Indoor human exposure to size-fractionated aerosols during the 2015 Southeast Asian smoke haze and assessment of exposure mitigation strategies

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Abstract

The 2015 smoke haze episode was one of the most severe and prolonged transboundary air pollution events ever seen in Southeast Asia (SEA), affecting the air quality of several countries within the region including Indonesia, Malaysia and Singapore. The 24 h mean outdoor PM$_{2.5}$ (particulate matter (PM) with aerodynamic diameter ≤ 2.5 μm) concentrations ranged from 72–157 μg m$^{-3}$ in Singapore during this episode, exceeding the WHO 24 h mean PM$_{2.5}$ guidelines (25 μg m$^{-3}$) several times over. The smoke haze episode not only affected ambient air quality, but also indoor air quality due to the migration of PM of different sizes from the outdoor to the indoor environment. Despite the frequent occurrence of smoke haze episodes over the years, their potential health impacts on indoor building occupants remain largely unknown in SEA due to the lack of systematic investigations and observational data. The current work was carried out in Singapore to assess human exposure to size-resolved PM during the 2015 smoke haze episode, and to evaluate the effectiveness of exposure mitigation measures in smoke-haze-impacted naturally ventilated indoor environments. The potential health risks associated with exposure to PM$_{2.5}$ were assessed based on the concentrations of redox active particulate-bound trace elements, which are known to be harmful to human health, with and without exposure mitigation. Overall, it was observed that human health exposure to PM$_{2.5}$ and its carcinogenic chemical components was reduced substantially by 62% ($p < 0.05$) while using an air cleaner. However, extremely small hazardous particles were only partially removed by the air cleaner and remain a matter of concern for public health.

1. Introduction

Smoke haze which results from uncontrolled forest and peatland fires in Indonesia is a significant source of PM in SEA [1–4, 48]. Annual land clearance activities in Sumatra and Kalimantan, which are provinces of Indonesia, cause transboundary smoke haze episodes, affecting the air quality of neighboring countries such as Malaysia, Singapore and Thailand [5]. Smoke haze not only affects atmospheric visibility, regional climate and economy, but also has an adverse impact on public health due to the increased incidence of asthma, chronic obstructive pulmonary disease (COPD), lung cancer and cardiovascular diseases [6–10, 56–57]. During the occurrence of smoke haze episodes, outdoor PM can migrate into naturally ventilated indoor environments where people typically spend approximately 80% of their time, thereby posing threats to the health of building occupants [11–13]. In 2015, SEA experienced one of the worst haze episodes ever seen (surpassing the haze events of 1997–98 and 2013) lasting for more than three months. Recent regional-scale modeling studies have raised public health concerns about this episode relating to short-term population exposure to degraded air quality [14, 15].

It is well established that particle size and chemical composition are key parameters that determine
the health effects of PM [16–19]. Previous studies have reported that ultrafine particles (UFPs) (PM with aerodynamic diameter (AED) ≤ 100 nm) are prevalent during haze episodes [20–24, 48]. However, there is a lack of research on these harmful and easily inhalable sub-micrometer particles in SEA. PM associated with biomass burning contains a complex mixture of organic and inorganic compounds such as carbonaceous materials, including carcinogenic and mutagenic polycyclic aromatic hydrocarbons (PAHs), inorganic ions, trace elements and endotoxins [25]. Trace elements are of particular importance in health risk assessment due to the potential health effects caused by these toxic and carcinogenic chemical constituents [26–30]; the definition of trace elements is given in the supplementary information (SI) available at stacks.iop.org/ERL/12/114026/mmedia. Moreover, UFPs have a higher trace elemental content [30, 31] than coarse particles—especially in health-relevant toxic species. Redox active elements have been reported to exhibit a high oxidative stress potential caused by the reactive oxygen species (ROS) [32–34], which is responsible for inducing adverse health effects. Recent studies quantified indoor particle exposure based on the PM number concentration during the 2013 haze episode in Singapore [11, 12]. However, these studies did not investigate human exposure based on the elemental composition of the PM. It is therefore essential to assess the human health risks due to exposure to particle-bound trace elements and possibly other toxic components originating from biomass burning as well.

