Chemical characterization, source identification and health risk assessment of particulate matter pollutants in indoor environment, as a case study of Hanoi, Vietnam

Vo Thi Le Ha

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LIST OF ABBREVIATIONS

AL: Alveolar
BF: Breathing Frequency
BW: Body weight
Car-Risk: Carcinogenic Risk
CDM: Count Median Diameter
DF: Deposition Fraction
DONRE: Department of Natural Resources and Environment
$d_{aero:}$ Aerodynamic Diameter
$d_{th\,:}$ Thermodynamic Diameter
EDI: Deposited Dose
ET: Extra-thoracic
FRC: Functional Residual Capacity
GSD: Geometric Standard Deviation
I: Indoor
I/O: Indoor/Outdoor
Infiltration factor: F_{if}
ILCR: Incremental lifetime cancer risk
IARC: International Agency for Research on Cancer
ICRP: International Commission of Radiation Protection
HRT: Human respiratory tract
HA: Head Airways
HI: Hazard index
HMW: High molecular weight
HQ: Hazard Quotient
LL: Left Lower of lobe
LU: Left Upper of lobe
LMW: Low molecular weight
MMAD: Mass Median Aerodynamic Diameter
MPPD: Multiple-Path Particle Dosimetry
Month: m

Non-Car-Risk: Non-Carcinogenic Risk

PL: Pulmonary

PAHs: Polycyclic Aromatic Hydrocarbon

PM: Particulate Matter

TLC: Total Lung Capacity

TV: Tidal Volume

TB: Tracheobronchial

TE: Trace Element

RfD: Reference Dose

RL: Right Lower of lobe

RM: Right Middle of lobe

RU: Right Upper of lobe

URT: Upper Respiratory Tract

O: Outdoor

S: Summer

US EPA: United State Environmental Protection Agency

W: Winter

WHO: World Health Organization

y: Year

ABSTRACT

Vietnam has recently faced serious problems related to air pollution, primarily due to increased rates of urbanization in centers with high population densities, particularly in indoor air quality (IAQ). Poor IAQ can have a wide range of health consequences, including respiratory and cardiovascular illnesses, allergy symptoms, cancer, and early death (WHO, 1998). In addition, IAQ, which has a greater potential impact on public health than outdoor pollution, is considered to have been responsible for more than 3.8 million premature deaths around the word in 2016 (WHO, 2022)

This dissertation systematically examined the impact of indoor particulate matter (PM) and its components on the indoor environment, and analyzed the situation of indoor pollution in Vietnam. Specifically, this study clarified: *1)* the characteristics of PM in houses, such as mass size distribution, link between indoor and outdoor PM, and influencing factors; *2:* The chemical composition of PM (polycyclic aromatic hydrocarbon (PAH) and trace element (TEs); *3)* What are possible sources of indoor PM in urban area - Hanoi as case study?*; 4)* How does PM affect health assessments? Specifically, this study sought to clarify the distribution of deposited doses of PM and their composition in the human respiratory system and valuate the health risk assessments of the occupants of in the residential houses in urban area via the inhalation pathway.

In a review of 65 studies on PM pollutants and VOCs (Volatile organic compounds) in indoor and outdoor environments, a total of 19 total studies reported on PM, VOCs in different indoor contexts, and current mitigation measures related to IAQ in Vietnam. In some cases, high concentrations of PM ($PM_{2.5}$, PM_{10}, $PM_{0.1}$) and BTEXs (benzene, toluene, xylene, ethylbenzene) were observed in indoor environments such as residential houses, kitchens, parking basements, and offices in urban areas in Vietnam, in which the levels of $PM_{2.5}$ and benzene exceeded WHO guidelines; these levels pose a significant risk to human health. A variety of mitigation efforts focusing on controlling indoor and outdoor pollutants have been published to date; however, these measures have generally been insufficient due to lack of regulations, technical standards, and effective interventions related to IAQ.

To determine the seasonal variations of the mass concentrations of size-fractionated PM, the relationships among indoor and outdoor PM, PM size distribution, and deposited dose estimation were therefore clarified in urban residential houses in Hanoi. A total of daily 1240 PM samples ($PM_{0.1}$, $PM_{0.1-0.5}$, $PM_{0.5-1}$, $PM_{1-2.5}$, $PM_{2.5-10}$, and $PM_{>10}$) was collected simultaneously at four residential houses in summer and winter by using a nano sampler (Model 3182, Kinomax, flowrate 40 L/min), both indoors and outdoors. The four sampled houses (K1, K2, K3, K4) were representative of residential buildings with natural ventilation. K1 (urban periphery, two-story house) was located near roads and industrial zones. K2 and K4 (urban multi-story houses) were located in densely populated areas. K3 (roadside multi-story house) was located adjacent to a high-traffic-density road.

The results showed that the average concentrations of indoor $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$, and PM_{10} ranged from 5.3 to 8.9 $\mu g/m^3$, 10.8 to 20.1 $\mu g/m^3$, 20.5- to 47.6 $\mu g/m^3$, 33.7 to 105.9 $\mu g/m^3$, and 44.7 to 135 $\mu g/m^3$ among the four houses, respectively. The concentrations of outdoor $PM_{2.5}$ and PM_{10} were considerately higher than those of indoor $PM_{2.5}$ and PM_{10}, whereas the differences in the concentrations of $PM_{0.1}$, $PM_{0.5}$ and PM_1 were negligible. Seasonal variation was observed for indoor PM_1, $PM_{2.5}$, and PM_{10}, but not for $PM_{0.1}$ and $PM_{0.5}$, and the majority of indoor fractions originated from outdoor sources. Unimodal distributions of indoor particles determined the super-micron size range (1 to 2.5 µm) with highest concentrations, the lowest concentration observed for $PM_{<0.5}$ or $PM_{>10}$. Fine particles (i.e., $PM_{0.5-1}$ and $PM_{1-2.5}$) contributed predominantly to coarse particles both indoors and outdoors, suggesting that they posed a serious threat to human health. A multi-path particle dosimetry model (MPDM) was applied to predict the deposited doses of PM at different fractions in the human respiratory tract (HRT). The total deposition fraction (DF) obtained for PM_{10} was higher than those observed for $PM_{2.5}$, PM_1, $PM_{0.5}$, and $PM_{0.1}$. However, the total lobar DF was highest for $PM_{0.1}$, followed by $PM_{2.5}$, PM_{10}, PM_1, and $PM_{0.5}$ for all age categories (except for case of adults (>21y) for PM_{10}). PM_{10} deposited the greatest in the head airways (HA), whereas $PM_{0.1}$ deposited the greatest in the alveolar region (AL) and lobe region in the HRT. The deposited doses of PM increased with increased ages and PM sizes and the adults (>21y) was considered to be the most vulnerable group in this study for exposure to chronic effects.

To determine the chemical composition, source identification and health risk assessment associated with $PM_{0.1}$ and $PM_{2.5}$, 320 daily $PM_{0.1}$ and $PM_{2.5}$ samples were collected at the three dwellings (K1, K2, and K3, representing the urban periphery house, the roadside house, and the urban house, respectively) in Hanoi in summer and winter. The samples were analyzed for ten trace elements (TEs) (Cr, Mn, Co, Cu, Ni, Zn, As, Cd, Sn, Pb) and 15 Polycyclic Aromatic Hydrocarbon (PAHs) (Nap, Acy, Ace, Flu, Phe, Ant, Flt, Pyr, BaA, Chr, BbF, BaP, Ind, DahA, BghiP). Samples of indoor and outdoor daily $PM_{0.1}$ were collected using quartz filters (55 mm in diameter) by two identical Nano Sampler II (Model 3182, Kinomax). Samples of indoor and outdoor daily $PM_{2.5}$ were also collected in quartz filters (47 mm in diameter) using two similar cyclone samplers with a fiber holder (URG-2000-30EH, University Research Glassware Co., Chapel Hill, NC, USA) at a flowrate of 16.7 L/min. The enrichment factors, diagnostic ratios, and principal component analysis (PCA) were used to identify the possible sources of PM. The International Commission on Radiological Protection (ICRP) model was used to calculate the distribution of deposited of TEs or PAHs bound to PM in the HRT during exposure to these elements via the inhalation. The health risk assessment model of USEPA was applied to evaluate non carcinogenic risk (non-car risk) and carcinogenic risk (car risk).

The obtained results showed that the average concentrations of indoor $PM_{0.1}$ and $PM_{2.5}$ ranged from 7.0 to 8.9 $\mu g/m^3$ and 43.3 to 105.8 $\mu g/m^3$, respectively. The average concentrations of TEs bound to indoor PM varied from 66 to 216 ng/m^3 for $PM_{0.1}$ and 391 to 2356 ng/m^3 for $PM_{2.5}$. Meanwhile, the average $\sum_{15}$PAHs concentrations ranged from 102.9 ng/m^3 to 155.6 ng/m^3 and 25.3 to 52.9 ng/m^3 for indoor $PM_{2.5}$ and $PM_{0.1}$, respectively. The average concentrations of BaP bound to $PM_{2.5}$ and $PM_{0.1}$ were 2.6 ±0.4 ng/m^3 and 1.6 ±0.2 ng/m^3, respectively, which exceeded the threshold of the European guidelines, resulting in damage to the health of the occupants at three investigated houses

Domestic coal burning, industrial, and traffic emissions were considered to be outdoor sources, whereas household dust and indoor combustion were considered to be indoor sources. The outdoor sources (traffic, coal, biomass combustion) contributed significantly to PM at the urban periphery house and the roadside house, while indoor sources (combustion of incense, candle, natural gas, camphor usage, evaporation of building materials, paint wall, movements etc.) were the main sources of PM at the urban house.

The majority of $\sum$TEs or $\sum_{15}$PAHs or $\sum BaP_{(eq)}$ bound to $PM_{2.5}$ were deposited in the head airways (HA), whereas the dominant proportion of $\sum_{15}$PAHs or $\sum BaP_{(eq)}$ or $\sum$TEs bound to $PM_{0.1}$ was deposited in the alveolar region (AL) region. Doses of $BaP_{(eq)}$ bound to PM deposited greater in main extra-thoracic region (ET2) than in anterior nasal region (ET1); and

bronchiolar region (bb) than in bronchial region (BB). The 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. However, the occupants of the urban house were exposed to the highest risk, as the intake of BaP and DahA can lead to an increase in the potential cancer risk to the elderly group considering for individual setting threshold for cancer risk of USEPA (10^{-6}), although the cumulative cancer risk was within acceptable levels (10^{-4}) for all occupants of the three houses. Sensitivity analysis revealed that the concentration of TEs or $\sum BaP_{(eq)}$ was the most influencing factors on ICLR (Incremental cancer lifetime risks) variation, which contributed approximately 87% to 98% of the ILCR variance in micro-environment. The other variables such as BW, ET, AT etc. contributed the insignificance. In the macro-environment, the calculated ILCR showed a high potential cancer risk when based on the WHO method, while those ILCRs based on the CalEPA and US EPA methods showed that cancer risk was within acceptable limits.

Key words: *Indoor air, Particulate matter, Nano particles, Trace elements, Polycyclic aromatic hydrocarbons, Source apportionment, human risk assessment, International Commission of Radiation Protection, Vietnam.*

CHAPTER 1: INTRODUCTION

1. BACKGROUND

Indoor air quality (IAQ) refers to the air quality inside and around buildings and structures, especially as it relates to the health and comfort of building occupants. The term indoor air pollutants refer to the chemical, biological, and physical pollutants in indoor air. Therefore, interpreting and controlling indoor air pollutants involves assessing the possible risks to indoor activities (USEPA, 2021). Poor IAQ can cause variety of ailments, including respiratory and cardiovascular morbidity, allergic symptoms, cancers, and premature mortality (USEPA, 2021). More than 900 potential pollutants from thousands of different sources have been identified in indoor environments, which are currently recognized as having greater potential impact on public health than outdoor pollution (Petty, 2017).

In modern society, urban inhabitants spend most of their lives indoors (e.g., homes, schools, offices), where particulate matter (PM) ($PM_{2.5}$, PM_{10}, $PM_{0.1}$, etc.), NO_2, CO, volatile organic compounds (VOCs), and polycyclic aromatic hydrocarbons (PAHs), among others, are considered to be emerging indoor pollutants and toxins (Kim et al., 2013; Li et al., 2017; Petty, 2017; Vardoulakis et al., 2020). Indeed, household air pollution was ranked as the 10th most serious risk factor for mortality in 2019 and responsible for 2.7% of the global burden of disease in 2020 (GBD, 2020). Approximately 4 million premature deaths associated with pneumonia, stroke, ischemic heart disease, chronic obstructive pulmonary disease, and lung cancer worldwide were attributed to household air pollution in 2016 (WHO, 2018). Therefore, it is vital to characterize indoor air pollutants, define possible sources, and conduct health risk assessments in indoor environments.

IAQ is affected by many factors, such as house characteristics (ventilation mechanisms, building configuration, building materials, etc.), outdoor air environment (outdoor sources), metrological conditions, geographic location, life styles of inhabitants (type of stove, nature of fuels, cooking methods, use of household products, burning incense, smoking, etc.), and different intervention strategies (Petty, 2017; WHO, 2021). Furthermore, IAQ is also dependent upon internal factors such as the physical and chemical properties of pollutants (e.g., gases or particulate phases, reactivity, deposition, size) of PM (Vardoulakis et al., 2020; WHO, 2021).

In this perspective, great attention has been given to the characterization of the different size fractions of PM and their chemical properties in the indoor environment, especially in fine particles ($PM_{0.5}$, PM_1, $PM_{2.5}$) and nanoparticles ($PM_{0.1}$), which penetrate deeply into the lungs of the human respiratory tract (HRT), from where they enter the bloodstream and other organs after translocating into the circulatory system and induce inflammation (Topi and Hilkka, 2019). These particles are even more toxic as they can absorb more poisonous compounds, such as trace metals and PAHs, which adversely affect human health (e.g., reproduction, development, metabolic activities) (Kim et al., 2013; Kim et al., 2015). These emerging challenges in the area of public health cannot be ignored.

As a developing country, Vietnam has been facing serious problems related to air pollution, especially in Hanoi city, which has undergone a period of intense urbanization and increase in population, which in turn has resulted in severe air pollution, particularly in PM_{10}, $PM_{2.5}$ and $PM_{0.1}$ (Phung et al., 2021, Nguyen et al., 2018). Exposure to ambient PM was responsible for more than 3000 additional deaths in 2009 (Hieu et al., 2013), and admissions to hospitals among young children with respiratory ailments increased markedly in Hanoi in 2010 to 2011 (Luong et al., 2017). Outdoor air quality can strongly influence the IAQ, particularly in the context of Vietnam, where most people employ natural ventilation for thermal comfort and air circulation in the houses. Indoor air pollution has emerged as a critical issue due to the potential adverse impacts on human health, especially among urban citizens. Scholars should therefore drive their attention on examining the IAQ in Hanoi, focusing on characterizing PM and their chemical properties, identifying potential sources and distribution of deposited doses in human respiratory doses, and performing health risk assessment for the inhabitants of urban areas. This study was therefore undertaken to address these issues using a case study of Hanoi, Vietnam.

2. OBJECTIVES

This study characterized the PM in residential houses in urban area as a study case of Hanoi, Vietnam. Specifically, this study determined the distribution of particle sizes, examined chemical properties of PM at different fractions, identified PM sources, clarified the dose distribution of PM and their chemical components in the HRT, and performed health risk assessments. It is hoped that these findings will contribute to IAQ management in particular, and air quality management, in general. These data also provide the scientific basis for authorities and stakeholders to propose intermediate and appropriate measures, as well as to develop policies for mitigating indoor air pollution for protection human health in urban areas in Vietnam, in general, and in Hanoi, as a case study. The following objectives were raised.

1. Characterize indoor air pollution status in Vietnam and mitigation measures. The findings may provide important data for the management of air pollution in Vietnam in the near future.

2. Measure the mass concentration of size-fractionated PM in residential houses in Hanoi in different seasons (summer and winter), define the mass-size particle distribution, clarify the relationship between indoor and outdoor PM, and estimate deposited doses in HRT

3. Measure TEs bound to $PM_{2.5}$ and $PM_{0.1}$, estimate the infiltration factors, identify potential sources, determine the distribution of deposited doses of TEs in HRT and conduct health risk assessments.

4. Measure PAHs bound to $PM_{2.5}$ and $PM_{0.1}$, identify potential sources, determine the distribution of deposited doses of PAHs in HRT and perform health risk assessments.

5. Propose efficient measures to mitigate the health risk from PM exposure in indoor environments.

As shown in Figure 1, the dissertation consists of the following seven chapters:

Chapter 1: Comprehensive introduction of research background, objectives, and dissertation structure.

Chapter 2: Literature review on PM, trace elements, and PAHs and studies relevant to IAQ and regulations.

Chapter 3: Describe the current circumstances of indoor air pollution in urban areas in Vietnam.

Chapter 4: Measure the mass concentrations of size-fractionated PM in residential houses in an urban area in Vietnam in different seasons, clarify the relationship between indoor and outdoor PM, focusing on how they are correlated, their infiltration factors, mass size particle distributions, and the distribution of deposited doses of PM in the HRT.

Chapter 5: Analyze the trace elements bound to $PM_{2.5}$ and $PM_{0.1}$ in summer and winter and in indoor and outdoor air, and to identify possible sources, determine the distribution of deposited doses of TEs in the HRT, and perform health risk assessments.

Chapter 6: Determine the PAH bound $PM_{2.5}$ and $PM_{0.1}$ in summer and winter and in indoor and outdoor air, and to identify possible sources, determine the distribution of deposited doses of PAHs, BaP_{eq} in the HRT and perform health risk assessments.

Chapter 7: Summarize the thesis, followed by an overall conclusion derived from this research. In addition, the implications, limitations, and future directions of this study are also discussed. Structure of Ph.D dissertation Fig. 1.1

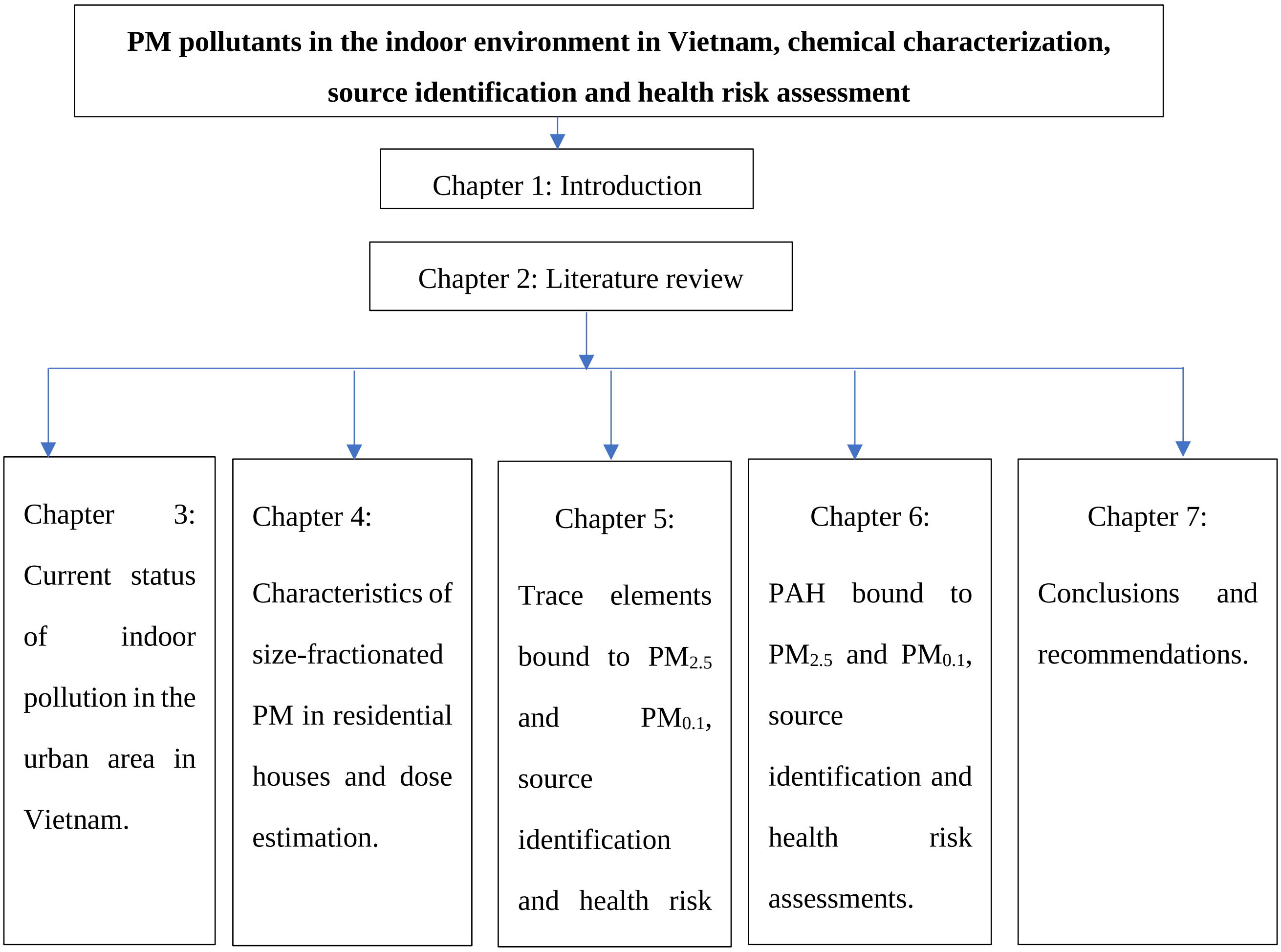


Fig 1. 1. Structure of dissertation

CHAPTER 2: LITERATURE REVIEW

1. CHARACTERISTICS OF ATMOPHERIC PARTICLES

1.1. The physical properties of particulate matter (PM)

Mass, number of particles, surface area, size distribution, and shape are all physical aspects of PM (Sanderson et al., 2014). The size of the particles is a significant factor that influences their behavior in the air as well as their physicochemical impacts on their surroundings. Particle size has a significant impact on human health, particularly in the respiratory system. Firstly, particle size influences particle deposition in the human respiratory tract (HRT) and affects the location of deposition in the HRT. Larger particles are deposited in the upper respiratory system, whereas smaller particles are deposited deeper within in the lower respiratory organs. PM deposition is highest when the particle size is <20 nm in diameter. Secondly, particle size determines surface area that comes into contact with tissue cells. The bigger the contact area, the greater the influence of the particles on the organs. Finally, particle size has an impact on particle removal; for example, only 20% of very small particles are removed after deposition in the pulmonary area, whereas up to 80% of particles >500 µm are removed by the body (Adachi, 2018; Kim et al., 2015).

Given this heterogeneity in PM size and shape, their kinetic behavior is frequently defined in terms of the diameter of an ideal sphere, also known as the aerodynamic diameter (AED), which is generally used to categorize particles by size. Although PM may be characterized in a variety of ways, one of the most important criteria for describing the capacity of PM to move in the atmosphere and/or inhaled is AED (Kim et al., 2015). The US Environmental Protection Agency (US EPA) has classified particles as coarse PM (PM_{10}), fine PM ($PM_{2.5}$), or ultrafine PM ($PM_{0.1}$) based on their expected penetration capability into the lungs (Kim et al., 2015). As illustrated in Fig. 1, the US EPA has grouped PM into size categories (segments) based on their size (Ivanova et al., 2016). The categorization of particles into modes in aerosol research is dependent on their diameter. Each mode has its own size range, formation mechanisms, origins, chemical properties, and route of deposition (Hinds, 1982).

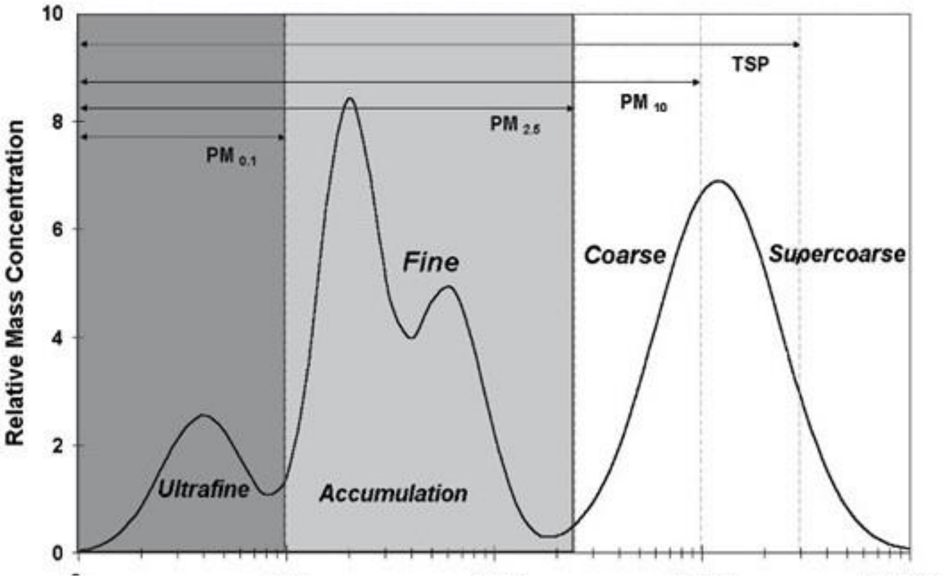


Fig.2. 1. Schematic representation of the size distribution of atmospheric particles (source: Ivanova et al., 2016).

