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ORIGINAL PAPER

Asbestos in the ambient air from rural, urban, residential, baseball and mining areas in South Korea

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Abstract

Asbestos is a naturally occurring fibrous silicate that has been widely used as electrical insulator and heat-resistant material in buildings, yet inhalation of asbestos fibers can lead to serious lung diseases such as asbestosis and cancer. Practically no research has been conducted on the size distribution and morphological characteristics of airborne asbestos, and airborne asbestos concentrations in South Korea are unknown. Here we studied type, concentration, size, morphology and composition of asbestos fibers in the ambient air of several regions in South Korea. Asbestos concentrations were analyzed in 7 urban areas, 7 rural areas including agricultural and fishing areas, 17 mines and their surrounding areas, 7 residential areas constructed with asbestos-containing stones near rivers, 2 baseball fields and 2 background sites. Results show that the highest air asbestos concentrations were 0.00161 for residential areas and 0.00122 for baseball fields according to phase-contrast microscopy, and 0.00057 for asbestos mines and 0.00055 for baseball fields, according to transmission electron microscopy. Asbestos types included chrysotile, tremolite, and actinolite. Chrysotile fibers measured 5.24–35.5 µm in length with aspect ratios of 12.6–202.6; tremolite fibers measured 6.07–40.2 µm in length with aspect ratios of 5.7–81.2; and actinolite fibers measured 5.01–28.5 µm in length with aspect ratios of 3.2–108.9. Chrysotile was distributed in bundles or single fibers, whereas tremolite and actinolite exhibited fibrous, acicular, and cleavage forms.

Keywords Asbestos exposure · Asbestos characterization · Ambient air · Transmission electron microscopy

Introduction

In regions with naturally occurring asbestos, asbestos fibers, particulate matter, and heavy metals can be dispersed in the air both naturally by weathering processes in rocks and soils containing asbestos and artificially by human activities, with substantial potential environmental impacts (Bayram and Bakan 2014; Brião et al. 2020; Das et al. 2020; de Gennaro et al. 2014; Hendrickx 2009; Lee et al. 2008; Mukherjee and Agrawal 2017, 2018). Artificial asbestos dispersal occurs due to asbestos mining and processing in mines and surrounding areas (Anastasiadou and Gidarakos 2007; Koumantakis et al. 2009; Meeker et al. 2003; Ryan et al. 2015). Mined rocks and soils are widely used in the construction of streets, parks, riverbanks and school grounds. Thus, asbestos exposure can be a hazard for local residents, as well as mine workers (Hansen et al. 1993; Reid et al. 2007; Jung et al. 2020). Moreover, asbestos can be introduced into the air by the natural weathering of as asbestos-containing roofing materials in rural areas (Buczaj et al. 2014; Pastuszka 2009; Spurny 1989; Tadas et al. 2011), as well as during the dismantling and demolishing of buildings containing asbestos (Campopiano et al. 2004; Chesson et al. 1990; Jung et al. 2015, 2021; Kangur 2007; Lee and Van Orden 2008). Other potential exposure settings include waste asbestos processing facilities (Gidarakos et al. 2008) and asbestos handling factories (Chang et al. 1999; Mensi et al. 2015).

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In South Korea, asbestos exposure via the ambient air has recently emerged as a social issue in several potential source settings Precision surveys in closed asbestos mines and surrounding areas have detected tremolite, actinolite, and chrysotile asbestos in soils (Korea Ministry of the Environment 2012, 2013, 2014). Furthermore, the Ministry of the Environment has raised concerns over potential airborne asbestos exposure from demolition of asbestos-containing buildings (Korea Ministry of the Environment 2009, 2010a, 2015), asbestos-containing landscape stones near urban rivers where residents enjoy leisure activities (Korea Ministry of the Environment 2010b), and at baseball grounds, where asbestos-containing crushed serpentine soils are used (Korea Ministry of the Environment 2011c).