As a mitigation measure to reduce exposure to haze particles, public health officials often recommend that residents in the haze-affected areas remain indoors, with doors and windows shut in naturally ventilated residences. However, there might still be infiltration of outdoor air through diffusion, which can cause the accumulation of particles indoors due to a lack of sufficient air circulation [35]. Furthermore, the lack of thermal comfort together with poor indoor air quality (IAQ) may lead to sick building syndromes among the building occupants. As per the United States Environmental Protection Agency, the term 'sick building syndrome' is used to describe situations in which building occupants experience acute health and comfort effects that appear to be linked to time spent in a building, but no specific illness or cause can be identified [58]. Portable air cleaners can be used under these circumstances to mitigate human exposure to PM in naturally ventilated buildings during haze periods [36–40]. The selection of appropriate air cleaners to be used depends on their clean air delivery rate (CADR), which defines the volume of indoor space for which the specific cleaner can efficiently work. Few studies have assessed the effectiveness of using air cleaners to improve IAQ during smoke haze episodes, based on a reduction of the indoor PM loading [35, 41]. Nevertheless, the effectiveness of these devices in reducing human exposure and the associated potential health risks based on the toxic elemental composition of PM indoors remains largely unknown. Additionally, previous studies reported in the literature [35, 41] have been carried out under mid-latitude environmental conditions and thus are not applicable to tropical climatic conditions in SEA, where the smoke-haze-impacted PM has unique physico-chemical characteristics [4, 5, 21, 25].

In view of the above-mentioned knowledge gaps, a pilot study was conducted in a typical naturally ventilated residential apartment under three different exposure cases during the 2015 haze episode in Singapore. During this episode, Singapore’s air quality was severely affected, with the 1 h PM$_{2.5}$ reaching a record high level of 471 μg m$^{-3}$ on 19 October 2015. Figure 1 shows the 24 h PM$_{2.5}$ concentration and 24 h PSI (Pollutant Standards Index) values reported by the National Environment Agency (NEA) of Singapore during the sampling days, with the PSI reaching an unhealthy range on several occasions during the haze period [42]; a description of the PSI is given in the SI. In addition, the PSI also reached a very unhealthy range and even close to hazardous range for two hazy days during sampling. Since the population density (8264 people km$^{-2}$) in Singapore is very high because of its small geographical area (710 km$^2$), exposure to PM$_{2.5}$ of smoke haze origin in multi-story residential buildings involving natural ventilation is a major concern [11, 12]. The key objective of the current study is to assess human exposure to size-resolved PM in indoor environments with and without exposure mitigation measures. The specific cases considered in the study include: Case 1—keep all windows open with a ceiling fan on as per the usual practice; Case 2—keep all windows fully closed as per the recommended practice during haze episodes; and Case 3—keep all windows fully closed with the operation of an indoor air cleaner. The indoor to outdoor PM concentration ratios (I/O ratios) of size-resolved PM were measured and the effectiveness of cleaning the air through filtration in the indoor environment was studied. A potential health risk assessment was performed in three exposure cases to identify a suitable strategy for reducing PM exposure indoors. To the best of our knowledge, this is the first comprehensive study focused on investigating indoor human exposure to various sizes of particles during smoke haze periods and identifying a suitable exposure mitigation measure in tropical and highly urbanized areas like Singapore. The results of this study provide a scientific basis for the enhancement of IAQ and improving public health during haze episodes.
2. Methodology

2.1. Sample collection
Size-fractionated PM was collected at a naturally ventilated multi-story residential building that is part of the National University of Singapore main campus. The PM samples were collected in Teflon fiber (polytetrafluoroethylene, PTFE) filters with the use of a pre-calibrated personal cascade impactor sampler (Sioutas™ PCIS, SKC Inc., Eighty-Four, PA, USA) [43] connected to SKC Leland Legacy pumps, each operating at 9 l min\(^{-1}\) (liters per minute). Each PCIS collected size-fractionated PM in the size range >2.5 \(\mu\)m, 2.5–1 \(\mu\)m, 1–0.5 \(\mu\)m, 0.5–0.25 \(\mu\)m and <0.25 \(\mu\)m. In this study, PM with a diameter of >2.5 \(\mu\)m is termed as coarse particles, 0.25–2.5 \(\mu\)m as accumulation mode particles and <0.25 \(\mu\)m as quasi-ultrafine particles (quasi-UFPs) [44]. Outdoor PM measurements were carried out concurrently on the 8th and 20th storeys of the naturally ventilated building for 24 h during hazy days (\(n = 5\)) to examine the vertical variation of PM. An intensive field study was conducted on the 20th storey (\(n = 13\)) from September to October 2015 both inside and outside an apartment by placing the samplers at a breathing zone height (∼1.5 m) from the floor in the three different exposure cases as mentioned earlier. Details of the gravimetric analysis of the PM samples are given in the SI.

