Size-fractionated Particulate Matter in Indoor and Outdoor Environments during the 2015 Haze in Singapore: Potential Human Health Risk Assessment

Ruchi Sharma, Rajasekhar Balasubramanian*

Department of Civil and Environmental Engineering, National University of Singapore, Singapore 117576, Singapore

ABSTRACT

Landscape fires in Indonesia during the 2015 resulted in large-scale emissions of airborne particulate matter (PM) that degraded ambient air quality of several countries in Southeast Asia (SEA) including Singapore. During this transboundary haze episode, the general public was advised to remain indoors as much as possible in order to mitigate their exposure to high concentrations of PM in the outdoor environment. To understand the quantitative relationship between outdoor and indoor air quality, we measured PM_{2.5} as well as the size-fractionated PM (coarse, accumulation and quasi-ultrafine (q-UF) particles) simultaneously inside and outside a naturally ventilated apartment and studied the potential health risk associated with exposure to PM of different sizes under the three levels of smoke haze (light, moderate and severe). PM mass concentrations increased with a decrease in particle size, and the q-UF particles (diameter ≤ 250 nm) were observed to be as high as 80 to 85 $\mu\text{g m}^{-3}$ both indoors and outdoors. Estimation of PM deposition patterns along the human respiratory tract revealed that q-UF particles were mainly deposited in the deeper alveolar region, thereby posing severe health threats. Potential human health risk assessment results based on bioavailable concentrations of toxic elements in PM raised further concerns about health impacts of q-UF particles deposited in the alveolar region. Moreover, uncertainty analysis of exposure parameters used in potential carcinogenic health risk assessment model indicated much higher exceedance of potential health risk than the threshold limit for 95th percentile values of the health risk (11.5 times higher for PM_{2.5}) during severe-haze episodes. The potential health risk estimated in this study indicates the need to conduct further studies focused upon mitigation of human exposure to achieve health benefits during haze episodes.

Keywords: Smoke haze; Size-fractionated particles; Bioavailable toxic elements; Potential human health risk; Uncertainty analysis.

INTRODUCTION

Severe smoke haze episodes have occurred frequently in SEA in recent years, degrading the ambient air quality of several countries in the region including Malaysia, Thailand and Singapore (e.g., Nichol, 1998; Balasubramanian *et al.*, 2003; Betha *et al.*, 2013; Othman *et al.*, 2014; Reddington *et al.*, 2014; Pongpiachan *et al.*, 2015; Pongpiachan, 2016). These transboundary smoke haze episodes are usually caused by uncontrolled biomass and peat burning in Indonesia, resulting in the emission of large amounts of airborne PM with unique chemical composition into the atmosphere (e.g., Nichol, 1998; Balasubramanian *et al.*, 2003; See *et al.*, 2006; Ryu *et al.*, 2007; Pongpiachan *et al.*, 2013; Pongpiachan and Paowa, 2014; Pongpiachan *et al.*, 2017). In 2015, Singapore experienced one of the worst and prolonged haze episodes,

lasting for three months with atmospheric visibility being less than 3 km on several occasions (NEA, 2015; The Strait Times, 2015; IBT, 2016). During this period, the 1 h PM_{2.5} concentration reached a record high level of 471 $\mu\text{g m}^{-3}$ on October 19, 2015 (Today, 2015).

Most of the public residential buildings in Singapore are designed for natural ventilation with windows being kept open for improved thermal comfort and air circulation. Under those conditions, indoor air quality (IAQ) tends to be strongly affected by the migration of air pollutants of outdoor origin (Massey *et al.*, 2013; Han *et al.*, 2015; Ji and Zhao, 2015; Choi and Kang, 2017). Since people spend typically 90% of their time indoors (Jenkins *et al.*, 1992; Schweizer *et al.*, 2007), IAQ studies are very important to assess human exposure to air pollutants. However, most of the previous PM exposure assessment studies conducted in Singapore during smoke haze episodes either relied on ambient PM concentrations (e.g., Betha *et al.*, 2014; Behera *et al.*, 2015a; Velasco and Rastan, 2015; Huang *et al.*, 2016; Karthik *et al.*, 2017; Urbancok *et al.*, 2017), or focused on IAQ in mechanically ventilated buildings (Zhou *et al.*, 2015; Chen *et al.*, 2016). Therefore, the health impact of

*Corresponding author.

Tel.: 65-65165135, Fax: 65- 68725483
E-mail address: ceerbala@nus.edu.sg

smoke haze episodes on occupants in naturally ventilated residential buildings in a densely populated country like Singapore remains largely unknown.

Airborne PM causes harmful health effects which are dependent on particle size, with different health effects being associated to different particle diameters (WHO, 2013). PM can be categorized as coarse (aerodynamic diameter (AED) $\geq 2.5 \mu\text{m}$), fine ($2.5 \mu\text{m} \geq \text{AED} \geq 100 \text{ nm}$) and ultrafine particles ($\text{AED} \leq 100 \text{ nm}$) (Seinfeld and Pandis, 1998). Smoke haze episodes increase the mass concentration of PM, especially particles in fine and ultrafine ranges, following their emissions, aerosol aging and mixing processes on the pathway between their sources and their inhalation by humans (Betha *et al.*, 2014; Behera *et al.*, 2015a, b). Previous epidemiological and toxicological studies have linked PM to a wide range of adverse health effects on the human cardiovascular and respiratory systems and therefore, premature mortality on a global scale (e.g., Pope III and Dockery, 2006; Russell and Bert, 2009; Cassee *et al.*, 2013; Apte *et al.*, 2015; Lelieveld *et al.*, 2015). Coarse PM has adverse impacts mostly on the upper respiratory system (Perez *et al.*, 2008), while ultrafine particles can translocate to the circulatory system and induce inflammation (Oberdorster, 2001). Therefore, it is important to study the distributions of size-resolved PM in both indoors and outdoors simultaneously, especially during severe air pollution episodes. Moreover, the PM size-resolved data improves our understanding of the deposition pattern of PM of different sizes in the human respiratory system (Haber *et al.*, 2003; Betha *et al.*, 2014; Behera *et al.*, 2015a).

