Workers’ Exposure to Indium Compounds at the Electronics Industry in Republic of Korea

Gwangyong Yi 1,7*, Jeeyeon Jeong 2, Yasung Bae 3, Jungah Shin 4, Hyelan Ma 5, Naroo Lee 1, Seung-Hyun Park 1, Dooyong Park 6,7

1 Occupational Safety & Health Research Institute, KOSHA, Ulsan, Republic of Korea
2 Yong In University, Youngin, Republic of Korea
3 Hsekorea Corporation, Incheon, Republic of Korea
4 Institute of Occupation and Environment, KCOMWEL, Incheon, Republic of Korea
5 Korea Industrial Hygiene Association, Seoul, Republic of Korea
6 Korea Occupational Safety & Health Agency, Ulsan, Republic of Korea
7 Department of Mechanical Systems Engineering, Hansung University, Seoul, Republic of Korea

Abstract

Objectives: The aim of this study was to provide baseline data for the assessment of exposure to indium and to prevent adverse health effects among workers engaged in the electronics and related industries in Republic of Korea.

Methods: Total (n = 369) and respirable (n = 384) indium concentrations were monitored using personal air sampling in workers at the following 19 workplaces: six sputtering target manufacturing companies, four manufacturing companies of panel displays, two companies engaged in cleaning of sputtering components, two companies dedicated to the cleaning of sputtering target, and five indium recycling companies.

Results: The level of exposure to total indium ranged from 0.9 to 609.3 μg/m³ for the sputtering target companies; from 0.2 to 2,782.0 μg/m³ for the panel display companies and from 0.5 to 2,089.9 μg/m³ for the indium recycling companies. The level of exposure to respirable indium was in the range of 0.02 to 448.6 μg/m³ for the sputtering target companies; 0.01 to 419.5 μg/m³ for the panel display companies; and 0.5 to 436.3 μg/m³ for the indium recycling companies. The indium recycling companies had the most samples exceeding the exposure standard for indium, followed by sputtering target companies and panel display companies.

Conclusions: The main finding from this exposure assessment is that many workers who handle indium compounds in the electronics industry are exposed to indium levels that exceed the exposure standards for indium. Hence, it is necessary to continuously monitor the indium exposure of this workforce and take measures to reduce its exposure levels.

1. Introduction

In 2001, a Japanese worker died of bilateral pneumothorax complicated with interstitial pneumonia, which was attributed to the worker’s exposure to indium. Since then, the adverse health effects of indium exposure have attracted substantial attention [1–10]. Indium and its compounds enter the body via inhalation and are deposited in the lungs. They are slowly eliminated from the body. Chronic exposure to indium is known to be associated with adverse health effects in humans. The solubility of indium compounds determines their serum concentrations [1,11]. Republic of Korea is one of the heavy indium-consuming countries due to the expansion of industries manufacturing panel screens using liquid-crystal displays, light-emitting diodes, and organic light-emitting diodes, as well as computers, tablets, and mobile phones embedding touch screens and touch panels. Republic of Korea has recently focused on expansion of its copper indium gallium selenide solar cell production capacity [12]. This trend...
suggests an increase in potential exposure to indium among workers engaged in indium smelting, indium tin oxide (ITO) and indium zinc oxide (IZO) sputtering target manufacture, panel display, and indium recycling industries [13,14]. However, the studies focused on indium exposure and the prevention of indium-induced health effects among workers in indium and indium compound handling industries have been limited. The relatively rapid emergence of the industry over a short period of time and the restricted access to information in indium-handling industries such as display manufacture might have contributed to the number of workers with adverse health effects [11]. Based on pulmonary diseases studied in animals, the American Conference of Governmental Industrial Hygienists set the threshold limit values of indium and indium compounds at 0.1 mg/m³ for total indium [15]. However, the permissible exposure limits set by the Occupational Safety and Health Administration in the USA were not defined [2]. Countries other than the USA established the occupational exposure limit (OEL) of workers to indium or indium compounds as 0.1 mg/m³ [16]. In Japan, cases of pulmonary diseases among workers handling indium or indium compounds were reported for the first time [7], and the OEL of respirable indium of 0.01 mg/m³ was based on the guidelines from the Ministry of Health, Labour and Welfare introduced in 2010 [11].

