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Indoor $PM_{0.1}$ and $PM_{2.5}$ in Hanoi: Chemical characterization, source identification, and health risk assessment

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ABSTRACT

This study attempted to provide comprehensive insights into the chemical composition, source identification, and health risk assessment of indoor particulate matter (PM) in urban areas of Vietnam. Three hundred and twenty daily samples of $PM_{0.1}$ and $PM_{2.5}$ were collected at three different types of dwellings in Hanoi in two seasons, namely summer and winter. The samples were analyzed for 10 trace elements (TEs), namely Cr, Mn, Co, Cu, Ni, Zn, As, Cd, Sn, and Pb. The daily average concentrations of indoor $PM_{0.1}$ and $PM_{2.5}$ in the city were in the ranges of 7.0–8.9 μg/m^3 and 43.3–106 μg/m^3, respectively. The average concentrations of TEs bound to indoor PM ranged from 66.2 ng/m^3 to 216 ng/m^3 for $PM_{0.1}$ and 391 ng/m^3 to 2360 ng/m^3 for $PM_{2.5}$. Principle component analysis and enrichment factor were applied to identify the possible sources of indoor PM. Results showed that indoor $PM_{2.5}$ was mainly derived from outdoor sources, whereas indoor $PM_{0.1}$ was derived from indoor and outdoor sources. Domestic coal burning, industrial and traffic emissions were observed as outdoor sources, whereas household dust and indoor combustion were found as indoor sources. 80% of $PM_{2.5}$ was deposited in the head airways, whereas 75% of $PM_{0.1}$ was deposited in alveolar region. Monte Carlo simulation indicated that the intake of TEs in $PM_{2.5}$ can lead to high carcinogenic risk for people over 60 years old and unacceptable non-carcinogenic risks for all ages at the roadside house in winter.

1. Introduction

The average person spends 90% of their time indoors (in homes, schools, offices, etc.), where ultrafine particles or nanoparticles ($PM_{0.1}$) and fine particles ($PM_{2.5}$) are known to cause health problems (Kearney et al., 2014; Madureira et al., 2020). Therefore, poor indoor air quality, including indoor particulate matter (PM) pollution, can pose significant health risks. Household air pollution was ranked as the 10th greatest risk factor for mortality in 2019 (GBD Global Burden Disease, 2020). Approximately 4 million premature deaths associated with pneumonia, stroke, ischemic heart disease, chronic obstructive pulmonary disease, and lung cancer worldwide in 2016 were attributed to household air pollution (WHO, 2018). Therefore, indoor air pollution has received increasing attention from researchers in recent decades. Many studies have investigated different aspects of indoor PM, including mass distribution, spatial variation, indoor/outdoor ratios, chemical composition, potential sources, and health risk assessment (Kearney et al., 2014; Kulshrestha et al., 2014; Madureira et al., 2020; Massey et al., 2012; Sidra et al., 2015; Sharma and Balasubramanian, 2018; Tran et al., 2021a; Zhao et al., 2020). However, studies on the chemical characterization of $PM_{0.1}$ are scarce, especially in Southeast Asian countries. To the best of our knowledge, few studies on indoor PM in this region are available in open databases, such as those by Aung et al. (2019), Choo and Jalaludin (2015), Klinmalee et al. (2009), Lomboy et al. (2015), Mengersen et al. (2011), and Tran et al. (2021a). Most of these studies focused on examining the characteristics of PM_{10} and $PM_{2.5}$ linked with health effects in different indoor environments, although recent studies by Tran et al. (2021a) and Sharma and Balasubramanian (2018) conducted the chemical characterization of $PM_{2.5}$ and other particle sizes in indoor environments in Singapore during haze periods. However, there is still a knowledge gap on the chemical characterization of indoor PM, especially $PM_{0.1}$, and their seasonal variation,

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source identification, and health risk assessment in this region.

Indoor air quality is governed by many factors, such as climate, the characteristics of the soil where the building is located, outdoor sources, building characteristics (building design, operation, and maintenance; construction materials; and ventilation), cultural tastes and human activities (cooking, incense burning, smoking, etc.), and different intervention strategies (Godish, 2001). Therefore, indoor air quality can vary from country to country, and even between different cities in the same country. Hanoi has a different climate than most other large cities in Southeast Asia, and even those in the south of Vietnam, because of its cold winters. For example, the average temperature in winter in Hanoi is below 20 °C (Nguyen and Nguyen, 2004), whereas in the south of Vietnam, it is hot throughout the year with an average annual temperature of approximately 20–35 °C (Kontgis et al., 2019). As a result, the indoor air quality in Hanoi can be affected differently than that in other cities in this region. Moreover, over the past two decades, Hanoi has shown high concentrations of outdoor PM, especially $PM_{0.1}$ and $PM_{2.5}$, compared with those of other cities in this region (Kim Oanh et al., 2006; Nguyen et al., 2018; Phung et al., 2021). In addition, it has been reported that serious air pollution caused more than 3000 deaths in Hanoi in 2009 (Hieu et al., 2013) and increased respiratory admissions among young children in the city from 2010 to 2011 (Luong et al., 2017). Therefore, indoor air quality in general and indoor PM pollution in particular in Hanoi need to be studied.

There are very few studies on indoor PM in Hanoi. The following are those found in open databases. Tran et al. (2017) focused on measuring the number concentration of $PM_{0.1}$ in different households in Hanoi without determining the chemical compositions. Another study by Tran et al. (2020) dealt with the mass distribution and elemental compositions of PM of different sizes in school environments in Hanoi. However, source identification and health risk assessments were not conducted in this study. Two other studies were performed on $PM_{2.5}$; one quantified the concentration of indoor $PM_{2.5}$ associated with incense burning in residential houses (Tran et al., 2021b), and the other analyzed polycyclic aromatic hydrocarbons (PAHs) in $PM_{2.5}$ in the school environment (Vo et al., 2020a). However, these studies mainly focused on the concentration of $PM_{2.5}$, and no attention was devoted to $PM_{0.1}$. To the best of our knowledge, no studies have been conducted on the quantification of the chemical distribution in respiratory tract regions and health risk assessments in different seasons. Thoroughly interpreting the exposure to indoor PM and their chemical compositions is vital for assessing indoor air quality and evaluating health risks to avoid adverse health impacts. Therefore, this study was designed to address this gap. The objective of this study was to characterize indoor $PM_{0.1}$ and $PM_{2.5}$ in Hanoi focusing on their chemical composition, source identification, and health risk assessment.

2. Methods

2.1. Description of the sampling area

Sampling was conducted in residential dwellings in Hanoi, which is the second largest city in Vietnam with a population of more than 7.5 million people and a maximum density of 40,300 inhabitants/km2 in the central districts (GSO, 2018). The climate of this region is characterized by two monsoon seasons, namely the northeast monsoon in winter (November to March) and the southeast monsoon in summer (May to September) (Phung et al., 2021; Vo et al., 2020b). Three buildings (K1, K2, and K3) in the city were selected to represent residential dwellings. K1, representing an urban periphery dwelling, was located in Long Bien inner district, northeast Hanoi, and strongly impacted by heavy traffic activities (upper ring road #2) and industrial activities (Sai Dong industrial zone B). K2, representing a roadside house, was located adjacent to roads with high traffic density (upper ring road #3 and lower road systems) in southern Hanoi. K3, representing urban houses in densely populated areas and surrounded by commercial centers, was located in the southwest of the city. The direct distance between these sampling sites was approximately 5–10 km. The locations of the three dwellings are shown in Fig. 1. The characteristics of the three dwellings are summarized in Table S1, and the layout of the sampling sites is shown in Fig. S1.

