscopic study.

The soil samples collected in this study contained tremolite/actinolite. Weathered serpentinites was an important source for asbestos-containing soils²⁾. The emission of asbestos from crushed serpentinite is a possible source of soil pollution by asbestos near serpentinite quarries.

Airborne asbestos concentration

Tremolite/actinolite fibers were predominant in ambient

air at the quarry property boundary in this study, whereas the majority of asbestos in the atmosphere was chrysotile^{6, 16, 17)}. It has been suggested that the emission of asbestos from crushed serpentinite is a source of air pollution by asbestos near serpentinite quarries in Japan⁹⁾. Fiber concentrations of tremolite/actinolite were higher than those of chrysotile in most air samples obtained at both the quarry property boundary and the serpentinite area in this study. Fibrous tremolite/actinolite was identified in the stone samples of the serpentinite quarries and the serpentinite area in this study. It was considered that the erosion of serpentinite was responsible for most of the asbestos in the serpentinite areas.

The geometric mean (range) concentrations of airborne asbestos in residential and agricultural areas determined by ATEM in Japan were 19.8 (<4~111) f/L and 21.8 (7~47) f/L, respectively⁶⁾. The range of airborne asbestos concentrations in Tokyo was 3.58~94.7 f/L¹⁸⁾. We previously

Table 5. Fiber size dimension of nonasbestos fibers in ambient air

Site No	Type of area	Nonasbestos fiber (combination of main components)												
		Total			Al + Si			S + Ca			Mg + Si			
		Length (μm)	Diameter (μm)	Aspect ratio	Length (μm)	Diameter (μm)	Aspect ratio	Length (μm)	Diameter (μm)	Aspect ratio	Length (μm)	Diameter (μm)	Aspect ratio	
1	Quarry property boundary ^{a)} (in operation)	1.7 (2.7)	0.23 (3.0)	7.3 (1.6)	1.9 (2.5)	0.25 (3.0)	7.6 (1.7)	0.49 (1.5)	0.077 (1.2)	6.5 (1.5)	2.1 (2.4)	0.27 (2.8)	8.0 (1.7)	
2		1.1 (2.7)	0.18 (2.5)	6.4 (1.6)	1.4 (2.7)	0.18 (3.2)	7.4 (1.6)	0.76 (2.0)	0.15 (2.1)	5.3 (1.1)	2.4 (1.9)	0.29 (1.9)	8.3 (1.4)	
	Arithmetic mean	1.40	0.203	6.9	1.61	0.215	7.5	0.63	0.111	5.9	2.28	0.279	8.1	
3	Quarry property boundary ^{a)} (in closed)	0.87 (2.2)	0.16 (2.0)	5.4 (1.6)	1.5 (2.9)	0.25 (2.9)	6.0 (1.4)	0.71 (2.0)	0.14 (1.9)	5.2 (1.4)	1.0 (1.9)	0.091 (1.6)	11.1 (1.7)	