In general, coarse mode particles were defined as those >2.5 µm in diameter. As shown by Fig. 2.1, coarse particles can be further classified into super-coarse and coarse categories. Typically, these particles settle out of the atmosphere within a few hours of formation (Ivanova et al., 2016). Coarse particles are primarily deposited in the HRT in head airways from where they are expelled from the body via the nose, by coughing or by swallowing. Due to their large size, super-coarse particles (i.e., >10 µm) are not regarded to be typically harmful to_respiratory system. However, total suspended particle matter may have an effect on the environment. The US EPA categorized particles (segments) according to particle size, as indicated in Table 2.1

Table 2. 1. Classification of particle segments according to particle size

N	Particle size	AED (µm)
1	Nano particle (NP)	AED ≤ 0.1
2	Fine particle (FP)	0.1<AED≤2.5
3	Coarse particle	0.1<AED≤10
4	Super coarse particle	AED >10

1.2. Chemical properties of PM

Airborne PM, which is a heterogeneous combination of particles with a diversity of chemical and physical properties, poses a danger to human health. These health concerns are affected by their size, concentration, and chemical components. PM can contain a wide variety of harmful agents, which are derived from industrial emissions, transportation emissions, domestic combustion, and unknown specific sources of human origin (i.e., anthropogenic sources), as well as the sea salt and dust (i.e., natural sources) (Ali, 2019; Karagulian et al., 2015). PM chemical components may be ions (NH_4^+, SO_4^{2-}, etc.), organic carbon (OC), elemental carbon (EC), trace elements (metalloid), and polyaromatic hydrocarbons (PAHs) (Ali et al., 2019; Cohen et al., 2010; Nguyen et al., 2018; Slezakova et al., 2014; Vo et al., 2022; Vu-Duc et al., 2021). Cohen et al., (2010) examined the chemical composition of fine PM in Hanoi, Vietnam, from 2001 to 2008. They found that the average concentration of $PM_{2.5}$ contained 29% ammonium sulfate, 8.9% soil, 28% organic matter, 0.6% salt, and 9.2% black carbon. The remaining 25% consisted primarily of nitrates and absorbed water. Ali et al. (2019) reported that the majority of coarse particles are formed of mineral from the earth's crust, biogenic material, and sea salt, whereas the majority of fine particles are composed of carbonaceous compounds with trace elements and organic substances. Ivanova et al.(2016) reported that PAHs were the most prevalent component of atmospheric nanoparticles in most settings. Some elements, such as potassium, calcium, and iron, were detected in high concentrations in nanoparticles from industrial activities. Biomass combustion is the primary source of potassium, whereas calcium is employed as an oil additive (Ali et al., 2019). Semi-volatile compounds were discovered in nanoparticles; these included organic substances, such as hopanes from engine oils, or condensed secondary organic aerosols, such as organic acids. Combustion-derived PM makes up the majority of EC (black carbon) and OC. PM has also been linked to a variety of trace elements, such as As, Pb, Ni, Cr, Cu, Sn, Mn, Co, Zn, and Cd (Ali et al., 2019). In indoor environments, Tunno et al. (2016) reported that chemical components in PM could be used as tracers of indoor sources, with Cd, BC, and K indicating smoking activities; Ca and Fe associated with cooking, As suggested domestic coal combustion; and Cu indicating personal activities. Al, Ca, Cu, Fe, and K were most frequently associated with soil and dust resuspension. On the other hand, the PAHs bound to PM served as tracers for indoor sources such as Naphthalene (Nap), which is associated with camphor and mothballs and other insect repellents (Batterman et al., 2012; Vardoulakis et al., 2020; Zhu et al., 2009);

Fluorene (Flu), Phenanthrene (Phe) , and Fluoranthene (Flt) are related to natural gas combustion and evaporation from building materials (Abdullahi et al., 2013; Vardoulakis et al., 2020); Benzo[b]fluoranthene (BbF), Benzo[a]pyrene (BaP), and Indeno(1,2,3-cd)pyrene (Ind) are indicative of burning of incense and candles (Derudi et al., 2012; Lin and Jhuang, 2012). However, these tracers have also been used for additional source identification (Derudi et al., 2012; Lin and Jhuang, 2012).

In summary, the physicochemical properties of atmospheric PM are highly variable, primarily due to the variability of the source, mechanism of formation, and geographical and meteorological conditions. The presence of hazardous components in PM, such as trace elements, PAH compounds, and other chemicals, increases their toxicity, causing a variety of adverse health effects, such as respiratory and cardiovascular morbidity, lung cancer, stroke, and asthma, as they penetrate deeply into the lungs (Ali et al., 2019; Kim et al., 2013).

1.3 PM Sources

PM originates from both natural and anthropogenic sources, both of which exhibit temporal and spatial variation. On a global scale, Karagulian et al.(2015) reported that urban ambient $PM_{2.5}$ and PM_{10} comprised 25% and 25% of traffic emissions, 15% and 18% of industrial emissions, 20% and 15% of domestic fuel combustion, 22% and 20% of unspecific sources, and 18% and 22% of natural sources, respectively. Unknown anthropogenic sources were mostly assigned to secondary particle formation, and natural sources were mostly natural dust and sea salt. As shown in Fig. 2.2, the contribution of PM-derived sources varied regionally. Traffic was discovered to be the primary cause of $PM_{2.5}$ in Southern Asia, India, Southern Asia, and Southwestern Europe, and PM_{10} in Africa and India, the United States, the rest of the Americas, and Northern China. For $PM_{2.5}$, industrial emissions were the dominant source in Japan, the Middle East and Southern Asia, and Turkey, whereas PM_{10} predominated in the rest of the Americas and Turkey. Domestic fuel combustion was a major contributor to $PM_{2.5}$ and PM_{10} pollutants in Central and Eastern Europe, as well as in Africa. Unknown sources emerged as a significant contributor to $PM_{2.5}$ and PM_{10} concentrations in Canada, the United States, the Republic of Korea, Western Europe, and Turkey. Natural sources were identified as the primary contributors to PM_{10} in the Middle East, Southwestern Europe, and Northwestern Europe, as well as of $PM_{2.5}$ in the Middle East and Japan (Karagulian et al., 2015).

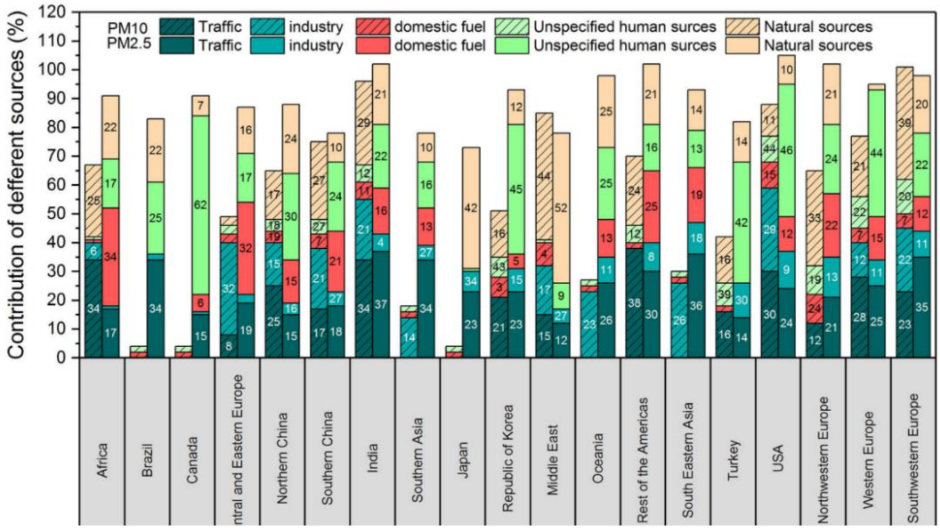


Fig.2. 2. Relative contribution of different anthropogenic sources (traffic, industry, domestic fuel, unspecified human sources) and natural sources to PM_{10} and $PM_{2.5}$ emissions worldwide.

Source: (Ali et al., 2019; Karagulian et al., 2015)

While a systematic global review of source contribution has not been conducted for indoor urban, the variations of the source contribution to PM have been reported. For example, Morawska et al. (2017) indicated that the primary sources of particle measurements vary according to the type of indoor setting. For households, outside air is the major source of both PM_{10} and $PM_{2.5}$, but indoor sources of nano particles (NP). Conversely, for schools and daycare centers, outdoor air is the origin of NP, but indoor sources are responsible for PM_{10} and $PM_{2.5}$. In offices, all three particle sizes come from outdoor air. These results are illustrated in Fig. 2.3.

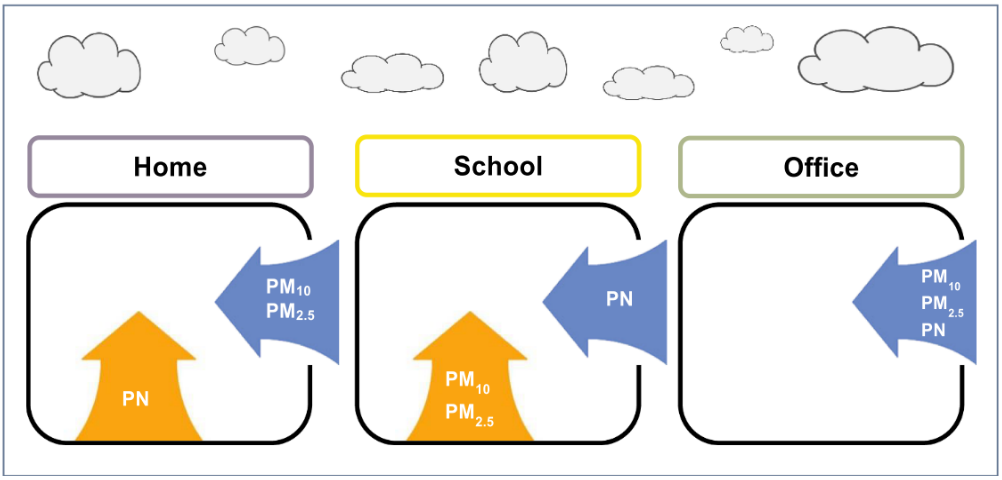


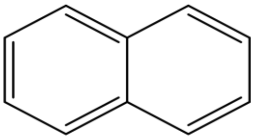
Fig.2. 3. Summary of indoor source versus outdoor origin of indoor particles (source: Morawska et al., 2017)

2. INTRODUCTION OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHS)

2.1. General information of PAHs

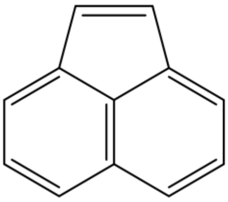
PAHs are a family of organic compounds that are composed of two or more aromatic rings directly attached to each other and in a structure that does not contain heterocyclic elements or substituents (Adeniji et al., 2018; Ali et al., 2016; Capuano et al., 2005). Hundreds of PAHs species have been discovered in recent years. However, of these, most attention has been focused on specific PAHs that have carcinogenic and mutagenic properties and are found in significant quantities in the environment. The US EPA has identified 16 PAHs that are of most concern in the air, seven of which have been categorized as carcinogenic substances, including chrysene, benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenz[a, h]anthracene, and seno[1, 2,3-cd]pyrene) (US EPA). The names and chemical structure of the 16 PAHs are shown in Fig. 2.4

PAHs are organic compounds with medium mass. Under normal conditions, they exist mainly as white or pale-yellow solids. PAHs have low volatility and low water solubility but high melting and boiling points (Adeniji et al., 2018; Capuano et al., 2005; Wilson and Jones, 1993). The melting temperatures of PAHs are distributed over a wide range, and high boiling points (>200°C) increase with an increase in their molecular weight. The evaporation pressure of PAHs decreases as molecular weight increases, and this has an effect on the absorption of PAHs in the particulate phase. Since low molecular weight (LMW)-PAHs are more volatile than high molecular weight (HMW)-PAHs under the same conditions, HMW-PAHs tend to be more bio-accumulative and environmentally persistent compared to LMW-PAHs (Adeniji et al., 2018). An increase in the number of benzenes will increase the hydrophobicity of PAHs. LMW-PAHs have high solubility and low bioaccumulation in organisms. Conversely, HMW-PAHs have low solubility, high biological accumulation, and low volatilities. PAHs are soluble in organic solvents and show affinity for lipids. PAHs are inert chemicals. Due to their benzene ring structure, PAHs exhibit aromatic features, including addition and displacement reactions, in which the photochemical oxidation reaction is critical for the degradation of PAHs in the air (Adeniji et al., 2018; Capuano et al., 2005; Wilson and Jones, 1993). The basic physical characteristics of some PAHs are described in Table 2.1 in the appendix.



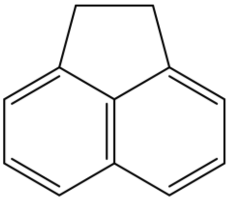
Naphthalene (Nap)

MW=128



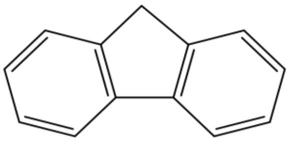
Acenaphthylene (Acy)

MW=152



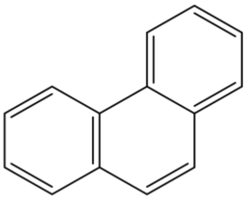
Acenaphthene (Ace)

MW=154



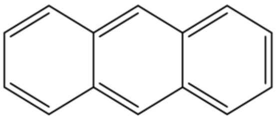
Fluorene (Flu)

MW=166



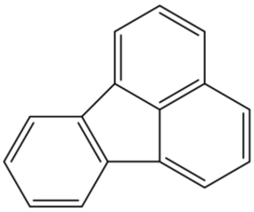
Phenanthrene (Phe)

MW=178

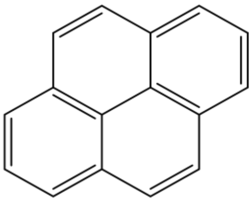


Anthracene (Ant)

MW=178

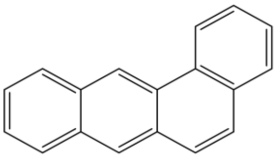


Fluoranthene (Flt) MW=20



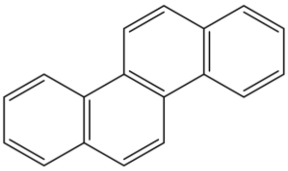
Pyrene (Pyr)

MW=202



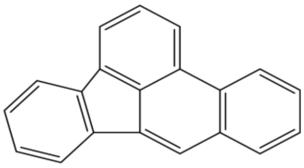
Benz[a]anthracene (BaA)

MW=228



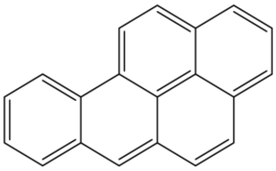
Chrysene (Chr)

MW=228



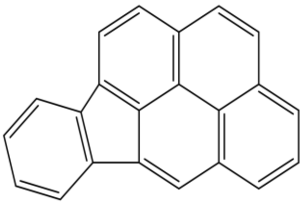
Benzo[b]fluoranthene (BbF)

MW=252



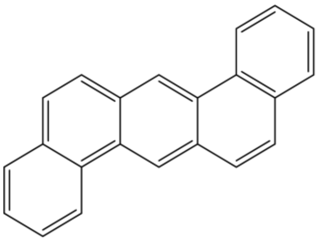
Benzo[a]pyrene (BaP)

MW=252



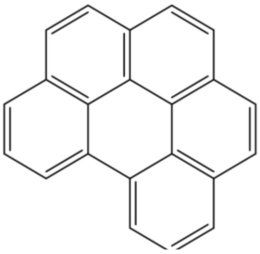
Indeno(1,2,3-cd) pyrene (Ind)

MW=276



Dibenz[a,h]anthracene (DahA)

MW=278



Benzo[g,h,i]perylene (BghiP)

MW=276

Fig.2. 4. Formulae of 16 PAHs identified by the US EPA as being harmful to human health

2.2. Toxicity of PAHs

To date, our understanding of the toxicity of PAHs is not sufficient to comprehensively assess the severity of their impacts on human health. A fundamental limitation on the risk assessment of PAHs is that the effects of PAHs in humans have been mainly evaluated from information on animal studies in experimental settings. Studies on the effects of PAHs on human health have focused on acute toxicity from short-term exposure, and chronic toxicity from long-term exposure. However, PAHs mainly exist at residue levels, so the main concern with PAHs is their potential to cause chronic impacts on the health of communities (Kim et al., 2013; Abdel-Shafy and Mansour, 2016). Humans are likely to be exposed to PAHs through three routes: inhalation via the respiratory system, dermal contact through the skin, and digestion via foods (Abdel-Shafy and Mansour, 2016). PAH toxicity is attributed to their capacity to interfere with cell membranes and also membrane-related enzyme systems. In addition, due to their high solubility in lipids, PAHs are easily absorbed by the gastrointestinal tracts of humans and other mammals (Kim et al., 2013; Abdel-Shafy and Mansour, 2016). The influence of PAHs on human health is primarily driven by exposure duration, environmental characteristics, and rate of exposure, as well as the concentration and toxicity of the individual PAHs. In addition, human factors such as health history, age, working habits, and daily life also determine the level of exposure to PAHs (Kim et al., 2013; Abdel-Shafy and Mansour, 2016). PAHs have been shown to have negative effects on human and animal health, such as causing skin allergies, asthma, reproductive abnormalities, hormonal imbalances, and neurological disorders, as well as being mutagenic, carcinogenic, and potent immunosuppressants (Kim et al., 2013; Abdel-Shafy and Mansour, 2016). Recognizing the serious potential human health risks and environmental threats associated with PAHs, the US EPA classified sixteen PAHs as being priority pollutants due to their health impacts. The International Agency for Research on Cancer (IARC) has also classified some PAHs as carcinogenic and mutagenic substances, as shown in Table 2.1 in the appendix

2.3. Sources of PAHs

PAHs are found in the environment principally as a result of incomplete combustion of carbon-containing fuels from natural, commercial, transportation, industrial, and residential sources (Kim et al., 2013). PAHs are also emitted by natural sources, such as volcanic eruptions, forest fires, savanna fires, and rock formation and erosion (Kim et al., 2013; Patel et al., 2020).

Anthropogenic sources PAHs result from incomplete combustion of coal, oil, and agricultural waste, as well as the production of cement, dyes, iron and steel, and rubber tires, and waste incineration (Patel et al., 2020). Transportation sectors also contribute to PAH emissions via off-road vehicles, light motor vehicles, ships, trains, and planes (Vega et al., 2021; Zheng et al., 2018). Domestic emission sources include garbage combustion, coal combustion for coking, wood fires, cooking on oil/gas burners and kerosene/wood stoves, and other home heating. (Abdullahi et al., 2013; Chen et al., 2016; Li et al., 2021). Combustion of open biomass and agricultural waste are two sources of agricultural emissions (Pham et al., 2019). Table 2.2 shows the PAH emission factors from various sources. PAHs are often discharged into the air in mixtures; however, they can be synthesized into separate compounds for scientific purposes or for application in specific manufacturing processes, such as the production of pharmaceuticals, dyes, plastics, and insecticides (Abdel-Shafy and Mansour, 2016). Some specific uses of PAHs are shown in Table 2.2 in the appendix.

2.4. Distribution PAHs in the air

PAHs in the air can be distributed in the gas phase or adsorbed onto suspended dust particles; this characteristic influences their mobility and form of existence. The distribution of PAHs between the two gas-particle phases is governed by atmospheric conditions (such as temperature and humidity), molecular weight, and physical and chemical parameters such as evaporation pressure and the partition coefficient Kow (Kim et al., 2013; Patel et al., 2020). LMW-PAHs are believed to have a strong affinity for the gas phase, while HMW-PAHs are mainly distributed in the particulate phase. Although LMW-PAHs are more typically identified in the atmosphere as vapor, they can also reside in the particulate phase as a result of condensation after their release. HMW PAHs, on the other hand, are predominantly found in the particulate phase (Kim et al., 2013). Since PAHs in the atmosphere can easily be inhaled by humans, considerable research on PAHs in the atmosphere has been conducted around the world during the last two decades (Kim et al., 2013; Li et al., 2021; Patel et al., 2020). Most of these studies have focused on PAH characteristics in indoor and outdoor environments in rural and urban areas, industrial zones and sources (Chen et al., 2016; Dat and Chang, 2017; Liaud et al., 2014; Palmisani et al., 2020; Romagnoli et al., 2014; Wang et al., 2016).

Table 2. 2. Emission factors of $PM_{2.5}$-bound PAHs (mg/kg fuel burned) and $PM_{2.5}$ (g/kg fuel burned) from various combustion sources (Source: Li et al., 2021)

PAH	Open combustion		Household biofuel combustion			Household coal combustion	Coal-fired industrial boiler (4 t/h)	Coal-fired industrial boiler (10 t/h)	Coal-fired power plant outlet of ESP	Coal-fired Power Plant Outlet of WFGD
	Wheat straw	Maize stover	Wheat straw	Maize stover	Wood	Bituminous coal	Bituminous coal	Bituminous coal	Bituminous coal	Bituminous coal
Nap	0.099±0.032	0.11±0.04	0.099±0.006	0.18±0.11	0.12±0.05	0.12[a]	0.013[a]	0.016[a]	0.0034[a]	0.002[a]
Ace	0.12±0.04	0.04±0.069	0.11±0.01	0.15±0.04	0.077±0.068	0.15	0.016	ND	ND	ND
Acy	ND	ND	ND	0.016±0.028	ND	ND	ND	ND	ND	ND
Flu	ND	0.089±0.155	0.12±0.11	0.064±0.11	0.057±0.098	ND	ND	ND	ND	ND
Phe	0.37±0.09	0.48±0.21	1.1±0.5	1.2±0.6	0.5±0.14	0.99	0.041	0.0057	0.0089	0.0053
Ant	0.011±0.019	0.027±0.046	0.093±0.072	0.099±0.094	0.017±0.029	0.1	ND	ND	ND	ND
Flt	0.8±0.32	0.58±0.15	2.2±0.7	2±0.3	0.86±0.46	4.7	0.039	0.0089	0.0067	0.0033
Pyr	0.79±0.31	0.56±0.16	1.8±0.6	1.7±0.2	0.71±0.37	4.2	0.028	0.0064	0.005	0.0022
BaA	1.7±0.4	1.9±0.4	1.6±0.2	1.7±0.4	1.6±0.3	2.6	0.14	0.017	ND	0.0015
BbF	0.22±0.12	0.56±0.75	1.2±0.02	1.2±0.4	1.3±0.4	2.1	ND	0.015	ND	0.0014
BkF	0.49±0.11	0.38±0.06	0.82±0.28	0.68±0.25	0.74±0.21	1.2	0.031	0.0038	0.0061	0.003
BaP	0.64±0.16	0.57±0.1	0.79±0.31	0.64±0.22	0.76±0.23	1.6	ND	0.0032	ND	ND
DahA	0.11±0.19	0.52±0.16	0.38±0.11	0.35±0.12	0.58±0.38	0.85	0.0049	ND	ND	ND
Ind	1.6±0.4	1.7±0.5	1.9±0.4	1.7±0.5	1.8±0.6	3.1	ND	ND	ND	ND
BghiP	0.43±0.09	0.32±0.07	0.8±0.29	0.62±0.22	0.72±0.18	1.7	ND	ND	ND	0.0025
∑PAH[b]	8±1.8	8.5±1.8	14±4	13±2	10±2	24	0.31	0.062	0.03	0.021
$\sum BaP_{eq}$	1.6±1	3.6±0.6	3.3±0.8	2.9±1	4.2±2	6.8	0.042	0.0068	0.00064	0.00063
$PM_{2.5}$	7.6±4.1	12±1	3.2±0.3	4.5±1	2.7±0.4	3.4	0.21	0.12	0.26	0.035

[a] Measurement results or combined analysis of several sampling filters, [b] Sum of emission factors of 16 PAHs

ND: Not detected or less than background

ESP: electrostatic precipitator

WFGD: wet flue gas desulfurization

It has been estimated that emissions of the 16 PAHs accounted for 520 Gg/y in 2004 in the world, with 56.7% of these emissions attributed to biofuel combustion, 17% to wildfires, and 6.9% to the utilization of consumer goods (Zhang and Tao, 2009). The top three ranked nations with the largest PAH emissions were China (114 Gg/y), followed by India (90 Gg/y) and the US (32 Gg/y). The sources of PAH in the various countries differed significantly from one another. For example, biofuel combustion was the most significant PAH discharge in India, whereas wildfire emissions were the most significant PAH source in Brazil, and consumer items were the most significant PAH source in the US. Coke ovens and biomass combustion were substantial sources of PAHs in China (Zhang and Tao, 2009).