PM consists of various chemical components, out of which trace elements (TEs) play a significant role in inducing health effects (Adamson *et al.*, 2000; Birmili *et al.*, 2006; Zhang *et al.*, 2017). Recent studies on *in vivo/in vitro* toxicity assessments and human historical data analyses suggest that TEs can produce reactive oxygen species (ROS) in lung fluids via the Fenton reaction which exhibit a high oxidative stress potential, leading to adverse health effects associated with PM (e.g., Lippmann *et al.*, 2007; Lippmann and Chen, 2009; Verma *et al.*, 2014; Lu *et al.*, 2011; Liberda *et al.*, 2015; Lu *et al.*, 2015). However, the overall health impacts depend on the severity of smoke haze, total concentrations of toxic elements and their bioavailability (Karthikeyan *et al.*, 2006a, b). The water-soluble fraction of TEs is considered to represent their bioavailability (Birmili *et al.*, 2006; Karthikeyan *et al.*, 2006a, b; Manousakas *et al.*, 2014). Estimating the bioavailability of size-fractionated particulate-bound TEs is an important step in assessing accurate potential human health risks. However, to the best of our knowledge, no scientific reports are available in the literature from SEA on the assessment of potential human health risk based on bioavailable concentrations of PM-bound TEs measured simultaneously both indoors and outdoors during transboundary haze episodes in SEA. Additionally, health risk evaluations are done based on a number of assumptions, which may introduce uncertainties in exposure parameters and limitations in the risk assessment (USEPA, 1992; Megido *et al.*, 2017; Shao *et al.*, 2017). Uncertainties exist in the estimation of exposure duration and individuals'

characteristics as well as exposure concentrations and toxicity data for the potentially toxic elements in PM. Therefore, uncertainty analysis is very crucial for exposure assessment so that health risk results can be more realistic. Generally, Monte Carlo (MC) technique is used for probabilistic uncertainty analysis of parameters and results can provide the next level of refinement as well as extreme values of output (USEPA, 1992; Gentry *et al.*, 2017). This can further facilitate the policymakers to make decisions.

In this study, simultaneous measurements of size-fractionated PM in indoor and outdoor air were conducted in a naturally ventilated residential apartment during the 2015 haze in Singapore. The key goal of our study was to investigate the relationship between indoor and outdoor size-fractionated PM and their potential health risk under the three different levels of haze i.e., light-haze (Pollutant Standards Index (PSI) 50–100), moderate-haze (PSI 100–150) and severe-haze (PSI > 150); the description of PSI is given in supplementary information (SI). The collected PM samples were analyzed for total and water-soluble toxic elemental concentrations to estimate size-fractionated potential human health risk associated with exposure to both indoor and outdoor PM. Uncertainty analysis of potential carcinogenic human health risk was also conducted, and extreme values were calculated during haze episodes. Additionally, airway deposition in human respiratory system was evaluated to get insights into the fate of inhaled size-fractionated PM during different smoke haze levels. This is the first study of its kind that provides a scientific evaluation of potential human health risk due to inhalation of PM_{2.5} and size-fractionated PM-bound TEs in naturally ventilated indoor environments during haze episodes with different intensities in Singapore. Based on our results, appropriate remedial actions can be taken to improve IAQ and mitigate its adverse health impacts.

METHODOLOGY

Description of Site and Sampling

Size-fractionated PM samples were collected on the 2nd floor of a residential apartment building located nearby the National University of Singapore (NUS) main campus in Singapore, approximately 50 m from a busy expressway. The PM sampling was conducted during October 2015 for seven days, out of which two days had light-haze levels (atmospheric visibility < 8 km); three days had moderate-haze levels (atmospheric visibility < 6 km); and the remaining two days had severe-haze levels (atmospheric visibility < 3 km). Personal Cascade Impactor Samplers (PCIS; Misra *et al.*, 2002) connected to SKC Leland Legacy pumps, operating at 9 L min⁻¹ (liters per minute), were used for collection of size-fractionated PM onto PTFE (Polytetrafluoroethylene) filters. All SKC Leland Legacy pumps were calibrated using Defender 510 calibrator before sampling to maintain the desired flow rate of 9 L min⁻¹.

Each PCIS collected size-fractionated PM in the five size ranges: > 2.5 μm (stage A), 1.0–2.5 μm (stage B), 0.50–1.0 μm (stage C), 0.25–0.50 μm (stage D) and < 0.25 μm (stage E). In this study, PM with diameter > 2.5 μm refers

to coarse particles, PM with diameter 0.25–2.5 μm and < 0.25 μm represent accumulation mode particles and q-UF particles, respectively (Viana *et al.*, 2015). The collection substrates were Pall PTFE or Teflon-fibre filters, 25 mm in diameter for coarse and accumulation stages, and 37 mm for the q-UF particle stage. Two PCIS units were placed simultaneously (one inside the apartment and the other one outside the apartment) and operated for 24 h for seven days. The indoor PM sampling was conducted in the living room area of the apartment (approximately 1.5 m above the floor) away from openings and nearby walls, and the outdoor sampling site was located outside the window. During the sampling period, the apartment windows were kept open and occupants were requested to fill questionnaire about their indoor activities.