4		0.90 (2.2)	0.15 (2.0)	6.0 (1.7)	0.94 (2.1)	0.091 (2.2)	10.4 (2.0)	0.91 (2.2)	0.17 (1.9)	5.2 (1.4)	1.3 (2.2)	0.21 (1.7)	6.0 (1.6)	
	Arithmetic mean	0.89	0.156	5.7	1.21	0.168	8.2	0.81	0.156	5.2	1.15	0.153	8.6	
5	Serpentinite area ^{b)}	1.2 (2.3)	0.23 (2.2)	5.4 (1.6)	1.9 (1.9)	0.28 (2.3)	6.8 (1.5)	1.1 (2.0)	0.25 (1.7)	4.4 (1.4)	—	—	—	
6		0.99 (2.4)	0.18 (2.0)	5.4 (1.7)	1.7 (2.8)	0.22 (3.1)	7.7 (1.7)	1.1 (2.2)	0.20 (1.9)	5.5 (1.5)	0.3	0.06	5.0	
7		1.1 (2.0)	0.20 (2.0)	5.6 (1.4)	1.4 (1.9)	0.22 (2.0)	6.4 (1.5)	1.8 (1.7)	0.30 (1.6)	6.1 (1.4)	0.9	0.25	3.6	
8		1.6 (2.2)	0.35 (2.1)	4.5 (1.5)	2.8 (2.9)	0.39 (3.7)	7.0 (1.7)	2.2 (1.9)	0.52 (1.7)	4.2 (1.2)	3.2 (3.3)	0.38 (3.0)	8.5 (2.8)	
9		0.95 (2.9)	0.16 (2.7)	6.1 (1.5)	1.5 (3.5)	0.19 (3.7)	7.9 (1.4)	2.1 (2.7)	0.32 (2.3)	6.6 (1.6)	—	—	—	
10		1.4 (2.5)	0.21 (2.3)	6.5 (1.6)	1.6 (2.8)	0.19 (3.9)	8.5 (1.6)	0.95 (1.8)	0.18 (1.9)	5.3 (1.2)	—	—	—	
11		1.5 (2.1)	0.30 (2.1)	5.2 (1.4)	1.9 (1.7)	0.40 (1.7)	4.9 (1.3)	1.4 (1.8)	0.27 (1.8)	5.2 (1.3)	—	—	—	
12		1.5 (2.3)	0.26 (2.2)	5.7 (1.5)	1.4 (3.2)	0.17 (3.6)	8.4 (1.6)	1.8 (2.2)	0.33 (1.9)	5.6 (1.4)	2.7	0.35	7.7	
13		1.3 (2.5)	0.22 (2.4)	5.9 (1.6)	2.0 (2.8)	0.26 (3.4)	7.9 (1.4)	1.1 (2.1)	0.17 (2.0)	6.6 (1.6)	—	—	—	
14		0.89 (2.0)	0.16 (2.0)	5.6 (1.5)	1.0 (2.5)	0.13 (3.9)	7.8 (1.9)	0.90 (1.7)	0.13 (1.5)	6.7 (1.5)	—	—	—	
		Arithmetic mean	1.25	0.226	5.6	1.72	0.244	7.3	1.45	0.267	5.6	1.79	0.260	6.2
15		Reference area ^{c)}	1.1 (2.9)	0.20 (2.7)	5.3 (1.4)	1.1 (3.1)	0.17 (3.3)	6.8 (1.3)	1.6 (3.2)	0.27 (2.4)	5.8 (1.5)	1.9 (3.2)	0.32 (2.2)	6.0 (1.7)
16			1.4 (2.6)	0.26 (2.6)	5.4 (5.4)	1.7 (2.2)	0.28 (2.9)	6.0 (1.7)	1.6 (2.5)	0.29 (2.4)	5.6 (1.3)	—	—	—
		Arithmetic mean	1.22	0.228	5.4	1.41	0.224	6.4	1.61	0.280	5.7	—	—	—
	Total (arithmetic mean)	1.22	0.215	5.7	1.57	0.212	7.4	1.17	0.214	5.5	1.77	0.223	7.9	

Geometric mean (geometric standard deviation). —: A fiber was not found. a) Property boundary of a serpentinite quarry producing crushed stone. b) Area within 10 km of an area with serpentinite outcrops. c) Area far from more than 10 km of an area with serpentinite outcrops.

reported that the mean (range) concentration of airborne asbestos in the outside of 21 buildings in Nagoya, Japan, was 16.2 (1.4~127) f/L¹⁹). The airborne asbestos concentrations in this study, except the quarry property boundary, were within the range of those in previous reports.