Recently, the incidence of pulmonary disease due to environmental asbestos exposure has emerged as a social issue in Korea, highlighting the need not only for research into asbestos occurrence and concentrations in the air, but also for studies related to asbestos exposure since the ban on asbestos use (Choi et al. 2013; Jung et al. 2012, 2020; Kim 2009; Park et al. 2009). South Korea implemented the Asbestos Injury Relief Act in 2011, which provides national compensation to individuals with asbestos lung diseases and their families affected by environmental asbestos exposure, as well as to residents living near asbestos mines and factories (Lippmann 1988). Diseases related to asbestos exposure are predominantly influenced by the asbestos dose, morphological characteristics of the fibers, including their lengths and diameters, and the durability or persistence of fibers in the lungs (Markowitz 2015). Experimentally derived asbestos distribution data typically suggest that longer and thinner fibers may have greater carcinogenic potency than shorter and wider fibers (Korea Ministry of the Environment 2011aMarkowitz 2015; Stanton et al. 1981; World Health Organization 1989). Stanton et al. (1981) reported that long (> 8 µm) and thin (< 0.25 µm) mineral fibers were extremely carcinogenic and induced the development of pleural mesothelioma in rats. Moreover, long, thin fibers have been observed to be positively and strongly associated with an increase in lung cancer incidence (Lippmann 2014; Loomis et al. 2010; Stayner et al. 2008). In an epidemiological and exposure-evaluation study including patients grouped by environmental exposure, Stayner et al. (2008) demonstrated a strong association between lung cancer and long, thin fibers (length > 10 µm, diameter < 0.25 µm) than with short (< 5 µm) or thick (> 3.0 µm) fibers. In addition, the main factors affecting the asbestos burden of lung cancer patients, who have been living in a city in Korea with 22 asbestos textile factories in the past, were asbestos exposure (environmental/occupational), gender, and old age. Notably, there was a significant difference in the length (4.26–91.7 µm vs. 4.06–37.6 µm) and aspect ratio (5.6–735.6 vs. 4.5–151.9) of asbestos fibers detected in the lung tissue between the environmentally exposed lung cancer patient and occupationally exposed lung cancer patient (Jung et al. 2020).

Research into airborne asbestos concentrations has been actively conducted elsewhere in the world. In 1989, the World Health Organization announced airborne asbestos concentrations of 0.001–0.0001 f/cc (World Health Organization 1989). Considering that a person inhales 14,400 L of air per day, this suggests a daily inhalation of 1,440–14,400 asbestos fibers. Fibers per cubic centimeter (f/cc) of air is a measurement that determines the permissible levels of asbestos exposure in the workplace and environment. The European Directive of the EC 2009/148 and the Occupational Safety and Health Administration (OSHA) have established acceptable airborne levels for the work environment. According to the European Directive of the EC 2009/148 and OSHA, permissible exposure limits for all types of asbestos are 0.1 f/cc, based on an 8-h weighted average (TWA) (EU 2009; OSHA 2014).

In contrast with other efforts worldwide (Axten and Foster 2008; Chiappino et al. 1993; Corn 1994; Howitt et al. 1993; Jaffrey 1990; Lajoie et al. 2003; Sakai et al. 2001) research on airborne asbestos concentrations in South Korea is relatively lacking. In 2004, the geometric mean concentration of asbestos was reported for urban and agricultural/fishing areas (Lim et al. 2004). In 2011, Kwon et al. (2013) determined the airborne asbestos concentrations in two serpentine quarries and one steelworks, and for individual workers. In 1996, Yu and Kim (1996) reported on airborne fiber concentrations in the ambient air of 14 large buildings in South Korea. Furthermore, the geometric mean concentrations of airborne asbestos in two mining areas in Chungcheongnam-do were reported by Lee et al. (2015), and the airborne asbestos concentration range of one mining area in Chungcheongbuk-do was reported by Shin et al. (2011). Both domestic and international studies, however, have been limited to measuring airborne asbestos concentrations in specific regions only. Furthermore, almost no research has been conducted on the size distribution and morphological characteristics of the airborne asbestos.

Therefore, this study evaluated the characteristics (types, concentrations, and morphological features such as lengths and widths) of asbestos fibers dispersed in the air across multiple regions of potential asbestos exposure, including urban areas, agricultural and fishing areas, asbestos mines, and residential environments. The findings of this study provide essential data for evaluating airborne asbestos exposure in South Korea, including for enforcing the Asbestos Injury Relief Act.