2.2. Chemical extraction and analysis
The PM samples collected on the 20th storey were further analyzed for a total of 24 particulate-phase trace elements (Al, As, Ba, Be, Bi, B, Cd, Cr, Cs, Cr, Co, Cu, Ga, Fe, Pb, Li, Mn, Ni, Rb, Se, Sn, Te, Ti, Sn, and Zn) using the acid digestion method as reported in previous studies [45, 46]. Further details about the analytical method and QA/QC protocols are provided in the SI (figure S1).

2.3. Human health risk assessment
A human health risk assessment was conducted for the three different cases based on the approach reported in the relevant literature [28, 47–50]. Two types of potential health risk, (1) non-carcinogenic (for Al, Be, Cd, Cr, Mn and Pb) based on the hazard quotient (HQ) and (2) carcinogenic (for As, Be, Cd, Co, Cr, Ni and Pb) based on the estimated lifetime cancer risk (ELCR), were examined [28, 51]. The steps involved to define HQ and ELCR, and details of the potential health risk assessment, are given in the SI (tables S1–S2).

3. Results and discussion

3.1. The 2015 haze episode
The 2015 haze event was very severe in the history of SEA, so it is important to analyze the sources and transport of smoke-haze-impacted air masses. Consequently, backward air trajectories were generated during the haze period in Singapore for the three cases by using the latest version of the hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model developed by the National Oceanic and Atmospheric Administration [52]. The air trajectories were constructed over 72 h at three different altitudes: (a) above the boundary layer (1500 m), (b) below the boundary layer (500 m), and (c) at surface level (100 m); further details about the HYSPLIT model are given in the SI. It can be seen from the haze map and trajectories for the Case 1 exposure experiment (figure 2) that the smoke haze originated mainly from Indonesia (Sumatra and
Kalimantan), as per the 1500 m trajectories. The 100 m and 500 m trajectories originated mainly from the open Java Sea and were therefore not significantly affected by the wildfires in Indonesia. These air masses mainly comprised airborne particles emitted from ships, sea spray and local emissions within Singapore. Furthermore, it was observed that the air trajectories only began shifting toward the biomass burning sources at 600 m and above. This suggests that smoke emissions from wildfires reached Singapore well above the surface levels and were intermixed with the clean air mass transported into Singapore at surface level by convective winds. Thus, the severity of the smoke haze was relatively low during the period for the Case 1 exposure experiment (PSI 92–139, PM$_{2.5}$ = 48–92 µg m$^{-3}$). However, it should be noted that during the period for Case 2 (PSI 211–279, PM$_{2.5}$ = 160–229 µg m$^{-3}$) and Case 3 (PSI 165, PM$_{2.5}$ = 117 µg m$^{-3}$) exposure experiments, both 500 m and 1500 m trajectories were found to originate in the Indonesian islands, creating relatively more severe haze situations during these cases (figures S2–S3).

3.2. Vertical variations of haze-influenced particles

Simultaneous measurements of outdoor PM concentrations on the 8th and 20th storeys were made to analyze the vertical distribution of the biomass burning-impacted aerosols, and the results are reported in figure S4. As such, no significant variations were observed in the PM concentrations across different size ranges with respect to the vertical height. The PM$_{2.5}$ mass concentrations measured in ambient air during the study period were in good agreement with the national mean PM$_{2.5}$ concentration (calculated from real-time measurements of PM$_{2.5}$ at 22 air quality monitoring stations, located at different heights within Singapore), as shown in figure S5. The lack of vertical variations in PM concentrations can be attributed to the severity and intensity of the regional smoke haze, characterized by the dominant advective inflow of biomass burning-derived PM into Singapore from Indonesia compared to the local urban emissions of PM. Therefore, it can be reasonably said that the smoke haze plume originating from Indonesia was uniformly spread out in a vertical direction across Singapore, causing equal amounts of public health concern on any floor above the 8th story. We made similar observations during previous smoke haze episodes, and discussed our findings based on the transport of biomass burning-impacted PM emissions [50].