2.5 Studies related to PAHs bound to PM in indoor air

Numerous scientific studies have shown that exposure to PAHs bound to PM can cause a variety of severe health outcomes, such as carcinogenicity, mutagenicity, and cardiovascular mortality (Durant et al., 1996; Kim et al., 2015; Lin et al., 2013; Zhu et al., 2009). Sources of PAHs included domestic heating, cooking, burning of incense and candles, evaporation, electronic devices, mothballs and camphol, and tobacco smoking in different urban indoor settings around the world (Abdullahi et al., 2013; Cai et al., 2018; Derudi et al., 2012; Krugly et al., 2014; Han et al., 2015). The main outdoor sources of PAH include vehicle emissions, industrial emissions, and coal and biomass combustion (Anh et al., 2019; Krugly et al., 2014; Zhu et al., 2009; Pham et al., 2019).

Several reports have focused on PAH bound to different size particles in different indoor environments globally. The distribution of particulate PAHs is influenced by PM particle size and PAH was found with higher concentrations than finer particles in previous studies such as Zhang et al.(2019); Chen et al.(2016); Zhu et al.(2009), and Liaud et al.(2014). It is likely that, since smaller particles have a greater relative surface area than large particles, they are enriched by the chemicals that condense and deposit on surfaces. Small particles, on the other hand, had a tendency to collide with one another and combine to form larger particles by condensation and migration (Lighty et al., 2000). According to Zhu et al.(2009) and Liaud et al.(2014), particulate PAHs were absorbed on $PM_{2.5}$ with a proportion of 59% to 97% to the total particulate PAH phase in Chinese residential dwellings, whereas PAHs were more concentrated in PM_1 in households and schools in rural and urban environments in France. Zhang et al. (2012) measured PAHs in different particles in residential houses, offices, and laboratories in China, and found that accumulation mode particles accounted for the majority of the total PAH loads (68.1% to 76.5%), followed by coarse particles (20.4% to 29.2%) and ultrafine particles (3.0% to 4.0%). PAH distribution on fine particles can be divided into two size ranges: $D<1$ µm and $D>1$ µm. For PAHs with low volatility, the distribution was mainly affected by the adsorption mechanism, whereas for highly volatile PAHs, their distribution was affected by the condensation mechanism, except for Flt (Pistikopoulos et al., 1990).

Numerous factors affect the distribution of particulate PAHs indoors, such as meteorological conditions, indoor activities (cooking methods, fuel types, indoor combustion, etc.), and building characteristics (building type, ventilation conditions, floor level) (Ansari et al., 2010; Chen et al., 2017; Downward et al., 2014; Ouyang et al., 2020; Romagnoli et al., 2014). Several studies have reported seasonal variations in PAHs bound to PM in indoor environments that was associated to meteorological conditions (Chen et al., 2017; Ouyang et al., 2020; Romagnoli et al., 2014). The highest concentrations of indoor and outdoor PAHs bound to $PM_{2.5}$ appeared in winter, followed by those observed in spring and autumn and lowest in summer in dormitories, offices, and residential homes in Beijing and Italy (Chen et al., 2017; Romagnoli et al., 2014), and the same trend was observed in middle schools in China (Ouyang et al., 2020). This obvious seasonal trend suggests that meteorological conditions, such as low temperatures, stable atmospheric layer, and conditions that favor the accumulation of PAH pollutants, lead to high PAHs during winter. Conversely, the high temperatures in summer may favor the desorption of PAHs, resulting in lower PAHs. Furthermore, domestic heating in the cold season is considered to be a significant contributor to seasonal variations in pollutants.

Fuel type also strongly influenced the indoor PAHs bound to PM (Ansari et al., 2010; Chen et al., 2016; Zhang et al., 2019). Zhang et al. (2019) reported that the total concentrations of PAHs in bedrooms and local kitchens using clean energy (electricity and LPG) were 2.2 and 1.7 times lower than those in houses using traditional biomass, respectively. The BaP_{eq} concentration in kitchens with biomass combustion was approximately 3.3 times that in kitchens using clean energy. The contribution of PAHs bound to fine particles (PM_1) to total particulate PAH emitted from utilizing clean energy (59.6%) was lower than the contribution of PM_1-bound PAHs to total particulate PAH produced by the combustion of firewood and crop residues (67.7%). The concentration of particulate PAHs increased in the order: electricity < LPG < firewood < crop residues. The finer particles ($PM_{0.25}$ and $PM_{0.25\text{-}1}$) contributed more PAHs from combustion of firewood and crop residue, whereas larger PM sizes ($PM_{1\text{-}2.5}$ and $PM_{>2.5}$) contributed more PAHs when using clean energy. The same trend in indoor particulate PAHs associated with fuel consumption was also observed in in rural residences in Shanxi, China by Chen et al. (2016). The majority of particulate PAHs was associated with particles with size (<1.0 µm), and higher concentrations of $PM_{0.25}$ bound to PAHs were observed in the kitchens using peat and wood as an energy source compared to houses using electricity, honeycomb coal briquettes, and LPG. HMW-PAHs showed higher tendencies to bind to PM_1. In rural homes in Northern India, levels of all respirable fractions (i.e., $PM_{2.5}$ and PM_{10}) and their PAH levels were almost two-fold higher in houses using plants and wood for cooking than houses using plants and cakes of cattle dung (Ansari et al., 2010). Downward et al.(2014) added that particle-bound PAHs (BaP, Flt, Chr) were between 3 and 8 times higher in houses that burned smoky coal compared to houses burning smokeless coal.

The effects of house configuration (i.e., of the enclosed space) on PAHs bound to PM indoors have also been examined in previous studies (Downward et al., 2014; Jung et al., 2011; Sharma and Jain, 2020). Jung et al.(2011) concluded that floor level and building type both influenced indoor particulate PAH concentration in residential houses. The $\sum_8 \mathrm{PAH}_{\mathrm{nonvolatile}}$ concentrations were lower at the same level floors in high-rise buildings compared to low-rise buildings in New York, whereas insignificant differences in $\sum_8 \mathrm{PAH}_{\mathrm{semivolatile}}$ concentrations were observed. In some instances, the concentrations of $\sum_8 \mathrm{PAH}_{\mathrm{nonvolatile}}$ measured in low-rise building were 10-30% higher than those in high-rise houses. Moreover, indoor $\sum_8 \mathrm{PAH}_{\mathrm{nonvolatile}}$ and $\sum_8 \mathrm{PAH}_{\mathrm{semivolatile}}$ were higher at the 3rd to 5th floors compared to either lower or upper floors only during the non-heating season. Sharma and Jain (2020) found that the level of PAHs bound to PM varied widely depending on kitchen characteristics. The application of improved cook-stoves resulted in a critical decrease in total particulate PAHs in 75%, 86%, and 90% of enclosed, semi-enclosed, and open kitchens, respectively, in houses with traditional cook-stoves in rural settings in India. Downward et al.(2014) reported that there was a 4- to 10-fold decrease in particulate PAHs in homes with ventilated stoves in comparison with unventilated stoves. The PAHs bound to $PM_{2.5}$ increased in switched-off ventilation systems in schools (Liaud et al., 2021). Indoor PM sources also affected the levels of particulate PAHs. For example, the concentrations of ∑PAHs in smoking residences were two-fold higher than those in non-smoking residences. Mothball emissions and cooking methods contributed 71.5% and 32.5% of indoor PAHs in residences in China, respectively (Zhu et al., 2009). Human activities inside schools increased PAHs bound to PM with diameter greater than 1 µm. However, most investigations focused on PAHs bound to fine and coarse particles, and very few studies were devoted to PAHs bound to ultrafine particles in indoor environments, which should be studied further

In terms of risk assessments, a few studies reported that PAHs are found predominantly in fine and finer particles, which can penetrate deeply into pulmonary alveoli, posing a serious hazard to human health. Despite their low mass, Kawanaka et al.(2004) found that ultrafine particles in the environment were major contributors to PAH deposition in the lung. Zhang et al.(2012) reported that accumulation particles produced the most PAHs in the pulmonary region in indoor urban environments. There is a lack of open data regarding fine and ultrafine particle-bound PAHs associated with health risk assessment in indoor urban areas, not only globally, but also for developing countries, particularly Southeast Asian countries, such as Vietnam, where indoor air quality (IAQ) has been challenging. Comprehensive studies are required to investigate the occurrence of PAH bound to fine and ultrafine particles in indoor urban environments, clarify seasonal variation and possible source apportionment, and then to estimate the distribution of deposited doses in the HRT and perform health risk assessments, all of which formed the goal of this dissertation.

3. INTRODUCTION OF TRACE ELEMENT (TE)

3.1. General information of TE

Trace elements (hereafter referred to as TEs) are minerals present in living tissues in small amounts. Some of them are known to be essential metals (e.g., Zn, Fe, Mn, Cu) and the rests are considered to be nonessential (e.g., Cd, Pb, Hg, As)(Council, 1989). Essential TEs are vital for sustaining living organisms and they are involved in processes such as growth, metabolism, and organ development. These elements are commonly utilized in trace concentrations of 10–15 ppm in various environmental matrices and are also referred to as micronutrients (Raychaudhuri et al., 2021). Non-essential TEs that are not needed by vegetation for any metabolic reactions, are known (Raychaudhuri et al., 2021).

3.2. Properties of TEs

TEs can form covalent bonds, which in turn affect their chemical characteristics. The ability to bind covalently to organic substances and nonmetallic elements of cellular macromolecules is the most notable consequence of these characteristic (Briffa et al., 2020). As a result, when TEs bind to organic compounds, lipophilic compounds are produced, which can be broadly transferred to the natural environment and induce a variety of different toxic responses. TEs can be absorbed by humans in three pathways: inhalation of TE bound to PM, digestion of TE-contaminated foods and liquids, and dermal exposure.

TEs are typically inert and non-biodegradable, which means that they tend to bioaccumulate. Living things can detoxify metallic ions by inactivating them within a protein or keeping them in an insoluble form within intracellular granules for disposal or long-term retention (Briffa et al., 2020; Potter et al., 2021). When living organisms inhale TEs bound to PM or digest TEs contaminated foods, these elements can bioaccumulate in their organs, potentially leading to acute and chronic effects (Briffa et al., 2020; Potter et al., 2021). The properties of TEs and their applications are summarized Table 2.3 in the appendix

3.3. Toxicity of TEs

TEs that are poisonous and neurotoxic, such as Mn, Hg, As Pb, Cu, and Ni, can adhere to PM surfaces and cause neurotoxic symptoms in human during the exposure. Epidemiological studies have found a link between exposure to Mn-bound PM and reduced dexterity in the elderly, as well as disrupted neurodevelopment in children (Potter et al., 2021) Inhaling PM bound to Cu can reduce motivation and change the basal ganglia in schoolchildren (Potter et al., 2021). Briffa et al. (2020) reported that metalloid ions react with DNA and nuclear proteins, causing DNA damage as well as disruption of the cell cycle, apoptosis, or cancer. The mutagenicity of TEs is caused by metal-mediated free radicals, indicating that there is a relationship between carcinogenesis and oxidative damage. Inhibition of DNA repair mechanisms by metals has been implicated in Cd, Ni, and As. As shown in Table 2.3, the IARC has classified metals into four categories based on their potential carcinogenicity.

Table 2. 3. Classification of heavy metal carcinogenicity (source: Briffa et al., 2020)

Category	Carcinogenic level in humans	Evidence	Heavy metal classification
Group 1	Carcinogenic	Sufficient evidence in humans	-Aluminum production (Al) -Arsenic and inorganic compounds (As) -Cadmium and cadmium compounds (Cd) - Nickel compound (Ni) -Chromium VI (CrVI)
Group 2A	Probably carcinogenic	Limited evidence in humans; sufficient evidence in animals	- Lead compounds inorganic (Pb)
Group 2B	Possibly carcinogenic	Limited evidence in humans; insufficient evidence in animals	- Nickel metallic and alloys (Ni) - Lead (Pb) - Cobalt (Co) - Methylmercury $(CH_3Hg)^+$
Group 3	Carcinogenicity not classifiable	Insufficient evidence in humans; insufficient evidence in animals	- Chromium III compounds (Cr(III) - Copper (Cu) - Mercury and inorganic mercury compounds (Hg) - Arsenic organic (As)
Group 4	Probably not carcinogenic	Evidence suggests carcinogenicity in humans and animals	- Manganese (Mn) - Silver; Zinc (Ag, Zn)

3.4. Sources of TE-bound PM

Heavy metals bound to PM can originate from both natural and anthropogenic sources. Natural sources include natural soil, geologic origin, and parent material, whereas anthropogenic sources comprised of industrial emissions, traffic emissions, coal combustion, pesticides and insecticides, construction activities and household wastes (Briffa et al., 2020; Hou et al., 2019). In terms of natural sources, Hou et al.(2019) reported that TEs, such as Cu, Zn, Cd, and Pb, in road dust could be attributed to 2% to geology, 6% to parent material, and 4% to natural soil in 53 cities in China from 1998 to 2008. Cr, Mn, and Co originated naturally from the Earth's crust (Nguyen et al., 2021; Wang et al., 2019). For man-made sources, traffic emissions contributed the greatest amount of TE in the form of road dust, followed by industrial emissions and coal combustion (Nguyen et al., 2021; Wang et al., 2019). The rapid growth in the number of vehicles and traffic jams were associated with TE enrichment in road dust (Nguyen et al., 2021). For example, Zn and Cu were released from tires and brake pads, as well as the corrosion of vehicles, while Cd and As were associated with gasoline combustion (Grigoratos and Martini, 2015; Nguyen et al., 2021). Hou et al. (2019) added that the second largest contributor of TE was industrial sectors, such as power plants, mines, metal smelters, and chemical plants. In another study from 1980 to 2008, coal combustion in four sectors (power plants, industrial sectors, domestic sectors and other sectors) in China induced the release of 230.38 t of Cd, 7574.28 t of Cr, and 9890.04 t of Pb into atmosphere (Tian et al., 2012). Among the four sectors, industrial activities were responsible for 88.3%, 86.7%, and 81.8% of total Cd, Cr, and Pb emissions, respectively, and power plants contributed 9.5%, 31.2%, and 16.4%, respectively. Indoor TE-PM can be generated by both indoor and outdoor sources over time. Indoor origins included smoking, cooking, fuel combustion, and decorative materials (E.g., paints), whereas outdoor origins included soil, mining, smelting, industrial activities, and vehicular emissions (Shi and Wang, 2021). Combustion of domestic coal was considered to be a significant indoor source of As and Cr in residential houses in urban areas (Vo et al., 2022); yellow pigments (Pb chromate) and other Pb pigments may play a major role in Pb levels in household (Shi and Wang, 2021). Smoking tobacco in a residential house can contribute to Cd and As (Yang et al., 2018), and wall dust and wooden furniture/paper products were major sources of Ca, Fe, Mn, Fe Ma, Cu, and Ni on a residential academic campus (Suryawanshi et al., 2016).

3.5. Distribution TEs in the atmosphere

Pollutants enter the atmosphere in various forms, and particulate TE is released from two types of source categories, namely anthropogenic and natural sources (Briffa et al., 2020; Hou et al., 2019). TEs bound to PM and deposited dust in both indoor and outdoor air were investigated around the world by Pacyna and Pacyna (2001). Their findings showed that total global emissions of trace metals through fuel combustion by point sources in 1995 were 1475 t of Hg, 809 t of As, 691 t of Cd, 10145 t of Cr, 7081 t of Cu, 9417 t of Mn, 86110 t of Ni, 11690 t of Pb, 3517 of Sn, and 9417 t of Zn. Accordingly, the total emissions of TEs in Asian countries accounted for 42.2% of global TE emissions in 1995. In the case of indoor dust, Shi and Wang (2021) reported that TE concentrations varied markedly across the globe and were closely linked to the human activities in a region, e.g., mining, smelting, e-waste recycling, Pb-related industries, and traffic emissions. Cu and Zn were the most pervasive pollutants, followed by Pb and Ni. In comparison to other indoor environments such as buses, residential houses, museums, offices, public places, schools, and industrial workshops, the contents of TEs in indoor dust in the e-waste workshop were the highest, with median concentrations of Pb, Cu, Ni, and Cd approximately 67, 63, 5.37, and 4.27 times higher than the permissible WHO limits for soils, respectively (Shi and Wang, 2021).

3.6. Studies related to TEs bound to PM in indoor air

TEs bound to PM is an emerging pollutant in different indoor environments. In residential houses in urban areas in Vietnam, average levels of As and Ni exceeded EU limits by approximately 2 to 5 times, and concentrations of Zn, Pb, and Cr were the most abundant of the TEs bound to $PM_{2.5}$ and $PM_{0.1}$ (Vo et al., 2022). In residential houses in China, Wang et al.(2018) reported that Fe, Zn, Ni, and Mn were the main constituents of the metal elements in indoor TE-$PM_{2.5}$, Cd was the most enriched element, and Ni and As exceeded EU thresholds. In a study by Wang et al. (2019) on dormitories in China, As was higher than EU guideline, although Fe, Zn, Pb, and Cr were predominant. The exceedance of Cd and Pb for EU thresholds was shown during the renovation of laboratory in Malaysia (Talib et al., 2011). During the heating and non-heating seasons in China, As and Cd bound to $PM_{2.5}$ in residential houses were higher than EU guideline (Yang et al., 2018). In a Portuguese hospital, As was accounted for the majority of $\sum$Car-TE bound to $PM_{2.5}$ and $PM_{2.5-10}$ (Slezakova et al., 2012). However, only

As bound to $PM_{2.5}$ was more than 16 times above EU limits, potentially causing serious damage to human health.

The particulate TE concentration can vary broadly due to differences in geography and human activities. Kulshrestha et al.(2014) reported that Pb and Ni at roadside houses were to 5 and 21 times higher than those in urban and rural houses, respectively**,** whereas Ni at rural houses was up to 36 times higher than that in roadside and urban houses. Compared to urban houses, the higher TEs observed at roadside house was closely related to traffic emissions (Vo et al., 2022). Mn, Ni, Pb, and Sb concentrations in school environments in Poland were higher in urban areas than in rural areas, although Cr concentrations were relatively high in rural schools (Mainka and Zajusz-Zubek, 2019). The burden of Ni, Pb, Cr, Cu, and Mn in urban areas in Vietnam was due to the heavy traffic and condense population (Nguyen et al., 2021). As a result of the close association between traffic and industrial activities, the greater concentrations of total suspended particulate matters (TSP) were obtained at industrial sites than at residential sites (Kwon et al., 2019). Indoor human activities have great influence on IAQ (Kelepertzis et al., 2019; Cheng et al., 2018; Yang et al., 2018). In residential houses in China, Yang et al. (2018) reported that the concentration of indoor Cd bound to $PM_{2.5}$ was closely associated with smoking activities, outdoor PM concentrations, and building age, whereas indoor Pb concentrations were governed by outdoor Pb levels, the time that windows were open, building age, and relative humidity. The levels of Cd and As in smoking homes were significantly higher than in non-smoking homes in Beijing, China, which was also observed in a smoking café in Iran (Masjedi et al., 2020; Yang et al., 2018). Masjedi et al. (2020) concluded that active waterpipe heads, type of tobacco, and the floor materials were all contributing factors to the generation of TE in the smoking café. The greater TE bound to $PM_{2.5}$ was found in waterpipe café in comparing to that in cigarette counterparts. The individuals consuming waterpipes with fruit-flavored tobacco are subjected to higher levels of TE exposure than individuals exposed to traditional cigarette smoke.

Heavy metal bound to PM in different indoor environments varied significantly according to the season. The levels of TE bound to $PM_{2.5}$ and $PM_{0.1}$ increased considerately in urban houses in winter, which was also observed in study of Vo et al.,(2022). This variation was attributed to different meteorological conditions (i.e., wind direction, wind speed, rainfall, relative humidity) between two seasons. Similar findings were reported by Schiavo et al.(2021), who concluded that the content of Mn in PM (<20 µm) at an urban school in Hermosillo Mexico varied significantly between pre- and post-monsoon seasons. Similar variation of TEs bound to $PM_{2.5}$ was also reported in the heating and non-heating seasons in indoor environments in China (Yang et al., 2018). However, there were only minor changes in Mn levels for coarse fractions (<45 µm, <63 µm, <125 µm). No seasonal trend was also observed in TE bound to $PM_{2.5}$ in summer and winter at childcare facilities in Korea, which was consistent with the seasonal trend in indoor $PM_{2.5}$ mass concentrations (Oh et al., 2019); however, clear seasonal variations were observed in outdoor $PM_{2.5}$ concentrations. As a result of the considerable variety of data obtained from different geographic locations and indoor activities, not enough information is available on the influence of seasons on indoor TE bound to PM to draw conclusions.

IAQ was highly affected by the characteristics of surveyed houses. For example, ventilation is a possible factor affecting indoor pollutants. The concentration of Pb was higher in residential houses with more open windows and/or less time operating an air conditioner during the non-heating season (Yang et al., 2018). The likely explanation for this observation is that air conditioners can filter indoor $PM_{2.5}$ via the deposition. Wang et al.(2018) confirmed that application of heating, ventilating, and air conditioning system (HVAC) improved IAQ considerably, with 22.1% and 29.1% decreases observed in TEs contents and mass concentrations of $PM_{2.5}$, respectively. The house age was also considered to be an influencing factor, with indoor Cd and Pb both increasing in old houses (Kelepertzis et al., 2019; Yang et al., 2018). The levels of Cr, Cd, Pb, Zn, Cu and Ni in dust house rose with the time for last paint and age of building, which was attributed to paint peeling off walls (Cheng et al., 2018). Different colors of wall paint were linked to different TEs in house dust; for example, Cd, Cu, Pb, and Zn were associated with yellow paint, Zn and Pb were associated with purple paint, and Cu was linked to green paint (Briffa et al., 2020; Cheng et al., 2018). Cheng et al. (2018) found that higher TE concentrations in house dust were observed at lower floors. Similarly, cafés in basements accumulate higher levels of TE bound to $PM_{2.5}$ than in first floor (Masjedi et al. 2020).

Particle size governs the distribution of TE bound to PM, which is extremely important for assessing the potential health impacts of TEs. Fe, Ca, K, Cr, and Cu all have a close association with coarse particles, whereas Mn, Mg, Zn, Al, Ni, and Pb tend to bond to fine particles in indoor environments in India (Rohra et al., 2018). The specific surface area of particles was proportional to the particle size, which can affect the amount of TE. Niu et al. (2010) reported that the levels of V, Mn, Ni, Cu, Zn, Se, and Cd increased as PM size decreased, which contributed the dominance of these TEs in the nanoparticle size range. Meanwhile, Fe, Sr, Mon, Sn, Sb, and Pb were most abundant in in fine particles. Elevated concentrations of metals in the nano and fine particle size ranges are important due to capacity of these PM to permeate into the alveolar region. Similarly, most of the TEs bound to $PM_{2.5}$ measured in barbeque restaurants had greater concentrations than in coarse particle ($PM_{>2.5}$). Similarly, V, Se, Zn, Cr, As, Cu, Ni, and Pb all bound to $PM_{2.5}$ (Taner et al., 2013), who also confirmed that PM with an AED < 0.8 µm concentrated the most trace elements, which was attributed to larger surface area

Many scientific studies have been undertaken around the world to identify potential sources of TEs in various indoor settings (Kulshrestha et al., 2014; Suryawanshi et al., 2016; Tunno et al., 2016; Vo et al., 2022; Vu et al., 2017; Wang et al., 2019; Yang et al., 2018). For example, As, Cr, and Cd are all indicators of indoor fuel combustion; Ca and Zn are indicators of indoor cooking; Pb, Cu, Ni, Co, and Cr are indicators of wall paint dust; Cd and As are indicators of smoking activities; and Cu is an indicator of cleaning activities. Traffic and industrial emissions (Cd, Zn, Ni, Pb, Cu) were identified as outdoor sources as well as being indicators of domestic combustion (As, Cd, Cr).

4. STANDARDS AND REGULATIONS RELATED TO PM IN THE ATMOSPHERE

Good air quality is regarded as a fundamental prerequisite for human health and well-being. To protect human health, many countries and international organizations have established air quality standards, regulations or guidelines which have become an important component of risk management and environmental strategies at both national and international scales. However, there is not enough evidence to devise safe limits or threshold levels for exposure below which there are no negative health effects. Since PM is pervasive, it plays a key role in human health determinants. PMs should be regulated in order to minimize their potential harm to human health and the environment, and guidelines and standards for protecting public health under a variety of circumstances (see Tables 2. 4 and 2.5) should be developed.

Table 2. 4. Air quality guidelines for particulate matter (WHO, 2021)

Particulate matter	Average	Interim target				AQG level
		1	2	3	4	
$PM_{2.5}$ µg/m^3	Annual	35	25	15	10	5
	24 h	75	50	37.5	25	15
PM_{10}, µg/m^3	Annual	70	50	30	20	15
	24 h	150	100	75	50	45

AQG: Air quality guideline

Table 2. 5 Air quality guidelines for potential toxic elements.