Gravimetric and Chemical Analysis

PTFE filters were equilibrated in a controlled chamber ($T = 20 \pm 1^\circ\text{C}$, $\text{RH} = 30 \pm 5\%$) for 24 h both before and after sampling, and then weighed by an electronic microbalance balance with a sensitivity of 1 μg (Sartorius MC5, German). All PTFE filters were analyzed for both total and water-soluble elements analysis. Half of each filter was subjected to microwave-assisted extraction to determine the contents of trace elements in size-fractionated PM by inductively coupled plasma mass spectrometry (ICP-MS) as reported in our previous studies (Karthikeyan *et al.*, 2006b; Behera *et al.*, 2014). The remaining half of PTFE filters was used for water-soluble analysis of trace elements. 24 elements (Al, As, Ba, Be, Bi, B, Cd, Cs, Cr, Co, Cu, Ga, Fe, Pb, Li, Mn, Ni, Rb, Se, Sn, Te, Tl, Sn, and Zn) were analyzed using ICP-MS (Agilent 7700 Series, Agilent Technologies, USA) to determine their total and water-soluble concentrations. Further details about the analytical method and QA/QC protocols are provided in SI (Eq. (S1)).

Estimation of Deposition Doses in the Respiratory System

The deposition patterns along the human respiratory system (i.e., head airways, tracheobronchial and pulmonary regions) were estimated using PM mass concentration data through Eq. (1), as reported by Azarmi and Kumar (2016); Kumar and Goel (2016) and Segalin *et al.* (2017):

$$RDD = (VI \times f) \times DF_{ij} \times PM_i \quad (1)$$

where, VT is a tidal volume ($\text{m}^3 \text{ breath}^{-1}$), f is the typical breath frequency ($\text{breath minute}^{-1}$), DF_{ij} is deposition fraction of a size fraction i for and a region j , and PM_i is the mass concentration of a size fraction i . The DF_{ij} was estimated using equations recommended by Hinds (1999) and Megido *et al.* (2016) and the details are provided in SI (Eqs. (S2)–(S5)). To simulate the deposition in the respiratory system of an average human adult, the following assumptions were made: breathing frequency of 20 $\text{breaths minute}^{-1}$ and $12.5 \times 10^{-4} \text{ m}^3$ of tidal volume for slight exercise conditions in both indoors and outdoors. Additionally, the deposition dose calculations were carried out for $\text{PM}_{2.5}$ as well as for size-fractionated PM.

Potential Human Health Risk Assessment

USEPA models, as used by earlier studies (Betha *et al.*, 2014; Lu *et al.*, 2014; Behera *et al.*, 2015b), were used to estimate potential human health risk for adults for the three haze levels for $\text{PM}_{2.5}$ as well as size-fractionated PM. The results are helpful to analyze health hazards associated with inhalation exposure to PM during the hazy days. Two types of potential health risk, (1) non-carcinogenic (for Al, As, Be, Cd, Co, Cr, Cu, Mn Ni, Pb and Zn) 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 (Lu *et al.*, 2014; ATSDR, 2017; USEPA, 2017). Further details about the calculation of HQ and ELCR and the assessment of potential health risk are given in SI (Tables S1–S2; Eqs. (S6)–(S10)).

Uncertainty Analysis for Potential Carcinogenic Health Risk

In order to refine the potential carcinogenic health risk and assess extreme values, uncertainty analysis of exposure parameters was conducted by applying MC simulations (1000 times) for probabilistic quantification (USEPA, 1992; Gentry *et al.*, 2017). Variability of exposure parameters is a very important factor for human health risk assessment. Therefore, five input exposure parameters (as defined in SI), namely, exposure frequency (EF), exposure duration (ED), average life time (AT), inhalation rate (IR) and body weight (BW) were considered variables. To estimate probabilistic health risk, these parameters were assumed to follow lognormal distribution and MC simulations were performed using Python language (Chiu and Slob, 2015; Shao *et al.*, 2017). Results obtained from the output file were then analyzed to estimate 50th and 95th percentile values of potential health risk. The uncertainty assessment was conducted for both $\text{PM}_{2.5}$ and size-fractionated PM. More details about uncertainty analysis are provided in SI (Table S3).

RESULTS AND DISCUSSION

Characteristics of the 2015 Haze Episode

To analyze the sources and transport of smoke haze-impacted air masses, backward air trajectories were generated using the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Hess, 1998; Draxler and Rolph, 2015) for the three haze levels. As shown in Fig. 1, the haze map and backward air trajectories showed that the air mass parcels arriving in Singapore originated from the Indonesian islands of Sumatra and Kalimantan during the severe-haze period. The backward trajectories were generated for the three haze levels over a period of 72 h back in time. For each haze level, trajectories can be clustered into three groups, marked by the red, blue and green lines corresponding to different altitudes. The red line represents above boundary layer (1500 m), blue line represents below boundary layer (500 m), and green line shows surface level (100 m). It can be observed from Fig. 1 that for the severe-haze period, both 1500 m and 500 m trajectories originated from Indonesian islands. Therefore, the air quality at those altitudes was significantly affected

Fig. 1. Haze map and backward trajectory analysis using HYSPLIT model for severe-haze level on 23 October 2015. The haze map is reproduced from weblink <http://asmc.asean.org/home/> (with permission from ASEAN Specialized Meteorological Centre (ASMC)).

by the wild fires in Indonesia while the 100 m trajectory originated from the open Java Sea. Similarly, haze maps and backward air trajectories for light-haze and moderate-haze levels were constructed and are shown in SI (Figs. S1 and S2). It can be observed in Figs. S1 and S2 that as the haze intensity decreased, 500 m trajectories showed deviations in their origin, shifting from land to sea areas. In addition, only 1500 m trajectories were observed to originate from Kalimantan Island for the light-haze, suggesting possible reduction in the haze intensity during this period.