Experimental

Survey sites and air sampling

We selected 42 asbestos survey sites that were categorized into urban areas, rural areas, mining areas, and residential environments. The locations and photographs of the selected survey regions are shown in Fig. 1. For urban areas, we selected 7 sites: Seoul, Busan, Incheon, Daegu, Ulsan, Gwangju, and Daejeon. For rural areas, we also selected 7 sites: Gangwon-do, Gyeongsangbuk-do, Chungcheongbuk-do, Chungcheongnam-do, Jeollabuk-do, Jeollanam-do, and Gyeonggi-do. For mining areas, we selected 14 asbestos mines and other mines that potentially contain asbestos, as well as three serpentine mines and their surrounding areas. For residential environments, we selected 7 artificial rivers constructed using asbestos-containing landscape stones, 2 baseball fields, and 2 background sites that did not have potential exposure components.

To survey the airborne asbestos in urban and rural areas, air quality monitoring stations were used. Airborne asbestos concentrations were measured at 4 stations in each of the 7 cities and at a total of 20 stations in the 7 rural areas. These air quality monitoring stations are generally installed in public use facilities (e.g., community service centers, elementary and middle schools) away from roads, and measurements were performed at representative locations of these public use facilities. In addition, the selected background sites were locations where the influence of artificial air pollutants is minimal and were chosen to examine the inflow and outflow of pollutants from/to other countries, as well as long-term pollutant transport. The airborne asbestos concentrations were measured at the 2 background sites for 2 days.

The airborne asbestos surveys in the mine areas were conducted in 17 asbestos mines and the surrounding villages. At each of the representative mine sites and the surrounding villages, which are located away from roads, 10–15 samples were collected and measured. For the surveys of river areas where landscape stones have been used, 1–3 samples were collected for three days and measured according to the length of the landscape stone distributions at locations where many residents enjoy walking and other activities. For the surveys of baseball fields, a total of 34 samples were collected over two days at the first and third base dugouts, at the front of the scoreboard located behind the catcher, at first and third bases, and in the spectator seats behind the catcher at two representative baseball fields, and in the outfield spectator seats at one baseball field. The sampling was performed during the day (12:00 to 16:00) on sunny days without rain for three days.