N	Element	Threshold	Agency
1	As	6 ng/m^3	EC (European Commission), Directive 2004/107/EC EC (European Commission), Directive 2008/50/EC for annual exposure ambient air
		1.5 ng/m^3	WHO guideline for air quality
		1 µg/m^3	WHO Air Quality Guidelines for estimation of cancer risk for a lifetime exposure.
		10 µg/m^3	OSHA - Legal limit for an 8-hour workday of exposure to airborne arsenic in places that use inorganic arsenic
2	Cd	5 ng/m^3	EC (European Commission), Directive 2004/107/EC EC (European Commission), Directive 2008/50/EC For annual exposure ambient air
		5 ng/m^3	WHO guideline for air quality
		5 µg/m^3	OSHA- Legal limit for an 8-hour workday of exposure to cadmium in air
3	Ni	38 ng/m^3	WHO guideline for air quality
		20 ng/m^3	EC (European Commission), Directive 2004/107/EC EC (European Commission), Directive 2008/50/EC for annual exposure
		1 mg/m^3	OSHA- Legal limit over an 8-hour workday of Ni in air
4	Pb	500 ng/m^3	EC (European Commission), Directive 2004/107/EC EC (European Commission), Directive 2008/50/EC For annual exposure ambient air
5	BaP	1 ng/m^3	EC (European Commission), Directive 2004/107/EC EC (European Commission), Directive 2008/50/EC For annual exposure ambient air
6	Nap	0.01 mg/m^3 (annual average concentration)	WHO guidelines for indoor air quality: Selected pollutants Annual exposure to indoor air

CHAPTER 3

INDOOR POLLUTANTS IN URBAN ENVIRONMENT IN VIETNAM

1. INTRODUCTION

Indoor air pollution has been a major threat to global public health as people spend much more time in enclosed spaces than outside. This is particularly the case in urban areas where approximately 80% to 90% of our time spent in indoor spaces (Martins and da Graça, 2018). The_World Health organization_(2016) reported that indoor air pollution leads to premature deaths for 4 million people each year in low-to-middle-income countries. Air quality in Vietnam, especially in Hanoi, has recently deteriorated with the high concentration of particulate matter (PM), especially fine particles ($PM_{2.5}$) (Anh et al., 2021; Nguyen et al., 2018) and nanoparticles ($PM_{0.1}$), (Vo et al., 2022), as emerging indoor pollutants in particular and air pollutants in general. Population growth coupled with the emissions from motorcycles and cars due to rapid urbanization makes air pollution increasingly severe, its negative impact is estimated to cost up to 5% of GDP._As a result, air pollution by PM has been identified as a major environmental concern in Hanoi (Anh et al., 2021; Nguyen et al., 2018; Vo et al., 2022) that was responsible for more than 60000 deaths from heart disease, stroke, lung cancer, chronic obstructive pulmonary diseases, and pneumonia in Vietnam in 2016 (WHO, 2016). Indoor PM is strongly influenced by both outdoor and indoor sources. Outdoor sources include traffic, industrial activities, and combusting fuels, all of which are increasing due to the growth of industrialization and high energy consumption and indoor sources (Martins and da Graça, 2018). Indoor sources are attributed to the combustion of fuel for cooking, cleaning, sweeping, burning incense, and smoking, as well as human activity and equipment operation (Abdullahi et al., 2013; Isaxon et al., 2015; Martins and da Graça, 2018; Wieslander et al., 2014). These particles may reach buildings interiors via penetration and infiltration through the cracks and ventilation systems (Dickerhoff et al., 1982). Dickerhoff et al. (1982) confirmed that infiltration air leakages from walls in air-tighten buildings accounted for 18% to 50%; whereas closed windows/doors were responsible for 6% to 22% of infiltration. In addition to PM, volatile organic compounds (VOCs) are also important indoor air pollutants (Delgado-Saborit et al., 2011; Vo et al, 2018; Tran et al., 2020b). VOCs evaporate readily at room temperature and the inhalation route of exposure is the primary pathway for breathing in these contaminants. The most common VOCs are benzene, toluene, ethylbenzene, and xylene (BTEX), all of which have been found at high concentrations in public and residential indoor environments due to their high vapor pressure (Dai et al., 2017; Delgado-Saborit et al., 2011). The primary sources of BTEX indoor include outdoor air, tobacco smoke, fuel combustion, household products (e.g., cleaners, solvents, mothballs), and building materials (e.g., floor and wall coverings, carpet, insulation, paint, wood finishing's) (Dai et al., 2017; Delgado-Saborit et al., 2011). These compounds can cause a variety of health effects, including irritation of the eye, nose, and throat; headaches; loss of coordination; nausea; and liver, kidney, and central nervous system damage

(Bahadar et al., 2014). Benzene is most toxic chemical within the BTEX family and long-term exposure to benzene can increase incidence of leukemia and aplastic anemia in humans (Bahadar et al., 2014). The International Agency for Research on Cancer (IARC) has classified benzene as carcinogenic to humans (group 1) and ethylbenzene as being potentially carcinogenic to humans (2B) (Forrest, 2018). To date, few local studies have been conducted on PM, BTEX levels and their sources in different indoor environments in Vietnam. Nonetheless, some studies have examined aspects of the indoor air environment, such as Vo et al. (2020 a,b,c,d), Vo et al. (2017), Quang et al.(2017), Pham et al.(2020), and Tran et al.(2020b). However, very little open access data has been collected and few systematic reviews have been conducted on the IAQ of different indoor environments in Vietnam. This chapter therefore aims to comprehensively review the current status of indoor air pollution in Vietnamese cities, with a focus on the distribution of indoor pollutants, sources and potential mitigation measures, and an exploration of new insights for future study. The findings may provide valuable data for the management of air pollution in Vietnam in the near future.

2. METHODOLOGY

This review summarizes PM pollutants such as NP, $PM_{2.5}$, and PM_{10}, and VOCs (mainly in BTEX compound) in indoor environments in Vietnam and around the world. Scientific studies published in the last decade (2002–2021) were examined using a variety of sources, including Google Scholar, the Institute for Scientific Information (ISI) web of science, Scopus, Science Direct, PubMed, Vietnamese academic journals, and Vietnamese regional reports. Additional articles appearing in the “related citations” and “cited” were also verified. Articles were then selected depending on whether they met the following criteria: studies of PM and BTEXs (VOCs) in indoor environments in Vietnam and elsewhere; current air management in Vietnam; and articles with a sufficient level of statistical analysis (i.e., mean, standard deviation, standard error). A total of 65 studies focused on PM indoor/outdoor environments and VOCs (BTEXs) in different indoor environments. Of these, 19 examined PM and VOCs in different indoor contexts and current mitigation measures employed in air management system in Vietnam. Only publications that were published in peer-reviewed journals in English or Vietnamese were considered for this part of the study. Keywords used for the review were “PM”, “BTEXs”, “VOCs”, “indoor environment”, “indoor sources”, and “Vietnam”.

3. RESULTS AND DISCUSSION

3.1. Indoor PM and their sources in Vietnam

3.1.1. PM in different indoor environments in Vietnam

3.1.1.1. Current Indoor PM pollution status

Airborne PMs are widespread in indoor urban environments in Vietnam. The concentration of indoor PM is dependent on indoor activities, building characteristics, and outdoor sources, all of which vary in a spatiotemporal manner. In instances where an internal source is absent, indoor PM levels are expected to be lower than outdoor levels. As shown in Fig. 3.1, high levels of PM (PM_{10}, $PM_{2.5}$, PM_1, $PM_{0.5}$, $PM_{0.1}$) were observed in some indoor environments in urban areas in Vietnam. The preliminary results of PM_{10} in the parking basements and commercial areas of three complex buildings in Hanoi were reported (Anh Le and Thuy Linh, 2019). In that study, higher mean concentrations of PM_{10} were seen in the parking basement areas, compared with those in commercial areas. The PM_{10} concentrations in the parking basements exceeded WHO guidelines (PM_{10} for 24 h (50 µg/m^3)) These high PM_{10} levels were likely attributed to the high density of cars and motorbikes, which release PM as they enter and exit. Additionally, parking basements are tight spaces with poor ventilation, and these conditions may have been insufficient to disperse PM_{10}. Tran et al.(2020a) also analyzed the mass concentrations of PM_{10}, $PM_{2.5}$, and NP in two Hanoi preschools for four weeks in 2019. Their findings showed that the daily mean concentrations of PM_{10} and $PM_{2.5}$ during teaching hours were 59.7 µg/m^3 and 49.4 µg/m^3, respectively, whereas those of $PM_{0.1}$ (NP) were 3.1 µg/m^3. Indoor PM_{10} slightly exceeded the upper daily threshold set by WHO (50 µg/m^3), while indoor $PM_{2.5}$ concentrations largely exceeded recommended limits (25 µg/m^3). These concentrations were lower than those measured at ten schools in Hanoi, which were 131 µg/m^3 and 144.1 µg/m^3 for $PM_{2.5}$ and PM_{10}, respectively (Anh Le and Quynh Linh, 2020). In contrast, they were slightly higher than those observed at 16 nursery schools in Hanoi (38.7 µg/m^3) (Vo et al., 2020b). The differences in the PM concentrations at different schools were attributed to differences in school activities, PM sources, sampling time, sampling locations, and outdoor characteristics. In residential environments, Tran et al. (2021) reported that 47% of the investigated homes had a daily $PM_{2.5}$ concentration greater than >50 µg/m^3, which was limits recommended by the Vietnam technical regulations on ambient air quality (QCVN05/2013) and WHO guidelines. In particular, higher indoor levels of $PM_{2.5}$ levels were observed in homes that burn incense. Vo et al. (2020a) also reported that the average concentration of indoor $PM_{2.5}$ and PM_{10} in residential apartment in urban area in Vietnam exceeded WHO guideline by 2 to 3 times, and concentrations of $PM_{0.1}$, $PM_{0.5}$, and PM_1 corresponded to 7.51 µg/m^3, 16.6 µg/m^3, and 37.23 µg/m^3, respectively. The concentrations of PM_{10} observed in residential houses and schools were significantly higher than that in an empty office, as reported by Thuy et al. (2018). These findings imply that there is a considerable potential health risk associated with ultrafine and fine particles, and that the children and occupants of these dwellings should be made aware of these risks. There are no guidelines for indoor air quality (IAQ) in Vietnam. Studies on indoor PMs and their indoor sources in Vietnam are summarized in Table 3.1.

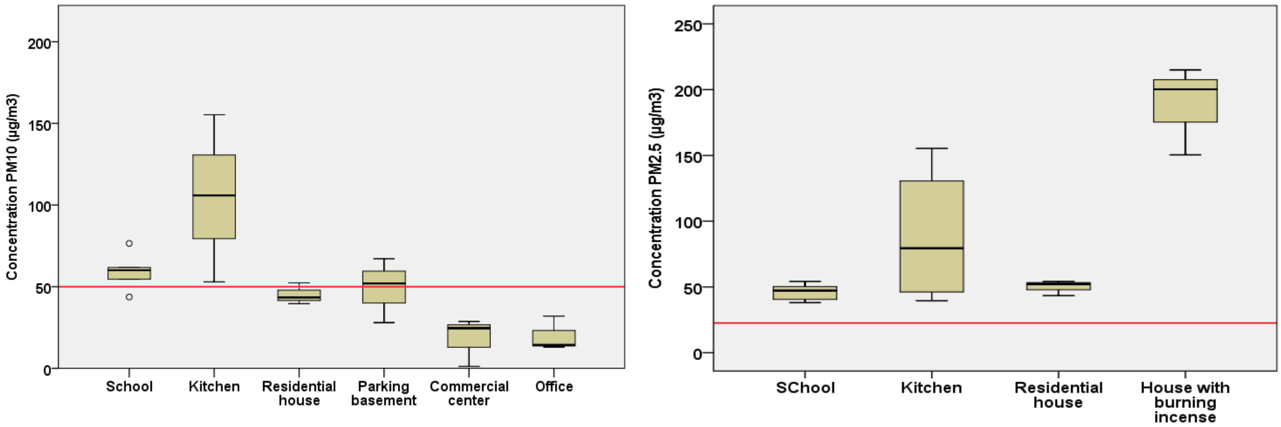


Fig 3. 1. Concentration of PM_{10} and $PM_{2.5}$ in different indoor environments in urban areas in Vietnam
Source: (Anh Le and Thuy Linh, 2019; Anh Le and Quynh Linh, 2020; Lê et al. 2018; Pham et al., 2020; Thuy et al. 2018; Tran et al, 2021; Vo et al, 2022)

3.1.1.2. Diurnal variation of indoor PM

Indoor PM pollution showed significant diurnal variation, and several studies have shown that indoor PM concentrations vary diurnally, which are strongly affected by human activities in the indoor environment. For example, Anh Le and Thuy Linh (2019) reported that the mass concentration of indoor PM_{10} increased sharply at weekends compared to weekdays at some parking basements and commercial areas in three building complexes. The maximum levels of PM_{10} in the parking basements and commercial areas reached 88.3 $\mu g/m^3$ and 40.7 $\mu g/m^3$ at weekends, which is more than twice the level on weekdays. These increases are attributable to the higher number of cars in the parking basements, higher levels of occupant activities, more intensive indoor sources in commercial areas at weekends. Quang et al.(2017) reported that there were great differences between indoor NP concentrations during rush hour (lunch time) and non-rush hour in six Hanoi households. During lunch break, the indoor NP count concentration increased transiently to 9.1×10^4 p/cm^3**,** which increased several folds compared to the non-rush hour when the count concentrations of indoor NP ranged from 1.5 to 3×10^4 p/cm^3. This relatively short peak in NP was primarily due to cooking activities.

3.1.1.3. Seasonal variation of indoor PM

To date, no large-scale surveys have been conducted to investigate the seasonal variation of PM in indoor environments in Vietnam. Thuy et al.(2018) investigated the seasonal variation of PM_{10} in several offices in Hanoi in 2015 and found that the mean mass concentration of PM_{10} was 13.3 $\mu g/m^3$ and 11.3 $\mu g/m^3$ in the wet and dry seasons, respectively. Thus, the mass concentration of PM_{10} showed very little variation between two seasons. Conversely, significant differences in the mass concentrations of PM_{10} and $PM_{2.5}$ were observed in offices in three Vietnamese cities (i.e., Hanoi, Ho Chi Minh, Da Nang) in summer and winter (Pham et al., 2020). For example, the mass concentrations of $PM_{2.5}$ and PM_{10} were higher in summer compared to those in winter. A total of 27% of the surveyed offices had PM_{10} and $PM_{2.5}$ levels that exceeded WHO recommended limits by 1.7 to 3 times in summer, whereas all offices had acceptable levels in winter. The tendency for these offices to close windows and use air conditioners in summer was the likely explanation for this phenomenon. In addition, clear seasonal variations were observed among particles of different sizes (PM_1, $PM_{2.5}$, PM_{10}, TSP) in a residential apartment in Hanoi, not for $PM_{0.1}$ and $PM_{0.5}$ in study of Vo et al. (2020a), in which their average concentrations of PM_1, $PM_{2.5}$, PM_{10}, TSP in winter was considerably higher than those in summer. In winter, the values of indoor $PM_{2.5}$ exceed the daily limit recommended by WHO (25 $\mu g/m^3$) by more than four times, whilst those of indoor PM_{10} exceed WHO limit (50 $\mu g/m^3$) by about three times. The likely explanation for high levels of PM is that these particles are resuspended in air by human activities, especially when these activities are conducted in a room with low ventilation rates due to closed doors and windows in winter periods. These high PM levels can be reduced to some extent in summer due to infiltration and higher air-exchange rates. This phenomenon supports the hypothesis of infiltration from outdoor PM, especially in houses with natural ventilation. The houses investigated in this study were naturally ventilated and IAQ was strongly influenced by infiltration of outdoor PM, which in turn was strongly correlated with meteorological conditions. These discrepancies in indoor PM concentrations were affected by outdoor PM status, indoor activities, ventilation conditions, windows/doors closed or opened, and the number of occupants between the summer and winter. In the context of Vietnam, the unique geographic location and climate of the region mean that PM concentrations are strongly affected by long-range transport pollution from the north and northeast monsoons, which may greatly influence on indoor PM during the dry and cold seasons.

3.1.2. Sources of indoor PM in Vietnam

The PM the indoor environment is derived from both outdoor and indoor sources (Fig. 3.2). There is a time lag for the outdoor PM concentration to affect the indoor PM.

3.1.2.1. Outdoor sources

In our modern society, people tend to spend more of their lives in enclosed spaces indoors, such as homes, offices, and schools, or inside other spaces (Martins and da Graça, 2018). In the urban areas of Vietnam, indoor PM is strongly influenced by outdoor PM sources, such as coal and biomass combustion, and vehicle and industrial emissions (Thuy et al., 2018; Pham et al., 2019; Vo et al., 2020b; Vo et al., 2022). These outdoor sources can penetrate into the indoor environment through filtration and infiltration. Outdoor air infiltration refers to the uncontrolled flow of air through cracks and leaks in the building envelope, resulting in the entry of outdoor particles. Air filtration refers to ventilation airflow, which can bring outdoor particles into the indoor environment through natural and mechanical ventilation (Dickerhoff et al., 1982; Quang et al., 2013). It has been reported that indoor-to-outdoor (I/O) concentration ratios of PM provide a rough indicator of pollution origin. If I/O value is less than the unity, then outdoor sources are considered to be the main source of indoor PM (Anh Le and Quynh Linh, 2020; Quang et al., 2017; Tran et al., 2020a; Vo et al., 2020b). Quang et al. (2017) reported strong correlations between indoor and outdoor concentrations of PM in naturally ventilated buildings when the I/O ratios of PM ranged from 0.7 to 0.9. These I/O ratios are comparable to those of naturally ventilated rooms at nursery schools (Vo et al., 2020b) and preschools in Hanoi (Tran et al., 2020a). These values imply that outdoor sources are the main contributor to indoor PM. Significantly lower I/O values were observed in tighter buildings and offices with air purifiers (Ly et al., 2018; Quang et al., 2017). It has been reported that air purifiers can reduce levels of PM_{10} by 23% to 43% in offices (Thuy, 2018). Thus, tighter buildings and the use of air purifiers can significantly reduce the contribution of outdoor PM sources to indoor PM concentrations (Ly et a., 2018; Quang et al., 2017).

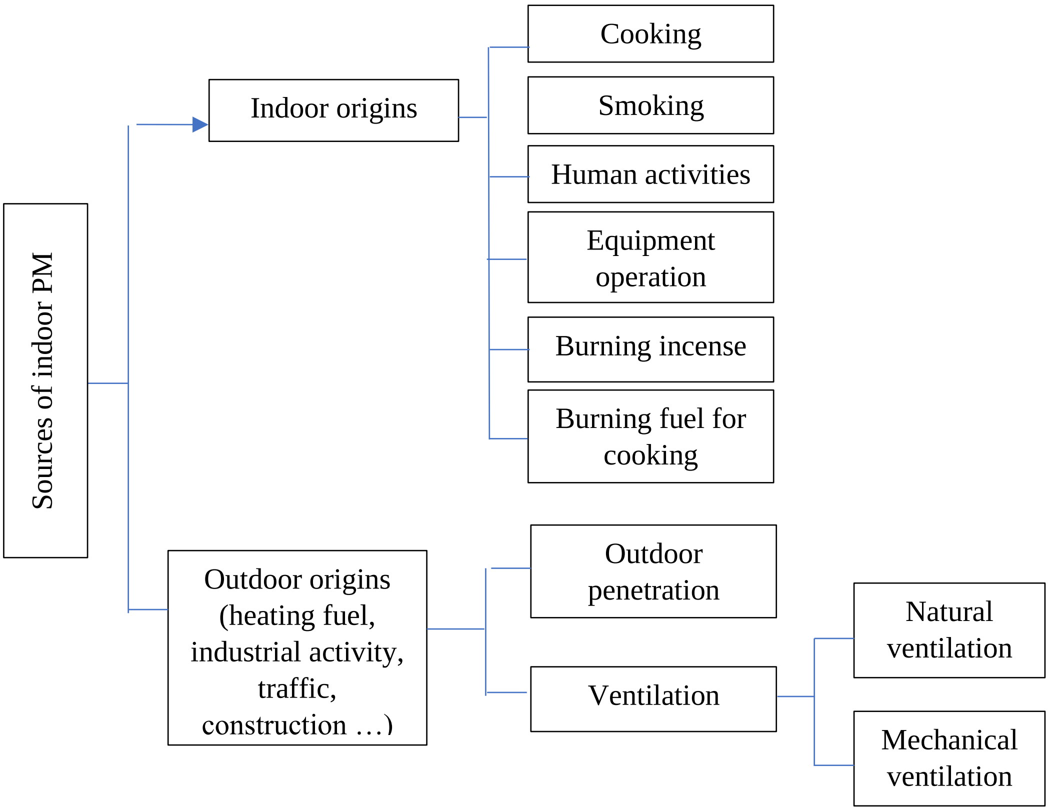


Fig 3. 2. Sources of indoor particulate matter

3.1.2.2. Indoor sources

There are numerous types of indoor PM sources, ranging over household fuel combustion, human activities, cleaning, cooking, and operating electronic equipment (e.g., printers, faxes and photocopiers) (Abdullahi et al., 2013; Lê et al., 2018; Martins and da Graça, 2018; Quang et al., 2017; Vo et al., 2022). Lê et al. (2018) reported that the combustion of various fuels for cooking, such as wood, coal, and liquid petroleum gas (LPG), in residential kitchens in Hanoi may release particles of various fractions (PM_1, $PM_{2.5}$, PM_{10}) at different concentrations. The highest concentrations of PM (PM_{10}: 305.7 µg/m^3; $PM_{2.5}$: 158.3 µg/m^3, and PM_1: 135.9 µg/m^3) were induced by wood combustion, followed by coal combustion and then LPG. Interestingly, when igniting coal in a stove, the peak concentration of $PM_{2.5}$ was 1537 µg/m^3. When wood was replaced with LPG, the PM concentrations in the kitchen were estimated to be reduced by approximately 70%. Tran et al.(2020a) examined the effect of children's activities on increased PM in preschools. Except for NP, their findings showed a significant difference in the mass concentration of PM_{10} and $PM_{2.5}$ between occupied periods (during teaching hours) and unoccupied periods (no children in the classroom). The mass concentrations of PM_{10} and $PM_{2.5}$ obtained during lessons were 2 to 3 times higher than when the classrooms were empty. The physical activities of students in the classrooms (running, playing with toys, movements, etc.) were responsible for the increased $PM_{2.5}$ and PM_{10} in the indoor environment. Similar results were also obtained in offices in Hanoi by Ly et al. (2018), who showed that an increased number of occupants and room sweeping could increase indoor PM_{10} by approximately 10-fold. Ngoc et al. (2017) reported that different cooking habits and cooking methods strongly affect PM generation. Tran et al. (2021) reported that burning incense was the highest contributor to total indoor $PM_{2.5}$, even higher than cooking, smoking, and cleaning activities in residential houses; for example, a 1-minute average peak of 825.5 µg/m^3 was observed for burning incense. The average concentrations of $PM_{2.5}$ were 1.3 to 3.3 times higher for intensive indoor sources (burning incense, cooking, smoking, cleaning) than for no indoor activities, respectively. Studies related to indoor PM and their sources in urban environments in Vietnam are summarized in Table 1.

Table 3. 1. Summary of indoor PM studies that have been conducted in Hanoi

Sampling site (date)	Sources	Average indoor concentration	Study
Office (2015)	Occupants' activities, sweeping floors, outdoor sources	Rainy season, PM_{10}: 13.3 μg/m^3 Dry season, PM_{10}: 11.2 μg/m^3	(Ly et al., 2018)
Households (2016)	Occupants' activities microwave for cooking, outdoor sources	NP: 1.9×10^4 p/cm^3	(Ngoc et al., 2017)
Building (2018)	Human and traffic activities	Commercial area, PM_{10}: 28.7 μg/m^3 Basement parking, PM_{10}: 67.1 μg/m^3	(Anh Le and Thuy Linh, 2019)
Nursery school (2018)	Pupils' activities and infiltration from outdoor sources	$PM_{2.5}$: 38.7 μg/m^3	(Vo et al., 2020b)
Residential houses (2018)	Occupants' activities Burning incense	Average $PM_{2.5}$: 51.2 μg/m^3 Burning incense: $PM_{2.5}$: 201.3 μg/m^3	(Tran et al., 2021)
Kitchen (2018)	Cooking with fuels	Wood stove: PM_{10}: 305.7 μg/m^3; $PM_{2.5}$: 158.3 μg/m^3; $PM_{1.0}$: 135.9 μg/m^3 Charcoal stove: PM_{10}: 96.8 μg/m^3; $PM_{2.5}$: 39.6 μg/m^3; $PM_{1.0}$: 41.6 μg/m^3 LPG: PM_{10}: 103.7 μg/m^3; $PM_{2.5}$: 52.9 μg/m^3; $PM_{1.0}$: 41.6 μg/m^3	(Lê et al., 2018)
Preschools (2020)	Traffic emissions, occupants' activities (moving around and sweeping the floor)	In the absence of children: $PM_{0.1}$: 1.4 μg/m^3; $PM_{0.5}$: 2.0 μg/m^3; $PM_{1.0}$: 10.3μg/m^3; $PM_{2.5}$: 15.7 μg/m^3; PM_{10}: 16.8 μg/m^3 In presence of Children: $PM_{0.1}$: 3.1 μg/m^3; $PM_{0.5}$: 7.9 μg/m^3; $PM_{1.0}$: 29.3 μg/m^3; $PM_{2.5:}$ 40.4 μg/m^3; $PM_{10:}$ 59.7 μg/m^3	(Tran et al., 2020a)
Apartment (2020)	Occupants' activities (moving around and sweeping the floor); domestic activities (cooking, cleaning and burning incense) and outdoor sources	In winter: $PM_{0.1}$: 8.08 μg/m^3; $PM_{0.5}$: 20.11 μg/m^3; PM_1: 47.63 μg/m^3; $PM_{2.5}$: 105.85μg/m^3; PM_{10}: 135μg/m^3; In summer: $PM_{0.1}$: 6.95μg/m^3; $PM_{0.5}$: 13.03 μg/m^3; PM_1: 26.83 μg/m^3; $PM_{2.5}$: 43.38 μg/m^3; PM_{10}: 59.27 μg/m^3	(Ha, 2020)

3.1.3. Other studies related to indoor PM

High mass concentrations of indoor PM and NP were observed recently in offices, preschools, nursery schools, and households in several short studies, suggesting that IAQ is poor in Hanoi (Anh Le and Quynh Linh, 2020; Ly et al., 2018; Quang et al., 2017; Tran et al., 2020a; ran et al., 2021; Vo et al., 2020b). However, the overall number of studies focused on indoor PM, PM sources, and I/O ratios in urban areas in Vietnam is low. In particular, studies on the spatiotemporal distributions of indoor PM and influencing factors are lacking. A comprehensive overview on indoor PM based on the findings of previous studies is urgently required for Vietnam.