2.2. Sample collection

Sampling at the three dwellings was conducted in two seasons, namely winter (from November 2018 to January 2019) and summer (from April to June 2019). These sampling seasons were selected to cover the periods of the lowest and highest PM concentrations in Hanoi throughout the year (Nguyen et al., 2018; Phung et al., 2021). At the sampling site, each sampling period lasted for 14 consecutive days. During each sampling season, sampling was conducted consecutively from one site to another. At each dwelling, daily samples of $PM_{0.1}$ and $PM_{2.5}$ in indoor and outdoor air were simultaneously collected. During sampling, the windows and doors were kept closed and only opened when going out or in and cleaning the house. Daily activities were performed as usual at the three dwellings during the sampling periods. The indoor samplers were placed in the bedrooms of K1 and K2 and in the master room of K3, which were on the 2nd floor of the dwellings. Outdoor particles were sampled on the balcony of the dwellings. The inlets of the samplers were placed at a height of 1.5 m from the floor to simulate the human breathing zone. The samplers were located at least 1 m from any obstacles (window, door, ventilation inlet, and wall). Samples of indoor and outdoor $PM_{0.1}$ were collected in quartz filters (55 mm in diameter) by two identical Nanosampler II devices (Model 3182, KINOMAX Japan Inc) with a constant flow rate of 40.0 L/min. Samples of indoor and outdoor $PM_{2.5}$ were collected simultaneously in quartz filters (47 mm in diameter) by two similar cyclone samplers with a fiber holder (URG-2000-30EH, University Research Glassware Co., Chapel Hill, NC, USA) at a flow rate of 16.7 L/min. Field blanks were also obtained. Before sampling, all samplers were calibrated to obtain the recommended flow rates using a TSI mass flow meter 4040 (TSI Incorporated, USA). Quartz filters were pre-baked at 900 °C for 4 h to remove possible contaminants. A total of 320 samples were collected.

2.3. Mass and chemical analysis

The collected filters, real samples, and blank samples were equilibrated in a desiccator in a balance room in which the relative humidity and temperature were controlled in the ranges of 30–40% and 25 ± 5 °C, respectively, for 48 h before weighing according to the reference method (EN12341:2014). An electronic microbalance with a detection limit of 10^{-6} g (AX26 DeltaRange Microbalance, Mettler Toledo Company, Switzerland) and a constant ionizing air blower (Model YIB01-ODR, Germany) were used for weighting. The quartz filters were subsequently stored in a refrigerator at 4 °C until further analysis. Half of each quartz filter was digested by a concentrated acid mixture (1HNO_3:3HCl, v/v) in a Mars 6 microwave according to United States Environmental Protection Agency (US EPA) IO-3.1 for 30 min. After digestion, the solutions were analyzed for 10 trace elements (TEs) (Cr, Mn, Co, Cu, Ni, Zn, As, Cd, Sn, and Pb) by inductively coupled plasma mass spectrometry (ICP-MS; X-series 2; Thermo Fisher Scientific, MA, USA) in the laboratory of Kyoto University, Japan, following US EPA method IO-3.5 (US EPA, 1999). The TEs in the digested solutions were quantified using Y and In as internal standards. Each sample was measured in triplicate, and the relative standard deviations were less than 5%. The method detection limit, which was estimated from seven spiked samples, was 0.1 ng/m^3 for individual TEs, except for Cr (0.2 ng/m^3) and Zn (0.5 ng/m^3). TE recovery tests were also conducted using spiked samples, and the recoveries of all the TEs were in the range of 85–120%. After analyzing each batch of 10 samples, the possible contamination and the operation of the ICP-MS instrument were checked using the blank sample and laboratory control sample.

Fig. 1. Locations of the three dwellings in Hanoi.

2.4. Infiltration factor determination

The infiltration factor (F_{inf}) is defined as the fraction of outdoor PM or their chemical compounds that enter indoors, which is estimated using Eq. (1) (Kearney et al., 2014; Wang et al., 2019).

$$F_{inf} = \frac{C_{in} - C_{ig}}{C_{out}} \quad (1)$$

where C_{in} and C_{out} are the concentrations of PM and their chemical components in the indoor and outdoor environments, respectively; C_{ig} is the concentration of PM or its chemical components generated from indoor sources; and C_{ig} intensity ($\%C_{ig}$) is the fraction of indoor-generated PM, which is calculated using Eq. (2), as follows:

$$\%C_{ig} = \frac{C_{ig} \times 100}{C_{in}} \quad (2)$$

2.5. Source identification

2.5.1. Enrichment factor

The enrichment factor (EF) is applied to differentiate whether atmospheric aerosol elements are derived from indoor or outdoor sources (Kulshrestha et al., 2014; Wang et al., 2019). The outdoor PM concentration is taken as a reference value in this term, and all atmospheric particles are assumed to be of outdoor origin. EFs can be calculated using the following formula (Eq. (3)):

$$EF_{indoor} = \frac{(C_{indoor}/PM_{indoor})}{(C_{outdoor}/PM_{outdoor})} \quad (3)$$

where C is the concentration of elements in the particle phase. Elements with an EF value less than or equal to 1 are considered "non-enriched" elements. This means that these elements are derived mainly from outdoor sources. Conversely, elements with an EF greater than 1 are considered "enriched" elements, which means that these elements originate from indoor sources in addition to outdoor sources (Kulshrestha et al., 2014).

2.5.2. Principal component analysis

Principal component analysis (PCA) is widely used to identify the possible sources of PM (Kulshrestha et al., 2014; Wang et al., 2019). PCA is a multivariate tool used to reduce the dimensionality of a dataset and transform the dependent variables into principal components (PCs). PCs are the eigenvectors of a covariance matrix, and each PC extracts a maximal share of the total variance. The number of PCs is determined according to the Kaiser criterion, which states that the eigenvalues of the correlation matrix are greater than 1 (Kulshrestha et al., 2014; Wang et al., 2019). Factor loadings equal to or greater than 0.7 are used for source apportionments.

2.6. Health risk assessment

2.6.1. Exposure assessment

The International Commission on Radiological Protection (ICRP) developed the human respiratory tract (HRT) model to determine the PM deposition fraction (DF) in the respiratory tract, which consists of three main parts, namely the head airways (HA), trachea-bronchial region (TB), and pulmonary/alveolar region (AL), and to estimate the internal dose of TEs. In this study, a simplified equation based on the ICRP model with Hind's parameterization was used to calculate the deposited dose (EDI) of inhaled TEs in PM in the HRT (Gao et al., 2017; Hinds, 1999; ICRP, 1994). The major mechanisms of PM deposition across the HRT include diffusion, sedimentation, and impaction. The deposition mechanism of PM in the HRT regions varies depending on the size of the PM and the anatomical and physiological properties of the respiratory system (Gao et al., 2017; ICRP, 1994). The total EDI of inhaled TEs in the form of $PM_{0.1}$ and $PM_{2.5}$ in the HRT are the sum of the EDI in the three regions, which are estimated by the following US EPA model (US EPA, 1989; US EPA, 2009) (Eq. (4)):

$$EDI = \sum_{i=1}^{3} \sum_{j=1}^{n} \frac{DF_i \times C_j \times IR \times ET \times ED \times EF}{24 \times AT} \quad (4)$$

where EDI is the total EDI of TEs (ng/d); DF_i is the particle deposition fraction of region i in the HRT (HA, TB, and AL); C_j is the concentration of TE j (ng/m^3); j = 1-n, in which n is the number of TEs; IR is the inhalation rate (m^3/d); ET is the exposure time (h/d); EF is the exposure frequency (d/y); ED is the exposure duration (y); and AT is the average lifetime (d). DF_i is estimated using Eqs. (1)–(4), as presented in Table S2.

2.6.2. Estimation of non-carcinogenic and carcinogenic risks

Non-carcinogenic (non-car) and carcinogenic (car) risks were assessed for seven age categories in winter and summer following the US EPA model, which has been widely applied to assess health risks in previous studies (Sharma and Balasubramanian, 2018; Wang et al., 2018).

To determine the non-car risk, the hazard quotient (HQ) for individual TEs and the hazard index (HI) are used, which are calculated using Eqs. (5) and (6), respectively.

$$HQ = \frac{EDI_j}{BW \times RfD_j} \quad (5)$$

$$HI = \sum_{j=i}^{n} HQ \quad (6)$$

To determine the car risk, the incremental lifetime cancer risk (ILCR) is quantified as the incremental cancer probability of an individual over their lifetime owing to potential exposure to car substances, which is calculated according to Eq. (7), as follows:

$$ILCR = \sum_{j=1}^{n} ILCR_j = \frac{EDI_j \times CSF_j}{BW} \quad (7)$$

where BW is the body weight (kg), RfD_j is the chronic reference dose [mg/(kg·d)] for element j, and CSF_j is the cancer slope factor for a specific element j [mg/(kg·d)]. To estimate the EDI and ILCR, variables including C_j, ET, and BW were determined from the sampling campaigns and questionnaires; other variables such as IR, EF, ED, AT, and CSF were referenced from the US EPA exposure handbook. Details of the data sources for calculation are shown in Tables S3 and S4. For public health protection from TEs, the acceptable HI value recommended by the US EPA for non-car risk is ≤ 1. In contrast, the HI value is unacceptable when >1. The ILCR $\leq 10^{-6}$ indicates "zero risk" or no adverse risk, the acceptable ILCR value for car risk is within $10^{-6} <$ ILCR $\leq 10^{-4}$. In contrast, the ILCR value is unacceptable when ILCR $>10^{-4}$ (Gao et al., 2017; US EPA, 2009).