This study aims to provide exposure data for workers engaged in ITO and IZO sputtering target manufacture, panel display, and indium recycling industries in Republic of Korea.

2. Methods

A preliminary survey of ITO and IZO sputter target manufacturing, panel display [maintenance of thin film transistor (TFT) and color filter (CF) sputtering equipment, replacement of sputtering targets, cleaning of sputtering component, and cleaning of sputtering target], and indium recycling industries was conducted during 2011. The worker exposure to indium was monitored in the following year. The total and respirable indium concentrations were monitored using personal air sampling in workers at the following 19 workplaces: six ITO and IZO sputtering target manufacturing companies, four manufacturing companies of panel displays, two companies engaged in cleaning of sputtering components, two companies dedicated to the cleaning of sputtering target, and five indium recycling companies. The exposure data were analyzed using SPSS 20 (SPSS, Chicago, IL, USA) and Sigma plot 11 Systat software, San Jose, CA, USA. A threshold limit value of 0.1 mg/m³ recommended by the American Conference of Governmental Industrial Hygienists [15] was used to assess total indium exposure. For respirable indium, an OEL of 0.01 mg/m³ set by the Japanese Ministry of Health, Labour and Welfare was applied [11].

2.1. Air sampling and analytical methods

Total indium was sampled using the National Institute of Occupational Safety and Health Method ID-121 [17,18]. A mixed cellulose ester membrane filter (diameter: 37 mm, pore size: 0.8 µm, Millipore, USA) was used to sample indium dust generated during the indium and indium compound handling processes. Respirable indium was collected using the National Institute of Occupational Safety and Health Method 500 [17] and a mixed cellulose ester membrane filter.

Total and respirable indium particles were analyzed using inductively coupled plasma spectrometry (Vista Pro, Varian, Australia) and inductively coupled plasma mass spectrometry (810MS, Varian, Australia).

3. Results

3.1. Statistical analysis

Total and respirable concentrations of the indium and indium compound industries were represented as arithmetic mean (AM) and standard deviation, geometric mean (GM) and geometric standard deviation, range (min, max), and excess percentage of samples exceeding indium exposure standard, as indicated in Tables 1 and 2. Total indium concentrations of the ITO and IZO sputtering target companies, the display companies, and the indium recycling companies showed log-normal distributions (Fig. 1). According to the results of analysis of variance using logarithmic transformed measurement data, statistically significant differences were found in total and respirable indium levels at all companies (p < 0.05).

3.2. Sputtering target manufacturing industry

The level of exposure to total indium (n = 182) was in the range of 0.9 to 609.3 µg/m³, and the values of GM and AM of the level of exposure were 33.3 µg/m³ and 73.3 µg/m³, respectively. The level of exposure to respirable indium by workers (n = 192) was in the range of 0.02 to 448.6 µg/m³, and the values of GM and AM were 5.2 µg/m³ and 14.3 µg/m³, respectively. A total of 23.1% (n = 42) of total indium samples exceeded the standard and 33.0% (n = 64) of respirable indium samples exceeded the exposure standard (Tables 1 and 2).

During the powdering process, the indium metal (99.99% or purer) is dissolved in nitric acid, which is supplemented with ammonia and deionized water. The indium nitrate [In(NO₃)₃] that is dissolved in nitric acid solution is converted into and precipitated as indium hydroxide [In(OH)₃] using ultrapure ammonia. The In(OH)₃ precipitate is subjected to washing, drying, and calcination until indium oxide (In₂O₃) powder is obtained. In this process, workers are at a potential risk of exposure to In(OH)₃ and In₂O₃. Workers’ exposure to total indium (n = 20) during the powdering process was estimated to range from 2.7 to 366.9 µg/m³, whereas exposure to respirable indium (n = 23) was estimated to range from 0.0 to 98.2 µg/m³. The percentage of excessive exposure to total indium was 15.0% (n = 3), whereas that of respirable indium was 39.1% (n = 9).

During the mixing process, In₂O₃ and tin oxide (SnO₂) or zinc oxide (ZnO) were mixed approximately in the ratio of 9:1. Water and a binder were added to the mixture to generate slurry, followed by milling and spray drying in a dryer at 400°C. The mixing process yielded In₂O₃ nanoparticles and SnO₂ or ZnO particles. Workers are at a potential risk of exposure to In₂O₃ and SnO₂ or ZnO. Workers’ exposure to total indium (n = 13) during the mixing process was estimated to range from 13.1 to 502.1 µg/m³, whereas exposure to respirable indium (n = 16) ranged from an estimated 1.1 to 448.6 µg/m³. The percentage of excessive exposure to total indium was 30.8% (n = 4), whereas that of respirable indium was 50.0% (n = 8).