Compared to previous studies, the mass concentrations of indoor $PM_{2.5}$ and PM_{10} in naturally ventilated preschool classrooms (Tran et al., 2020a) and nursery schools (Vo et al., 2020b) in Hanoi are significantly lower than those in New Delhi and Italy (Chithra and Shiva Nagendra, 2014; Schibuola and Tambani, 2020). However, the levels of these PM are higher compared to those in school environments in US (Carrion-Matta et al., 2019), Malaysia (Mohd Zahid et al., 2018), Korea (Rim et al., 2017), and Poland (Błaszczyk et al., 2017a). Mean count concentrations of NP at 6 Hanoi households correspond to those measured at three high-rise apartments in Beijing, China (Mullen et al., 2011). These values are also comparable to concentrations reported for residential houses in Denmark (Bekö et al., 2013). Table 2 summarizes some of the literature related to indoor PM and sources.

Spatial and temporal variations in indoor PM levels were observed in urban and rural areas as a direct consequence of the PM source and local climate. The concentrations of PM varied spatially (i.e., house location) and temporally (i.e., hourly, daily, seasonally). For example, in some offices, indoor PM_{10} levels in Hanoi were transiently worse during summer than they were in winter (Ly et al., 2018; Pham et al., 2019), and no data are available of the seasonal variation in PM in the offices in Hanoi. However, levels of indoor PM_{10} and $PM_{2.5}$ increased significantly in winter in residential apartments in Hanoi, Vietnam (Vo et al., 2020a), and a similar seasonal effect on indoor PM (PM_1, $PM_{2.5}$, PM_{10}) was also observed in residential homes in India and schools in China (Cao et al., 2019; Massey et al., 2012). In urban homes in Beijing, higher indoor $PM_{2.5}$ concentrations were seen in the early morning (5:00–8:00) and evening (19:00–23:00), which is attributed to heavy traffic emissions during the morning/evening rush hour as well as a lower mixing height in the atmosphere. The diurnal variation in residential indoor $PM_{2.5}$ concentrations tracked the ambient counterpart (Huang et al., 2015). In addition, Chithra and Shiva Nagendra (2014) reported that the maximum concentrations of PM during morning (8:00 to 9:00) and afternoon (14:00 to 16:00) in urban schools in India was attributed to widespread movements of the occupants at the start/end of the school day and lunch time. Compared to weekdays, the indoor concentrations of $PM_{2.5}$ and PM_1 decreased by 26% to 29% on the weekends. Błaszczyk et al. (2017a) added that the indoor concentrations of $PM_{2.5}$ at urban sites in Poland increased compared to those at rural sites.

Concentrations of PM in the indoor environment were also affected by meteorological factors, such as winds, humidity, and temperature. Chithra and Shiva Nagendra (2014) reported that wind speed was associated with changes in the concentrations of $PM_{2.5}$ and PM_{10} in the indoor environment. For example, high wind speeds decreased concentrations, as a consequence of mixing, dilution, and dispersion of the air pollutants. Increased outdoor humidity was associated with the precipitation, causing the washing out or absorption of pollutants. This lowered outdoor PM concentrations, and consequently, decreased indoor PM concentrations due to filtration and infiltration. Increased outdoor temperatures enhance particle migration into the indoor environment. It may be attributed to the temperature gradient established between indoor and outdoor locations, which favors the motion of particles. In contrast, higher indoor temperatures can force air out of a building (exfiltration), resulting in decrease of indoor PM concentrations (Chithra and Shiva Nagendra, 2014; Martins and da Graça, 2018). Similar effects of meteorological parameters were also observed in residential houses in urban areas such as Hanoi by Vo et al.(2020a), who reported that the concentrations of indoor PM_1, $PM_{2.5}$, PM_{10}, and TSP decrease by approximately 50% on rainy and windy days compared to days when there is no rain or wind in the winter. On rainy days in summer, the concentrations of indoor PM_1, $PM_{2.5}$, PM_{10}, and TSP decreased by approximately 40%.

The ventilation system also affects indoor PM concentrations as outdoor PM can penetrate indoor spaces through the ventilation system. I/O values, which have been used as indicators of indoor PM pollution, depend on building topology, ventilation strategy, and the amount of indoor PM. In naturally ventilated buildings with few indoor PM sources, average I/O ratios are close to unity, which means that there is equivalent between indoor and outdoor PM concentrations (Deng et al., 2015). Indoor PM concentration is are strongly influenced by outdoor sources when the I/O ratio is significantly lower than 1 (Deng et al., 2015; Han et al., 2016), which has been reported in some offices, households, and preschools in urban areas of Hanoi city. Indeed, in Hanoi, where the citizens prefer using natural ventilation systems (i.e., opening ventilation ducts, windows, doors), air infiltration and filtration occur easily, which leads to elevated levels of indoor PM. However, when indoor PM sources are present as major sources in indoor environments such as in shopping centers, basketball stadiums, and kitchens and residential houses, the I/O ratios can markedly exceed 1 (Quang et al., 2013; Vo et al., (2020a). The I/O values in naturally ventilated rooms have been shown to be higher than in tighter rooms (Chithra and Shiva Nagendra, 2014; Quang et al., 2013). In the presence of indoor PM sources, I/O values increased markedly in mechanically ventilated buildings compared to buildings without such sources (Deng et al., 2015; Quang et al., 2013). In these cases, the indoor PM concentrations could be reduced by up to 70% to 75% using air filters in ventilation systems (Cheng, 2017).

Indoor activities, such as cooking and cleaning, smoking, and burning incense/candles, and residents moving can increase indoor $PM_{2.5}$ concentrations (Chakraborty et al., 2014; Chen et al., 2018; Kang et al., 2019; Quang et al., 2017; Tran et al., 2021; Vu et al. 2017; Wallace et al., 1996). Cooking also contributes to indoor $PM_{2.5}$ and PM_{10} emissions in homes, restaurants since food preparation at high temperatures leads to the emission of water vapor and other solid and liquid particles (Kang et al., 2019). Wallace (1996) reported that $PM_{2.5}$ released from cigarette smoking was the main contributor to PM_{10} mass concentrations. Vu et al. (2017) reported that cleaning activities could emit 98.2% of NP, and that particles (90 to 150 nm) were released predominantly from cigarette smoking and burning incense. Tran et al.(2020a) concluded that coarse particles may be resuspended due to classroom activities, such as the movement of students and teachers inside the classroom, sweeping, etc. In residential houses, biomass/coal combustion and vehicle and industrial emissions (i.e., outdoor sources) were the main contributors to indoor PM in urban areas in Vietnam (Vo et al., 2022; Vo et al., 2020a; Vo et al., 2020b). At an annual scale, the increased consumption of coal and biomass for heating, electricity production, and cooking elevates PM pollutants in regions such as Chinese, India, and Vietnam (Chakraborty et al., 2014; Chen et al., 2018; Donre, 2017; Li et al.,2016). Li et al. (2016) reported that indoor $PM_{2.5}$ concentrations in the heating season were higher than the non-heating season in China and higher indoor $PM_{2.5}$ concentrations were observed in households using coal, compared to houses that used gas and electricity. In India and Vietnam, indoor $PM_{2.5}$ concentrations were higher in households that combusted wood, compared to houses that used agricultural residuals and gas (Lê et al., 2018; Li et al. 2016; Sidhu et al., 2017).

In many Asian countries, households in rural areas mostly use biomass and fossil fuels for cooking or heating simple stoves, which release a variety of pollutants into the indoor environment. However, beehive coal stoves are used for daily cooking in some households, restaurants, and street food vendors in urban area such as in Hanoi, Vietnam. Coal consumption for residential cooking in Hanoi was reported to be 528.2 t/day (Van, 2020), which is considered to be a significant contributor to indoor PM. In the short term, some Asian countries partially shared several characteristics related to indoor PM pollution, PM sources, and climate conditions. PM concentrations have tended to increase in the indoor urban environment in Asian countries. The levels of indoor PM also exceeded WHO recommendations in many Asian countries, which should be mitigated in order to protect human health.

3.2. BTEX compounds and their sources in indoor environments in Vietnam

3.2.1. Status of BTEX compounds in indoor environments

In Hanoi, large variations in the concentrations of BTEX compounds have been observed in different indoor environments, such as residential houses, nursery schools, and indoor parking areas (Tran et al., 2020b; Vo et al., 2017; Vo et al., 2020d). Vo et al. (2017) reported that the concentrations of BTEX compounds (benzene, toluene, xylene and ethylbenzene) in new/renovated houses were significantly higher than those in old houses and that indoor BTEX concentrations exceeded those in outdoor air. In new/renovated houses, the average levels of indoor benzene, toluene, and xylene were 9.9 $\mu g/m^3$, 197.2 $\mu g/m^3$, and 818.4 $\mu g/m^3$, respectively. Meanwhile, the corresponding values in old houses ranged from 5 to 17.3 $\mu g/m^3$ for benzene, 3.8 to 313 $\mu g/m^3$ for toluene, and 199 to 1559 $\mu g/m^3$ for xylene. Ethylbenzene was not detected in either new/renovated or old houses. Toluene and xylene appeared to be the most abundant of BTEX compounds, which was attributed to their widespread use for interior decorations, replacing benzene in solvents or for dilution in civil engineering applications in Vietnam. In another study on VOCs in the urban environment in Vietnam, Do et al. (2013)

reported that aromatic hydrocarbons are the most prominent group of total volatile organic compounds (TVOCs) in both indoor and outdoor environments in Hanoi. In that study, the average concentration of aromatic hydrocarbons was 43.7 µg/m^3 inside residential apartments and 268 µg/m^3 in streets outdoors. In the residential apartment, the concentrations of benzene and toluene were 7.8 µg/m^3 and 19.9 µg/m^3, respectively. In other indoor environments, Vo et al.(2020d) reported that high levels of BTEX were measured in underground parking lots in high-rise buildings, with BTEX concentrations ranging from 58.3 to 1232.7 µg/m^3 when the BTEX concentrations outdoors ranged from 61.9 to 981.3 µg/m^3. The average BTEX and benzene concentrations in parking lots were 842.6 µg/m^3 and 16.9 µg/m^3, respectively, with higher BTEX levels observed in underground parking lots with >1000 vehicles. Interestingly, the highest benzene concentrations were explored in underground parking areas in new/renovated high-rise buildings with high vehicle densities, even though the new/renovated buildings had better ventilation than older buildings. In such cases, the higher benzene concentrations were likely due to new buildings having more cars and newly painted walls. Vo et al. (2020d) also added that BTEX concentrations increased on weekends when the number of vehicles at these sites increased. The high BTEX levels in underground parking lots was attributed to BTEX accumulation resulting from low light conditions and, consequently, decreased rates of photochemical degradation, as well as these sites having less air circulation and numerous confined spaces. In another study, Tran et al.(2020b) reported that BTEX compounds were detected in Hanoi nursery schools in winter and summer at levels that were significantly lower than those observed in residential homes (Vo et al., 2017) and underground parking lots (Vo et al., 2020d). During class, the average levels of benzene, toluene, ethylbenzene, and xylene indoors were 3.5, 13.7, 3.12, and 5.96 µg/m^3, respectively, while the corresponding values obtained in the absence of children were 3.87, 20.4, 4.72, and 6.4 µg/m^3, respectively. These confirm that children’s activities during lessons did not contribute significantly to the concentrations of BTEX compounds. Pham et al. (2020) investigated the IAQ inside offices in three cities (Hanoi, Da Nang, Ho Chi Minh) during the hottest summer month and coldest winter month. Their results showed that VOCs were only detected in Hanoi offices, ranging from 3.3 to 8.7 ppm in building A and 0.1 to 0.3 ppm in buildings B and C. The highest concentration of VOCs was observed at building A (5.5 ppm), where there were numerous smokers, printers, and photocopy machines. Among the BTEX compounds, benzene is more widely used as an indicator in public health studies. WHO guidelines state that an upper threshold of benzene of 1.7 µg/m^3 is still acceptable, but the levels in the underground parking lots, residential houses, and schools in Vietnam, as shown above, largely exceeded these limits, implying that the poor IAQ at these locations poses a significance to human health. Compared to the national regulations for ambient air in Vietnam (QCVN 06:2009/BTNMT), all of the indoor benzene, toluene, xylene concentrations at the studied schools, residential houses, and underground parking lots were lower than the suggested limit for 1 h of exposure (22 µg/m^3 for benzene, 500 µg/m^3 for toluene, 1000 µg/m^3 for xylene). Regarding the indoor working environment, benzene and toluene were detected in the indoor air of a packaging production factory by Pham et al.(2018). The concentrations of these compounds varied markedly, with benzene concentrations ranging from 40 to 2010 µg/m^3 and those of toluene ranging from 100 to 25560 µg/m^3. However, these values complied with the limits promulgated as part of Decision No. 3733/2002/QDD-BYT on safety and hygiene standards in the section on chemical exposure limits in the working environment.

3.2.2. Seasonal variation of indoor BTEX concentrations

To date, there is gap in our knowledge of seasonal variations in BTEX concentrations in indoor environments in Vietnam. The only publication on this topic examined the seasonal variation of BTEX concentrations in indoor and outdoor air at nursery schools in Hanoi, by Tran et al.(2020b). The findings of that study showed that there was a clear seasonal trend in BTEX levels in outdoor air and no seasonal variation in BTEX levels in indoor air. Compared to values in summer, outdoor BTEX concentrations in winter increased approximately 2 times when children were present and 2.6 times in the absence of children. The higher concentrations of outdoor BTEX in winter were attributed to a reduction in photodegradation of BTEX compounds, i.e., less reactions between pollutants and OH radicals, which are more abundant in summer. It is considered that the negligible difference in indoor BTEX concentrations between seasons was due to the indoor activities of children being similar in both seasons, and that indoor BTEX concentrations increased during teaching periods because of the use of solvents and glues for handicraft work.

3.2.3. Sources

The indoor concentrations of BTEX compounds were highly dependent on the type of fuel used and extend of ventilation, type of chemical reagents used for cleaning, amount of furniture, cooking style and smoking habits, whereas outdoor BTEX concentrations were affected by vehicle emissions, fugitive emissions from petrol stations and industrial sources, and coal combustion (Delgado-Saborit et al., 2011; Dai et al., 2017; Hazrati et al., 2016; Vo et al., 2017). Vo et al. (2017) reported that increased levels of indoor BTEXs were attributed to cooking with gas in comparison to cooking using induction hobs. Among the houses investigated, new furniture and smoking activities contributed the most indoor BTEXs (benzene: 15 µg/m^3; toluene: 167 µg/m^3; xylene: 785 µg/m^3). The mean concentrations of benzene, toluene, and xylene in new/renovated houses were 1.2 to 1.4 times higher than those of old houses, which may be attributed to more new furniture and solvents used to decorate or paint the walls. This finding corroborates the studies of Demirel et al. (2014) and Liu et al. (2008) who found that the use of solvent-based products (furniture adhesives, thinner, paints, etc.) increases indoor BTEXs in the new/renovated houses.

I/O ratio can be used to identify the possible sources of VOCs. Vo et al. (2017) reported a wide variation in I/O ratios for BTEXs among residential houses. Lower I/O values were found in old houses, whereas I/O values greater than 1 were observed in new/renovated houses, implying the strength of indoor sources. In underground parking lots, the I/O values of BTEX compounds were significantly higher than the unity (1.4), suggesting the predominance of indoor sources. Indoor sources were attributed to vehicle emissions released when entering and leaving the parking lots and evaporation from the fuel tanks of vehicles when the vehicles were at rest, resulting in the accumulation of BTEX compounds in underground parking areas (Vo et al., 2020d). The same profile of potential BTEX sources was observed at schools. Indoor and outdoor ratios of BTEX compounds ranged from 0.4 to 14.2, implying the presence of indoor emission sources, such as paint solvents, glues, and cleaning agents. Outdoor BTEX compounds originated from common sources, mainly automobile traffic (Tran et al., 2020b).

3.2.4. Related studies on indoor BTEX compounds

Although very few studies on BTEX compounds have been conducted in residential houses, nursery schools, offices, and parking lots in Vietnam in recent years, it appears high concentrations of BTEXs as merging indoor pollutants in different indoor environments, implying poor air quality in urban environment in Vietnam. A brief summary of studies on BTEXs in different indoor environments is shown Table 3.2 and Fig. 3.3

In comparison with previous studies at newly/renovated houses, the concentrations of benzene and xylene in houses in Vietnam were significantly higher than those in Shanghai, China (Dai et al., 2017) and Yongcheng, South Korea (Shin and Jo, 2012), whereas higher levels of toluene and ethylbenzene were found in houses in Shanghai. The levels of benzene in residential houses in Vietnam was lower than those observed in houses in Guangzhou, China (Du et al., 2014) and in Melbourne, Australia (Brown, 2002), whereas notably higher concentrations of xylene were observed in houses in Vietnam. Compared to old houses, a large variation of BTEX concentrations was observed in Vietnam, in which values of benzene and ethylbenzene were lower than those in residential homes in Ardabil City, Iran and in Northern India, whereas the concentrations of toluene and xylene were higher (Hazrati et al., 2016; Masih et al., 2017). In addition, residential homes in England and Turkey had considerably lower levels of BTEXs in comparison with homes in Vietnam (Delgado-Saborit et al., 2011; Demirel et al., 2014). In school environments, BTEX concentrations in nursery schools in Vietnam were higher than those in Poland (Mainka and Kozielska, 2016) and Italy (Lucialli et al., 2020), but notably lower than those in Turkey (Sofuoglu et al., 2011). In underground parking lots, the mean concentrations of benzene (16.9 $\mu g/m^3$) and toluene (200.36 $\mu g/m^3$) in underground parking lots in high-rise buildings in Hanoi were lower than those of benzene and toluene in underground parking lots in Rio De Janeiro, Brazil (de Castro et al., 2015) and Greece (Soldatos, 2003), while the values of xylene were greater. The concentrations of these compounds in underground parking lots in Vietnam were considerably higher than those in parking lots in Thailand (Loonsamrong et al., 2015) and a multi-story car parking lot in Italy (De Gennaro et al., 2015). The similar temporal trend in BTEX variation was observed at parking lots in Hanoi and Thailand, where BTEX concentrations were higher on weekdays than on weekends (Loonsamrong et al., 2015). In Thailand and Greece, the mean BTEX concentrations in underground floors were higher than those on upper floors (Loonsamrong et al., 2015; Soldatos et al., 2003).

Hazrati et al. (2016) investigated numerous factors related to home’s categorizes governing the influencing BTEX levels in residential buildings. For example, an approximately two-fold decrease in the level of benzene was observed in homes that use central heating systems instead of natural gas heaters. Benzene levels in homes located on the ground floor were approximately

2.5 times higher than those from the 2nd floor up. Higher benzene and toluene concentrations were observed in homes_turn on air conditioner compared to turning off. Alsbou and Omari (2020) examined the relationships between different fuel types for heating and chimney condition on indoor BTEX concentrations. They found that kerosene heaters released the most BTEX (290 µg/m^3), followed by diesel pot-bellied heaters with chimneys (120 µg/m^3), unfluted gas heaters (84 µg/m^3), wood pot-bellied heaters with chimneys (31 µg/m^3), and electric heaters (16 µg/m^3). The majority of BTEX released from kerosene heaters and diesel pot-bellied heaters with chimneys was due to fuel evaporation, while BTEX compounds emitted from unfluted gas heaters were due to the combustion of LPG.

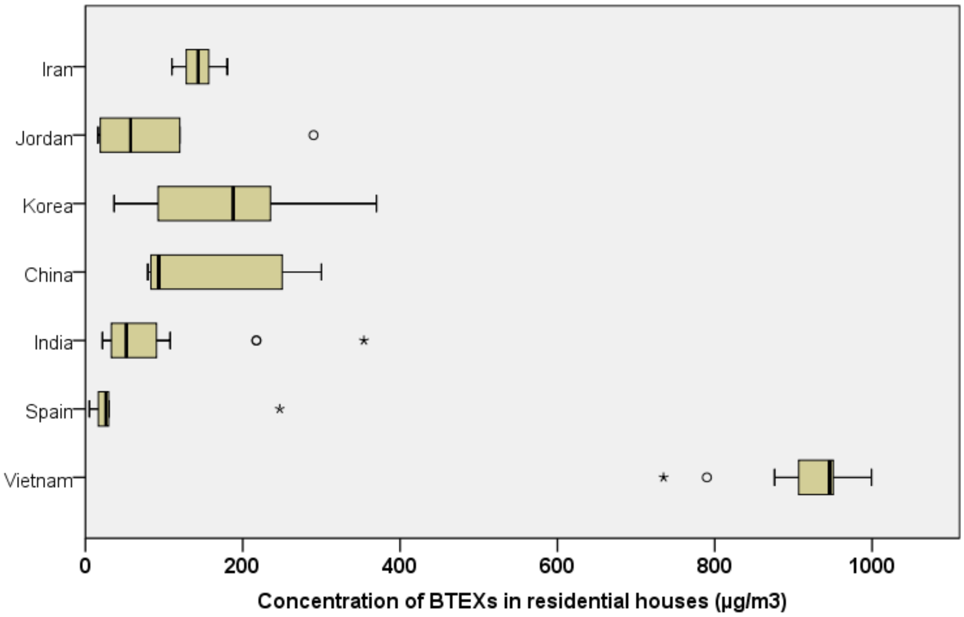


Fig 3. 3 BTEX concentrations in residential houses in different countries
Vietnam (Vo et al., 2017); Spain (Esplugues et al., 2010); India (Masih et al., 2017; Majumdar et al., 2012); China (Dai et al., 2017; Du et al., 2014; Liu et al., 2013); Korea (Lee et al., 2018; Shin and Jo, 2012); Jordan (Alsbou and Omari, 2020); and Iran (Hazrati et al., 2016).

Moreover, meteorological condition linked to seasonal trend was regarded as the one of governing factors influencing the indoor level of BTEX. Slightly higher indoor BTEX levels were observed in winter compared with summer, but no clear seasonal variation was observed in schools in Vietnam (Tran et al., 2020b) and in Italy (Lucialli et al., 2020). Conversely, a clear seasonal trend in BTEX concentrations was performed in the homes in India (Masih et al., 2017) and in nursery schools in Poland (Mainka and Kozielska, 2016), where BTEX levels in winter were markedly higher than in summer It was well-known that the higher temperatures in summer can promote the degradation of BTEX due to reactions between pollutants and OH radicals in the atmosphere (Masih et al., 2017). In addition, lower BTEX concentrations could be attributed to lower emissions from sources and enhanced ventilation in summer.

Spatial variations in indoor BTEX levels were examined in urban, suburban, and rural areas as a direct consequence of the BTEX source and socio-economic status of the inhabitants.

Masih et al. (2017) reported that the levels of benzene, toluene, xylene, and ethylbenzene in rural areas in India were 9.8, 2.2, 3.3, and 4.1 times higher than those in urban areas, which was attributed for improper ventilation and use of biomass for fuel. The highest BTEX levels were observed in rural houses, followed by houses in close proximity to industrial areas and roads. The variation of BTEX levels in different sites was attributed to local sources. For example, biomass fuel was the primary source in rural areas, traffic activities and manufacturing plants were the primary sources in houses near industrial sites, and automobile traffic was the primary source in roadsides houses; further, additional indoor sources at all sites also contributed to indoor BTEX levels. In residential areas, indoor activities such as cooking and smoking can contribute to BTEX levels. For example, Du et al. (2014) found that BTEX concentrations in urban houses were 1.5 to 2.2 times higher than they were in suburban houses, which was attributed to the higher socio-economic status of the inhabitants of these areas. In addition, Sofuoglu et al. (2011) reported that benzene and toluene levels in urban schools were 2.6 to 4.4 times higher than in suburban schools in fall, whereas there is an increase of 1.4- to 2.0-fold in toluene and benzene levels in suburban schools in winter and spring. The presence of toluene and benzene in urban and suburban schools suggested that water-based paints used to paint indoor walls in the schools were responsible.