Assessment of Indoor-Outdoor PM Relationship under Different Haze Levels *PM_{2.5}*

Indoor and outdoor $PM_{2.5}$ concentrations measured were analyzed for the three haze levels together with reference to the 24 h WHO $PM_{2.5}$ air quality guidelines as shown in Fig. 2. 24 h average indoor $PM_{2.5}$ concentrations during light-haze, moderate-haze and severe-haze levels were observed to be 42, 88 and $121 \mu g m^{-3}$, respectively while the 24 h average outdoor $PM_{2.5}$ concentrations for the respective haze levels were 47, 101 and $134 \mu g m^{-3}$. It is thus clear that the $PM_{2.5}$ concentration increased both indoors and outdoors with an increase in haze intensity. Another important finding is that the 24 h average $PM_{2.5}$ levels exceeded the 24 h mean $PM_{2.5}$ WHO guidelines of $25 \mu g m^{-3}$ indoors and outdoors by several folds during the three haze levels, raising concerns over potential public health impacts. It should be noted that the exceedance from the WHO guidelines was about 5-folds during severe-haze levels for both indoors and outdoors. Furthermore, the pie charts clearly show that more than 50% of $PM_{2.5}$ particles comprised the lowest size particles ($PM < 0.25 \mu m$) which are most harmful to humans upon inhalation (Betha *et al.*, 2014; Behera *et al.*, 2015a, b). Additionally, Fig. S3 shows variations of indoor to outdoor particle mass ratios (I/O ratios) for $PM_{2.5}$ on different hazy days with the mean I/O ratio being 0.9. The I/O ratio being closer to 1 indicates that remaining indoors in

naturally ventilated buildings may also be as harmful as remaining outdoors during the haze period.

Size-fractionated PM

Using PCIS, size-fractionated PM was collected both indoors and outdoors during the three haze levels and their variations are shown in Fig. 3. The graphs in Fig. 3 demonstrate that haze particles were more dominant both indoors and outdoors in stage D ($PM_{0.25-0.5}$) and stage E ($PM_{<0.25}$) compared to other stages. Also, a higher proportion of submicron-sized particles was observed with an increase in haze intensity, as reported during previous haze episodes (Betha *et al.*, 2014; Behera *et al.*, 2015a). Furthermore, box plots in Fig. S4 for size-resolved particle mass concentrations for the entire sample set show that the PM mass concentration increased with a decrease in particle size and the highest PM concentration was observed in the stage E (q-UF particles): $80 \mu g m^{-3}$ for indoors and $85 \mu g m^{-3}$ for outdoors. Elevated levels of q-UF particle are a matter of concern due to their deleterious health effects on humans (Betha *et al.*, 2014; Behera *et al.*, 2015a, b).

Fig. 4 illustrates a general trend for average I/O ratios as a function of different particle sizes. It can be observed that the I/O ratio increased as the particle size decreased. In addition, the I/O ratio was found almost 1 for stage E i.e., q-UF particles, suggesting that even remaining indoors may not be safer for occupants of naturally ventilated buildings and remedial actions are required during haze periods. Detailed variations of I/O ratios with the particle size during each hazy day are shown in Fig. S5.

Deposited Doses in the Respiratory System *PM_{2.5}*

$PM_{2.5}$ deposited doses were calculated for the three regions of human respiratory system i.e., head airways, tracheobronchial and alveolar regions as illustrated in Fig. S6. Overall, $PM_{2.5}$ deposited doses were found higher for outdoors than indoors and increased with an increase in

Fig. 2. PM_{2.5} mass concentration for the three haze levels for indoors and outdoors with its comparison to the WHO guideline. B, C, D and E are stages in PCIS which refer to PM_{1.0-2.5}, PM_{0.5-1.0}, PM_{0.25-0.5} and PM_{<0.25}, respectively.

Fig. 3. Indoor and outdoor PM concentrations in various sizes under different levels of haze as per PSI values. A, B, C, D and E are PM_{>2.5}, PM_{1.0-2.5}, PM_{0.5-1.0}, PM_{0.25-0.5} and PM_{<0.25}, respectively.

haze intensity due to larger emissions of particles during moderate-haze and severe-haze events (Lai *et al.*, 2009). Furthermore, it was observed that the head airway had larger deposition fractions of PM_{2.5} as compared to the other two regions during the three haze levels (100% in light-haze, 80% moderate-haze and 80% in severe-haze). The head deposition happens due to a combination of sedimentation and the impaction of particles onto the larynx and airway bifurcations (Zhang and Yu, 1993) so these processes are likely to be more influential ones than interception and diffusion. More interestingly, the fraction of PM_{2.5} deposited in alveolar region increased with an

increase in haze intensity due to higher sub-micron particle concentration in higher haze periods.

Size-fractionated PM

Estimation of deposited doses of size-fractionated PM was also conducted for the three regions of human respiratory system as presented in Fig. 5. Again, the deposited dose was observed higher for outdoors than indoors and showed an increase with an increase in haze intensity. Additionally, it is interesting to note that as the particle size decreased, the contribution of deposition in the head region to the total deposition decreased while it increased for the alveolar region

Fig. 4. Average I/O ratios in various sizes. A, B, C, D and E are $PM_{>2.5}$, $PM_{1.0-2.5}$, $PM_{0.5-1.0}$, $PM_{0.25-0.5}$ and $PM_{<0.25}$, respectively.