3.3. Size-fractionated PM concentrations

In order to examine the impact of smoke haze on indoor human exposure with and without mitigation strategies, size-fractionated PM was collected concurrently in both outdoor and indoor environments for three different cases on the 20th storey. The purpose of the concurrent PM measurements was to assess the migration of PM of different sizes from the outdoor environment to indoors, and the related exposure of building occupants to particulate pollution under different exposure cases. The outdoor PM$_{2.5}$ data measured in the vicinity of the building was compared to that of the entire country at a network of 22 air quality monitoring stations maintained by the NEA, and was found to be in good agreement, as mentioned earlier (figure S5).

The Case 1 exposure experiment was investigated for a period of four hazy days while keeping the windows of the living room area open, as usually done in the absence of haze and with a ceiling fan on for the uniform mixing of indoor air; this is the normal practice in naturally ventilated apartments in Singapore for improving thermal comfort. The goal of the study was to analyze the concentration of PM of different sizes
indoors without putting in place any mitigation measure to reduce human exposure to smoke haze; this case represents the worst-case scenario. The indoor and outdoor size-fractionated PM mass concentrations are shown in figure 3. The 24 h mean outdoor PM$_{2.5}$ mass concentration observed was $94 \pm 34 \mu g m^{-3}$, while it was $71 \pm 20 \mu g m^{-3}$ in the indoor air. These PM$_{2.5}$ levels exceeded the WHO 24 h mean PM$_{2.5}$ guidelines ($25 \mu g m^{-3}$) by a factor of four outdoors and three indoors, showing the severity of haze and its potential health impacts on the exposed population. Another important finding in the study was that the mass concentration of the PM in both outdoor and indoor air increased as the particle size decreased. This observation suggests that the smoke haze plume contained submicron-sized particles of high concentrations, which were also reported in several previous studies during the past smoke haze episodes [21, 22, 49].

The Case 2 exposure experiment was carried out by keeping the windows of the living room area fully closed and with a ceiling fan on for a period of eight hazy days. The goal was to find out the effectiveness of keeping the windows fully closed in improving IAQ while staying indoors during the severe haze. The outdoor and indoor size-fractionated PM mass concentrations are shown in figure 4. An increase in the mass concentration of the PM was noticed as the particle size decreased, as also observed in Case 1. The 24 h mean PM$_{2.5}$ mass concentrations were found to be $157 \pm 107 \mu g m^{-3}$ and $98 \pm 54 \mu g m^{-3}$ in outdoor and indoor air, respectively. It is interesting to note that the PM$_{2.5}$ levels were six and four times higher in outdoor and indoor air, respectively, than the WHO 24 h mean PM$_{2.5}$ guidelines ($25 \mu g m^{-3}$). Elevated indoor PM levels in Case 2, despite having the windows closed, can be attributed to more severe hazy conditions observed during this time period; the PSI was greater than 200 (i.e. in the very unhealthy range) as shown in the haze map (figure S2). Nevertheless, the effectiveness of this mitigation strategy over Case 1 is discussed in section 3.4.

The Case 3 exposure experiment was carried out by keeping the windows of the living room area fully closed, as done in Case 2, but with the notable difference that an air cleaner was kept in operation for 24 h. The idea behind this case was to examine the effectiveness of using a commercial air cleaner (Panasonic Model F-PXH55A equipped with a HEPA (high-efficiency particulate arrestance) filter, while keeping the windows fully closed during a severe haze episode. Based on the recommended room size of $42 m^2$ for the air cleaner, the CADR was calculated as $292 m^3 h^{-1}$ for tobacco smoke particles, so the air cleaner can clean up to 80% of $292 m^3$ of tobacco-smoke-contaminated indoor air in 1 h [53, 54]. One air cleaner was found to be sufficient in the study room where it was operated. Figure 5 shows the outdoor and indoor size-fractionated PM mass concentrations for Case 3. The 24 h outdoor PM$_{2.5}$ mass concentration was observed to be $72 \mu g m^{-3}$ while the 24 h indoor PM$_{2.5}$ mass concentration was $23 \mu g m^{-3}$. The latter was within the WHO 24 h mean PM$_{2.5}$ guideline of $25 \mu g m^{-3}$, suggesting that one air cleaner was sufficient for reducing the PM$_{2.5}$ levels to an acceptable indoor concentration and to provide protection to the apartment occupants against harmful exposure to PM$_{2.5}$.