2.7. Questionnaire method

Questionnaires were distributed to households in Hanoi to collect basic information about family members and their daily activities during the sampling campaigns. The collected data, including exposure parameters (body weight, age, and time spent indoors and outdoors), are summarized in Table S3 and were used in the health risk assessment. At the three surveyed dwellings, questionnaires including additional information on the characteristics of the houses (ventilation system, floors, walls, window structures, and building age), daily indoor activities (cooking, cleaning, smoking, incense burning, etc.), types of cookstoves, and number of occupants and daily reports were conducted during the sampling campaigns.

2.8. Statistical analysis

Statistical analysis was performed using IBM SPSS Statistics 20 software. Normal distribution was tested using the Shapiro–Wilk test. *T*-tests and linear regression were performed. The Mann–Whitney and Kruskal–Wallis tests were used for nonparametric tests. All results were considered statistically significant at $p < 0.05$. A Monte Carlo simulation was used to perform probabilistic risk assessment. This probability-based method can generate the output as a cumulative distribution function from the random input variables instead of using fixed single values, which can minimize the uncertainty of the expected outputs. Accumulative distribution functions were determined using a Monte Carlo simulation with 100000 iterations run using the Oracle Crystal Ball software (Version 11.1.2.4.850, Oracle, Inc., USA).

3. Results and discussion

3.1. Mass concentrations of $PM_{0.1}$ and $PM_{2.5}$

The mass concentrations of indoor $PM_{2.5}$ and $PM_{0.1}$ in the three dwellings (K1, K2, and K3) are presented in Fig. 2. The average concentration of indoor $PM_{2.5}$ measured in the three dwellings was 59.9 ± 23.6 μg/m^3, which is approximately four times higher than the World Health Organization (WHO) guideline (15.0 μg/m^3). Meanwhile, the average concentration of $PM_{0.1}$ was 8.2 ± 0.7 μg/m^3. The concentration of indoor $PM_{2.5}$ in this study was at the same level as that in residential apartments in China, which have the same characteristics as houses (natural ventilation and type of house) (Wang et al., 2016). However, the $PM_{2.5}$ concentration in this study was higher than that in houses with mechanical ventilation in China and Canada (Kearney et al., 2014; Wang et al., 2016). Air cleaning systems can enhance indoor air quality. Meanwhile, the concentration of indoor $PM_{0.1}$ in this study was considerably higher than that in schools in Hanoi, in which no indoor sources such as cooking and burning incense were present (Tran et al., 2020). The high concentrations of indoor $PM_{0.1}$ and $PM_{2.5}$ in this study may result in an occupant disease burden, including adverse effects on microvascular function, inflammation, and lung cell integrity (Karottki et al., 2015).

Significant seasonal variations were observed for indoor $PM_{2.5}$, but not for indoor $PM_{0.1}$. The indoor concentrations of $PM_{2.5}$ were in the range of 43.4–49.5 μg/m^3 in summer, whereas they varied from 50.6 μg/m^3 to 106 μg/m^3 in winter. The indoor $PM_{2.5}$ concentration in winter was 2.5 times higher than that in summer. The seasonal variation in indoor $PM_{2.5}$ in this study was closely associated with the seasonal variation in outdoor $PM_{2.5}$, which has also been reported by Phung et al. (2021) and Nguyen et al. (2018). This can be explained by the ratio of indoor/outdoor concentrations (I/O) of $PM_{2.5}$, which was less than 1, thereby suggesting that indoor $PM_{2.5}$ is strongly influenced by outdoor $PM_{2.5}$. The trends of seasonal variations in indoor $PM_{2.5}$ were also observed in residential houses in north-central India and Lahore, Pakistan (Massey et al., 2012; Sidra et al., 2015). Meanwhile, the

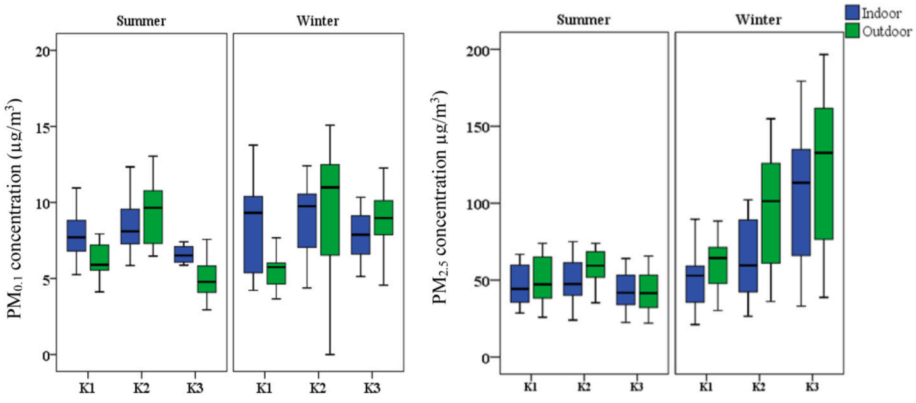

Fig. 2. Indoor and outdoor concentrations of $PM_{2.5}$ and $PM_{0.1}$ in two seasons.

concentration of indoor $PM_{0.1}$ was almost unchanged in different seasons. The indoor $PM_{0.1}$ concentration varied from 7.2 μg/m^3 to 8.7 μg/m^3 in summer, whereas it ranged from 8.1 μg/m^3 to 8.9 μg/m^3 in winter. The ratio of I/O for $PM_{0.1}$ was slightly higher than 1, thereby implying that the indoor $PM_{0.1}$ was affected by indoor sources. However, the seasonally unchanged trend of indoor $PM_{0.1}$ was likely mainly attributed to the seasonal stability of outdoor $PM_{0.1}$ in Hanoi, as observed in this study and the study conducted by Nguyen et al. (2018).

The contribution of particles among the three houses was also evaluated to better understand the PM distribution. The ratios of indoor $PM_{0.1}/PM_{2.5}$ were 18%, 17%, and 13% at K1, K2, and K3, respectively, whereas the ratios of outdoor $PM_{0.1}/PM_{2.5}$ were 13%, 14%, and 11% at K1, K2, and K3, respectively. At each house, the ratio of $PM_{0.1}/PM_{2.5}$ in the indoor air was slightly higher than that in the outdoor air, thereby revealing that there was a larger contribution of $PM_{0.1}$ to $PM_{2.5}$ in the indoor air. Morawska et al. (2017) suggested that cooking, incense burning, and cleaning activities release particles smaller than 300 nm, the majority of which are $PM_{0.1}$, and indoor $PM_{2.5}$ mainly originates from outdoor sources. In addition, the ratios of indoor $PM_{0.1}/PM_{2.5}$ at K1 and K2 were almost the same, thereby implying that indoor PM in these houses originated from the same type of indoor sources. However, this ratio at K3 was slightly different from those at K1 and K2, thereby indicating different types of indoor sources and their contributions at K3.

3.2. Concentrations of TEs in $PM_{0.1}$ and $PM_{2.5}$

The total concentrations of 10 TEs (∑10 TEs) bound to $PM_{0.1}$ and $PM_{2.5}$ at the residential dwellings (K1, K2, and K3) are presented in Table 1 and Fig. S2. The average concentrations of ∑10 TEs in indoor $PM_{0.1}$ and $PM_{2.5}$ were 83.8 ± 22.2 ng/m^3 and 763 ± 324 ng/m^3 in summer and 193 ± 31.8 ng/m^3 and 1510 ± 736 ng/m^3 in winter, respectively. In general, higher concentrations of TEs in indoor $PM_{0.1}$ and $PM_{2.5}$ were observed in the roadside house; these values increased by 1.1–1.4 times for $PM_{0.1}$ and 1.5 to 2.4 times for $PM_{2.5}$ in comparison with those at the urban periphery house and urban house, respectively. This result is similar to that reported by Kulshrestha et al. (2014), which also indicated that the concentrations of TEs in indoor $PM_{2.5}$ at roadside houses are higher than those in urban and rural houses.