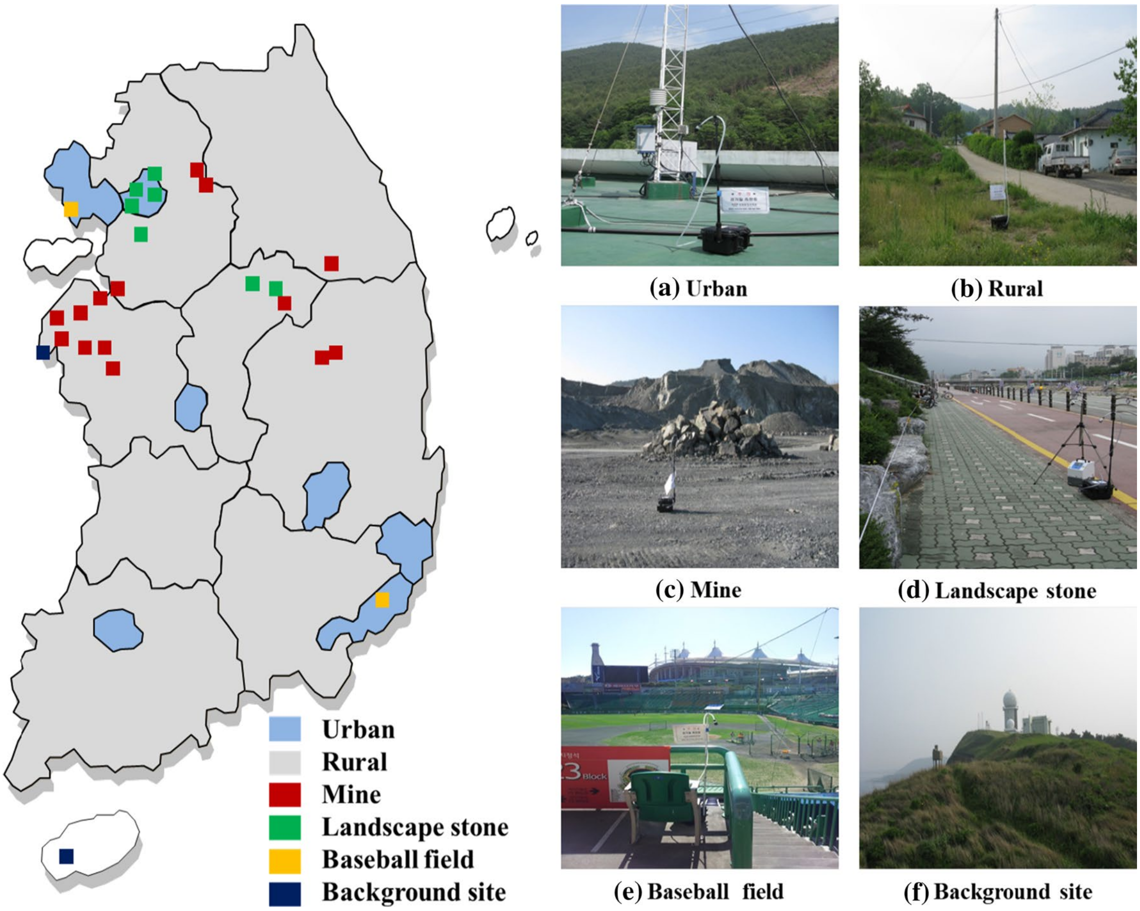

Fig. 1 Map of South Korea and photographs showing the locations and sampling environments of the selected survey sites

For airborne samples, we sampled a total of 2,400 L of air using mixed cellulose ester (MCE) filter cassettes (25 mm diameter, 0.8 μm pore size) for 4 h, at a flow rate of approximately 10 L/min, 1.5 m above the ground using a high-flow pump, according to the air pollution process test standard (Korea Ministry of the Environment 2007).

Sample preparation and analysis

Phase-contrast microscopy

After vitrifying 1/4 of the filter using acetone, 2–3 drops of triacetin were added to the center of the filter to fix it to the slide. Asbestos fibers were counted in accordance with the indoor air quality process testing standard (Korea Ministry of the Environment 2010c) at 100 counting fields of view. All fibrous shapes satisfying the counting criteria (length > 5 μm, aspect ratio > 3) were counted using phase-contrast microscopy (PCM) (BX51TF, OLYMPUS, Tokyo, Japan).

Transmission electron microscopy

The filter containing the captured sample was vitrified with a vitrification solution (35% dimethyl formamide, 15% acetic acid, and 50% distilled water). Then, approximately 10% of the filter surface was etched using a plasma etcher (EMITECH K1050X, Quorum, Lewes, UK) before coating the filter with carbon in a sample coating machine (EMITECH K950X Turbo Evaporator, Quorum, Lewes, UK). The filter was then melted using a Jaffe washer containing acetone solution and dried to produce a grid for analysis. The operation parameters for transmission electron microscopy (TEM) included an acceleration voltage of 100 kV or higher. Then, asbestos fibers were counted and analyzed according to the phase-contrast microscopy equivalent (PCME) counting criteria (length > 5 μm, aspect ratio > 3) at 10,000 or higher magnifications, in accordance with ISO 10312 (International Standard Organization 1995). Furthermore, the asbestos crystal structure was analyzed using selected area electron diffraction (SAED). Additional qualitative evaluation included analysis of the chemical components using energy-dispersive X-ray spectrometry (EDS, EDAX, Mahwah, USA) and calculation of the asbestos concentrations.

Statistical analyses

The characteristics of the collected airborne asbestos and fibrous dust samples (concentrations, types, lengths, widths, aspect ratios) were analyzed using SPSS version 12.0. The airborne asbestos concentrations are presented here using the arithmetic mean, arithmetic standard deviation, geometric mean, geometric standard deviation, and minimum and maximum values. If no asbestos was detected during the TEM analysis (denoted by N.D.), half of the analyzed sensitivity was calculated and applied as the asbestos concentration for the statistical analysis. The Kolmogorov–Smirnov test was used to determine the normality of the data, which did not satisfy a normal distribution. Thus, we verified intergroup statistical differences by performing non-parametric tests, i.e., the Kruskal–Wallis (K–W) test and Mann–Whitney (M-W) test with the Bonferroni correction post-test.