3.4. Ratios of indoor to outdoor PM concentrations (I/O ratios)
I/O ratios were plotted against the particle size for the three cases, as shown in figure 6. Overall, the I/O ratios were less than 1 and found to decrease in general as
Figure 4. Indoor and outdoor size-fractionated PM concentrations for Case 2.

Figure 5. Indoor and outdoor size-fractionated PM concentrations for Case 3 (6 October 2015).
Figure 6. I/O ratios for different PM sizes for the three cases.

The particle size increased for Cases 1 and 2. This could be attributed to the gravitational settling of larger particles in the confined indoor environment following their penetration from the outdoor environment. However, the I/O ratios followed a different trend for Case 3 with a peak for the 0.5–1 μm PM. This peak in the I/O ratio seems to be related to the limitation of the filter contained in the air cleaner in terms of its efficiency in the removal of such particles. While coarse and ultrafine particles are captured efficiently by the filter medium through the impaction and diffusion mechanisms respectively, intermediate size particles are collected based on the interception mechanism, which is relatively weak, hence their accumulation indoors with the increased I/O ratio. The latter mechanism becomes effective with the formation of the filter cake after its prolonged use.

A notable observation was that the I/O ratios were found to be the lowest for Case 3 for all particle sizes, implying that using the air cleaner was the most effective strategy for reducing the PM levels indoors and achieving good IAQ during severe haze episodes. In the event of not having access to an air cleaner, Case 2 is still better than Case 1, as it provides partial protection against exposure to PM with a diameter < 100 nm. The overall effectiveness of the air cleaner was quantified using equation (1) [35], and it was found to be 73% ± 27%, 54% ± 2% and 23% ± 21% for coarse, accumulation and quasi-UFPs, respectively. Therefore, it is evident that quasi-UFPs were less efficiently removed, and that the performance of the existing air cleaner needs to be improved with the use of a better air filter than the commercial HEPA filter to remove easily inhalable and harmful UFPs.

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\text{% PM Reduction} = 100 \times \left[1 - \frac{\text{Indoor PM w Air Cleaner}}{\text{Indoor PM w/o Air Cleaner}} \right] / \text{Outdoor PM}.
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3.6. Total elemental characterizations

Figures S6–S8 show the mean values and standard deviations of the measured concentrations of 24 elements in coarse, accumulation and quasi-UF size ranges outdoors and indoors for the three cases, respectively. For the three cases, the data sets were divided into three distinct categories based on their respective concentrations in the different PM size ranges [46]. This classification showed that (i) trace elements with concentrations >80 ng m\(^{-3}\) were the major elements, (ii) trace elements with concentrations between 1–80 ng m\(^{-3}\) were the sub-major elements, and (iii) trace elements with a concentration <1 ng m\(^{-3}\) were the minor elements.
For all three cases, B, Al, Fe and Sn were found to be the major elements. Fe, which originates mainly from soil dust (peat soil), was dominant in the coarse mode [25, 50]. However, some other major elements such as B and Sn were thought to be derived from anthropogenic sources with higher concentrations in the accumulation mode compared to the coarse mode. All other elements were quantified either as sub-major or minor elements for the different cases, as shown in figures S6–S8. Among the sub-major and minor elements, Sr was believed to be derived mainly from soil dust (peat soil), as the concentration in the coarse mode was higher than or equal to the concentration in accumulation mode indoors and outdoors [25, 50]. However, for most of the other sub-major and minor elements, their concentrations were dominant in the accumulation or quasi-UF mode. These trends suggest that the emission of these elements could be associated with anthropogenic sources of combustion, such as smoke haze and/or on-road vehicles, industries (power plants, chemical and other industries) and other domestic sectors [25, 50]. It is noteworthy that all the elements in Case 3 were found to be predominant in the accumulation and quasi-UF modes, especially indoors. This trend may be attributed to the efficient removal of coarse particles by the air cleaner.

Overall, Case 3 showed the lowest concentrations and I/O ratios of almost all the 24 elements, indicating that it was the most effective approach for PM elemental exposure reduction. Also, most of the indoor elemental concentrations were found to be lower than those outdoors, implying that remaining indoors is safer than being outdoors during the smoke haze period.