3.3. Strategies to reduce exposure to indoor PM

In recent years, the ambient air quality in urban areas in Vietnam has deteriorated and this has had a strong influence on IAQ, had a variety of adverse impacts on public health and the economy. With efforts to improve the air quality, Vietnam has been taking steps to control PM pollution by implementing numerous interventions at both the macro to micro scales. Rational and effective methods play important roles in minimizing indoor pollution and for protecting human health. Based on the characteristics and sources of indoor PM, control strategies can be targeted at two sources: ambient PM and indoor PM.

3.3.1. Control strategies for ambient PM

Since traffic, industrial activities, and biomass/coal combustion are the main contributors to outdoor PM, numerous efforts to reduce urban PM pollution have been implemented in Vietnam so far.

i. Traffic emissions: Progressively stringent emission regulations have forced vehicle manufacturers to control exhaust emissions. Many measures related to emissions inspections for different vehicles have been implemented to check compliance with fuel emissions standards. Inspection routes on compliance to emission standards have been implemented; for example, compliance with European 2 standards (1999, 2006, 2008) has been applied for cars, Euro 4 standards for cars manufactured, assembled, or imported after January 1, 2017. Further, 92-research octane number (RON 92) (A92) gasoline was replaced by 95 RON (A95) gasoline. Trials of Selective Non-Catalytic Reduction (SNCR) systems and use of exhaust gas recirculation system (ERG) to reduce NO_x emissions from engines have been conducted (Hoang et al., 2017). Encouraging the use of bio-fuel through increasing community awareness has been integrated into a national program focusing on energy saving and efficiency. Several policies have been implemented, such as reduce VAT on output of the plant for bio-ethanol and biodiesel plants, the use of modified lecithin from soy oil has been researched for use as an additive for reducing diesel exhaust emissions. The initial stage of a program for urban busses to use clean natural gas (CNG) fuels instead of diesel has been implemented in in Ho Chi Minh city. Some city bus routes have also started using electrical engines instead of diesel engines. Accordingly, the metro systems in Hanoi and Ho Chi Minh will improve traffic and decrease emissions in the near future. These measures are being scaled up nationally as part of improved public transportation strategies to create walking and cycling networks in green cities in urban areas.

ii. Control of industrial emissions

Vietnam is currently focusing on controlling industrial emissions through control of emissions standards, especially in industrial sectors with high pollution levels (steel processing, chemical fertilizer production, the operation of coal-powered thermal plants and cement plants, etc.). The industrial facilities must register their exhaust gas emission sources with the Ministry of Natural Resources and Environment (MONRE), and install automatic–continuous emission monitoring systems to inspect emissions. The MONRE has a responsibility to approve the registration of emissions sources, receive monitoring data from Department of Resources and Environment (Donre, 2017), and develop inventories of industrial emissions. In addition, the government encourages enterprises to employ cleaner production technologies to save energy and material resources and minimize waste production.

iii. Control of biomass and domestic coal combustion

Vietnam has periodically tackled biomass/coal combustion. Firstly, it is crucial to discourage farmers (stakeholders) from burning biomass (e.g., crop byproducts, rice straw) in the fields or to dry the straw on the roads. Secondly, to support and encourage farmers to apply environmentally friendly farming methods, such as process rice straw into organic fertilizer, employ biological methods to decompose rice straw and crop byproducts in the field, process foods for livestock, produce mushrooms, and promote organic fertilizers for certain crops.

For domestic coal and biomass combustion, Directive No. 15 of the Hanoi People's Committee was launched in 2019 and 2020 to replace and completely eliminate the use of coal stoves. burning straw, and other agricultural wastes toward 2021. However, the regulations still need more time for enforcement. It’s still early to conclude the policy impacts. In conjunction with such measures, propagating and supporting inhabitants to replace their coal stoves to more environmentally friendly stoves is currently underway.

3.3.2. Control Strategies for Indoor PM

Controlling indoor PM sources is essential for reducing PM exposure among occupants.

i. Cooking: Methods for controlling the PM generated by cooking is imperative. For example, firewood, coal, and agricultural residuals for cooking must be replaced with cleaner fuels, such as natural gas or LPG, or with electric equipment; kitchens must be ventilated to promote air exchange during cooking; and exhaust hoods must be installed to control indoor

PM. Stoves need to be optimized for maximum efficiency. Cooking habits need to change and operation and maintenance of stoves needs to be improved. The findings of an investigation showed that reducing 56670 charcoal stoves to 15418 stoves in Hanoi city resulted in a decrease of 696 t of $PM_{2.5}$, 18711 t CO, and 382444 t CO_2 emissions, respectively (Donre, 2020).

ii. Smoking: the government and relevant stakeholders should actively promote a smoking ban and raise the public's health awareness. Smoking zones and non-smoking zones have been established in public areas (e.g., designated areas in supermarkets, airports, hospitals, offices).

iii. Indoor activities: It is a good habit to clean house regularly, decrease the burning of incense indoors and decorate reasonably to minimize indoor PM.

iv. Optimizing building design and use

The building envelope, as an interface between the indoor and outdoor environments, plays an important role in reducing indoor PM. Firstly, the building should be designed focusing on energy conservation, good ventilation and improving the building's air tightness. The latter prevents the penetration of outdoor PM into buildings and supports exfiltration, leading to a reduction in indoor PM concentrations. It is preferable to use natural ventilation instead of artificial ventilation if the ambient air quality is good. Secondly, the use of filters in mechanical ventilation systems should be employed as a common method to reduce indoor PM if the ambient air quality deteriorates. Especially in the case of hospitals, clinics, and other areas where PM reduction is important, highly efficient filters have been used to capture PM in the ventilation airflow.

4. CONCLUSION

This chapter reviews current studies on indoor air pollution in Vietnam as well as interventions, focusing on the mitigation of PM and BTEXs compounds. High concentrations of PM of different sizes (e.g., PM_{10}, $PM_{2.5}$, NPs) and BTEXs were found in different indoor environments in Vietnam, such as in schools, residential homes, offices, basement parking lots, and commercial areas. In many cases, the PM levels in these areas exceeded acceptable WHO standards. These findings showed that PM had a direct impact on human health due to poor IAQ. The findings also showed that the origins of indoor PM and BTEXs were complex and comprised indoor and outdoor sources. The primary outdoor sources included vehicle and industrial emissions and the main indoor sources were attributed to the combustion of domestic coal and biomass. In addition, indoor sources may be included human activities such as moving, sweeping and cleaning; cooking; smoking; and burning incense. Factors such as meteorology, ventilation mechanisms, and daily human activities have a marked effect on IAQ. From a practical standpoint, indoor air-purifier technologies and natural ventilation are most commonly adopted to control concentrations of indoor PM. Additionally, regular indoor cleaning, suitable interior decorations, and air purifiers also significantly influence IAQ and reduce indoor pollution. Environmentally friendly trends in the transportation and industrial sectors should be promoted to achieve sustainable development in urban areas, as these measures indirectly enhance IAQ. Given the lack of technical regulations or standards on IAQ, combined with gaps in human capacity in the area of environmental protection, the general awareness about IAQ should be improved and government should formulate detailed technical regulations and control standards as soon as possible.

Table 3. 2. Indoor PM levels in previous studies

Sampling location	Building type	Source	Average indoor concentrations	Reference
Poland (2010)	Kindergarten	Outdoor source Fuel combustion	Rural area $PM_{2.5}$: 25.1 µg/m^3 Urban area $PM_{2.5}$: 36.1 (20 to 41.9) µg/m^3	(Błaszczyk et al., 2017)
India (2011)	Residential Home	Household activities like cooking on stoves, indoor smoking, and outdoor vehicular traffic and garbage combustion	Roadside houses: I/0: PM_{10}: 0.97; $PM_{5.0}$: 0.93; $PM_{2.5}$: 1.01; $PM_{1.0}$: 1.03 Winter: PM_{10}= 304 µg/m^3; $PM_{5.0}$= 253 µg/m^3; $PM_{2.5}$= 207 µg/m^3; $PM_{1.0}$= 138 µg/m^3 Summer: PM_{10}= 218 µg/m^3; $PM_{5.0}$= 185 µg/m^3; $PM_{2.5}$= 119 µg/m^3; $PM_{1.0}$ = 87 µg/m^3 Urban houses: I/0: PM_{10}: 0.92; $PM_{5.0}$: 0.91; $PM_{2.5}$: 0.94; $PM_{1.0}$: 0.96 Winter: PM_{10}= 254 µg/m^3; $PM_{5.0}$= 200 µg/m^3; $PM_{2.5}$= 146 µg/m^3; $PM_{1.0}$= 135 µg/m^3 Summer: PM_{10}= 137 µg/m^3, $PM_{5.0}$= 108 µg/m^3, $PM_{2.5}$= 80 µg/m^3, $PM_{1.0}$= 67 µg/m^3	(Massey et al., 2012b)
China (2013)	Household	Domestic combustion and outdoor penetration	Heating season: Kitchen: Coal $PM_{2.5}$: 204 µg/m^3; Gas $PM_{2.5}$: 114 µg/m^3; Electricity $PM_{2.5}$: 107 µg/m^3 Bedroom: Coal $PM_{2.5}$: 159 µg/m^3; Gas $PM_{2.5}$: 109 µg/m^3; Electricity $PM_{2.5}$: 139 µg/m^3 Non-heating season: Kitchen: Coal $PM_{2.5}$: 213 µg/m^3; Gas $PM_{2.5}$: 65 µg/m^3; Electricity $PM_{2.5}$: 55 µg/m^3 Bedroom: Coal $PM_{2.5}$: 102 µg/m^3; Gas $PM_{2.5}$: 79 µg/m^3; Electricity $PM_{2.5}$: 68 µg/m^3	(Li et al., 2016)
India (2014)	Classroom	Outdoor sources Pupils' activities	Winter: PM_{10}: 30 to 403 µg/m^3; $PM_{2.5}$: 14 to 146 µg/m^3; PM_1: 10 to 117 µg/m^3 Summer: PM_{10}: 51 to 286 µg/m^3; $PM_{2.5}$: 15 to 53µg/m^3; PM_1: 6 to 20 µg/m^3 I/O: PM_{10}: 2.3; $PM_{2.5}$: 1.2; PM_1: 0.88	(Chithra and Shiva Nagendra, 2014)
China (2014)	Buildings	Cleaning, indoor activities, outdoor sources	Before air cleaning: Apartment $PM_{2.5}$: 154.64 µg/m^3; I/0 = 0.49 Detached villa $PM_{2.5}$: 97.63 µg/m^3; I/0 = 0.56 Shopping Centre $PM_{2.5}$: 56.88 µg/m^3; I/0 = 1.27 Hotel $PM_{2.5}$: 74.38 µg/m^3; I/0 = 0.72 Research Centre $PM_{2.5}$: 61.1 µg/m^3; I/0 = 0.34 Commercial office $PM_{2.5}$: 10.4 µg/m^3; I/0 = 0.12 Basketball stadium $PM_{2.5}$: 123.6 µg/m^3; I/0 = 1.25 After air cleaning: Apartment $PM_{2.5}$: 75.83µg/m^3; I/0= 0.76 Detached villa $PM_{2.5}$: 49.67 µg/m^3: I/0= 0.29 Shopping Centre $PM_{2.5}$: 80.67 µg/m^3; I/0: 1.74 Hotel $PM_{2.5}$: 131.33 µg/m^3; I/0: 1.16 Research Centre $PM_{2.5}$: 28.83 µg/m^3; I/0= 0.16 Commercial office $PM_{2.5}$: 29.17 µg/m^3; I/0= 0.47	(Deng et al., 2015)

			Basketball stadium $PM_{2.5}$: 133.83 μg/m^3; I/0=1.65	
China (2015)	Apartment building	Mainly from outdoor sources	$PM_{2.5}$: 55.2 ± 47.3 μg/m^3 I/0: 0.55	(Han et al., 2016)
China (2015-2016)	Classrooms	Students' activities, outdoor sources	Spring $PM_{2.5}$: 41.4 ± 18.2 μg/m^3; I/0 = 0.42 Summer $PM_{2.5}$: 35.0 ± 12.4 μg/m^3; I/0 = 2.35 Autumn $PM_{2.5}$: 64.3 ± 31.3 μg/m^3; I/0 = 1 Winter $PM_{2.5}$: 44.7 ± 22.5 μg/m^3; I/0 = 0.31	(Bi et al., 2018)
India (2017)	Household	Domestic cooking (combustion fuel)	In kitchen during cooking: SBF $PM_{2.5}$: 1865.7 μg/m^3; LPG $PM_{2.5}$: 511.86 μg/m^3 In living room: SBF $PM_{2.5}$: 412.6μg/m^3; LPG $PM_{2.5}$: 235.4 μg/m^3 In kitchen during non-cooking: SBF $PM_{2.5}$: 492.4 μg/m^3; LPG $PM_{2.5}$: 100.59 μg/m^3 In living room: SBF: $PM_{2.5}$: 286.1 μg/m^3, LPG $PM_{2.5}$: 83.6 μg/m^3	(Sidhu et al., 2017)
Italy (2018)	Classroom	Students' activities	PM_{10} = 168 to 388 μg/m^3, $PM_{2.5}$ = 51 to 117μg/m^3,	(Schibuola and Tambani, 2020)

Table 3. 3. Comparison of mean concentration of BTEXs in worldwide (μg/m^3)

Sampling site (dates)	Characteristics of building	Average indoor concentration	Reference
Vietnam (2017)	Residential houses	New/renovated houses B: 9.9; T: 197.2; X: 785.1 Old house B: 1-17.3; T: 3.8-313; X: 199-1559	Vo et al. (2017)
Ardabil city - Iran	Residential houses	Indoor B: 15.18; T 68.7; E: 12.07; X: 48.08 Natural gas heaters: BTEX: 19.8 Central heating systems: BTEX: 8.8 Gas stoves: B: 19.2; T: 99.9 Off gas stoves: B: 12.1; T: 46 Ground floor: B: 21.5 First floor: B: 11.8 >1st floor: B: 9.2	(Hazrati et al., 2016)
Melbourne, Australia	New/Established buildings	B: 30; T: 250; X: 30	(Carrion-Matta , 2002)
Shanghai, China (2015)	Newly renovated houses	B: 2.32; T: 200.13, X: 72.27; E: 26.33	(Dai et al., 2017)
Daegu, Ulsan Yong-cheon, Korea (2012)	Newly built apartment	B: 3.9; T: 184; X: 16.8; E: 8.2	(Shin and Jo, 2012)
Guangzhou, China (2012)	Residential house (renovated/newly/decorated houses)	B: 18.5; T: 173.2; X: 98.9	(Du et al., 2014)
Northern India (2014)	Old homes	Urban area: B: 12.68; T: 28.93; X :3.11; E: 4.11 Rural area: B: 124.67; T: 65.47; E: 17; X: 10.2 Agriculture area: BTEX: 54.3 Industrial area: BTEX: 18.2 Roadside: BTEX: 12.3 Residential: 6.1 Winter: BTEX: 32. 56 Monsoon: BTEX: 19.9 Summer: BTEX: 14.44	(Masih et al., 2017)
England (2005-2007)	Residential homes	B: 1.97; T: 17.53; E: 1.74; X: 5.84	(Delgado-Saborit et al., 2011)
Turkey (2008)	Residential homes Schools	Homes: B: 2.29, T: 26.55; E: 0.73; X:1.95 Schools: B: 0.92; T: 42.01; E: 0.39; X: 1.25	(Demirel et al., 2014)
Italia (2014-2015)	Schools	Winter: B: 1.31; T: 3.27; E: 1.65; X: 1.99 Summer: B: 0.9; T: 2.12: E: 2.87: X: 2.86	(Lucialli et al., 2020)
Vietnam (2020)	Schools	Teaching period B: 3.5; T: 13.7: E: 3.12; X: 5.96 Non-teach period B: 3.87, T: 20.4; E: 4.72 and X: 6.4	(Tran et al., 2020b)
Poland (2013-2014)	Schools	B: 1.37; T: 1.19; E: 2.11; X: 4.03	(Mainka and Kozielska, 2016)
Turkey	Schools	B: 10.4; T: 18.7	(Sofuoglu et al.,

(2009)			2011)
Hanoi, Vietnam 2017	Underground parking	B: 16.99; T: 200.36; X: 625.22	Vo., 2020d)
Athens Greece (2003)	Underground car parking	Floor 1: B: 406; T: 458: E: 77.7; X: 377 Floor 2: B: 418; T: 390; E: 129: X: 440 Floor 3: B: 360; T: 249; E: 100; X: 312	(Soldates, 2003)
Rio de Janeiro, Brazil (2010)	Confined parking	B: 54.14; T: 209.24; E: 45.87; X:118.93	(Castro et al., 2015)
Bangkok, Thai Lan (2014)	Parking structure	B: 11.28; T: 56.13; E: 7.17; X: 10.59	(Loonsamrong et al., 2015)
Salerno, southern Italy	Multi-story car parking	B: 7.22; T: 13.79; E: 3.71; X: 14.47 BTEX: 39.19	(De Gennaro et al., 2015)

CHAPTER 4

SIZE-FRACTIONATED PARTICULATE MATTER IN URBAN RESIDENTIAL HOUSES IN VIETNAM: RELATIONSHIPS AMONG INDOOR AND OUTDOOR PM, MASS SIZE DISTRIBUTION AND DEPOSITED DOSE ESTIMATES

1. INTRODUCTION

In recent years, great attention has been focused on the air quality and associated health risks of indoor environments, as this is where the majority of humans spend most of their time. Numerous studies have demonstrated that certain particulate matter (PM) fractions (PM_{10}, $PM_{2.5}$, $PM_{0.1}$) cause neurodegenerative, respiratory, cardiovascular problems, as well as being mutagenic and carcinogenic (Karottki et al., 2015; Li et al., 2017). Among these different types of PM, $PM_{0.1}$ (NP) is the most harmful to human health (Karottki et al., 2015; Li et al., 2017). NP has high surface area and reactivity, which have great ability to absorb toxic substances and capable of penetrating deeply into cells (Karottki et al., 2015). Indoor size-fractionated PM can originate from a variety of outdoor sources (e.g., transportation and manufacturing activities) and indoor sources (e.g., cooking, cleaning, incense burning, smoking and the operation of electric appliances) (Li et al., 2017; Morawska et al., 2017; Tran et al., 2021). Morawska et al. (2017) concluded that the majority of $PM_{2.5}$ and PM_{10} in residential houses was generated outdoors, and that NP in houses was mainly originated by indoor sources. In contrast, at schools, the origin of NP was outdoor air, whilst PM_{10} and $PM_{2.5}$ were from indoor sources. Several studies have explored the relationship between the levels of indoor and outdoor PM_{10}, $PM_{2.5}$, and PM_1 in urban areas to ascertain how outdoor air can influence indoor PM (Massey et al., 2012; Sangiorgi et al., 2013). In particular, houses in developing countries are naturally ventilated, allowing the particles from outdoor sources to enter the house via cracks and openings (Massey et al., 2012). In addition, seasonal variations in the different fractions and particle mass distributions in indoor environments should be studied further.

As a developing country, Vietnam has faced serious air pollution problems, especially in Hanoi city, which has experienced rapid urbanization in recent decades. Scholars should therefore focus on examining indoor air quality in Hanoi. Despite the extensive literature on the characterization of PM in the indoor environment around the world, to the best of our knowledge, there is a notable lack of studies in Vietnam, especially in Hanoi. Several previous studies have been conducted to identify and assess the health impacts of indoor PM pollutants in several indoor environments in Hanoi (Anh Le and Thuy Linh, 2019; Tran et al., 2021; Tran et al., 2020; Vo et al., 2020a). However, there has been a shortage of comprehensive studies focusing on the size distribution of such particles, the relationship between indoor and outdoor PM, source implication and dose estimations of PM in the residential environment on a seasonal basis. Therefore, thoroughly clarifying PM exposure to different particle fractions, particle distributions, and potential sources is vital for assessing indoor air quality in urban areas. To fill this knowledge gap, this study examined the seasonal variation in the mass concentration of size-fractionated PM in residential houses in Hanoi to analyze the relationship between the indoor and outdoor particles, determine the mass size distribution, and estimate the doses deposited in the human respiratory tract (HRT). These findings will facilitate initiatives that can be implemented to improve indoor air quality, especially in urban areas.

2. METHODOLOGY

2.1. Description of sampling

Measurements were performed in four residential houses in the center of Hanoi (capital of Vietnam) in winter (November 2018 to January 2019) and summer (from April to June 2019), to cover the periods with the lowest and highest PM concentrations in Hanoi throughout the year (Nguyen et al., 2018). The city, which is located in the northern part of Vietnam, has more than 7.5 million inhabitants (GSO, 2018). Hanoi has a tropical monsoon climate with two rainy seasons that are strongly influenced by the southeastern monsoon in summer and northeastern monsoon in winter (Nguyen et al., 2018; Vo et al., 2020a). The four houses in this study (K1 to K4) were typical of residential buildings with natural ventilation and are shown in Fig. 4.1; K1 (urban periphery two-story house) was located close to roads and industrial zones; K2 and K4 (urban multi-story houses) were located in densely populated areas; and K3 (road-site multi-story house) was located adjacent to a busy road.

2.2. Sampling campaign

Samples of different sizes ($PM_{0.1}$, $PM_{0.1\text{-}0.5}$, $PM_{0.5\text{-}1}$, $PM_{1\text{-}2.5}$, $PM_{2.5\text{-}10}$ and $PM_{>10}$) were collected indoors and outdoors simultaneously for 24 hours on the 2nd floor of each house using a Nano sampler II (Model 3182, Kinomax, Shimizu Suita City, Japan ; flowrate 40 L/min). The indoor sampler was placed in bedrooms (K1, K2, K3) and the master room (K4) at a height of 1.5 m above the floor (breathing zone), and the outdoor sampler was placed on a balcony. Simultaneously, questionnaires concerning daily indoor activities in each home were used to characterize the activities of the occupants. Mass concentrations were determined using a microbalance (AX26, Mettler-Toledo GmbH Company, Germany) to an accuracy of 10^{-6} g. The filters were weighed in a balance room (temperature: 20–25°C; relative humidity 30-40%). Each sampled filter was passed through an ionizing air blower (YIBO1-ODR, Sartorius) to limit the effects of static electricity. Meteorological data were collected at the Lang meteorological station in Hanoi.

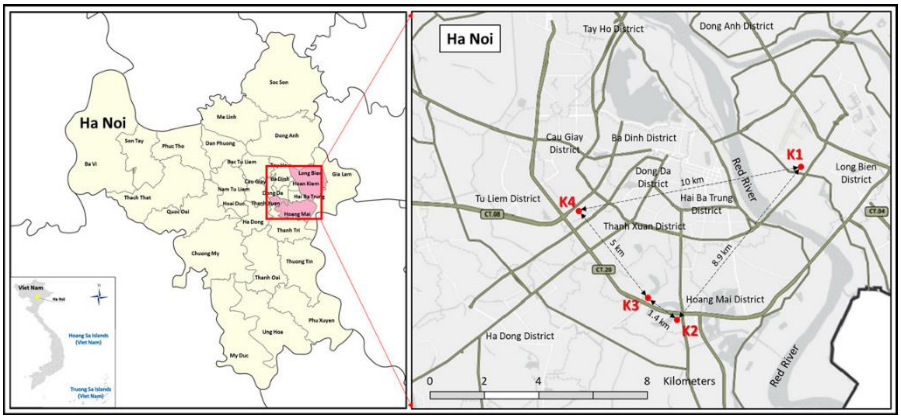


Fig 4. 1. Sampling sites in Hanoi

2.3 Indoor-outdoor relationship

Relationship between indoor and outdoor size-fractionated PM at residential houses was characterized by the indoor/outdoor (I/O) ratio for PM and regression lines (Sangiorgi et al., 2013; Vo et al., 2020a). The I/O ratio was used extensively to estimate whether indoor PM was enriched to PM or their compositions from outdoor sources. Linear equations were used to determine the correlation between the indoor and outdoor PM mass concentrations in naturally ventilated buildings using equation (1) (Sangiorgi et al., 2013; Vo et al., 2020a):

$$C_{in} = F_{inf} \times C_{out} + C_{ig} \quad (1)$$

where F_{inf} is the infiltration factor representing the ambient PM at equilibrium that penetrates indoors and remains suspended. C_{in} and C_{out} are the concentrations of indoor and outdoor PM at different fractions, and C_{ig} indicates concentration of particles generated indoors.