During the sintering process, the mixed powder is press molded into a body to fit ITO and IZO sputtering target specifications (i.e., dimensions and thickness) and compacted with water pressure. Workers are potentially exposed to In₂O₃, SnO₂, or ZnO powder during the sintering process. The concentrations of total indium (n = 15) and respirable indium (n = 19) ranged from 1.4 to 376.0 µg/m³ and 1.0 to 74.9 µg/m³, respectively. The percentage of excessive exposure to total indium was 40.0% (n = 6), whereas that of respirable indium was 26.3% (n = 5).

During the sintering process, the resulting compact is sintered in a furnace at high temperature (approximately 1,500–1,600°C) for one week under a constant supply of liquid oxygen to form a dense ITO and IZO ceramic target. Workers’ exposure to indium
Total indium concentrations of the indium and indium compound industries

| Industry/process                      | n  | AM         | SD | GM          | GSD | Range | Excess | p    |
|---------------------------------------|----|------------|----|-------------|-----|-------|--------|------|
| ITO and IZO sputter target companies  |    | Total      |    |             |     |       |        |      |
| Powder                                | 20 | 61.1       | 84.3 | 32.3        | 3.2 | 2.7–366.9 | 15.0  | 0.0000 |
| Mixing                                | 13 | 113.0      | 144.0 | 58.6        | 3.3 | 13.1–502.1 | 30.8  |      |
| Molding                               | 15 | 107.0      | 111.0 | 45.8        | 5.8 | 1.4–376.0 | 40.0  |      |
| Sintering                             | 21 | 19.4       | 21.0  | 12.9        | 2.4 | 4.5–92.4  | 0     |      |
| Polishing                             | 45 | 107.0      | 123.0 | 56.0        | 3.7 | 3.1–609.3 | 37.8  |      |
| Bonding                               | 39 | 31.9       | 45.6  | 18.5        | 2.8 | 0.9–239.4 | 2.6   |      |
| Finishing                             | 29 | 87.9       | 101.0 | 43.3        | 3.9 | 4.0–438.2 | 37.9  |      |
| Total                                 | 182| 73.3       | 99.7  | 33.3        | 3.8 | 0.9–609.3 | 23.1  |      |
| Panel display companies                |    | Total      |    |             |     |       |        |      |
| Target replacement and maintenance    | 57 | 13.1       | 29.7  | 4.4         | 4.3 | 0.2–206.8 | 1.8   | 0.0001 |
| Sputter parts cleaning                | 46 | 195.0      | 500.0 | 47.3        | 4.7 | 3.5–2782.0 | 26.0  |      |
| Target cleaning                       | 16 | 29.1       | 56.7  | 6.7         | 8.7 | 0.2–235.0 | 6.3   |      |
| Total                                 | 119| 85.6       | 322.3 | 11.7        | 7.0 | 0.2–2782.0 | 11.7  |      |
| Indium recycling companies             |    | Total      |    |             |     |       |        |      |
| Pulverization                         | 11 | 901.5      | 776.1 | 583.6       | 2.9 | 81.0–2089.9 | 90.0  | 0.0000 |
| Dissolution                           | 4  | 153.8      | 118.6 | 105.7       | 3.3 | 19.7–308.2 | 75.0  |      |
| Substitution                          | 24 | 94.0       | 205.4 | 34.0        | 3.6 | 5.9–1003.5 | 25.0  |      |
| Electrolysis                          | 13 | 124.6      | 187.3 | 32.4        | 8.1 | 0.5–643.4 | 38.5  |      |
| Casting                               | 16 | 59.3       | 97.4  | 28.4        | 3.1 | 5.6–343.9 | 12.5  |      |
| Total                                 | 68 | 225.8      | 451.1 | 54.7        | 5.7 | 0.5–2089.9 | 38.3  |      |

* Arithmetic mean.
† Standard deviation.
‡ Geometric mean.
§ Geometric standard deviation.
The percentage of samples exceeding indium exposure standard (10 μg/m³).