Fig. 5. Deposited dose in the three zones of respiratory system for size-fractionated PM under three haze levels for indoors and outdoors. L, M and S refer to light-haze, moderate-haze and severe-haze levels, respectively.

for a particular haze level. In the case of severe-haze, the deposited doses were 75% in head, 7% in tracheobronchial and 18% in alveolar for $PM_{2.5-1}$ while the composition was 14% in head, 13% in tracheobronchial and 73% in alveolar for $PM_{<0.25}$ for both indoors and outdoors. The main reason for more deposition of submicron-sized particles in the alveolar region was due to the flow path of smaller particles in the human respiratory system, which is primarily governed by Brownian diffusion leading to preferential deposition in the alveolar region (Ingham, 1984, Martins *et al.*, 2010, Ham *et al.*, 2011). Another important observation was that the deposition of $PM_{2.5-1}$, $PM_{1.0-0.5}$, $PM_{0.5-0.1}$, and $PM_{<0.25}$ in tracheobronchial and alveolar regions increased, while it decreased in the head airway region during the severe-haze levels as compared to light-haze and moderate-

haze levels, which can be attributed to enrichment of submicron-sized particles in the accumulation mode during the severe-haze period.

Chemical Composition of Indoor-Outdoor PM

Altogether 24 TEs in coarse, accumulation and q-UF PM modes were measured for indoors and outdoors for the three haze levels. Their mean concentrations and the corresponding standard deviations are shown in Figs. S7–S9. For this purpose, the datasets were divided into three distinct categories based on their respective concentrations in the different PM size ranges (Behera *et al.*, 2015b). This classification assumed that (i) TEs with concentrations $> 80 \text{ ng m}^{-3}$ were the major elements, (ii) TEs with concentrations between 1 and 80 ng m^{-3} were the sub-major

elements, and (iii) TEs with concentration $< 1 \text{ ng m}^{-3}$ were the minor elements.

For all the three haze levels, it was observed that most of the TEs were predominantly found either in accumulation or q-UF PM modes. These trends suggest that the emission of these TEs could be associated with anthropogenic sources of combustion such as smoke haze and/or on-road vehicles, industries (power plants, and chemical and other industries) and other domestic sectors (See *et al.*, 2007; Betha *et al.*, 2014; Kalaiarasan *et al.*, 2017). However, Sr was mostly present in the coarse mode and the reason for this discrepancies may be due to the difference in its emission sources, since Sr is thought to be derived from soil dust (peat soil) (See *et al.*, 2007; Betha *et al.*, 2014). It is noteworthy that Zn, which is observed as a major element with anthropogenic origin, showed the highest concentration during light-haze and moderate-haze levels while its concentration was relatively lower during severe-haze levels. This finding suggests that Zn is likely to originate due to local traffic emissions, considering that the sampling location was not too far from the local expressway (Zhang *et al.*, 2017). However, the contribution of local traffic emissions to the PM loading in urban air should be less significant during the severe-haze period, leading to a decrease in its concentration. Furthermore, a number of major elements were observed during moderate-haze and severe-haze levels, indicating that smoke haze increases the concentration of PM-bound TEs.

Enrichment factor (EF) analysis has been done for selected trace elements in the size-fractionated PM samples collected indoors and outdoors during light-haze, moderate-haze and severe-haze levels for differentiating sources of PM (Mason, 1966; Balasubramanian and Qian, 2004). Relevant details about the analysis have been provided in SI (Figs. S10–S11). It is noteworthy that the EF value for Zn was found to be quite high ($\text{EF} > 100$) (Fig. S10), suggesting that it has dominant non-crustal sources across all size ranges indoors and outdoors for the three haze levels. Furthermore, Cd, Cu and Pb were found to be enriched in almost all the size-fractions during the three haze levels indoors, indicating that non-crustal sources are the major contributors for these elements (Fig. S11). However, Cd, Cu and Pb showed the $\text{EF} > 10$ in q-UF particles predominantly for outdoors during all haze levels, suggesting that smaller particles originate from non-crustal sources. Fe and Mn showed EF close to 1 for both indoors and outdoors for the three haze levels, suggesting that they are predominantly of crustal origin. However, Cr and Ni appear to be of mixed origin (crustal and non-crustal sources) as they showed $\text{EF} < 10$ for coarse and accumulation modes while $\text{EF} > 10$ for their q-UF particle mode for both indoors and outdoors for the three haze levels as demonstrated in Fig. S11. Since the primary sources of smoke haze in Indonesia are peat and agricultural land fires, Cr and Ni could have been emitted from peat soil as well as biomass burning.

Probability distribution function (PDF) analysis, as reported by (Pongpiachan and Iijima, 2016), was performed for 24 selected elements in $\text{PM}_{2.5}$ collected indoors and outdoors

during the light-haze, moderate-haze and severe-haze levels in Singapore. The PDF results are shown in Figs. S12–S13. $\text{PM}_{2.5}$ was selected for the temporal and spatial distribution analysis as ambient air quality standards/indoor air quality guidelines currently exist for this criteria air pollutant. More details about the analysis are provided in SI. As shown in Fig. S13, elements such as Cr, Fe, Ni, Mn (indoors) and Cs displayed a sharp symmetrical bell-shaped curve for both indoors and outdoors. As the observed values of the parameter are more concentrated in the middle than in the tails, it seems reasonable to assume a considerably strong homogeneous distribution of these elements of crustal origin in $\text{PM}_{2.5}$, which were not significantly influenced by human activities. This observation is consistent with their corresponding EF values shown in Fig. S11, confirming the crustal emissions of these elements. Furthermore, Al, Be, Mn (outdoors) and Rb showed relatively broad peaks of positively skewed distributions, emphasizing the higher degree of means than modes. The Al and Rb curves with positive skewedness are consistent with their temporal distribution patterns as illustrated in Fig. S12, indicating the common sources of their natural emissions, associated with the geographic location of Singapore. The findings of this study are consistent with those from a previous study reported by Pongpiachan and Iijima, (2016).