3.7. Human health risk assessment

Using the measured concentrations of toxic elements, the potential human health risk was assessed for all three exposure cases due to inhalation exposure to PM$_{2.5}$ observed both indoors and outdoors in this study. PM$_{2.5}$ rather than the size-fractionated PM was considered in the risk assessment as we only have air quality standards for the former at this point in time. This risk assessment would help in understanding the effectiveness of mitigation measures explored in this study during hazy days. Tables 1 and 2 present the potential non-carcinogenic and carcinogenic health risk estimates, respectively, in the three cases for outdoors as well as indoors.

For non-carcinogenic risk assessment, HQ values for both indoors and outdoors were observed to be less than the acceptable upper limit of 1 in all three cases. However, there might be long-term chronic health effects due to continued outdoor exposure, if hazy atmospheric conditions persist in the future. The health risk was higher in the outdoor environment compared to the indoor environment, showing that it is better to remain indoors during haze to reduce exposure to PM$_{2.5}$ mass concentrations and the related health effects. Al and Mn showed the highest HQ values among all the elements. Of the three cases, the health risk for Case 3 showed the lowest value, as expected.

In the carcinogenic risk assessment, the ELCR values for both indoors and outdoors were greater than the acceptable upper limit of 1 × 10$^{-6}$ in all three cases, indicating that the carcinogenic risk associated with exposure to PM$_{2.5}$ is of concern. As and Cr showed the highest ELCR values out of the 24 elements.

### Table 1. Potential non-carcinogenic health risk estimates of PM$_{2.5}$ for the three cases indoors and outdoors.

| Element | Reference dose (mg kg$^{-1}$ day$^{-1}$) | Case 1 | Case 2 | Case 3 |
|---------|---------------------------------|--------|--------|--------|
|         |                                 | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor |
| Al      | 1.43 × 10$^{-3}$                | 7.99 × 10$^{-3}$ | 1.22 × 10$^{-2}$ | 1.62 × 10$^{-2}$ | 1.22 × 10$^{-2}$ | 7.91 × 10$^{-3}$ | 8.00 × 10$^{-3}$ |
| Be      | 5.71 × 10$^{-6}$               | 4.52 × 10$^{-4}$ | 2.91 × 10$^{-4}$ | 2.22 × 10$^{-4}$ | 1.67 × 10$^{-4}$ | 8.23 × 10$^{-5}$ | 1.03 × 10$^{-4}$ |
| Cd      | 5.71 × 10$^{-6}$               | 4.70 × 10$^{-4}$ | 5.40 × 10$^{-4}$ | 4.80 × 10$^{-4}$ | 6.09 × 10$^{-4}$ | 7.75 × 10$^{-4}$ | 6.30 × 10$^{-4}$ |
| Cr      | 2.86 × 10$^{-5}$               | 1.82 × 10$^{-3}$ | 4.33 × 10$^{-3}$ | 2.71 × 10$^{-3}$ | 4.84 × 10$^{-3}$ | 4.10 × 10$^{-3}$ | 2.87 × 10$^{-3}$ |
| Mn      | 1.43 × 10$^{-5}$               | 8.13 × 10$^{-3}$ | 8.26 × 10$^{-3}$ | 1.13 × 10$^{-2}$ | 1.74 × 10$^{-2}$ | 5.89 × 10$^{-3}$ | 1.12 × 10$^{-2}$ |
| Pb      | 4.29 × 10$^{-5}$               | 2.15 × 10$^{-5}$ | 1.62 × 10$^{-5}$ | 2.78 × 10$^{-5}$ | 3.45 × 10$^{-5}$ | 6.77 × 10$^{-4}$ | 1.48 × 10$^{-3}$ |
| Σ       |                                 | 1.89 × 10$^{-2}$ | 2.56 × 10$^{-2}$ | 3.09 × 10$^{-2}$ | 3.53 × 10$^{-2}$ | 1.58 × 10$^{-2}$ | 2.43 × 10$^{-2}$ |