The mean ratios of ∑10 TEs between indoor $PM_{0.1}$ and $PM_{2.5}$ were 12%, 11%, and 18% at K1, K2, and K3, respectively. The relative similarity of these ratios was observed at K1 and K2, thereby revealing the same types of indoor sources in these houses. However, this ratio was slightly higher at K3, thereby suggesting additional indoor sources at this site, which is consistent with the discussion in Section 3.1. Furthermore, the contents of TEs in indoor PM were 2%, 2%, and 1% for $PM_{0.1}$, whereas they were 2%, 3%, and 1% for $PM_{2.5}$ at K1, K2, and K3, respectively. TEs accounted for 3%, 3%, and 2% of outdoor $PM_{0.1}$ and 3%, 4%, and 2% of outdoor $PM_{2.5}$ at K1, K2, and K3, respectively. For each type of PM and at each sampling site, the content of TEs in outdoor PM was higher than that in indoor PM, thereby implying that the TEs in indoor PM predominantly originated from outdoor sources. The average concentration of indoor $PM_{0.1}$ was higher than that of outdoor $PM_{0.1}$. Therefore $PM_{0.1}$ generated by indoor sources had a lower TE content than $PM_{0.1}$ derived from outdoor sources. The different contents of TEs in PM in the houses might have been caused by the different contributions of sources, although the houses could be influenced by the same types of sources.

Table 1
Concentrations of trace elements (TEs) bound to particulate matter (PM) at the three houses in two seasons.

Type of PM	Season	Type of environment	TE concentration in PM (ng/m3) (1 atm, 25 °C)		
			K1	K2	K3
$PM_{2.5}$	Summer	Indoor	990 ± 825	907 ± 627	391 ± 144
		Outdoor	1260 ± 1050	1410 ± 1190	516 ± 271
	Winter	Indoor	1180 ± 1070	2360 ± 1350	1000 ± 252
		Outdoor	1630 ± 1620	5030 ± 2900	1480 ± 527
$PM_{0.1}$	Summer	Indoor	66.2 ± 29.0	108 ± 115	76.5 ± 28.2
		Outdoor	123 ± 89.0	116 ± 65.5	86.9 ± 33.9
	Winter	Indoor	216 ± 54.3	205 ± 81.5	156 ± 49.2
		Outdoor	203 ± 54.4	214 ± 48.0	214 ± 102

The concentrations of individual TEs in PM are shown in Tables S5 and S6. The concentrations of Cd, As, and Ni in this study exceeded the respective EU limits on air quality for individual TEs (As: 6.0 ng/m^3, Ni: 20.0 ng/m^3, Cd: 0.5 ng/m^3, and Pb: 500 ng/m^3) (EC, 2005; EC, 2008) in almost all cases, although these elements were not predominant among the 10 TEs measured. For instance, for TEs in indoor $PM_{2.5}$, the average concentrations of As, Cd, and Ni in the three houses were 1.4 to 4.6, 3.8 to 6.9, and 1.5 to 4.5 times higher than the EU limits. The exceedances of As, Ni, and Cd at the roadside house were the highest, followed by those at the urban periphery house and urban house. Wang et al. (2018) also pointed out that the concentrations of Ni and As were higher than the EU limits at residential houses in Nanjing, China in winter, which was also found at the urban house in our study.

Among the 10 TEs, Zn, Pb, and Cr were the most abundant in both particle sizes in both seasons. The proportions of Pb and Zn to the total concentration of TEs bound to $PM_{2.5}$ were 8%–12% and 74%–77%, respectively, whereas those of Zn, Pb, and Cr to the total concentration of TEs bound to $PM_{0.1}$were 34%–53%, 9% to 11%, and 17% to 20%, respectively. These findings are almost consistent with those of previous studies, in which Zn, Cr, Pb, and Fe were the predominant elements in fine and coarse particles in urban environments in India (Kulshrestha et al., 2014) and in residential homes in China (Wang et al., 2018). The concentrations of individual TEs and further comparisons with related studies in different indoor environments are summarized in Table S7.

3.3. F_{inf} and indoor generated particles

3.3.1. F_{inf} and indoor generated particles for PM concentration

The F_{inf} values of $PM_{0.1}$ and $PM_{2.5}$ in the three houses in summer and winter are shown in Fig. S3. The average F_{inf} was 1.0 and 0.9 for $PM_{0.1}$ and $PM_{2.5}$ in summer, respectively, whereas the average F_{inf} was 1.0 and 0.8 for $PM_{0.1}$ and $PM_{2.5}$ in winter, respectively. The results revealed that almost no seasonal difference was observed for F_{inf} of $PM_{0.1}$ and $PM_{2.5}$. This can be explained by the similar sampling conditions in both seasons. In fact, outdoor $PM_{0.1}$ and $PM_{2.5}$ can enter the buildings with closed doors and windows (Chatoutsidou et al., 2015; Dickerhoff et al., 1982). However, due to its larger size, the infiltration of $PM_{2.5}$ is worse than that of $PM_{0.1}$. This may be the reason for the fact that F_{inf} value of $PM_{2.5}$ was slightly smaller than that of $PM_{0.1}$.

The contribution of indoor-generated PM to indoor PM (%C_{ig}) in this study varied widely among the houses with the seasons, thereby reflecting the complex and dynamic features of indoor environments. However, based on the values of %C_{ig} obtained, it can be estimated that approximately 2%–21% of indoor $PM_{2.5}$ in the three houses originated from indoor sources. This highlights the importance of outdoor sources for $PM_{2.5}$ in indoor environments. It was estimated that 18% and 54% of indoor $PM_{0.1}$ at K2 and K3, respectively, were derived from indoor sources. In other words, at K2 and K3, 82% and 46% of indoor $PM_{0.1}$ originated from outdoor sources, respectively. In contrast, %C_{ig} of $PM_{0.1}$ was negative at K1, which might have been attributed to the decomposition of some species of $PM_{0.1}$ (Sangiorgi et al., 2013; Vu et al., 2017).

3.3.2. F_{inf} and C_{ig} for TE concentrations

The mean F_{inf} values of $\sum$10 TEs were 0.6 and 0.5 for $PM_{2.5}$, whereas they were 0.8 and 0.8 for $PM_{0.1}$ in summer and winter, respectively. There was no clear seasonal variation in F_{inf} of TEs in $PM_{0.1}$ and $PM_{2.5}$, which was consistent with the F_{inf} value of PM, as discussed in section 3.3.1. The contributions of indoor sources to indoor TEs ($\%C_{ig}$) differed significantly among the houses, thereby reflecting the complex and dynamic features of the indoor environment. However, the obtained results revealed that approximately 12%–41% and 3%–34% of indoor TEs in $PM_{0.1}$ and $PM_{2.5}$, respectively, were derived from indoor sources.

Further investigations were conducted to determine F_{inf} and C_{ig} of individual TEs at each house, as shown in Fig. S4. In general, As, Zn, and Co appeared to have the highest C_{ig} intensities ($\%C_{ig,As} = 71\%$ in $PM_{0.1}$ in winter at K1; $\%C_{ig,Co} = 56\%$ in $PM_{2.5}$ in summer at K3; $\%C_{ig,Zn} = 65\%$ in $PM_{0.1}$ in winter at K3), thereby showing that more than 50% of As, Zn, and Co in these cases was generated from indoor sources. However, the C_{ig} intensities of the remaining cases were all less than 50%, e.g., for Ni ($\%C_{ig} = 45\%$), Co ($\%C_{ig} = 32\%$), As ($\%C_{ig} = 35\%$), and Cd ($\%C_{ig} = 34\%$) at K2. Thus, except for the predominance of indoor sources for As at K1 and Zn and Co at K3, outdoor sources were the main contributors to most of the TEs in the three houses. Indoor sources in these houses may include coal briquette combustion (As), wall paints, construction materials (Pb, Cd, Ni, and Co) (Latif et al., 2011; Mielke et al., 2001; Suryawanshi et al., 2016), and vacuum cleaners (Cu) (Vu et al., 2017). Coal briquettes were used for domestic cooking in K1, whereas wall paints, construction materials, and vacuum cleaners were common indoor sources in all three houses. Other indoor TEs are associated with outdoor sources, including traffic, industrial emissions, and domestic combustion from surrounding households (Cohen et al., 2010; Kulshrestha et al., 2014; Kummer et al., 2009; Tunno et al., 2016).