As, B, Ba (outdoors), Bi, Cd, Cu, Pb, Sn, Sr (outdoors), Te, Tl and Zn demonstrated a sharp positive skewedness in their PDFs, indicating that these PM-bound elements existed during the study period with their concentrations being less than their corresponding mean values most of the time. These asymmetrical distribution curves are comparable for most of the elements with their own temporal distribution patterns and thus underline the importance of anthropogenic activities such as the emission of these elements from the local traffic (Figs. S12 and S13). These findings are consistent with the values of EFs > 10 obtained previously for Cd, Cu, Pb and Zn (Figs. S10–S11) which also showed that these elements were emitted from non-crustal sources. Moreover, elements such as Co, Ga (indoors), Li, Se and Sr (indoors) showed their PDFs with negative skewedness, suggesting that they may arise either from smoke haze or from some indoor activities such as cooking and sweeping.

ANOVA test has been performed to find out if there is any statistically significant difference in the relative abundance of the 24 selected trace elements among the three different sampling periods (i.e., light, moderate and severe-haze pollution). Results revealed that there was no significant variation in the PM-bound elemental concentrations with respect to the three haze levels across different size ranges. The lack of variations can be attributed to the fact that other local urban emissions sources of PM also make a substantial contribution to the PM-bound elemental concentrations. Student T-test was also conducted to investigate if there was any statistically significant difference in the abundance of the 24 PM-bound TEs collected at outdoor and indoor environments. However, no significant difference was observed ($p > 0.05$). This implies that remaining indoors may not be safer than being outdoors during the smoke haze period in terms of exposure to PM-bound TEs. Additionally,

the I/O ratios for the trace elements were also examined using ANOVA. There was no significant difference observed among the three haze levels for coarse, accumulation and q-UF particles. Overall, it can be reasonably stated that smoke haze and other local PM emission sources contribute to PM-bound elemental concentrations and exposure to toxic elements indoors and outdoors may lead to serious public health concern, if no mitigation measure is taken into account.

Total Potential Human Health Risk for Indoor-Outdoor PM

The following section deals with the potential health risk results based on constant input parameters values assumed for adults. Two types of potential health risk i.e. non-carcinogenic and carcinogenic are discussed. Moreover, as explained earlier, PM was analyzed for the bioavailable concentrations of toxic elements to obtain a more realistic estimate of potential human health risk.

Potential Non-carcinogenic Health Risk for PM_{2.5}

The results of potential non-carcinogenic health risk assessment for the three haze levels due to inhalation exposure to PM_{2.5} in both indoor and outdoor environments are shown in Table 1. PM_{2.5} is especially important to consider since it is a commonly regulated criteria air pollutant as part of ambient air quality standards. The potential health risk assessment will be helpful in understanding the health implications associated with exposure to PM_{2.5} under different haze levels so that appropriate mitigation measures can be suggested. The potential non-carcinogenic risk assessment showed that the HQ values obtained for PM_{2.5} for total potential health risk were well within the acceptable upper limit. However, there might be a long-term chronic health effect due to continued indoor and outdoor exposure, if hazy atmospheric conditions persist in the future. The potential non-carcinogenic health risk was found to increase with increase in haze intensity. Moreover, Mn and Cr showed relatively higher HQ values, indicating their contributions to higher potential non-carcinogenic health risk.

Potential Carcinogenic Health Risk

PM_{2.5}

Since PM_{2.5} is a regulated air pollutant, as explained in the previous sections, therefore it was also analyzed for potential carcinogenic health risk assessment. The results for this assessment for the three haze levels due to inhalation exposure to PM_{2.5} in both indoor and outdoor environments are summarized in Table 2. It can be observed that for total carcinogenic risk assessment, ELCR values exceeded the threshold value of 1×10^{-6} under all the three haze levels. At the same time, individual potential cancer health risks for indoors and outdoors were also observed to exceed the allowed threshold. Therefore, there is potential life time cancer risk due to the continual exposure to PM_{2.5} under haze conditions. The total potential human health risk estimated for occupants during the severe-haze levels was 5.24×10^{-6} , indicating 5 to 6 people out of a population of 1 million individuals exposed to the indoor and outdoor air during severe-haze events in Singapore are likely to develop cancer in their lifetime due to inhalation of toxic elements in PM_{2.5} and thus have shorter life expectancy. Similarly, the total potential cancer health risk was 4.4 and 2.8 times higher than the threshold value during moderate-haze and light-haze levels, respectively. It is important to note that the ELCR was observed to be the highest for Cr followed by As. Moreover, the potential carcinogenic health risk for Cr exceeded the threshold limit for the three haze levels indoors and outdoors. This finding is in agreement with that reported in a previous study by Jiang *et al.* (2017) which indicated higher carcinogenic health risk due to PM_{2.5}, especially due to Cr. The results of this study are also comparable to those from the 2015 haze study in Malaysia by Sulong *et al.* (2017). However, the latter study showed health risk results somewhat higher which may be due to only outdoors concentration being taken into account with no consideration of deposition fraction for PM_{2.5}. The potential carcinogenic health risk estimates for PM_{2.5} reflected higher health risk with an increase in the intensity of smoke haze. The exceedance of the potential health risk

Table 1. Total potential non-carcinogenic health risk estimates for indoors and outdoors of PM_{2.5} for the three haze levels.