Results and discussion

Airborne asbestos and fibrous dust concentrations in South Korea

We studied the characteristics, such as the type, concentration, size, morphology, and composition, of asbestos fibers detected in the ambient air at multiple potential asbestos exposure regions in South Korea. The concentrations of asbestos and fibrous dust at each survey site were determined by phase-contrast microscopy (PCM) and transmission electron microscopy (TEM) (Table S1 in Supplementary Materials). Asbestos was detected in the air at the survey sites in 20 out of 227 samples (0.0009–0.0305 f-TEM/cc) at mines and the surrounding areas, 1 out of 39 samples (0.0009 f-TEM/cc) at rivers using landscape stones, 3 out of 34 samples (0.0009–0.0074 f-TEM/cc) at baseball fields, and 1 out of 28 samples (0.0009 f-TEM/cc) in urban areas (Table S1 in Supplementary Materials).

The geometric mean (GM) concentration of airborne asbestos and fibrous dust for all surveyed regions in South Korea was 0.00045 f-PCM/cc and 0.00054 f-TEM/cc, according to PCM and TEM analysis, respectively. The PCM concentrations of airborne fibrous dust in all survey sites were significantly lower than 0.01 f/cc, which is the asbestos standard value for indoor air quality in public facilities (Korea Ministry of the Environment 2010c) and airborne clearance level of asbestos according to the Asbestos in Schools Rule (EPA 1987). However, the TEM concentration of airborne asbestos in some mines exceeded the standard value. The PCM GM concentration was highest at rivers built using landscape stones, followed by baseball fields, rural areas, background sites, mines, and urban areas. Statistical differences in concentrations were observed among the survey sites ($P_{\text{Kruskal–Wallis}} < 0.0001$). Conversely, the TEM GM concentration of airborne asbestos was highest at the mines and surrounding areas, followed by baseball fields, rivers built using landscape stones, urban areas, rural areas, and background sites ($P_{\text{Kruskal–Wallis}} < 0.0001$). As asbestos mines are located in naturally occurring areas that possibly contain asbestos in rocks and soil, it is highly likely that asbestos will be dispersed into the atmosphere due to natural and anthropogenic influences such as agricultural activities. Consequently, more asbestos fibers were detected and high asbestos concentration was confirmed compared to other regions.

In other parts of the world, reported PCM concentrations in urban areas were in the range 0.0001–0.0034 f-PCM/cc (Damiani et al. 2006; Whysner et al. 1994; Kakooei et al. 2013, 2009). Reported scanning electron microscopy (SEM) concentrations were in the range 0.0001–0.1 f-SEM/cc (Damiani et al. 2006; Kakooei et al. 2009, 2013). The reported TEM concentration in urban Italy was 0.0002 f-TEM/cc (Chiappino et al. 1993). In rural areas, airborne asbestos concentration was recorded as 0–0.00065 f-PCM/cc (Corn 1994) and 0.0001–0.014 f-TEM/cc (Axten and Foster 2008). For natural asbestos regions, reported concentrations were <0.000869–0.00197 s-TEM/cc and 0.0001–0.7987 s-TEM/cc (Environmental Protection Agency 2005, 2011). The corresponding values based on activated based sampling (ABS) were 0.0004–0.0336 s-TEM/cc and 0.0012–2.4832 s-TEM/cc. Studies at asbestos mines reported concentrations in the range 0.08–0.18 f-PCM/cc (Gidarakos et al. 2008) and 0.005 f-TEM/cc (Lajoie 2003). Although there are limitations in directly comparing airborne asbestos concentrations between South Korea and other countries due to differences in the total air sampling flow and analytical sensitivity, all concentration ranges determined in this study are similar to or less than those previously reported in other countries.

Types of asbestos detected in airborne samples

Furthermore, the TEM analyses detected chrysotile, tremolite, and actinolite types of asbestos, all of which were detected at the mine sites. Ten out of the 20 samples in which asbestos was detected were collected from operational serpentine mines, with the other 10 collected from asbestos mines and the surrounding areas. This indicates the effect of both mining and naturally occurring geological asbestos. Chrysotile and actinolite asbestos were also detected in rocks in Korean serpentine quarries by Kwon et al. (2013), and all three types were detected in asbestos mines in Chungcheongnam-do (Song et al. 2008).