During the polishing process, the sintered ITO and IZO target is fabricated to meet the product specifications including size, shape, and thickness followed by polishing of the surfaces. Indium particulates are mechanically generated during the polishing phase, which exposes workers to ITO and IZO particulates. Workers’ exposure to total indium (n = 45) during this process was estimated to range from 3.1 to 609.3 μg/m³ and respirable indium (n = 54) was distributed in the range 0.2 to 61.1 μg/m³. A total of 37.8% (n = 17) of samples from total indium exposure and 40.7% (n = 22) of samples from respirable indium exposure were found to be above the respective exposure limits.

During the bonding process, indium metal bonding is used to affix sputtering target to the backing plate. Theoretically, the likelihood of workers’ exposure to indium is very low as indium metal melts at 200°C before bonding. However, incomplete washing of the fabricated indium target during the prior stage of polishing can lead to exposure to ITO and IZO dust. Workers’ exposure to total indium (n = 39) ranged from 0.9 to 239.4 μg/m³ and respirable indium (n = 36) ranged from 0.8 to 11.9 μg/m³. The percentage of excessive exposure to total indium was 2.6% (n = 1), whereas that of respirable indium was 13.8% (n = 5).

During the finishing process, the ITO and IZO sputtering target surface and the bonding surface are polished and smoothed before the delivery of the finished product to display manufacturers. Workers are exposed to ITO and IZO particulates, as well as indium metal dusts, from many different sources. Workers’ exposure ranged from 4.0 to 438.2 μg/m³ to total indium (n = 29) and from 0.2 to 68.6 μg/m³ to respirable indium particles (n = 24). The percentage of excessive exposure to total indium was 37.9% (n = 11), whereas that of respirable indium was 45.5% (n = 11).

3.3. Panel display industry

The level of exposure to total indium (n = 119) was in the range of 0.2 to 2,782.0 μg/m³ (GM = 11.7 μg/m³, AM = 85.6 μg/m³) and
respirable indium by workers \((n = 121)\) was in the range of 0.01 to 419.5 \(\mu g/m^3\) (GM = 2.9 \(\mu g/m^3\), AM = 13.6 \(\mu g/m^3\)). The rate of excessive exposure to total indium was 11.7\%(n = 14), whereas that of respirable indium was 23.9\%(n = 29) (Tables 1 and 2).

The workers engaged in maintenance of TFT and CF sputtering equipment and replacement of sputtering target in companies manufacturing panel displays are exposed to particulates of ITO or IZO while they separate the sputtering components and exchange and clean the sputtering target. The concentration level of total indium \((n = 57)\) and respirable indium \((n = 59)\) was estimated to range from 0.2 to 206.8 \(\mu g/m^3\) (GM = 4.4 \(\mu g/m^3\), AM = 13.1 \(\mu g/m^3\)) and 0.01 to 34.2 \(\mu g/m^3\) (GM = 1.2 \(\mu g/m^3\), AM = 4.7 \(\mu g/m^3\)), respectively. The percentage of excessive exposure to total indium was 1.8\%(n = 1), whereas that of respirable indium was 13.5\%(n = 8).

Sputtering components (shield, mask, and carrier), which are delivered to external cleaning company, are distinguished into components of TFT and CF, and workers are exposed to ITO or IZO particulates in the course of manual disassembly of each component. Disassembled components are then classified according to respective characteristics to clean the contaminants using nitric acid or via aluminum oxide blasting. The concentration level of total indium \((n = 46)\) and respirable indium \((n = 46)\) was estimated to range from 3.5 to 2,782.0 \(\mu g/m^3\) (GM = 47.3 \(\mu g/m^3\), AM = 195.0 \(\mu g/m^3\)) and 1.1 to 419.4 \(\mu g/m^3\) (GM = 8.6 \(\mu g/m^3\), AM = 23.7 \(\mu g/m^3\)), respectively. A total of 26.0\%(n = 12) of samples from total indium exposure and 36.9\%(n = 17) of samples from respirable indium exposure exceeded the respective standards.