Element	RfD (mg kg ⁻¹ day ⁻¹)	HI for PM _{2.5}					
		Light-Haze		Moderate-Haze		Severe-Haze	
		Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
Al	1.40×10^{-3}	7.42×10^{-5}	4.97×10^{-5}	9.82×10^{-5}	6.62×10^{-5}	1.07×10^{-4}	9.47×10^{-5}
As	3.01×10^{-4}	3.26×10^{-5}	1.30×10^{-5}	5.33×10^{-5}	2.64×10^{-5}	5.45×10^{-5}	2.90×10^{-5}
Be	5.71×10^{-6}	1.42×10^{-5}	2.72×10^{-6}	9.77×10^{-5}	1.99×10^{-5}	1.90×10^{-6}	2.80×10^{-6}
Cd	5.71×10^{-6}	2.32×10^{-4}	7.81×10^{-5}	2.18×10^{-4}	1.03×10^{-4}	4.26×10^{-4}	3.12×10^{-4}
Co	5.71×10^{-6}	1.32×10^{-4}	7.46×10^{-5}	1.33×10^{-4}	7.31×10^{-5}	1.04×10^{-4}	8.37×10^{-5}
Cr	2.86×10^{-5}	1.20×10^{-3}	8.68×10^{-4}	2.02×10^{-3}	1.22×10^{-3}	2.59×10^{-3}	1.35×10^{-3}
Cu	4.02×10^{-2}	1.09×10^{-6}	6.81×10^{-7}	1.36×10^{-6}	7.67×10^{-7}	3.29×10^{-6}	2.17×10^{-6}
Mn	1.43×10^{-5}	2.99×10^{-3}	1.21×10^{-3}	4.13×10^{-3}	2.10×10^{-3}	5.05×10^{-3}	3.47×10^{-3}
Ni	2.06×10^{-2}	2.77×10^{-6}	1.62×10^{-6}	3.03×10^{-6}	1.73×10^{-6}	1.84×10^{-6}	1.61×10^{-6}
Pb	3.52×10^{-3}	2.20×10^{-6}	4.99×10^{-7}	1.73×10^{-6}	7.87×10^{-7}	2.62×10^{-6}	1.85×10^{-6}
Zn	3.00×10^{-1}	7.83×10^{-7}	3.87×10^{-7}	1.05×10^{-6}	5.16×10^{-7}	2.03×10^{-6}	1.31×10^{-6}
Σ =		4.69×10^{-3}	2.30×10^{-3}	6.76×10^{-3}	3.61×10^{-3}	8.35×10^{-3}	5.35×10^{-3}
Total Potential Non-Carcinogenic Health Risk		6.99×10^{-3}		1.04×10^{-2}		1.37×10^{-2}	

Table 2. Total potential carcinogenic health risk estimates for indoors and outdoors of PM_{2.5} for the three haze levels.

Element	SF (kg day mg ⁻¹)	ELCR for PM _{2.5}					
		Light-Haze		Moderate-Haze		Severe-Haze	
		Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
As	15.1	1.48×10^{-7}	5.92×10^{-8}	2.42×10^{-7}	1.20×10^{-7}	2.48×10^{-7}	1.32×10^{-7}
Be	8.4	6.83×10^{-10}	1.30×10^{-10}	4.68×10^{-9}	9.56×10^{-10}	9.13×10^{-11}	1.34×10^{-10}
Cd	6.3	8.35×10^{-9}	2.81×10^{-9}	7.86×10^{-9}	3.70×10^{-9}	1.53×10^{-8}	1.12×10^{-8}
Co	31.5	2.37×10^{-8}	1.34×10^{-8}	2.39×10^{-8}	1.31×10^{-8}	1.87×10^{-8}	1.51×10^{-8}
Cr	42	1.45×10^{-6}	1.04×10^{-6}	2.43×10^{-6}	1.46×10^{-6}	3.11×10^{-6}	1.62×10^{-6}
Ni	0.84	4.79×10^{-8}	2.80×10^{-8}	5.24×10^{-8}	2.99×10^{-8}	3.18×10^{-8}	2.78×10^{-8}
Pb	0.042	3.26×10^{-10}	7.37×10^{-11}	2.56×10^{-10}	1.16×10^{-10}	3.88×10^{-10}	2.74×10^{-10}
$\Sigma =$		1.67×10^{-6}	1.15×10^{-6}	2.76×10^{-6}	1.63×10^{-6}	3.43×10^{-6}	1.81×10^{-6}
Total Potential Carcinogenic Health Risk		2.82×10^{-6}		4.39×10^{-6}		5.24×10^{-6}	

values from the threshold limit for the three haze levels suggests that it is harmful to stay in naturally ventilated buildings during the haze period. Consequently, appropriate mitigation measures need to be developed and implemented to decrease human exposure to PM_{2.5} mass concentrations and related health effects during haze episodes.

Size-fractionated PM

Since the potential carcinogenic health risk or ELCR values estimated for PM_{2.5} exceeded the threshold limit, ELCR values were also calculated for size-fractionated PM to get further useful insights. Results obtained from this analysis for the three haze levels for total potential health risk are illustrated in Fig. 6. Again, an increase in potential health risk was observed with the increase in haze intensity. Overall, potential health risk due to exposure to coarse particles was found to be within the threshold limit while submicron-sized particles (accumulation and q-UF modes) showed higher potential health risk than the threshold value, except for the accumulation mode during light-haze. Furthermore, it is important to note that the potential health risk was the highest for q-UF particles, followed by accumulation mode particles and then coarse particles. Since these submicron-sized particles are found in higher concentrations during haze episodes, their effective removal indoors through mitigation methods such as air filtration is warranted. Therefore, suitable mitigation measures such as the use of suitable air cleaners indoors and respirators outdoors can be employed for effective air filtration during smoke haze episodes. However, using effective air cleaners in homes is considered to provide greater health benefits than using respirators outdoors as reported by Qi *et al.* (2017).