The air sample with the highest asbestos concentration was collected inside an operational asbestos mine, indicating the effect of mine processing operations after pulverizing the serpentine. Asbestos was detected in only one sample from the urban survey sites; however, there were no asbestos dismantling or removal sites in the surrounding areas, suggesting a negligible effect from the surrounding areas. At the baseball fields, both chrysotile and actinolite were detected, which were likely introduced from the ground that contained less than 1% asbestos. At the areas near rivers constructed using landscape stones, tremolite asbestos was detected in the stones, suggesting introduction into the air in trace amounts.

Characterization of airborne asbestos

Chrysotile asbestos in the air was detected in both bundles and single-fiber forms. Tremolite and actinolite asbestos detected in the air satisfied the length >5 μm and aspect ratio $>3{:}1$ counting criteria, and exhibited fibrous, acicular, prismatic, and cleavage forms (Fig. 2). The main components of chrysotile were Mg and Si, as well as trace amounts of Fe. The main components of tremolite and actinolite were Mg, Si, Ca, and Fe. Additional details are provided as Fig. 2 and Figure S1 in Supplementary Materials.

Morphologies of airborne asbestos fibers

Among the 66 detected asbestos fibers in 25 asbestos samples that had lengths higher than μm, 28 were chrysotile, 9 were tremolite, and 29 were actinolite (Table S2 in Supplementary Materials). Chrysotile exhibited a larger aspect ratio than tremolite and actinolite, indicating longer and thinner fibers (Fig. 3; Table S2 in Supplementary Materials). The aspect ratios of all detected chrysotile samples exceeded 20:1, according to the PCME counting criteria. However, only 44.4% of tremolite samples and 31.0% of actinolite samples had aspect ratios greater than 20:1. Thus, the aspect ratios of the amphibole asbestos types (tremolite and actinolite) dispersed in the air are smaller than those of standard asbestiform fibers, from a mineralogical perspective. According to the US EPA, an aspect ratio above 20:1 corresponds to commercial asbestos that occurs artificially in building materials (Environmental Protection Agency 1982) and is not relevant for asbestos that is naturally generated and dispersed in the air. Therefore, considering the possibility of asbestos dispersal in the air and associated health effects, it is reasonable to apply an aspect ratio of 3:1 or higher, i.e., the current counting criteria for asbestos in air. The widths and aspect ratios of the 66 asbestos fibers are shown in Fig. 4 and Table S2 in Supplementary Materials, which indicate that chrysotile fibers were much thinner than the tremolite and actinolite fibers.

Comparison of asbestos chemical composition with the United Kingdom Health and Safety Executive reference sample

The energy-dispersive X-ray spectroscopy (EDS) analytical results of 28 chrysotile fibers detected in the air in South Korea showed proportions of Mg and Si of 53.5% and 43.7%, respectively, with trace amounts of Fe (2.8%). In contrast, the chemical composition of chrysotile according