At companies engaged in cleaning of ITO and IZO sputtering targets, the workers were exposed to ITO and IZO particulates during the course of aluminum oxide blasting to remove contaminants on the surfaces of sputtering target primarily and during the course of grinding surfaces of sputtering target to remove the remaining contaminants secondarily. Workers’ exposure to total indium \((n = 16)\) ranged from 0.2 to 235.0 \(\mu g/m^3\) (GM = 6.7 \(\mu g/m^3\), AM = 29.1 \(\mu g/m^3\)) and respirable indium \((n = 16)\) ranged from 0.1 to 113.8 \(\mu g/m^3\) (GM = 2.3 \(\mu g/m^3\), AM = 12.5 \(\mu g/m^3\)). The percentage of excessive exposure to total indium was 6.3\%(n = 1), whereas that of respirable indium was 25.0\%(n = 4).

3.4. Indium recycling industry

The exposure to total indium ranged from 0.5 to 2,089.9 \(\mu g/m^3\) (GM = 54.7 \(\mu g/m^3\), AM = 225.8 \(\mu g/m^3\)) exhibiting a broad range of exposure. In regard to respirable indium \((n = 69)\), the exposure also appeared with a distribution over a broad range of 0.5 to 436.3 \(\mu g/m^3\) (GM = 15.4 \(\mu g/m^3\), AM = 63.8 \(\mu g/m^3\)) as for total indium. The results showed that the level exceeded the set standards for exposure by 38.3\%(n = 26) of total indium and 52.1\%(n = 36) for respirable indium (Tables 1 and 2).

Raw materials comprising the sputtering target, scraps, and sludge are disintegrated into powder in the pulverization process, at which time workers are naturally exposed to ITO or IZO particulates. Workers’ exposure to total indium \((n = 11)\) ranged from 81.0 to 2,089.9 \(\mu g/m^3\) and respirable indium \((n = 14)\) ranged from 0.5 to 253.7 \(\mu g/m^3\). A total of 90.9\%(n = 10) of samples from total indium exposure and 85.7\%(n = 12) of samples from respirable indium exposure exceeded the respective standards.

In the dissolution process, workers are exposed to total and respirable indium particulates from ITO, IZO, and In2O3, and aerosols are created during dissolution when workers take measurements, transfer, and put powders into the melting furnace. Workers’ exposure to total indium \((n = 4)\) was distributed in the range 19.7 to 308.2 \(\mu g/m^3\) and respirable indium \((n = 3)\) was distributed in the range 15.4 to 264.2 \(\mu g/m^3\). A total of 75.0\%(n = 3) of samples from total indium exposure and 100\%(n = 3) of samples from respirable indium exposure exceeded the respective standards.

The workers are exposed to indium during substitution via aerosols (mists) that are created through chemical reactions in the refining process of indium by melting the zinc or aluminum in the substitution tank containing the indium solution. Workers’ exposure to total indium \((n = 24)\) was distributed in the range 5.9 to 1,003.5 \(\mu g/m^3\) and respirable indium \((n = 24)\) was distributed in the range 1.8 to 317.9 \(\mu g/m^3\). A total of 25.0\%(n = 6) of samples from total indium exposure and 41.7\%(n = 10) of samples from respirable indium exposure exceeded the respective standards.

Electrolysis can be wet or dry. Workers’ in the wet electrolysis process are exposed to indium by aerosols, and workers in the dry-electrolysis process are exposed to fumes generated from the indium ingots melting in the furnace. Workers’ exposure to total indium \((n = 13)\) appeared as a distribution in the range of 0.5 to 643.4 \(\mu g/m^3\) and respirable indium \((n = 13)\) was distributed in the range of 1.9 to 436.3 \(\mu g/m^3\). A total of 38.5\%(n = 5) of samples from total indium exposure and 59.9\%(n = 7) of samples from respirable indium exposure exceeded the respective standards.

The casting process of indium is divided into primary casting process and secondary casting process. Ingots of indium (below 99% of purity) are prepared in the primary casting process by eliminating impurities contained in indium sponges obtained by refining during substitution. In the secondary casting process, the indium
made from the electrolysis and indium of high purity (over 99.99%) to control the indium content are placed in the melting furnace to produce high-purity indium ingots. Workers are then exposed to indium fumes. Workers’ exposure to total indium (n = 16) was distributed in the range of 5.6 to 343.9 μg/m³ and respirable indium (n = 15) was distributed in the range of 1.3 to 128.8 μg/m³. A total of 12.5% (n = 2) of samples from total indium exposure and 26.7% (n = 4) of samples from respirable indium exposure exceeded the respective standards.