### Table 2. Potential carcinogenic health risk estimates of PM$_{2.5}$ for the three cases indoors and outdoors.

| Element | SF (kg day mg$^{-1}$) | Case 1 | Case 2 | Case 3 |
|---------|----------------------|--------|--------|--------|
|         |                      | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor |
| As      | 15.05                | 3.04 × 10$^{-6}$ | 2.11 × 10$^{-6}$ | 1.68 × 10$^{-6}$ | 1.36 × 10$^{-6}$ | 5.17 × 10$^{-7}$ | 3.87 × 10$^{-7}$ |
| Be      | 8.4                  | 2.17 × 10$^{-6}$ | 1.04 × 10$^{-5}$ | 1.07 × 10$^{-6}$ | 8.02 × 10$^{-7}$ | 3.95 × 10$^{-9}$ | 4.95 × 10$^{-9}$ |
| Cd      | 6.3                  | 1.69 × 10$^{-6}$ | 1.94 × 10$^{-6}$ | 1.73 × 10$^{-6}$ | 2.19 × 10$^{-8}$ | 2.79 × 10$^{-8}$ | 2.29 × 10$^{-8}$ |
| Cr      | 31.5                 | 2.87 × 10$^{-7}$ | 3.36 × 10$^{-7}$ | 1.15 × 10$^{-7}$ | 1.59 × 10$^{-7}$ | 3.91 × 10$^{-8}$ | 9.87 × 10$^{-8}$ |
| Mn      | 42                   | 2.19 × 10$^{-6}$ | 5.19 × 10$^{-6}$ | 3.25 × 10$^{-6}$ | 5.80 × 10$^{-6}$ | 4.92 × 10$^{-7}$ | 3.44 × 10$^{-6}$ |
| Ni      | 0.84                 | 1.09 × 10$^{-7}$ | 1.41 × 10$^{-7}$ | 1.06 × 10$^{-7}$ | 1.59 × 10$^{-7}$ | 1.72 × 10$^{-8}$ | 2.94 × 10$^{-8}$ |
| Pb      | 0.042                | 3.17 × 10$^{-9}$ | 2.38 × 10$^{-9}$ | 4.09 × 10$^{-9}$ | 5.06 × 10$^{-9}$ | 9.93 × 10$^{-10}$ | 2.17 × 10$^{-9}$ |
| Σ       |                      | 5.67 × 10$^{-6}$ | 7.82 × 10$^{-6}$ | 5.19 × 10$^{-6}$ | 7.52 × 10$^{-6}$ | 1.10 × 10$^{-6}$ | 3.98 × 10$^{-6}$ |
The overall health risk was higher in the outdoor environment compared to the indoor environment, reflecting that remaining indoors is safer during haze episodes. The indoor human health risk for Case 1 was $5.67 \times 10^{-6}$, indicating that about six people out of a population of one million individuals exposed to indoor air with the windows open during haze events in Singapore are likely to develop cancer in their lifetime due to the inhalation of toxic elements in PM$_{2.5}$, and thus have a shorter life expectancy. Therefore, by considering the current population in Singapore to be 5.78 million [59] at least 33 individuals will be adversely affected by exposure to the smoke haze indoors. In the outdoor scenario for Case 1, it was observed that the estimated cumulative ELCR for PM$_{2.5}$ increased to $7.82 \times 10^{-5}$, indicating the greater impact of haze events on public health with about eight people out of a population of one million individuals likely to develop cancer in their lifetime. This potential health risk is about 2.3 times higher than that due to traffic-related ambient PM$_{2.5}$ in Singapore, as mentioned in a recent study by Zhang et al [55], which reflects the severity of haze-related particles. The number of individuals in Singapore adversely affected due to exposure to smoke haze outdoors will be at least 45. Similarly, this adversely affected population fraction for Case 2 will be at least 30 and 43 people due to indoor and outdoor smoke haze exposure, respectively, while for Case 3, the fraction will be at least 6 and 23 people for indoors and outdoors, respectively. Furthermore, the I/O ratios for Case 1, Case 2 and Case 3 were observed as 0.73, 0.69 and 0.28, respectively, for PM$_{2.5}$ with a carcinogenic health risk, indicating that the potential carcinogenic health risk reduction was substantial for Case 3 (62%) at $p < 0.05$.

The results of this study are found to be different from those estimated by Koplitz et al [15] and Crippa et al [14], based on the use of chemical transport models and dose-response relationships with some assumptions. The latter studies estimate the substantial health impacts of the 2015 haze event on the Singaporean population by considering the outdoor exposure to PM$_{2.5}$ over a period of two months (September–October 2015). The key differences between the current study and the two modeling studies are as follows.