3.4. Source identification

3.4.1. EF estimation

The EFs of TEs in $PM_{2.5}$ and $PM_{0.1}$ in the three dwellings in summer and winter are shown in Fig. S5. There were significant differences in the TE EFs between the three houses and the two seasons. At the urban periphery house (K1), the EF values of As and Cu were 1.2 ± 0.0 and 1.2 ± 0.1, respectively, in $PM_{2.5}$, and the EF of Cu was 1.3 ± 0.1 in $PM_{0.1}$ in summer. The EF values of Cu, Zn, and Pb were 1.3 ± 0.1, 1.5 ± 0.1, and 1.3 ± 0.0, respectively, in $PM_{2.5}$ in winter. At the roadside house (K2), EF values of Zn (1.3 ± 0.1) and Pb (1.3 ± 0.0) in $PM_{2.5}$ and Ni (1.4 ± 0.1), Zn (1.3 ± 0.1), and As (1.4 ± 0.1) in $PM_{0.1}$ in summer were obtained. Those of Co (1.6 ± 0.1) in $PM_{2.5}$ and Cr (1.3 ± 0.0) and As (1.3 ± 0.1) in $PM_{0.1}$ in winter were also determined. At the urban house (K3), the EF of Co in $PM_{2.5}$ in summer was 1.4 ± 0.1, and those of Zn and Co in $PM_{0.1}$ in winter were 1.4 ± 0.1 and 1.6 ± 0.2, respectively. These results suggest that the above TEs were influenced by indoor sources, such as domestic burning and other indoor activities. Other TEs appeared to be "non-enriched" in either $PM_{2.5}$ or $PM_{0.1}$ in both seasons at the three houses, thereby implying that outdoor sources were the main sources, which are discussed in detail in Section 3.4.2.

3.4.2. PCA

The results of PCA for source identification are presented in Fig. S6. The source distribution is summarized in Fig. S7.

Urban periphery dwelling (K1): In winter, three factors were extracted from PCA for $PM_{2.5}$, which explained 93% of the sources. Factor 1 accounted for 60% of indoor $PM_{2.5}$, which originated from mixed sources including domestic burning of coal briquettes (As, Cd, Pb and Mn) (Nghiem et al., 2020; Shao et al., 2016; Tunno et al., 2016), and vehicle non-exhaust (Zn) (Kummer et al., 2009). Factor 2 accounted for 20% of indoor $PM_{2.5}$, which was derived from industrial emissions (Cu and Ni) (Cao and Nguyen, 2013; Nghiem et al., 2020). Factor 3 accounted for 13% of indoor $PM_{2.5}$, which represented household dust (Co) (Suryawanshi et al., 2016). Three factors in $PM_{0.1}$ could explain 79% of sources in winter at this house. Factor 1 was industrial emissions (Mn, Cu, Cd and Ni), which accounted for 40%, and factor 2 was vehicle non-exhaust (Zn), which accounted for 15%. These sources are considered outdoor sources, and are responsible for 55% of indoor $PM_{0.1}$. Factor 3 was household dust (Co) and indoor combustion (As), which accounted for 24% of indoor $PM_{0.1}$. In summer, stable source apportionment was found for both particle sizes, in which two factors accounted for 80% of the variation in indoor $PM_{2.5}$ and 74% of the variation in indoor $PM_{0.1}$. For instance, the combustion of coal briquettes (As, Cd and Mn) and traffic emissions (Sn, Cu, and Zn) were extracted from factor 1 as outdoor sources, which could explain 58% and 57% of sources of indoor $PM_{2.5}$ and $PM_{0.1}$, respectively. In addition, 22% of indoor $PM_{2.5}$ and 17% of indoor $PM_{0.1}$ were explained by factor 2, which were attributed to household dust (Cu) (Vu et al., 2017) and indoor combustion (Cr) (Tunno et al., 2016).

The overall results implied that indoor $PM_{2.5}$ and $PM_{0.1}$ at this house were derived mainly from outdoor sources in both seasons. For instance, outdoor sources accounted for 55% and 80% of indoor $PM_{0.1}$ and indoor $PM_{2.5}$, respectively, in winter, and 57% and 58% of indoor $PM_{0.1}$ and indoor $PM_{2.5}$, respectively, in summer. Indoor sources were responsible for 24% and 17% of indoor $PM_{0.1}$ and 13% and 22% of indoor $PM_{2.5}$ in winter and summer, respectively. These results confirmed the dominant influence of coal combustion, traffic and industrial activities as outdoor sources at this house. Industrial sources contributed to 40% of indoor $PM_{0.1}$ and 20% of indoor $PM_{2.5}$ at K1, which is close to the Sai Dong industrial zone. This industrial zone has many different facilities, such as for electronics, computer accessories, mobile phones, electric appliances, non-ferrous metals, and precious metals, which release numerous heavy metals into the environment (Wu et al., 2018). TEs such as Mn, Cu, and Ni are also considered markers of industrial sources in Hanoi, accounting for approximately 6% of ambient $PM_{0.1}$ or $PM_{2.5}$ in studies conducted by Nghiem et al. (2020) and Cao and Nguyen (2013) and 17% of ambient $PM_{2.5}$ in the study conducted by Cohen et al. (2010). The indoor PM in this house, which is close to ring road #2 (upper) and embankment road with a high density of passenger vehicles, buses, and heavy trucks, is strongly associated with traffic sources. Indoor PM at this house was also affected by the burning of coal briquettes for domestic cooking in the house itself (indoor source) and surrounding households (outdoor source). Coal briquettes are still used in food stalls, small restaurants, and households in Hanoi (Vo et al., 2020c). The burning of coal briquettes in Hanoi contributes to 18% of ambient $PM_{2.5}$ according to Cohen et al. (2010), 16% of ambient $PM_{2.5}$ according to Cao and Nguyen (2013), and 12% of ambient $PM_{0.1}$ according to Nghiem et al. (2020). A mixture of coal, traffic, and industrial sources has also been identified as a common source at urban houses near industrial zones (Tunno et al., 2016).

Roadside dwelling (K2): In winter, two PCA factors explained 83% and 80% of the variation in indoor $PM_{0.1}$ and $PM_{2.5}$, respectively. The highest loadings of Sn, Pb, As, Cd, Zn, and Cu in $PM_{0.1}$ and Sn, Pb, As, Cd, and Zn in $PM_{2.5}$ were attributed to domestic coal burning (As, Pb and Cd), and vehicle non-exhaust emissions from brake or tire abrasion (Zn, and Sn) in factor 1. Cobalt, Ni, and Cr were dominant in factor 2, thereby suggesting that household dust was emitted from wall paints and construction materials (Mielke et al., 2001; Ogilo et al., 2017; Suryawanshi et al., 2016). These findings were consistent with the EFs of TEs in section 3.4.1. The sources contributing to TEs in $PM_{2.5}$ and $PM_{0.1}$ were similar, but the contribution of each source was different. For example, 64% of indoor $PM_{0.1}$ originated from domestic coal combustion and traffic emissions in factor 1 and 19% of indoor $PM_{0.1}$ originated from household dust in factor 2, whereas 52% of indoor $PM_{2.5}$ originated from coal combustion and traffic emissions in factor 1 and 28% of indoor $PM_{2.5}$ originated from household dust in factor 2. In summer, three PCA factors extracted from $PM_{0.1}$ and two PCA factors extracted from $PM_{2.5}$ could explain 79% and 76% of the sources, respectively. The outdoor source was attributed to traffic emissions, which was responsible for 64% of indoor $PM_{0.1}$, as found in factors 1 and 2. Meanwhile, 63% of

indoor $PM_{2.5}$ originated from this outdoor source, as found in factor 1. Household dust from wall paints and construction materials (Cd, Cu, and As) contributed to 15% and 13% of indoor $PM_{0.1}$ (factor 3) and $PM_{2.5}$ (factor 2), respectively.