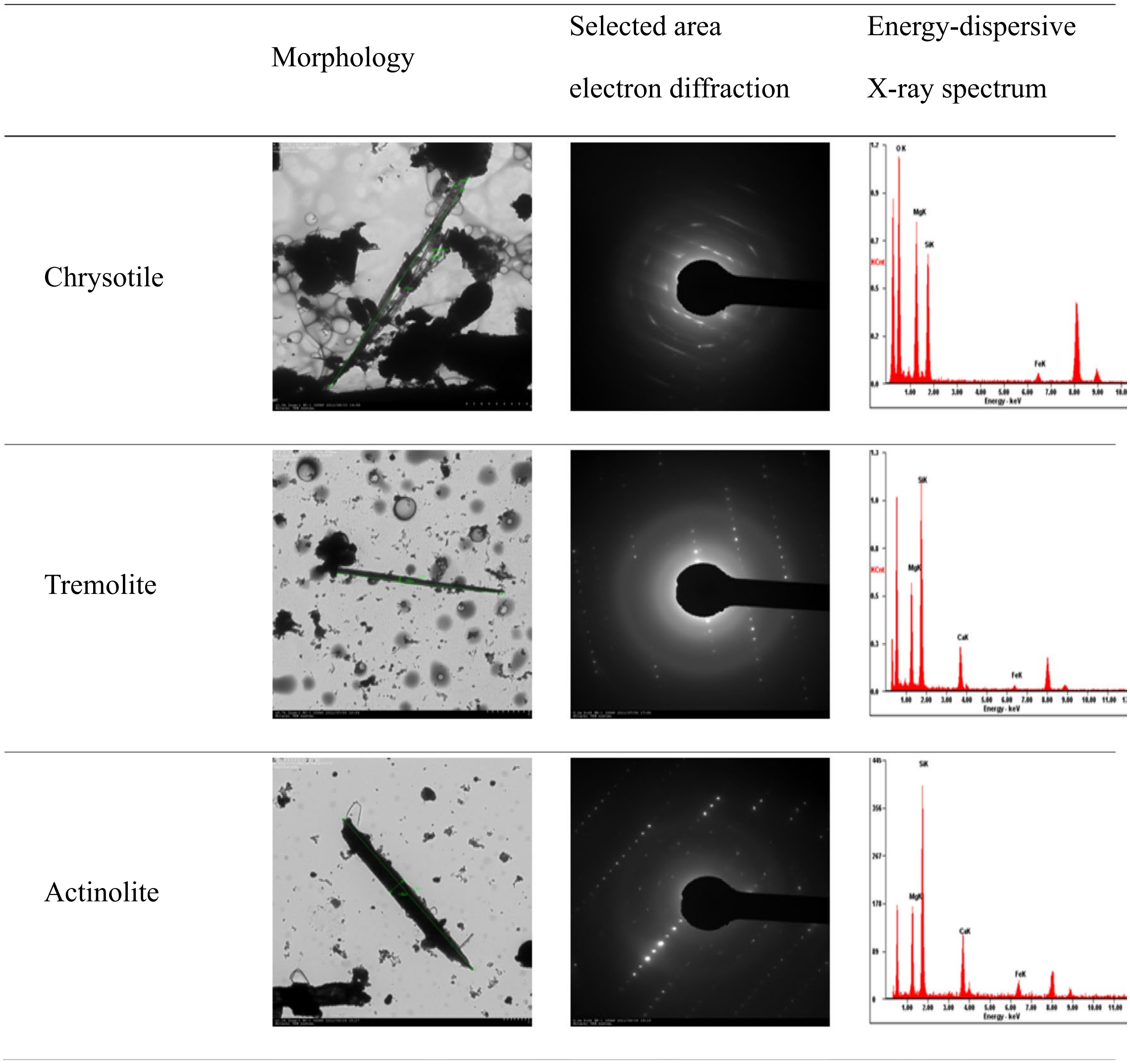

Fig. 2 Morphologies, selected area electron diffraction, and chemical compositions, obtained by the energy-dispersive X-ray spectroscopy, of asbestos detected in the ambient air. Chrysotile asbestos in the air was detected in both bundles and single-fiber forms. Tremolite and actinolite asbestos exhibited fibrous, acicular, prismatic, and cleavage forms. The main components of chrysotile were Mg, Si, and Fe, and of tremolite and actinolite were Mg, Si, Ca, and Fe

to the United Kingdom Health and Safety Executive (HSE) standard specifies proportions of Mg, Si, and Fe of 56.7%, 41.5%, and 1.7% (Table S3 in Supplementary Materials), respectively. Therefore, the asbestos detected in this study contained approximately 3% less Mg and approximately 2% more Si. When compared with 10 Si, the chrysotile detected in this study contained 12.4 ± 1.6% Mg and 0.6 ± 0.2% Fe, whereas the HSE standard contains 13.7 ± 0.7% and 0.4 ± 0.0%, respectively (Table S3 in Supplementary Materials).

The proportions of Mg, Si, Ca, and Fe in the detected tremolite fibers were 31.5%, 55.3%, 12.3%, and 0.9%, respectively, compared to 31.5%, 55.6%, 11.7%, and 1.2% in the HSE standard tremolite sample (Table S3 in Supplementary Materials). Furthermore, when compared with 10 Si, no differences were found between the chemical composition of the airborne tremolite asbestos detected in South Korea and the HSE standard sample. Similarly, no differences were found between the chemical composition of actinolite asbestos and the standard sample (Table S3 in Supplementary Materials).