Uncertainty Analysis for Potential Carcinogenic Health Risk

To refine the potential health risk assessment and perform extreme value analysis, the uncertainty analysis was conducted with variable exposure parameters for potential carcinogenic health risk assessment for PM_{2.5} as well as size-fractionated aerosols. For this purpose, five input exposure parameters i.e., EF, ED, AT, IR and BW were considered as variables as mentioned before in the methodology section.

Moreover, only potential carcinogenic health risk was considered in the uncertainty analysis as it was found to exceed the threshold limit.

PM_{2.5}

The analysis was conducted for the three haze levels for PM_{2.5} for total potential carcinogenic health risk values at 50th and 95th percentiles, as shown in Fig. 7. Overall, it was observed that the 50th percentile values for potential health risk exceeded the threshold limit by 2.7, 4.3 and 5.1 times for light-haze, moderate-haze and severe-haze levels, respectively. These values are quite close to the ones obtained in previous section for PM_{2.5} since mean values of distributions of parameters were assumed to be the same as constant values assumed in an earlier section. The values calculated for the 95th percentile were found to be 4.3, 9.2 and 11.5 times higher than the threshold limit; all these values are considerably higher than the allowed threshold. The values associated with the 95th percentile showed extreme values and are important for policy making. These results further indicate heightened concerns about public health impacts associated with exposure to PM_{2.5} during smoke haze episodes.

Size-fractionated PM

In addition to PM_{2.5}, the uncertainty analysis in potential carcinogenic health risk was also conducted for size-fractionated PM, and the obtained results are shown in Fig. S14. The results showed that 50th and 95th percentiles potential health risks were elevated by several folds for the three particles modes under the three haze levels. Again, the patterns showed the highest potential carcinogenic health risk for q-UF particles followed by accumulation mode and then coarse mode particles. It is noteworthy that the 95th percentile potential health risk was observed to be higher than the threshold limit for the severe-haze case in coarse mode, indicating that coarse mode particles can also cause deleterious health effects in the case of extreme values analysis. The uncertainty analysis provides more realistic evaluation of potential health risk, highlighting the need for implementation of effective mitigation efforts to reduce exposure of occupants in naturally ventilated buildings to PM, especially q-UF particles during smoke haze episodes.

Fig. 6. Total potential carcinogenic human health risk for indoors and outdoors for size-fractionated aerosols (coarse, accumulation and q-UF) for adults and its comparison with threshold value for the three haze levels.

Fig. 7. Uncertainty analysis for total potential carcinogenic human health risk for PM_{2.5} for indoors and outdoors and its comparison with threshold value for the three haze levels.

CONCLUSIONS

Ambient and indoor air quality in SEA were severely affected during the 2015 haze episode with a 5-fold increase in the 24 h mean PM_{2.5} concentration compared to the 24 h mean WHO PM_{2.5} guidelines. Size-fractionated analysis of PM_{2.5} showed that the increment in PM_{2.5} levels was mainly due to higher concentrations of q-UF particles. The I/O ratios of size-fractionated PM showed higher values for smaller-sized particles during the haze episodes compared to larger particles. This observation indicates that exposure to such PM may pose severe health impacts since small particles contain relatively higher levels of toxic elements than coarse particles. The estimation of deposited dose of PM in the human respiratory system showed that as the particle size decreased from PM_{2.5} to PM_{<0.25}, the contribution from head to alveolar regions to the total deposited dose

increased, suggesting that higher concentrations of q-UF particles can lead to their deeper penetration, thereby causing harmful health impacts during haze events. Total elemental analysis was performed for the 24 selected elements. EF and PDF analyses were used to differentiate sources of PM. Results showed that Cr, Mn Fe and Ni mainly originated from crustal sources while other elements such as Cd, Cu, Pb and Zn are mainly associated with non-crustal sources. Total potential carcinogenic health risk results estimated during the severe-haze period, based on bioavailable toxic elemental concentrations in PM_{2.5}, exhibited exceedance from the threshold limit by 5.2 times for constant exposure parameters based analysis for adults. The exceedance was observed to be 11.5 times higher compared to the allowed threshold for the extreme value (95th percentile) analysis conducted for potential health risk considering the uncertainty or the variability associated with exposure parameters.

Moreover, the results obtained for size-fractionated potential health risk revealed that the health risk increased with the decrease in particle size with accumulation and q-UF particles exceeding the threshold limit.

Overall, it was observed that the potential health risk increases with the increase in haze intensity. However, it should be noted that the potential health risk exceeded from the threshold limit for the three haze levels, implying that remaining in naturally ventilated environments may not be safer during haze events. Therefore, effective mitigation of indoor exposure to PM is much needed during the occurrence of transboundary smoke haze episodes. Mitigation measures such as the use of portable air cleaners with sufficient CADR (clean air delivery rate) indoors and suitable respirators equipped with micro-ventilators in outdoor environments during haze period can help achieve good health benefits. Additionally, the current study considers only toxic elements for potential health risk assessment since they make a significant contribution to the total health risk. However, organic components in PM such as polycyclic aromatic hydrocarbons (PAHs), nitrated-PAHs and quinones may also contribute toward the overall toxicity of PM and therefore should also be included in the overall health risk assessment. Consequently, more comprehensive studies focused on PM chemical speciation together with the use of simulated lung fluids containing antioxidants in place of water as the leaching agent and a comparative evaluation of various mitigation strategies are warranted to improve our understanding of the potential health effects of haze aerosols and to recommend a suitable indoor PM exposure mitigation method. Such studies will be helpful for policymakers to make appropriate decisions for maintaining good IAQ and minimizing public health impacts of haze aerosols.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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