Our findings indicated that outdoor sources are the main contributors to indoor PM at roadside houses. Outdoor sources contributed to 64% and 52% of indoor $PM_{0.1}$ and $PM_{2.5}$ in winter, respectively, and 64% and 63% of the corresponding PM in summer. Indoor sources contributed to 19% and 28% of indoor $PM_{0.1}$ and $PM_{2.5}$ in winter, respectively, and 15% and 13% of the corresponding PM in summer. Accordingly, traffic emissions were considered to be the dominant source contributing to indoor PM in this study, which was in line with the results of previous studies. It has been reported that traffic activities in Hanoi contribute to 46% of ambient $PM_{0.1}$ (Nghiem et al., 2020) and 40% of ambient $PM_{2.5}$ (Cohen et al., 2010). The dominance of traffic emissions at the roadside house can be explained by the strong influence of intensive traffic roads with light-duty and heavy-duty vehicles in upper ring road #3 and nearby Nguyen Huu Tho road. This house is also affected by condensed traffic activities at Giai Phong road and Nuoc Ngam and Giap Bat bus stations. However, the burning of coal briquettes occurs routinely at nearby eateries and small restaurants.

Urban dwelling (K3): There were considerable discrepancies in the source contributions between the two particle sizes at this site. In winter, two PCA factors could explain 88% of indoor $PM_{0.1}$ and 88% of indoor $PM_{2.5}$. TEs (As, Cr, Co, Cu, and Mn) extracted from factor 1 for $PM_{0.1}$ and TEs (Cu and Co) extracted from factor 2 for $PM_{2.5}$, which were attributed to household dust, were responsible for 74% and 20% of indoor $PM_{0.1}$ and $PM_{2.5}$, respectively. Regular indoor activities (domestic cooking, vacuum cleaning, incense burning, etc.), which usually occur during the sampling periods, might have contributed to indoor PM at this site. Traffic emissions and domestic coal combustion, as the outdoor sources, accounted for 14% of indoor $PM_{0.1}$ in factor 2 and 68% of indoor $PM_{2.5}$ in factor 1. In summer, traffic emissions and domestic coal combustion accounted for 57% of indoor $PM_{0.1}$ in factor 1 and 19% of indoor $PM_{2.5}$ in factor 2, whereas household dust contributed to 17% of $PM_{0.1}$ in factor 2 and 72% of $PM_{2.5}$ in factors 1 and 3. This house is highly affected by traffic emissions, domestic coal combustion and household dust in both seasons. However, their contributions to $PM_{0.1}$ and $PM_{2.5}$ varied by season. In winter, household dust was the main contributor to $PM_{0.1}$, whereas traffic emissions and domestic coal combustion were the primary sources of $PM_{2.5}$. In contrast, in summer, the majority of $PM_{0.1}$ originated from traffic emissions and domestic coal combustion, whereas household dust was the primary source of $PM_{2.5}$. Wang et al. (2019) reported that indoor $PM_{2.5}$ in academic dormitories in Nanjing, China originated from a mixture of coal combustion, industrial activities, traffic emissions, and student activities.

3.5. Estimation of EDI in the respiratory tract

The total EDI of non-car and car TEs was calculated using Eq. (4), which is the sum of the EDI in three respiratory regions (EDI_{HA}, EDI_{TB}, and EDI_{AL}). The values of the total EDI of car TEs ($EDI_{car\text{-}TEs}$) and non-car TEs ($EDI_{non\text{-}car\text{-}TEs}$) in the two particle sizes according to different age categories of inhabitants are shown in Tables S8 and S9, respectively. The age categories were classified into seven groups, namely infants (0–1 y), toddlers (1–3 y), preschool children (3–6 y), children (6–11 y), adolescents (11–21 y), adults (21–60 y), and elderly adults (>60 y). Overall, the $EDI_{non\text{-}car\text{-}TEs}$ and $EDI_{car\text{-}TEs}$ values varied largely among age groups, particle sizes, seasons, and houses. The indoor $EDI_{non\text{-}car\text{-}TEs}$ and $EDI_{car\text{-}TEs}$ values were in the ranges of 1680–22500 ng/d and 4.8–1880 ng/d in $PM_{2.5}$, respectively, and 63.1–458 ng/d and 0.2–79.2 ng/d in $PM_{0.1}$, respectively. Additionally, the difference in the EDI of non-car TEs and car TEs between indoor and outdoor air ranged from 2 to 16 times. The indoor EDI was higher owing to the longer exposure time, although the outdoor TE concentration was higher. Our results agree with those found by Gao et al. (2017), in which the EDI for indoor $PM_{2.5}$ was several times higher than that for outdoor $PM_{2.5}$; for example, the EDI of TEs in the residential environment was approximately 9–32 times higher than that in student offices, laboratories, and outdoor environments. However, our results contrasted those of Sharma and Balasubramanian (2018), which stated that a higher EDI of TEs was found for outdoor air and that EDI increased as the haze intensity increased.

The EDI of TEs in both $PM_{0.1}$ and $PM_{2.5}$ were estimated in the HA, TB, and AL respiratory tract regions of dwellers in the three houses, and are presented in Fig. 3. In general, the EDI values were distributed as 80% in HA, 7% in TB, and 13% in AL for $PM_{2.5}$, whereas they were distributed as 11% in HA, 14% in TB, and 75% in AL for $PM_{0.1}$. The proportions of EDI for $PM_{2.5}$ in different respiratory tract regions in our study were comparable to those for $PM_{2.5–1.0}$, whereas those for $PM_{0.1}$ were similar to those for $PM_{<0.25}$ (Sharma and Balasubramanian, 2018). Greater deposition of TEs-$PM_{2.5}$ was observed in HA, whereas the majority of TEs-$PM_{0.1}$ was deposited in AL, followed by TB and HA. The greater deposition of $PM_{0.1}$ in AL was due to the flow path in the HRT, which is primarily governed by Brownian motion, thereby leading to preferential deposition in AL (Adachi, 2008; Chatoutsidou et al., 2015; Sharma and Balasubramanian, 2018). Similar results have been reported in several previous studies. Vu et al. (2017) reported that $PM_{0.1}$ is deposited predominantly in AL, followed by TB. Madureira et al. (2020) stated that PM_{10} is primarily deposited in HA (86–97%), followed by $PM_{2.5}$ (60–75%) and $PM_{0.1}$ (31–40%), whereas $PM_{0.1}$ deposition mainly occurs in AL. Thus, the EDI in HA decreased as the particle size decreased, whereas that in TB and AL increased when the particle size decreased. Although the total EDI of TEs-$PM_{0.1}$ was significantly lower than that in $PM_{2.5}$ (ratios of EDI of TEs-$PM_{0.1}$/TEs-$PM_{2.5}$ was approximately 3% and 4% for non-car TEs and car TEs, respectively), the majority of TEs-$PM_{0.1}$ intrudes deeply in AL, which causes severe damage to human health.

The overall results in Tables S8 and S9 show the wide variations in EDIs with seasons and age categories. For both non-car TEs and car TEs, the corresponding EDIs of indoor $PM_{0.1}$ and $PM_{2.5}$ in winter were approximately 1.2–2.3 times and 1.6 to 2.3 times higher than those in summer, respectively. Among the age categories, $EDI_{car\text{-}TEs}$ also increased significantly as age increased. Elderly adults (>60 y) had the highest EDI of TEs, whereas children, including infants (0–1 y), toddlers (1–3 y), and preschool children (3–6 y), had the lowest EDI during the exposure to TEs in both particle sizes. Although children experienced the lowest EDI for both particle sizes, they, especially infants and toddlers, are considered to be vulnerable to potential health impacts induced by toxic chemical components in the air because their respiratory, immune, reproductive, central nervous, and digestive systems are not fully developed. Oliveira et al. (2019) concluded that children have a high tendency to deposit particles in the lower respiratory tract owing to less efficient uptake of particles in HA. Therefore, this should be kept in mind to protect the health of children and elderly adults. Our finding was similar to that of Madureira et al. (2020), which showed that the EDI is higher in mothers than in infants (0–3 months) for $PM_{0.1}$ and $PM_{2.5}$. These estimations provide insights into the effects of inhalation of TEs in PM in residential houses in different respiratory tract regions and the urgency of protecting human health because people spend almost 90% of their life indoors.