Asbestos was found in ambient air at a rock quarry which had produced crushed stone containing asbestos²⁰). Although the airborne asbestos concentrations in the vicinity of the serpentinite quarry producing crushed stone were relatively higher than those in the surrounding area, the asbestos concentrations rapidly decreased to background levels (20~30 f/L) at a distance of about 1 km from the serpentinite quarry⁹). Although the airborne asbestos concentrations at the quarry property boundary were much higher than those in the serpentinite area in this study, the airborne asbestos concentrations in the serpentinite area were similar to those in urban areas. Our observation was not compatible with previous reports.

The geometric mean concentration of airborne asbestos at the quarry property boundary was higher than that in the serpentinite area. This indicates that airborne asbestos concentrations inside the serpentinite quarries producing crushed stone would be very high and that the quarry workers also would be greatly exposed to asbestos.

Airborne nonasbestos fiber concentrations

There is no detailed study of airborne nonasbestos fiber concentration near serpentinite quarries producing crushed stone. The airborne concentrations of both fibrous magnesium silicate and fibrous aluminum silicate at a quarry property boundary (in operation) were not significantly different from those in other areas, but their geometric means were more than 7 times higher at the quarry property boundary (in operation) higher than in other areas. Fibrous magnesium silicate was a minority of nonasbestos fibers in urban atmospheres, whereas fibrous aluminum silicate was prevalent in urban atmospheres¹⁹). On the other hand, the geometric mean of fibrous calcium sulfate concentrations at the quarry property boundary (in operation) was similar to those in other areas. Calcium sulfates have also been frequently found in urban atmospheres^{18, 21, 22}). Fibrous magnesium silicate and aluminum silicate were found in the stone samples, but fibrous calcium sulfate was not found in any bulk samples collected in this study. These results suggest that serpentinite is one of the emission sources of both fibrous magnesium silicate and aluminum silicate as well as asbestos.

We previously reported that the mean (range) concentration of airborne nonasbestos fiber in the outside of 21 buildings

in Nagoya, Japan, was 332 (43.2~1716) f/L¹⁹). The airborne nonasbestos fiber concentrations at the sites investigated in this study were within those in the urban areas.

Fiber size distributions of asbestos and nonasbestos fibers

The mineral fibers in the atmospheres might be affected by natural crushing that breaks fiber bundles into thin fibers and/or shorter fibers. Consequently, it was supposed that the size of airborne asbestos fibers in the vicinity of emission sources is different from that in distant areas, but we could not find a significant difference between the quarry property boundary (in operation) and other areas in size distributions of airborne fibers in both asbestos and nonasbestos fibers.

We previously reported that the most potent mineral fibers in producing mesothelioma found by Stanton *et al.*¹²) was 0.13% in total mineral fibers in urban atmospheres¹⁹). In this study we could not find any mineral fibers of this size. The airborne concentration of mineral fibers of this size may be negligible even in the vicinity of natural sources of asbestos. The airborne size distributions of mineral fibers in this study were not very different from those in the urban area¹⁹).

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References

- 1) Yoshida K (1993) Outlines of mineral products business, revised and enlarged edition, 490–508, Thusyou-sangyou-tyousakai, Tokyo (in Japanese).
- 2) Burilkov T, Michailova L (1970) Asbestos content of the soil and endemic pleural asbestosis. *Environ Res* **3**, 443–51.
- 3) Rohl AN, Langer AM, Selikoff IJ (1977) Environmental asbestos pollution related to use of quarried serpentine rock. *Science* **196**, 1319–22.
- 4) Cooper WC, Murchio J, Popendorf W, Wenk HR (1979) Chrysotile asbestos in a California recreational area. *Science* **206**, 285–8.
- 5) Douglas V, Gomez J, Negrete L (1991) Control measure for asbestos-containing serpentine rock in surfacing applications. *Proc AWMA Ann Meet (Air Waste Manage Assoc)* 84th (15A) 91.135.2, 1–9.
- 6) Kohyama N (1989) Airborne asbestos levels in nonoccupational environments in Japan, In: *Nonoccupational exposure to mineral fibres (IARC Sci*