2.4. Human respiratory tract

The deposition fraction (DF) was estimated for different particle sizes in each region of the HRT (Head airways: HA, tracheobronchial region: TB and pulmonary or alveolar region: AL) illustrated in Fig. 4.2., as well as in the lobar-specific deposition (LU: left upper; RU: right upper; LL: left lower; RL: right lower; and RM: right middle) using a multiple-path particle dosimetry (MPPD) model (version 3.04). The MPPD model of the human lung is a mathematical lung deposition model, which has been used widely to calculate the DFs and internal doses for PM in three human lung regions and five lung lobe regions (Manojkumar et al., 2019; Sánchez-Soberón et al., 2015; Sarigiannis et al., 2015. Input parameters included PM characteristics (size distribution, shape and density); activity pattern (sitting, sleeping etc.); and respiratory physiological parameters (tidal volume (TV), breathing frequency (BF), functional residual capacity (FRC) and exposed subject characteristics (age and ventilation rate). An age-specific five-lobe model was adopted. Particles were considered to be spherical with a shape factor of 1 and a particle density of 1 g/cm^3. Respiratory physiological parameters were defined as recommended by the International Commission on Radiological Protection (ICRP, 1994) and the defaults of the model were used. Inhalation via the nose was assumed in this study in an upright position with constant exposure. Only deposition was within the scope of this study. A detailed schematic representation of the model used in this study, which was adopted by Manojkumar et al. (2019).

The deposited doses of inhaled $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$ and PM_{10} in the HRT or five lung lobes are the sums of the deposited doses in different regions, respectively and were estimated using the following US EPA model (USEPA, 1989, US-EPA, 2009):

$$\mathrm{EDI} = \sum_{i=1}^{n} \frac{DF_j \times C_j \times IR \times ET \times ED \times EF}{24 \times AT} \quad (1)$$

where EDI is the total daily deposited dose of PM (µg/day); DF_i is the particle deposition fraction in the *i* region of the HRT (HA, TB, AL), or the lobar-specific region (LU, RU, LL, RL, RM); i=1-n; C_j is the concentration of particle *j* (µg/m^3); IR is the inhalation rate (m^3/day); ET is the exposure time (hours/day); EF is the exposure frequency (days/year); ED is the exposure duration (years); AT is the average lifetime (days). DF_j is deposition fraction of particle *j*.

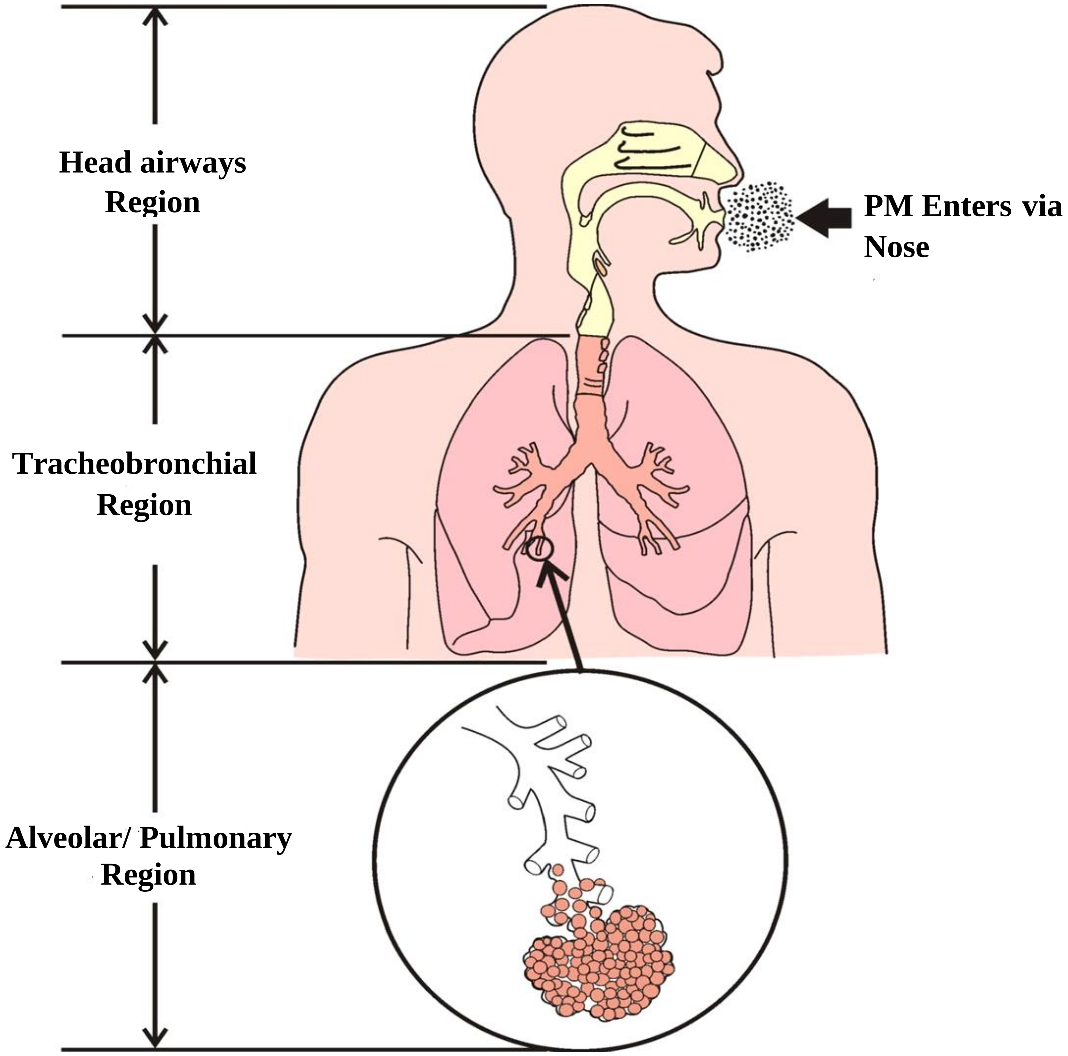


Fig 4. 2. Regions of the human respiratory tract; Source: Cheng, Y.S., 2014

2.5. Data analysis

Statistical analysis was performed using the SPSS IBM20 software package. The descriptive statistics (mean, standard derivation) were initially investigated for concentrations of size-fractionated PM. Pearson's coefficient was used to analyze the correlation between PM concentrations and metrological factors ($p<0.05$; $P<0.01$).

3. RESULTS AND DISCUSSIONS

3.1. Mass concentrations of indoor and outdoor PM

Table 4.1 shows the concentrations of $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$ and PM_{10} in the four houses examined in this study. The average concentrations obtained for the different PM were 7.6±1.3 µg/m^3 for $PM_{0.1}$; 15.5±3.3 µg/m^3 for $PM_{0.5;}$ 30.2±8.2 µg/m^3 for PM_1; 54.8 ±5.3 µg/m^3 for $PM_{2.5}$ and 71.8±27.5 µg/m^3 for PM_{10}. There was negligible difference between indoor and outdoor concentrations of NP ($PM_{0.1}$) and submicron particles ($PM_{0.5}$ and PM_1). However, for both $PM_{2.5}$ and PM_{10}, the outdoor levels were significantly higher than the indoor levels, suggesting that outdoor sources had a strong effect on indoor levels. Indoor PM levels exceeded WHO recommended standards ($PM_{2.5}$ of 15.0 µg/m^3; PM_{10} of 45 µg/m^3) by approximately 3.7-fold for $PM_{2.5}$ and 1.5-fold for indoor PM_{10}, and outdoor levels were 2.4- to 4.4-fold above recommended values in both seasons, implying that poor air quality in urban areas. Although, the indoor $PM_{0.1}$, $PM_{0.5}$ and PM_1 concentrations were significantly lower than those of the other fractions, these particles can permeate deep into the lungs and accumulate in different organs, causing serious diseases that require medical attention. In naturally ventilated buildings, the indoor $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$ and PM_{10} concentrations were significantly higher than those in a preschool environment in Hanoi, where there were no indoor domestic sources (Tran et al., 2020). However, the indoor levels of PM_1, $PM_{2.5}$, and PM_{10} observed in this study were lower than those in kitchens, where charcoal and wood were used for cooking (Lê et al., 2018). The combustion of biomass, in the form of charcoal and coal briquettes, is considered to be the dominant source of indoor air pollution in Hanoi. This source contributes significantly to the total burden of disease, which should not be ignored (Vo et al., 2020c). In addition, the $PM_{2.5}$ values obtained in this study were similar to those measured in residential houses by Tran et al. (2021), who reported that daily domestic activities included cooking, sweeping and burning incense. In addition, the results of indoor $PM_{2.5}$ measurements were significantly higher than those measured in a residential apartment using purifier systems in Canada (Kearney et al., 2014).

3.2. Seasonal variation of indoor and outdoor size-fractionated PM

In general, statistically significant differences were observed for concentrations of PM_1, $PM_{2.5}$ and PM_{10} between summer and winter *($p<0.05$),* but not between indoor and outdoor $PM_{0.1}$ and $PM_{0.5}$ *($p>0.05$).* The mean concentrations of indoor PM_1, $PM_{2.5}$, and PM_{10} were 26.5±4.2 µg/m^{3,} 43.3±6.8 µg/m^3 and 52.1±10.4 µg/m^3 in summer, whereas the outdoor concentrations were 33.9±10.1 µg/m^3, 47.1±8.7 µg/m^3 and 79.5±14.9 µg/m^3, respectively. The average levels for these PM categories increased 1.5-fold (indoors), and approximately 2-fold (outdoors) in winter, respectively. The seasonal variation of indoor size-fractionated PM in our study was closely linked with outdoor PM variation, which agreed with studies conducted previously in Hanoi (Nguyen et al., 2018; Vo et al., 2022). Similar seasonal trends in indoor PM_1, $PM_{2.5}$, PM_{10} levels have also been reported in India and Pakistan (Massey et al., 2012; Sidra et al., 2015). However, no seasonal variations in indoor $PM_{0.1}$ or $PM_{0.5}$ were observed in this study.

The levels of indoor $PM_{0.1}$ and $PM_{0.5}$ were 7.5±1.2 µg/m^3 and 13.9±2.6 µg/m^3 in summer, and 7.7±1.7 µg/m^3 and 16.9±3.6 µg/m^3 in winter, respectively.

Table 4. 1. Mass concentrations of size-fractionated indoor and outdoor PM at four houses in Hanoi

Concentration of PM (µg/m^3) (P= 1at; T= 25°C)										
	Indoor					Outdoor				
Winter	$PM_{0.1}$	$PM_{0.5}$	PM_1	$PM_{2.5}$	PM_{10}	$PM_{0.1}$	$PM_{0.5}$	PM_1	$PM_{2.5}$	PM_{10}
K1	8.4	16.7	31.4	53.7	70.1	6.3	13.8	31.7	71	121.5
K2	5.3	11.9	23.3	41.9	54.3	5.8	12.5	26.4	50.4	80.4
K3	8.9	18.9	33.6	63.5	78.1	10.5	24.4	44.3	94.7	153.4
K4	8.1	20.1	47.6	105.9	135.	8.7	21.7	50.7	117.9	173.9
Summer										
K1	8.4	15.6	28.2	46.5	64.3	6.7	14.4	25.1	49.4	85.4
K2	6.2	10.8	20.5	33.7	44.7	5.1	10.3	17.8	37.6	66.1
K3	8.7	16.6	30.5	49.5	68.5	9.4	18.1	33.4	57.9	97.3
K4	7.0	13	26.8	43.4	59.3	5.3	10.4	24.1	43.3	69.2

Meteorological parameters, including rainfall (Pr), temperature (T), relative humidity (RH), and wind speed (Ws), were examined to determine their influence on the seasonal variation in PM concentrations, which are shown in Fig.4.3-4. 6. Table 4.2 shows that negative correlations were observed between Ws and mass concentration for the different particle-size categories. These findings show that, high wind speeds can blow out the particles, leading to a decrease in the concentrations of $PM_{0.5}$, PM_1, $PM_{2.5}$ and PM_{10} in both indoor and outdoor environments, but not the $PM_{0.1}$ concentration. These correlations support our hypothesis about the impact of PM infiltration from outdoors to indoors. Weak correlations were also observed between T and outdoor $PM_{0.5}$, PM_1, $PM_{2.5}$ and PM_{10} concentrations, and RH and outdoor $PM_{2.5}$ and PM_{10} concentrations. High RH can facilitate the agglomeration of smaller particles to larger particles (Chithra and Shiva Nagendra, 2014), and a positive correlation was defined between RH and larger particles ($PM_{2.5}$ and PM_{10}); however, no such correlation was observed between RH and smaller PM. In addition, low temperatures in winter may promote the formation of PM, and extend its residence time. Conversely, high temperatures in summer favor height mixing, enhancing dispersion and, consequently, decreasing the PM concentration. Briefly, Ws, T, and RH had weak to moderate influences on specific PM fractions. However, Ws, T, and HR had a negligible impact on $PM_{0.1}$ concentration, which was also reported previously (Nguyen et al., 2018; Vo et al., 2020a). The results of this study showed that the lower variation observed for smaller particles compared to larger particles could be partly explained by meteorological factors. No correlation was observed between Pr and PM concentrations in this study, which was attributed to limited Pr during sampling periods.

Table 4. 2. Correlation between PM concentration and meteorological parameters at residential houses

	RH	T	Pr	Ws
PM$_{0.1}$. I	-0.1	0.1	-0.1	- 0.2
PM$_{0.1}$. O	0.1	-0.2*	-0.1	- 0.3**
PM$_{0.5}$. I	0.1	-0.2	-0.2	- **0.4****
PM$_{0.5}$. O	0.2	**-0.3****	-0.2	- **0.4****
PM$_{1}$. I	0.1	-0.2*	-0.1	- **0.4****
PM$_{1}$.O	0.3*	**-0.4****	-0.1	- **0.5****
PM$_{2.5}$. I	0.2*	-0.3**	-0.1	- **0.4****
PM$_{2.5}$. O	**0.3****	**-0.4****	-0.2	- **0.5****
PM$_{10}$. I	0.2	-0.2*	-0.2	- **0.4****
PM$_{10}$. O	**0.4****	**-0.4****	-0.2	- **0.5****

**. *Bold values: Correlation is significant at the 0.01 level (2-tailed).*

*. *Values: Correlation is significant at the 0.05 level (2-tailed).*

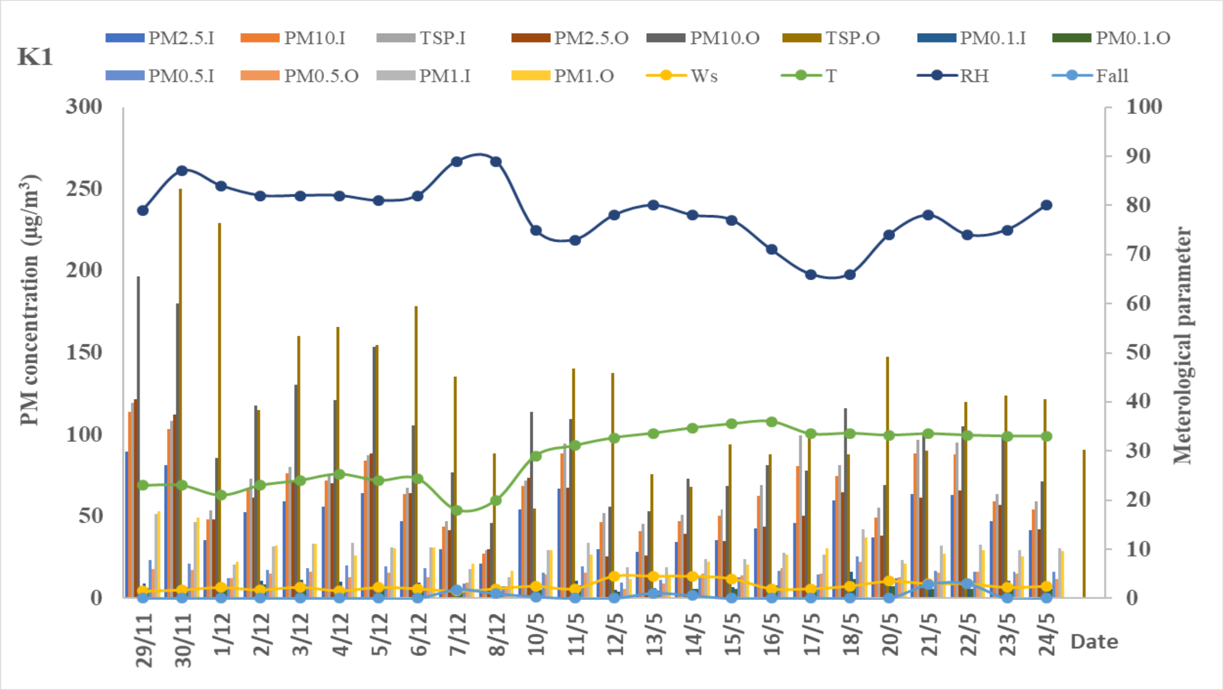


Fig 4. 3. Correlation between PM concentrations and meteorological factors at K1

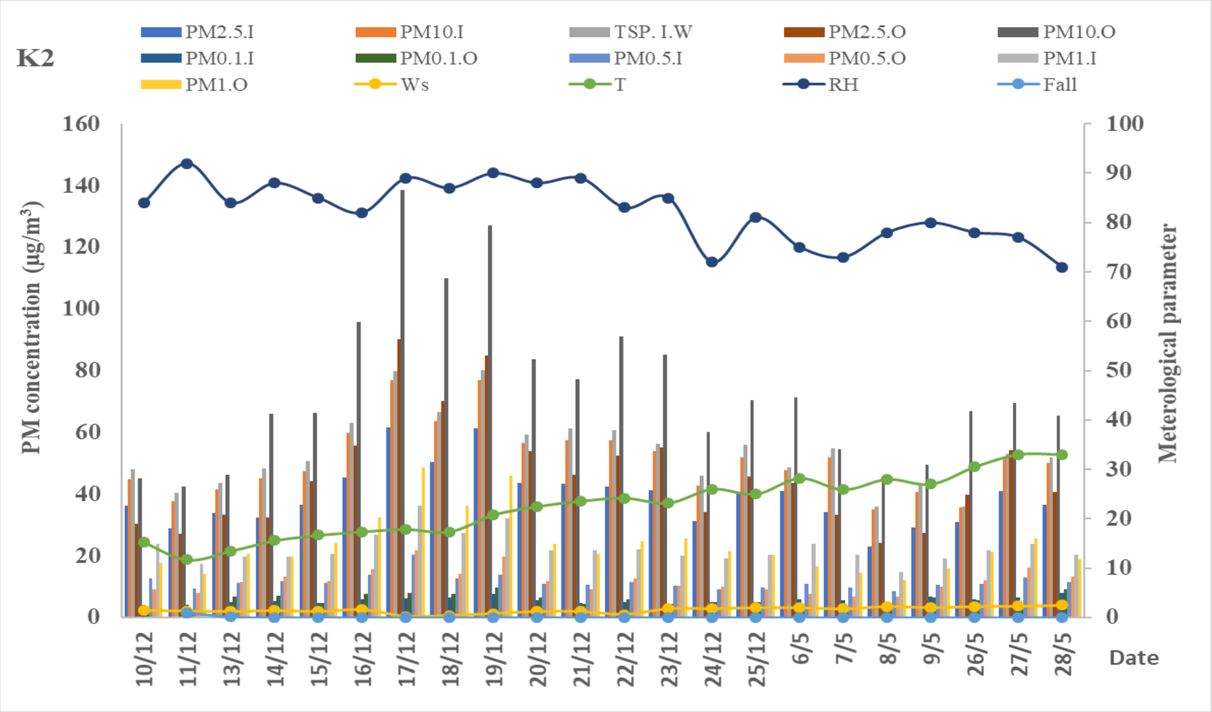


Fig 4. 4. Correlation between PM concentrations and meteorological factors at K2

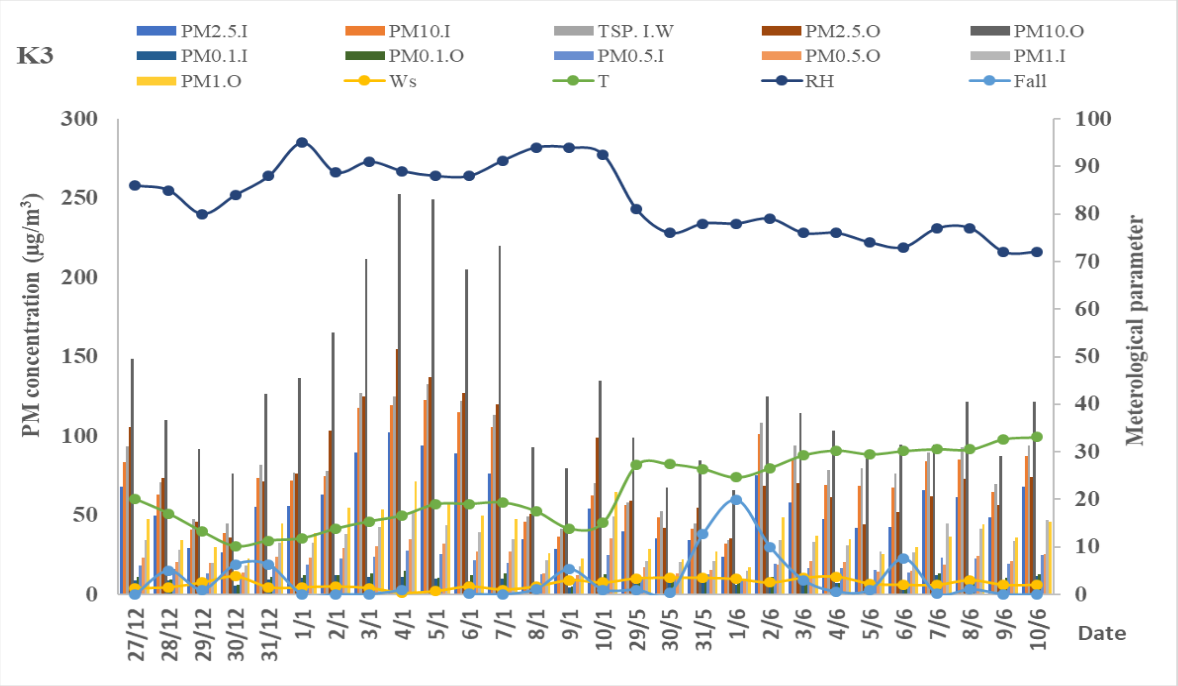


Fig 4. 5. Correlation between PM concentrations and meteorological factors at K3

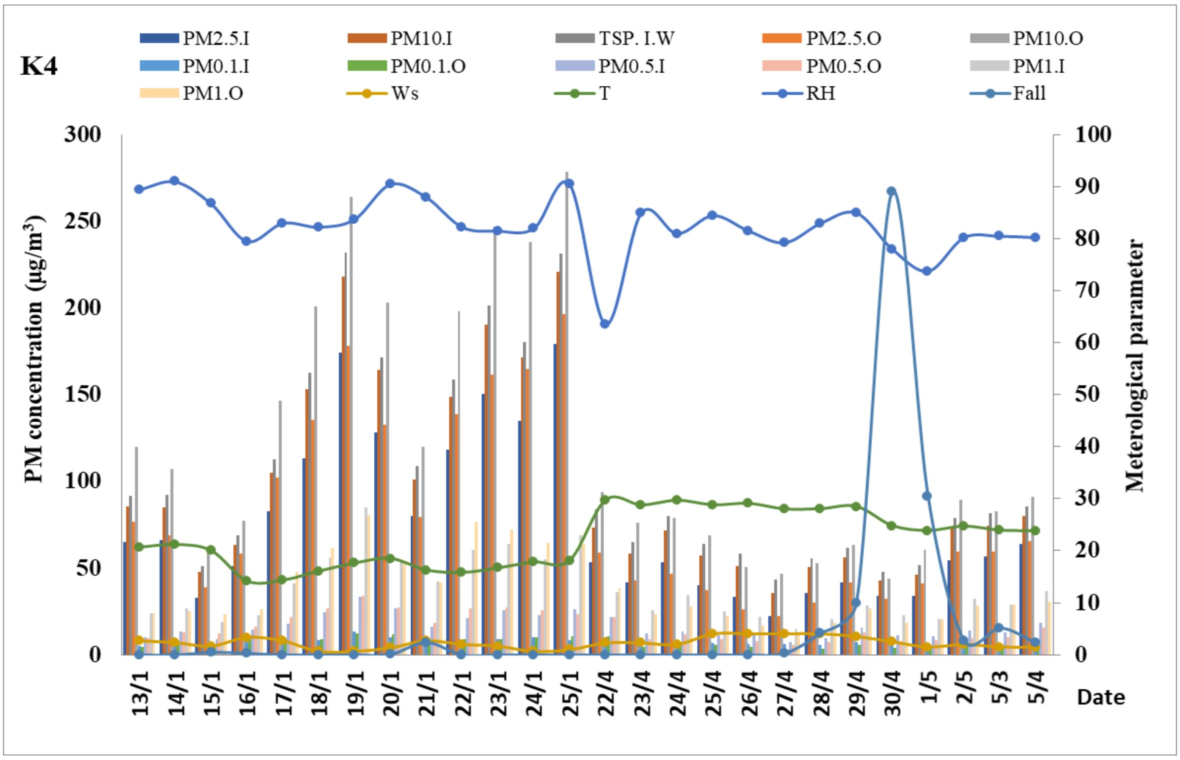


Fig 4. 6. Correlation between PM concentrations and meteorological factors at K4

3.3. Relationship indoor and outdoor PM concentrations, correlations and source implications

3.3.1. Indoor and outdoor ratios (I/O)

I/O ratios have been used widely as an indicator for evaluating the difference between the concentrations of indoor and outdoor size-fractionated particles. Fig.4.7 shows the average I/O ratios for different particle fractions in two seasons. The average I/O ratios of $PM_{0.1}$, $PM_{0.1-0.5}$, $PM_{0.5-1}$, $PM_{1-2.5}$, $PM_{2.5-10}$, $PM_{>10}$ in winter were 1±0.3, 0.9±0.2, 0.9±0.12 0.7±0.2, 0.4±0.1 and 0.2±0.1, respectively, whereas those in summer were 1.2±0.3, 1±0.3, 1.1±0.2, 0.8±0.2, 0.5±0.2, and 0.3±0.3. I/O ratios in summer were slightly higher than those in winter due to the concentrations of outdoor PM being relatively higher in winter. The I/O ratios of NP particles, submicron particles (0.1-1 µm), and fine particles ($PM_{1-2.5}$) were significantly higher than those of larger particles, which was reported in previous studies in which the infiltration of these airborne particles was more prevalent from outdoor air into houses (Massey et al., 2009; Sangiorgi et al., 2013). Thatcher and Layton.(1995) reported that the deposition velocities of coarse particles were greater than those of fine particles and NP, which resulted in reduced indoor concentrations. A significant decrease in the I/O ratios of coarse indoor fractions ($PM_{2.5-10}$; $PM_{>10}$) was found in both seasons.