3.6. Estimation of health risk

3.6.1. Non-car risk

Assessment of non-car risks for TEs bound to $PM_{0.1}$ and $PM_{2.5}$ in the three residential dwellings was conducted for seven age categories. The HI was determined to estimate the non-car risk associated with TE inhalation according to Eq. (6), and is presented in Fig. 4. The estimated average HIs of TEs bound to $PM_{0.1}$ and $PM_{2.5}$ ranged from 1.0 E−02 to 6.0E-2 and 3.0E-1 to 7.0E-1 in summer, whereas they varied from 3.0E-2 to 1.3E-1 and 6.0E-1 to 1.4E+0 in winter for all ages, respectively. The average HI values increased twice in winter in the three houses. Infants and toddlers were exposed to higher HIs for both particle sizes owing to

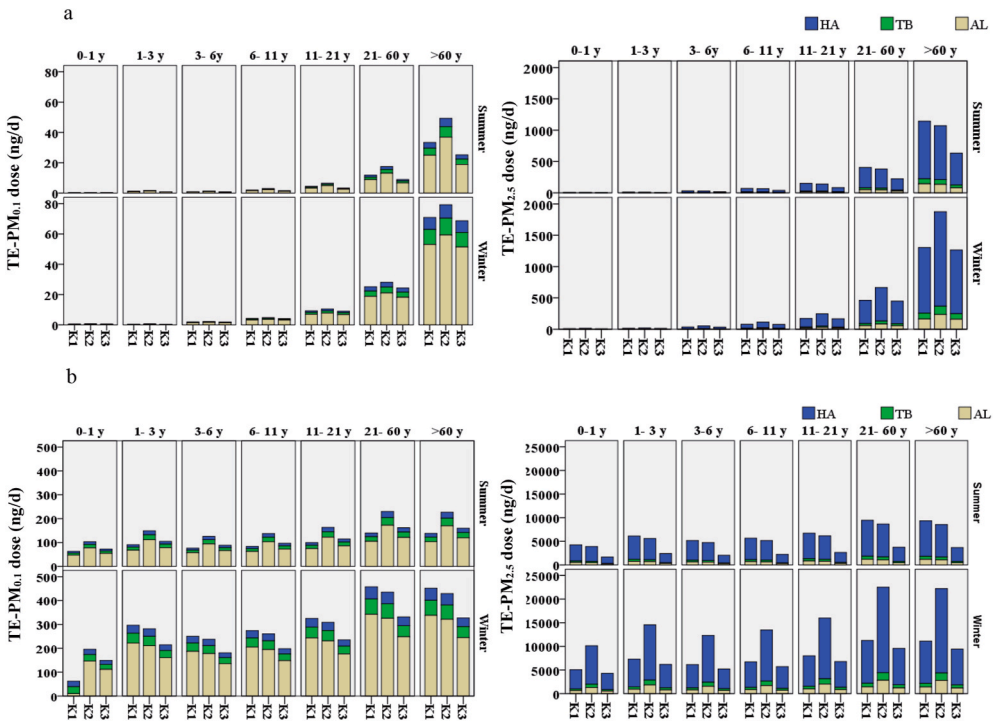

Fig. 3. Deposited doses of carcinogenic trace elements (TEs) (a) and non-carcinogenic TEs (b) in different respiratory tract regions (HA: head airways; TB: tracheobronchial; AL: alveolar/pulmonary).

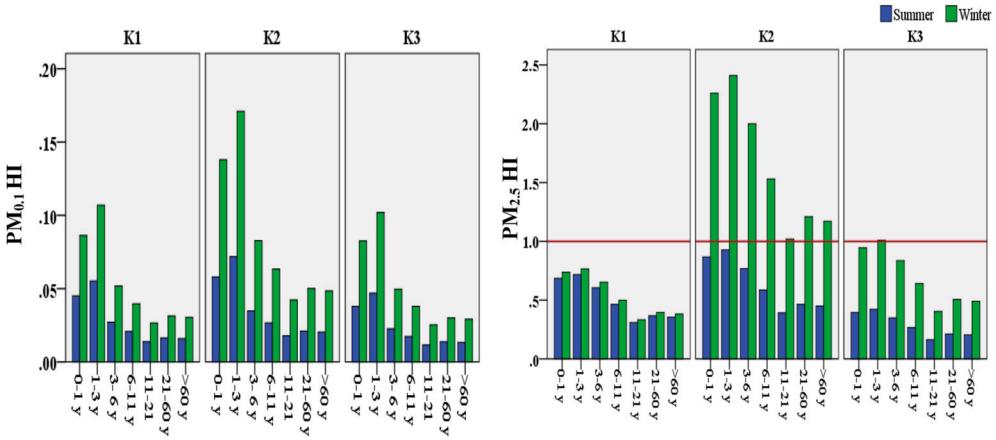

Fig. 4. Non-carcinogenic risk induced by trace elements in different age groups categories.

their lower body weight. It is estimated that the HI values for these age groups are two times higher than those for adults, thereby implying a greater potential non-car risk for infants and toddlers. Overall, the average HI of the three houses exceeded the threshold recommended by the US EPA, which was observed for infants, toddlers, and preschool children (3–6 y) in winter for $PM_{2.5}$. These estimations suggest that these groups are the most vulnerable and have a high non-car risk. Among the age categories, the HI values were in the following order (least to most vulnerable): (11–21 y)<(>60 y)≈(21–60 y)<(6–11 y)<(3–6 y)<(1–3 y)≈(0–1 y). Although the HQ of individual elements was <1 for $PM_{2.5}$, the HI values were >1 for all ages at the roadside house (K2) in winter, thereby suggesting that the accumulative risk posed by the inhalation of non-car TEs in $PM_{2.5}$ is important for health risk assessment. Similar conclusions have been reported by Gao et al. (2017) and Slezakova et al. (2014). These studies showed that the accumulative risk from non-car TEs in $PM_{2.5}$ is unacceptable for patients in hospitals and students at university campuses despite the acceptable risk induced by individual TEs. Except for the above cases, the HI values for $PM_{0.1}$ and $PM_{2.5}$ for other age categories were lower than the acceptable level, thereby indicating that the exposure of these age groups to TEs bound to PM via the inhalation pathway is within the safe limit.

Among individual TEs, Mn was the greatest contributor to non-car TE risk, accounting for 83%–92% for $PM_{2.5}$ and 68%–88% for $PM_{0.1}$. Other elements contributed to a negligible proportion. Although Zn and Pb were the most abundant elements, the low HQs of Zn and Pb were due to the low RfD values for non-car TEs. The dominant contribution of Mn to non-car risk was also found in dormitories in China (Gao et al., 2017; Wang et al., 2019) and residential homes in China (Wang et al., 2018). Arsenic is one of the most important contributors to non-car TE risk in hospitals in Portugal (Slezakova et al., 2014). The high non-car risk of Mn cannot be ignored. Because of its high solubility, Mn can cause toxic effects on multiple organs and the cardiovascular, reproductive, immune, and central nervous systems (Lu et al., 2015). However, Mn is an essential element in preventing metabolic diseases (Li, 2018).

3.6.2. Car risk

The International Agency for Research on Cancer has classified Cd, Cr, As, Ni, and Co as car substances because of their high toxicity, which raises potential health risks. Fig. 5 and Table S10 show the ILCRs estimated for car TEs in $PM_{2.5}$ and $PM_{0.1}$ via the inhalation pathway for the seven age categories in summer and winter. In general, the average ILCRs in $PM_{2.5}$ were in the ranges of 2.3E-06–4.9E-05 in summer and 4.8E-06–1.1E-04 in winter, whereas those in $PM_{0.1}$ varied from 2.0E-07 to 4.3E-06 in summer and 4.6E-07 to 9.4E-06 in winter among the three houses. The US EPA set an ILCR threshold of 1E-06 (1 out of case per 1000000 individuals developing cancer over a human lifetime) for the individual elements, which is considered no adverse car risk. However, they recommend the accumulative car risk for car TEs was set at 1E-04 (1 out of 10000 individuals developing cancer over a human lifetime) as an acceptable level (Gao et al., 2017; US EPA, 2009). It is estimated that the ILCR increases as age increases, and decreases in the following order: (>60 y)> (21–60 y)>(11–21 y)>(6–11 y)>(3–6 y)>(1–3 year)>(0–1 year). The highest ILCR was obtained in elderly adults (>60 years) owing to lifetime exposure extension, which was 3–22 times higher than that of other ages. Although the ILCR of children, especially infants and toddlers, was much lower than that of adults, children are still growing and their organ system is developing, so it is necessary to protect this group from chronic effects.