- Pub No. 90). eds. by Bignon J, Peto R, Saracci R, 262–76, IARC, Lyon.
- 7) Laamanen A, Noro L, Raunio V (1965) Observations on atmospheric air pollution caused by asbestos. *Ann NY Acad Sci* **132**, 240–5.
 - 8) Singh B, Thouez JP (1985) Ambient air concentrations of asbestos fibers near the town of Asbestos, Quebec. *Environ Res* **36**, 144–59.
 - 9) Kohyama N, Hiraoka T, Kurimori S (1995) Environmental asbestos pollution by an old asbestos mine and factory in Kumamoto Prefecture, seen from the asbestos concentration in the lung tissues of the residents. *J Occup Health* **37** supplement, S515 (in Japanese).
 - 10) Kohyama N, Hiraoka T, Kurimori S (1997) Environmental asbestos pollution by an old asbestos mine and factory in Kumamoto Prefecture, seen from the asbestos concentration in the lung tissues of the residents (2). *J Occup Health* **39** supplement, S470 (in Japanese).
 - 11) Suta BE, Levine RJ (1970) Non-occupational asbestos emissions and exposures, In: *Asbestos. Properties, applications, and hazards* Vol. 1, eds. by Michaels L, Chissick SS, 171–205, A Wiley-Interscience Publication, Chichester.
 - 12) Stanton MF, Layard M, Tegeris A, Miller E, May M, Morgan M, Smith A (1981) Relation of particle dimension to carcinogenicity in amphibole asbestoses and other fibrous minerals. *J Natl Cancer Inst* **67**, 965–75.
 - 13) Inomata M (1978) Geology of the Mikabu Belt and ultrabasic complexes in Mt.Ufu-san and Mt.Tonmakyama area to the north of Lake Hamana, Central Japan. *Chikyu Kagaku (Earth Science)* **32**, 336–44 (in Japanese, partly in English).
 - 14) Ortiz LW, Isom BL (1974) Transfer technique for electron microscopy of membrane filter samples. *Am Ind Hyg Assoc J* **35**, 423–5.
 - 15) Kamens R, Lee C, Wiener R, Leith D (1991) A study to characterize indoor particles in three non-smoking homes. *Atmo Environ* **25A**, 939–48.
 - 16) Selikoff IJ, Nicholson AM, Langer AM (1972) Asbestos air pollution. *Arch Environ Health* **25**, 1–13.
 - 17) Sakai K, Hisanaga N, Mitani K, Tsuchiya H, Haung J, Shibata E, Ono Y, Tanabe E, Takeuchi Y (1990) Airborne asbestos concentrations in rooms with sprayed-on asbestos materials. *Jpn J Pub Health* **37**, 109–17 (in Japanese, partly in English).
 - 18) Watanabe T, Kamataki H, Akiyama K, Asakuno K, Adachi H, Yoshino N (1994) Asbestos concentration in Tokyo. *Annual Report of the Tokyo Metropolitan Research Institute for Environmental Protection 1993*, 7–13 (in Japanese, partly in English).
 - 19) Sakai K, Kohyama N, Hisanaga N, Yamanaka K, Takeuchi Y (1996) Airborne fiber concentrations and fiber size dimensions in buildings determined by analytical transmission electron microscopy. *Indoor Air* '96 **2**, 973–8.
 - 20) Carter LJ (1977) Asbestos: trouble in the air from a Maryland rock quarry. *Science* **197**, 237–40.
 - 21) Middleton AP (1978) On the occurrence of fibres of calcium sulphate resembling amphibole asbestos in samples taken for evaluation of airborne asbestos. *Ann Occup Hyg* **21**, 91–3.
 - 22) Spurny KR, Stober W, Opiela H, Weiss G (1979) On the evaluation of fibrous particles in remote ambient air. *Sci Total Environ* **11**, 1–40.