4. Discussion

Indium is an interesting chemical that is surprisingly thinly applied to electronic products such as liquid-crystal displays, light-emitting diodes, and organic light-emitting diodes. Although this study focused on active development to improve performance of devices such as mobile phones and televisions, it remains critical to be vigilant against the hazards of exposure to toxic chemicals and their health effects, which are sometimes unknown. The development of the electronics industry has increased the indium or indium compound handling volume, and the number of workers in related industries is also increasing. However, measures to accurately evaluate and manage the risk of exposure to indium remain insufficient.

Workers are exposed to various types of indium or indium compounds. In(OH)₃, In₂O₃, ITO, IZO, In₂S₃, and indium fume are typical. It starts with the production of In₂O₃ of nanoparticles to make a sputter target used in the electronics industry. In₂O₃ is produced and used as a sputter target through physical bonding and chemical reaction and is recovered as indium metal. The workers of the sputter target company are exposed to In(OH)₃, In₂O₃, ITO, IZO, and indium fume, the workers of panel display companies and related companies are exposed to ITO and IZO, and workers of indium recycling companies are exposed to ITO, IZO, In₂S₃, and indium fume. Therefore, workers are exposed to indium or indium compounds which are in the form of particulates, aerosols, and fumes.

There are few published articles related to indium exposure, and it is difficult to compare directly with this article because the measurement targets and exposure evaluation strategies are different. The indium exposure concentration of ITO grinding workers at ITO production workplaces published by Blakley et al. [19], is judged to be comparable for the exposure concentration of polishing workers in this study. Among the data of the Blakley et al. [19], the geometric mean exposure concentrations of aspirating indium (n = 2) and respirable indium (n = 6) of the ITO grinder at the ITO production site, which were judged to be the same job as this study, were 90.5 μg/m³ and 14.6 μg/m³, respectively. This exposure level was about 2 times higher than the GM concentration of total indium concentration (n = 45, 56 μg/m³) and respirable indium concentration (n = 54, 5.5 μg/m³) of polishing workers in this study. According to the results of Hines et al. [2], there is no classification of detailed jobs, but the exposure concentration of total indium concentration (n = 6) of workers at sputter production plant was 17 μg/m³ to 390 μg/m³, which was lower at high concentration than 0.9 μg/m³ to 609.3 μg/m³ in this study.

Among indium-related industries, workers of indium recycling companies were found to be exposed to significantly higher levels of indium than workers at companies that make or use sputter targets for both total and respirable indium (p < 0.05). This increased exposure at recycling companies is likely due to small workplaces, closed structures, and the lack of awareness of the risks of exposure to indium among managers and workers. As a result, workplace management was very poor, and workers were exposed to high concentrations of indium. The problems experienced by workers in the indium recycling industry are similar to those experienced by workers in other industries.

Post-hoc analysis comparison indicated that indium exposure concentrations by process in the sputtering target manufacturing industry, the mixing, polishing, molding, and bonding process of total indium, and mixing process of respirable indium were significantly higher than those of other processes (p < 0.05). The total indium of the panel display industry was significantly higher in the sputtering components cleaning companies, and the respirable indium was significantly lower in the panel display companies (p < 0.05).

Respirable indium has been found to exceed exposure standard in all processes in all three industries. In particular, the rate of exceeding the exposure standard in the indium recycling industry was higher than that of the other two industries. This also showed the same trend in total indium concentration. In the case of the indium recycling plant, a high concentration of indium was generated in the process of physically crushing the sputter target to recover indium. Despite the presence of high concentrations of indium, these recycling plants, which were the subject of this study, were small business sites compared with the other two industries, and ventilation facilities such as local exhaust systems were not installed in most cases. In some workplaces, local exhaust systems were installed, but their efficiency was weak and management was not well performed.

Workers in indium or indium compounds industries are at high risk of exposure to indium and its associated health effects, but there are no data on exposure assessment, work environment management, and workers’ serum levels, which is in the blind spot of legal management.

The most important finding from this exposure assessment is that many workers who handle indium compounds in the electronics industry are exposed to indium levels (both total and respirable) that exceed the exposure standards for indium. Hence, it is necessary to continuously monitor the indium exposure of this workforce and take measures to reduce its exposure levels.

Conflicts of interest

All authors have no conflict of interest to declare.

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