The modeling studies in general considered population-weighted average smoke (the carbonaceous fraction of PM$_{2.5}$) exposure, neglecting the spatial variability in exposure within a receptor region. Koplitz et al [15] used the concentration response to estimate the health impacts based on studies done in high-income countries with different baseline health characteristics and air pollution sources. Crippa et al [14] calculated excess all-cause mortality due to short-term acute exposure to PM$_{2.5}$ using simulated 24 h PM$_{2.5}$ and exposure–response functions from recent and comprehensive epidemiological studies linking the health impacts to short-term exposure to outdoor PM$_{2.5}$. The current study considered the chemical speciation of PM$_{2.5}$ and calculated the long-term chronic health risk based on the relative abundance and toxicity of trace elements in PM$_{2.5}$, since several previous studies have reported that trace elements and PAHs account for most of the toxicity of PM$_{2.5}$ [26–30, 55].

Since the potential carcinogenic health risk was found to be greater than the threshold value, the size-fractionated I/O ratios for the ELCR were examined for the measured elements and across three cases to give a deeper understanding of the variability in the health risk, as shown in figure 7. It was observed that for all the elements, the I/O ratios for the potential carcinogenic health risk were highest in either the accumulation or quasi-UF size range across the three cases. Overall, this analysis shows that there is a substantial reduction in I/O ratios for almost all the elements in the coarse and accumulation size ranges in response to the use of PM exposure mitigation measures, with the exception of As and Cd. The relatively high concentrations of these two elements, as observed in Case 3, may be due to their presence in certain chemical states that have less affinity for the filter material and/or some specific indoor activity enhancing their concentrations. Moreover, quasi-UF particles did not show any significant reduction in the I/O ratios while using the air cleaner in Case 3, which again reflects the inefficiency of the air cleaner for the removal of ultrafine particles.

4. Conclusions

The 2015 transboundary smoke haze had a significant impact on the regional air quality of SEA due to the emission of primary PM and the formation of secondary PM. Analysis of the size-fractionated PM showed higher I/O ratios for smaller-sized particles during the haze events, suggesting that exposure to such PM may have a severe health impact, since they contain relatively higher levels of toxic elements than coarse particles. The results of this study involving the potential health risk assessment of PM in different exposure cases showed that without using an air cleaner, keeping all windows closed can provide only partial protection to exposure to indoor PM. However, keeping all windows closed with the operation of an indoor air cleaner with a sufficient CADR results in the highest reduction in the potential carcinogenic health risk in naturally ventilated apartments. It should be noted that the air cleaner used in the study was less efficient at removing smaller particles—especially ultrafine particles—due to the limitation of the filter used for air cleaning. Further research and development are needed to develop better commercial filters to trap smaller particles more efficiently. Moreover, the results of this study reveal different findings compared to recent modeling studies that considered short-term, acute outdoor PM$_{2.5}$
exposure over a period of two months (September–October) during the 2015 haze [14, 15].

The three haze exposure experiments were conducted in this study on different hazy days due to the lack of availability of the required instruments in multiple units, as well as other practical constraints, including our inability to predict the occurrence and duration of sporadic haze events and the necessity of conducting the field experiments for 24 h in order to collect a sufficient mass of PM for the intended chemical analysis. Therefore, it was not possible to make a quantitative comparison of the health risks resulting from different haze exposure scenarios across the different haze periods. The current study considered the total elemental concentrations for potential health risk calculation, which tends to overestimate the risk, as the bio-accessible fraction of PM is smaller than the total concentration of elements. Moreover, other particle-bound chemical components such as PAHs and quinones may also contribute to the overall toxicity of the PM and therefore merit consideration for health risk assessment. However, a recent study by Zhang et al [55] indicates that the health risk due to toxic elements is more significant than that due to PAHs. In any case, more comprehensive PM chemical speciation studies are required to improve our understanding of the potential health effects of haze and urban aerosols. Such studies will be beneficial for policymakers in implementing stringent environmental regulations for maintaining good IAQ and minimizing the impacts of haze aerosols on public health.

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Figure 7. Variations of I/O ratios for potential carcinogenic health risk assessment for the measured elements across different size ranges for the three cases.
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