The accumulative risks of car TEs in $PM_{2.5}$ and $PM_{0.1}$ varied widely among the three houses. Roadside dwellers were exposed to the highest risk, whereas the lowest risk was observed in urban dwellers, which was consistent for all ages and both particle sizes and seasons. At the roadside house, elderly adults (>60 years) had an accumulative risk of 1.4E-04 in winter, thereby exceeding the US EPA threshold. This finding indicates that the probability of car risk is 1–2 people per 10000 individuals exposed to TEs in $PM_{2.5}$, which is an unacceptable level. Except for the above cases, the accumulative car risks for all age categories in our study were less than the acceptable level of 1E-04, thereby suggesting a safe level for all ages at the three dwellings. It is well known that besides the size of PM, the chemical composition (heavy metals, PAHs, organic carbon, etc.) of PM strongly affects the toxicity of PM, which may cause synergistic effects. In this study, the accumulative risks were estimated for TEs in PM only. Therefore, the results of this study may underestimate the accumulative risks because the synergistic effects of other chemical species were not taken into account (Sharma and Balasubramanian, 2018; Slezakova et al., 2014). Our results were similar to those of a study in hospitals in Portugal, in which adult patients (55–64 y) and (>65 y) had a high car risk associated with car TEs bound to $PM_{2.5}$ (Slezakova et al., 2014). Chromium and As were the most crucial contributors to the accumulative car risks, which were responsible for approximately 90% of the total risk at all ages in the three houses. Some previous studies also reported that As and Cr appear to be the most car substances in residential indoor environments in China (Wang et al., 2018), university dormitories in China (Wang et al., 2019), and hospital environments in Portugal (Slezakova et al., 2014). The toxicity of substances strongly depends on chemical speciation, which is vital for evaluating health risks. In this study, we estimated the human car risk of Cr(VI) species instead of total Cr. Therefore, 14% of the determined total Cr concentration was used to estimate the concentration of Cr(VI) with the consumption of 1:6 of Cr(III):Cr(VI) (Slezakova et al., 2014; US EPA, 2013). Almost all ILCRs of Cr(VI) and As bound to $PM_{2.5}$ exceeded the US EPA threshold of 1E-06, thereby implying high car risk for all ages, whereas high car risk due to inhalation of Cr(VI) and As in $PM_{0.1}$ was only obtained in the elderly adult group (>60 y). These risks may be underestimated or overestimated owing to additional exposure factors such as the toxicity of other components of PM, lifestyle, smoking habits, diet, outdoor exposure, and uncertain outputs. These initial results highlight the urgent concern about health risks associated with indoor PM exposure in urban areas of Vietnam.

3.6.3. Sensitivity analysis and limitations

The determination of car risk depends on exposure factors as input variables (C, ET, IR, EF, ED, AT, and BW). Generally, the mean values of the input variables are used to estimate the single average risk value, which may cause uncertainties. A Monte Carlo simulation was applied in our study to determine the probability density functions assigned to each variable. The simulation results for car risks are summarized in Table S10 and Fig. S8. The probabilities of lifetime car risk for TEs in $PM_{0.1}$ and $PM_{2.5}$ in winter were significantly higher than those in summer, with 90% of ILCR ranging from 2.9E-07 to 6.2E-05 for $PM_{0.1}$ and 3.2E-06 to 6.8E-05 for $PM_{2.5}$ in summer, whereas the values in winter varied from 6.6E-07 to 1.4E-05 for $PM_{0.1}$ and 6.5E-06 to 1.4E-04 for $PM_{2.5}$. The sensitivity analysis identified that the most influential factor was the TE concentration, which contributed to approximately 85%–96% of the ILCR variance, as presented in Figs. S9 and S10. BW, EF, ED, AT, and IR had negligible significance ranging from 1% to 6%, and BW and AT had negative values. This study had several potential limitations, namely (1) the limited specific building configurations in urban areas of Hanoi, (2) the small number of samples, and (3) overestimated or underestimated risk values due to the assumptions following the US EPA. Notwithstanding these limitations, this study provides valuable insights for developing effective measures for controlling indoor PM and protecting health, especially in winter in urban areas.

4. Conclusions

A comprehensive study of indoor PM in urban areas was conducted in Hanoi for both seasons, summer and winter. High levels of indoor $PM_{0.1}$ and $PM_{2.5}$ were found in the city, in which the average concentration of indoor $PM_{2.5}$ was about four times higher than the WHO

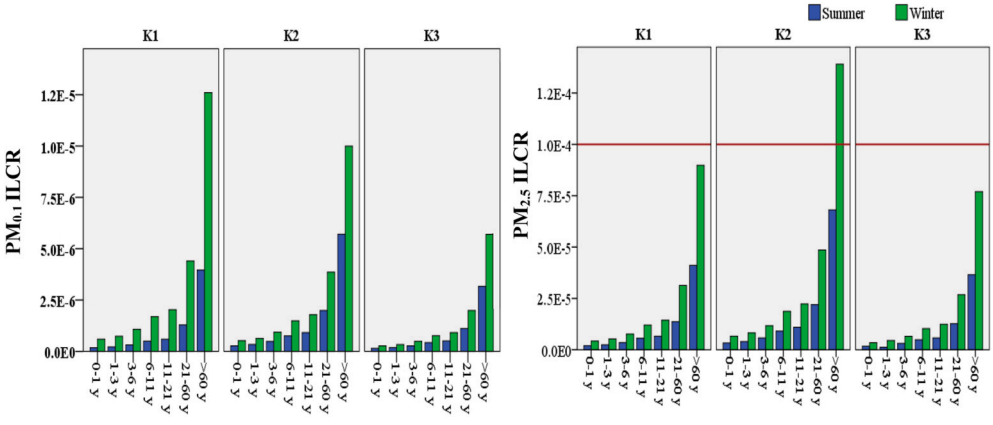

Fig. 5. Carcinogenic risk induced by trace elements in different age groups categories.

guideline. Seasonal variation was observed for indoor $PM_{2.5}$, but not for indoor $PM_{0.1}$. The average concentrations of TEs bound to indoor PM were 1140 ± 653 ng/m^3 and 138 ± 64.5 ng/m^3 for $PM_{2.5}$ and $PM_{0.1}$, respectively. Among the 10 investigated TEs, Zn, Pb, and Cr were the most abundant, and toxic elements such as Ni, Cd, and As exceeded the respective EU limits. This can result in a high potential for health risks as well as a large burden of disease for individuals who are exposed to TEs bound to PM.

Source apportionment indicated that indoor $PM_{2.5}$ was mainly derived from outdoor sources, whereas indoor $PM_{0.1}$ was derived from indoor and outdoor sources. Domestic coal combustion, traffic and industrial emissions were the main sources of TEs bound to both indoor $PM_{0.1}$ and $PM_{2.5}$ at the urban periphery dwelling (K1), whereas traffic emissions were the dominant sources of these elements at the roadside dwelling (K2). Traffic and domestic coal combustion were primary contributors to TEs bound to indoor $PM_{2.5}$ in winter and indoor $PM_{0.1}$ in summer, whereas indoor sources such as household dust and indoor combustion accounted for the majority of TEs bound to indoor $PM_{2.5}$ in summer and indoor $PM_{0.1}$ in winter at the urban dwelling (K3).

In winter, the deposited doses of TEs bound to PM, HI and ILCR at the roadside dwelling were significantly higher than those at other sites. The distribution of EDI of TEs in the human respiratory tract varied largely with particle sizes, in which, the majority of TEs bound to $PM_{2.5}$ was deposited in HA, whereas the dominant proportion of TEs bound to $PM_{0.1}$ was deposited in AL. The EDI in the HA region decreased as the particle size decreased, and increased in the TB and AL regions as the particle size decreased. Among the three investigated houses, the dwellers at the roadside house were exposed to the highest risk, where the intake of TEs in $PM_{2.5}$ can lead to high car risk for people over 60 years old and unacceptable non-car risks for all ages at the roadside house in winter. There were negligible effects of non-car and car risk, when exposed to indoor $PM_{0.1}$ and $PM_{2.5}$, for almost all ages, except for the above cases. Therefore, the findings of this study can provide a scientific basis for indoor air quality management, especially for the development of appropriate measures to protect human health in winter.

Credit author statement

Le-Ha T. Vo - Conceptualization, Methodology, Invetstigation, Formal analysis, Writing – Original Draft. Trung-Dung Nghiem - Conceptualization, Methodology, Writing - Review & Editing. Yoneda Minoru - Resources, Supervision, Methodology, Validation. Shimada Yoko - Supervision, Validation. Thuong T. Nguyen - Investigation, Validation. Thu-Hien T. Nguyen – Investigation. Dieu-Anh Van - Visualization.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apr.2022.101324.

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