Conclusion

This study analyzed airborne asbestos and fibrous dust concentrations and characteristics of potential asbestos exposure regions in South Korea, including urban, rural, mining, and residential areas. The geometric mean concentration of asbestos was highest in mines and surrounding areas

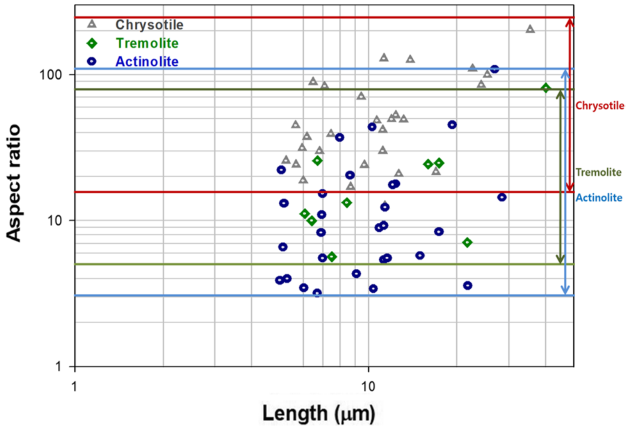

Fig. 3 Lengths and aspect ratios of the detected asbestos types. Chrysotile fibers exhibited a larger aspect ratio than tremolite and actinolite, indicating longer and thinner fibers

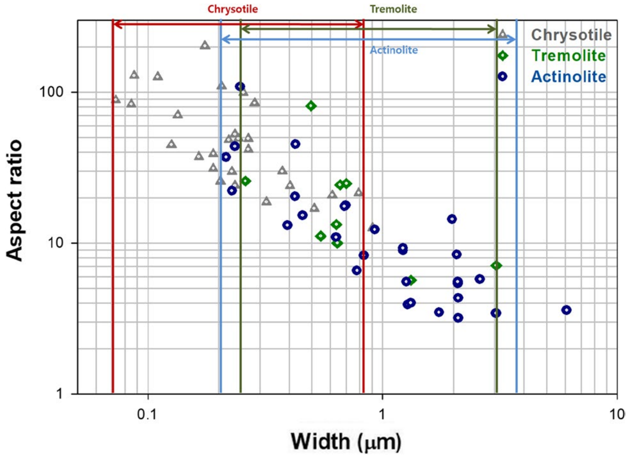

Fig. 4 Widths and aspect ratios of the detected asbestos types. Chrysotile fibers were much thinner than the tremolite and actinolite fibers

(0.00032 f-PCM/cc and 0.00057 f-TEM/cc), followed by baseball fields (0.00122 f-PCM/cc and 0.00055 f-TEM/cc), landscaped stones (0.00161 f-PCM/cc and 0.00046 f-TEM/cc), urban areas (0.00032 f-PCM/cc and 0.00046 f-TEM/cc), rural areas (0.00056 f-PCM/cc and 0.00045 f-TEM/cc), and background sites (0.00034 f-PCM/cc and 0.00045 f-TEM/cc). The types of asbestos detected in the air included chrysotile (serpentine), tremolite, and actinolite (amphiboles). Chrysotile exhibited bundled or single-fiber forms, whereas tremolite and actinolite exhibited fibrous, acicular, and prismatic forms. The aspect ratios of all detected chrysotile fibers were higher than 20:1, compared to 44.4% and 31.0% of the tremolite and actinolite fibers, respectively.

The present results are significant because they reveal the characteristics (types, concentrations, lengths, widths, aspect ratios, and compositions) of airborne asbestos in multiple potential asbestos exposure regions in South Korea. These data can be used in the evaluation of airborne asbestos exposure and health risks for the public. Moreover, the results provide a scientific basis in representing regional background airborne asbestos concentrations in the enforcement of the 2011 Asbestos Injury Relief Act in South Korea. Further studies on asbestos background concentrations and characteristics are required particularly for asbestos fibers with lengths of 0.5–5 μm. Moreover, further studies should also attempt determining asbestos concentrations in other locations which have the potential to introduce asbestos into the air, such as building demolition sites and waste asbestos treatment facilities.

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Author's contribution Hyun-Sung Jung, Hyunwook Kim were involved in conceptualization and methodology; Hyun-Sung Jung, Jinyoung Jang, Yangseok Cho contributed to formal analysis and investigation; Hyun-Sung Jung was involved in writing – original draft preparation; Hyun-Sung Jung, Jinyoung Jang, Jong-Chun Lee, Hyunwook Kim contributed to writing—review and editing; Jong-Chun Lee was involved in funding acquisition; Jong-Chun Lee, Hyunwook Kim contributed to supervision.

Declarations

Competing interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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