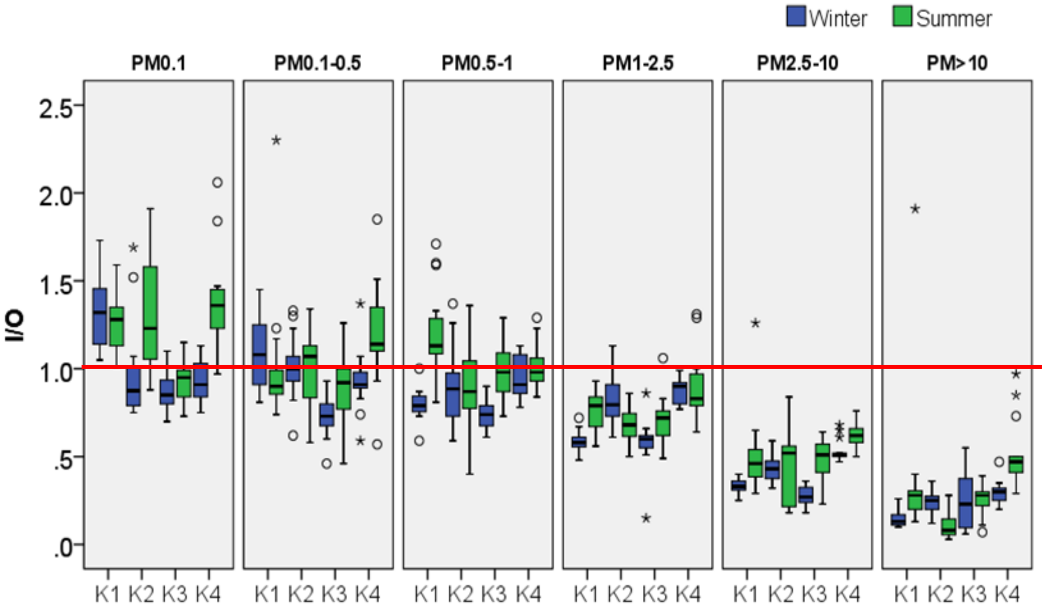


Fig 4. 7. I/O ratios of size-fractionated particulate matter in residential houses in Hanoi.

Additionally, the I/O values of $PM_{0.1}$ and submicron particles ($PM_{0.1\text{-}1}$) were greater than 1, suggesting the influence of indoor sources, in addition to outdoor sources. Conversely, the I/O values of coarse particles were considerably less than unity (<1), implying that outdoor sources were more pervasive. The decrease in indoor coarse particles was attributed to gravitational settling or deposition on windows, doors and furnished surfaces (Thatcher and Layton, 1995). Vu et al. (2017) found that cleaning activities could result in the emission of 98.2% of NP ($PM_{0.1}$), and that particles measuring 90-150 nm were attributed predominantly to smoking cigarettes and burning incense. Wallace**.**(1996) reported that $PM_{2.5}$ released from cigarette smoking was the main contributor to the mass concentration of PM_{10}. Kang et al. (2019) reported that concentrations of $PM_{2.5}$ and PM_{10} peaked during broiling fish (cooking activities), with levels of 1256 µg/m^3 for $PM_{2.5}$ and 1269 µg/m^3 for PM_{10} associated with this activity. Wang et al. (2006) found that the combustion of incense released $PM_{2.5}$ with concentrations in the range of 501 to 6024 mg/m^3. Resuspension of coarse fractions also occurred in response to the occupants' movements (Tran et al., 2020). Traffic emissions contributed 40% and 50% of $PM_{0.1}$ and $PM_{2.5}$ in Hanoi, respectively, which directly enters the indoor environment via infiltration (Cohen et al., 2010; Nghiem et al., 2020).

3.3.2. Infiltration factors and indoor-generated indoor PM

A linear regression analysis was used to investigate the relationship between different particle sizes indoors and outdoors at the four houses. The determination coefficient (R^2) was determined by linear regression with $P<0.05$. Except for $PM_{>10}$ and $PM_{2.5\text{-}10}$, the overall results revealed strong correlations between indoor and outdoor PM, with R^2 values ranging from 0.7 to 0.96. These findings imply that outdoor PM can explain approximately 70% to 96% of the variation in the indoor PM observed in the investigated houses.

Seasonal infiltration of size-fractionated particles was also examined among the houses. In winter, the average infiltration factors for $PM_{0.1}$, $PM_{0.1-0.5}$, $PM_{0.5-1}$, $PM_{1-2.5}$, and $PM_{2.5-10}$ were 0.8, 0.9, 0.8, 0.6 and 0.3, respectively, and in summer, these values were 0.8, 0.8, 0.9, 0.7 and 0.5, respectively. The infiltration factors for $PM_{0.1}$ and submicron particles (0.1-1 µm) were higher than those for the larger fractions ($PM_{1-2.5}$; $PM_{2.5-10}$). It is likely that the NP and submicron particles can be more easily to enter indoor environments, irrespective of whether the windows are open or closed. In contrast, the infiltration of larger sized particles was limited due to their sizes and building envelope. Dickerhoff et al. (1982) reported that infiltration of PM through air leakages from a wall of air-tight buildings accounted for 18-50% and closed windows/doors were responsible for 6-22%. There was a negligible variation in the infiltration factor (F_{inf}) for $PM_{0.1}$ between the two seasons, whereas a slightly higher F_{inf} was obtained for $PM_{0.5-1}$, PM_{1-25}, and $PM_{2.5-10}$ in summer. However, no apparent seasonal variation in F_{inf} was observed for these particles. It was likely due to windows/doors being closed during sampling, thus limiting air exchange between indoors and outdoors, this may explain for the lack of a clear seasonal variation in F_{inf} in this study.

There was a wide variation in the relative contributions of indoor-generated PM among the four houses (%Cig), reflecting the complexity and dynamic nature of the indoor environment. It was estimated that 16.6% to 63% of indoor $PM_{0.1}$, 5 to 63% of $PM_{0.1-0.5}$, 5 to 48% of $PM_{0.5-1}$, 3 to 32% of $PM_{1-2.5}$ and 9 to 18% of $PM_{2.5-10}$ were generated by indoor sources. These estimates excluded $PM_{>10}$, which had a low R^2 value and negative values for C_{ig}, which is attributed to the decomposition of chemical species in PM. Nevertheless, it was the presence of indoor-generated PM source, the majority of indoor fractions originated from outdoor sources, which was consistent with previous publications (Chatoutsidou et al., 2015; Sangiorgi et al., 2013; Vo et al., 2020a).

3.3.3 Correlation analysis

Pearson's coefficients were determined to estimate the extent of inter-correlation among indoor PM at residential houses in winter and summer, as shown in Tables 4.3-4.6. In general, positive correlations (0.8 to 0.9) were found among indoor fractions and seasons in the four houses. However, there were minor differences in correlations among the particles in the two seasons. The indoor coarse particles correlated well with fine particles in winter, and a strong correlation between coarse particles in summer was observed at K1. For example, in winter, $PM_{0.1-0.5}$ was strongly correlated with $PM_{0.5-1}$ and $PM_{2.5-10}$, and $PM_{1-2.5}$ was strongly correlated with $PM_{2.5-10}$. In summer, a strong correlation between $PM_{2.5-10}$ and $PM_{>10}$ was performed. These results imply that indoor fine and coarse particles likely originated from the same sources in winter, and indoor coarse particles were possibly derived from the same sources in summer at K1. The correlation among indoor particles observed at K2 and K3 showed similar trends, where good correlations were observed between $PM_{0.1}$ and fine particles ($PM_{0.1}$ and $PM_{0.1-0.5}$); fine particles ($PM_{0.1-0.5}$ and $PM_{0.5-1}$; and $PM_{0.5-1}$ and $PM_{1-2.5}$); fine and coarse particles ($PM_{1-2.5}$ and $PM_{>10}$) and coarse particles ($PM_{2.5-10}$ and $PM_{>10}$). However, there is somehow difference in correlations at K4, where all fractions were strongly correlated in winter, whereas only $PM_{0.1}$ was strongly correlated with $PM_{0.1\text{-}0.5}$ in summer. These findings suggest that the majority of indoor particles were likely generated from the same sources at K2, K3, whereas $PM_{0.1}$ and $PM_{0.1\text{-}0.5}$ originated from the same sources in summer at K4. The weaker correlation between $PM_{0.1}$ and other fractions indicated that the source of $PM_{0.1}$ might be different from other fractions. The discrepancies in the correlations among fractions reflected the complex and dynamic features in the indoor environment and between indoor and outdoor sources.

Table 4. 3. Correlation matrix for indoor size-fractionated particles in summer and winter at K1

Correlations

Winter. Indoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.5	1.0				
PM0.5.1	0.4	0.9^{**}	1.0			
PM1.2.5	0.6	0.9^{**}	0.8^{**}	1.0		
PM2.5.10	0.6	0.9^{**}	0.9^{**}	0.9^{**}	1.0	
PM10	0.3	0.4	0.5	0.4	0.4	1.0

Correlations

Winter. Outdoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.5	1.0				
PM0.5.1	0.3	0.9^{**}	1.0			
PM1.2.5	0.4	0.9^{**}	0.8^{**}	1.0		
PM2.5.10	0.7^{*}	0.9^{**}	0.8^{**}	0.9^{**}	1.0	
PM>10	0.6	0.9**	0.8^{**}	0.7^{*}	0.8^{**}	1.0

Correlations

Summer.Indoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.5	1.0				
PM0.5.1	0.7^{**}	0.6^{*}	1.0			
PM1.2.5	0.4	0.6^{*}	0.6^{*}	1.0		
PM2.5.10	0.0	0.3	0.2	0.5	1.0	
PM10	0.1	0.0	0.2	0.2	0.9^{**}	1.0

Correlations

Summer. Outdoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.5	1.0				
PM0.5.1	0.4	0.5	1.0			
PM1.2.5	0.2	0.5	0.5^{*}	1.0		
PM2.5.10	0.7^{**}	0.7^{**}	0.4	0.7^{**}	1.0	
PM10	0.5	0.4	0.2	0.5^{*}	0.8^{**}	1.0

Table 4. 4. Correlation matrix of indoor size-fractionated particles in summer and winter at K2

Correlations

Summer. Indoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.9^{**}	1.0				
PM0.5.1	0.4	0.6^{*}	1.0			
PM1.2.5	0.4	0.5	0.8^{**}	1.0		
PM2.5.10	0.5	0.5	0.6^{*}	0.5	1.0	
PM>10	0.5	0.4	0.2	0.1	0.7^{*}	1.0

Correlations

Summer. Outdoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.9^{**}	1.0				
PM0.5.1	0.5	0.5^{*}	1.0			
PM1.2.5	0.4	0.5	0.8^{**}	1.0		
PM2.5.10	0.5	0.3	0.5	0.2	1.0	
PM>10	0.1	0.3	0.1	0.0	0.6^{*}	1.0

Correlations

Winter. Indoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.2	1.0				
PM0.5.1	0.8^{**}	0.5	1.0			
PM1.2.5	0.7^{**}	0.3	0.8^{**}	1.0		
PM2.5.10	0.7^{*}	0.3	0.7^{**}	0.8^{**}	1.0	
PM>10	0.1	0.2	0.2	0.1	0.3	1.0

Correlations

Winter. Outdoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0				$.739^{**}$	
PM0.1.0.5	0.5^{*}	1.0				
PM0.5.1	0.7^{**}	0.8^{**}	1.0			
PM1.2.5	0.6^{*}	0.6^{*}	0.9^{**}	1.0		
PM2.5.10	0.7^{**}	0.7^{**}	0.8^{**}	0.8^{**}	1.0	
PM>10	0.5	0.7^{**}	0.8^{**}	0.6^{**}	0.7^{**}	1.0

Table 4. 5. Correlation matrix for indoor size-fractionated particles in summer and winter at K3

Correlations

Winter. Indoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0			.568*		
PM0.1.0.5	0.6**	1.0				
PM0.5.1	0.8**	**0.9****	1.0			
PM1.2.5	0.6*	.7**	.6*	1.0		
PM2.5.10	0.4	0.5	0.4	**0.9****	1.0	
PM>10	0.2	0.3	0.3	0.4	0.5*	1.0

Correlations

Winter. Outdoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.7**	1.0				
PM0.5.1	0.9**	**0.8****	1.0			
PM1.2.5	0.7**	0.6*	0.7**	1.0		
PM2.5.10	0.6*	0.6*	0.5*	**0.9****	1.0	
PM>10	0.1	0.1	0.2	0.1	0.1	1.0

Correlations

Summer. Indoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.8**	1.0				
PM0.5.1	**0.9****	**0.9****	1.0			
PM1.2.5	0.4	0.5	0.5	1.0		
PM2.5.10	0.3	0.5	0.5	0.6*	1.0	
PM10	0.2	0.5	0.4	0.3	**0.9****	1.0

Correlations

Summer. Outdoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.4	1.0				
PM0.5.1	0.6*	0.8**	1.0			
PM1.2.5	0.4	0.6*	0.3	1.0		
PM2.5.10	0.2	0.7**	0.4	0.7**	1.0	
PM>10	0.1	0.6*	0.4	0.4	**0.9****	1.0

Table 4. 6. Correlation matrix for indoor size-fractionated particles in summer and winter at K4

Correlations

Winter. Indoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.8**	1.0				
PM0.5.1	0.8**	0.9**	1.0			
PM1.2.5	0.7**	0.9**	0.9**	1.0		
PM2.5.10	0.8**	0.9**	0.9**	0.9**	1.0	
PM>10	0.8**	0.9**	0.9**	0.8**	0.9**	1.0

Correlations

Winter. Outdoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.8**	1.0				
PM0.5.1	0.7*	0.9**	1.0			
PM1.2.5	0.7**	0.7*	0.8**	1.0		
PM2.5.10	0.7*	0.8**	0.9**	0.9**	1.0	
PM>10	0.6*	0.9**	0.9**	0.8**	0.9**	1.0

Correlations

Summer. Indoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.9**	1.0				
PM0.5.1	0.4	0.6*	1.0			
PM1.2.5	0.4	0.5	0.8**	1.0		
PM2.5.10	0.5	0.5	0.6*	0.5	1.0	
PM>10	0.5	0.4	0.2	0.1	0.7*	1.0

Correlations

Summer. Outdoor	PM0.1	PM0.1.0.5	PM0.5.1.	PM1.2.5	PM2.5.10	PM>10
PM0.1	1.0					
PM0.1.0.5	0.9**	1.0				
PM0.5.1	0.5	0.5*	1.0			
PM1.2.5	0.4	0.5	0.8**	1.0		
PM2.5.10	0.5	0.3	0.5	0.2	1.0	
PM>10	0.1	0.3	0.1	0.0	0.6*	1.0

***. Correlation is significant at the 0.01 level (2-tailed).*

**. Correlation is significant at the 0.05 level (2-tailed).*

3.3.4. Particle mass-size distribution

The unimodal distributions of particle sizes at four residential houses were determined in Fig. 4.8. There was a negligible difference in the particle size distribution between two seasons at the four houses. However, the particle size distribution indoors differed from that outdoors at all four houses.

The highest concentrations of outdoor particles were gained in the super-micron sizes (1 to 10 µm), and the lowest concentrations were found in small particles ($PM_{<0.5}$) and coarse particles ($PM_{>10}$). Meanwhile, the size distributions of indoor PM shifted to $PM_{1-2.5}$, which had the highest concentrations, and small sizes ($PM_{<0.5}$), which were present at the lowest concentrations. Our results were partially consistent with the study of Oh et al.(2020), in which the lowest particle count concentrations in indoor environments (child care center, school, commercial building, and residential houses) were observed at coarse particles and the highest concentrations were observed for small particles ($PM_{<4}$) had the highest concentrations (Oh et al., 2020). Hussein et al. (2021) also found a unimodal size distribution in Jordanian residential environments, with the highest mass concentrations being at the super-micron particles (1 to 10 µm), which was similar to our findings. The wide variation in the particle size distribution found in different indoor environments in observed studies was due to the differences in indoor sources, ventilation conditions, and outdoor conditions.

The contribution of other PM sizes to fine and coarse particles indoors and outdoors was also examined in this study. In general, the contribution proportions of $PM_{0.1-0.5}$, $PM_{0.5-1}$, $PM_{1-2.5}$ to $PM_{2.5}$, and PM_{10} were relatively similar between summer and winter, which is shown in Fig. 9 and 10. However, fine particles (FP) with size intervals ($PM_{0.5-1}$ and $PM_{1-2.5}$) contributed larger proportions to $PM_{2.5}$ and PM_{10} than NP ($PM_{0.1}$). Indoor NP was responsible for 14.9% of $PM_{2.5}$ and 11.3% of PM_{10}, whereas outdoor NP accounted for 11.8% of $PM_{2.5}$ and 7.2% of PM_{10}. The contribution of indoor NP to FP in this study was similar to that in the school environment in Hanoi (Tran et al., 2020), whereas the contributions of outdoor $PM_{0.1}$ to $PM_{2.5}$ and PM_{10} were consistent with an earlier study on ambient air in Hanoi (Nguyen et al., 2018). In addition, the ratios of $PM_{0.1}/PM_{2.5}$ and $PM_{0.1}/PM_{10}$ indoors were higher than those outdoors, whereas the indoor ratio of $PM_{2.5-10}/PM_{10}$ was lower than the outdoor ratio. The findings indicated that indoor NP and fine particles are typically influenced by indoor sources, whereas coarse particles are typically attributed to outdoor sources (Massey et al., 2009; Sangiorgi et al., 2013; Tran et al., 2020). Indoor NP was associated with indoor activities, such as cooking, cleaning, and incense burning, and coarse particles were attributed to occupants' movements, which was reported during sampling.

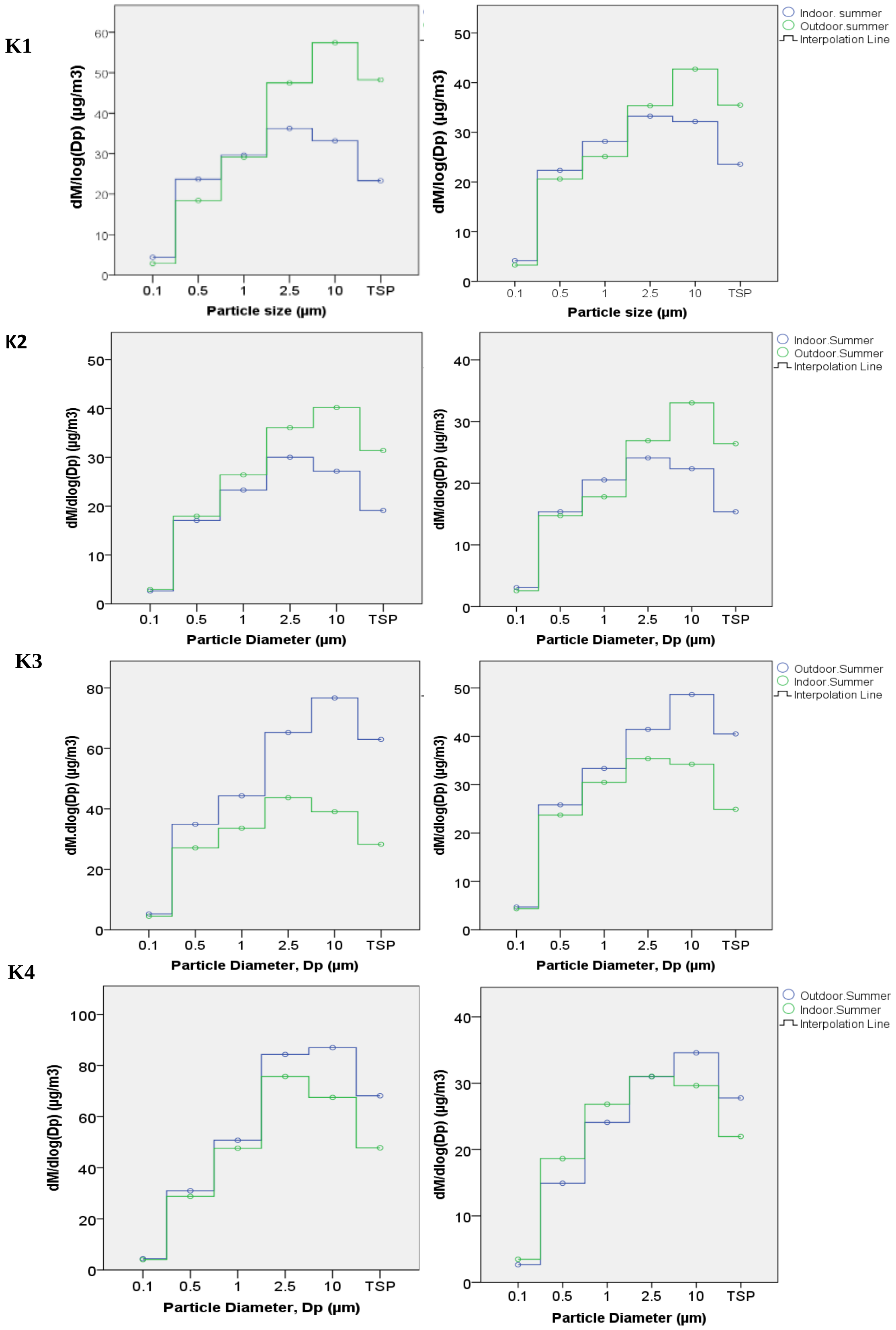


Fig 4. 8. Size distributions of airborne particles at four houses during summer and winter

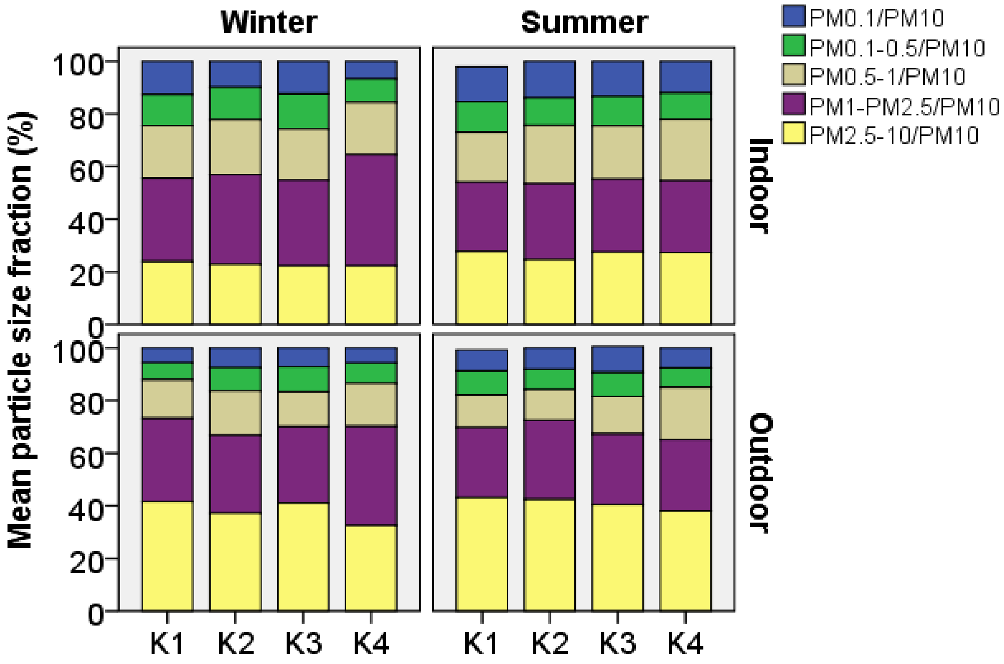


Fig 4. 9. Mass-size distribution of indoor and outdoor airborne particles at four houses (case)

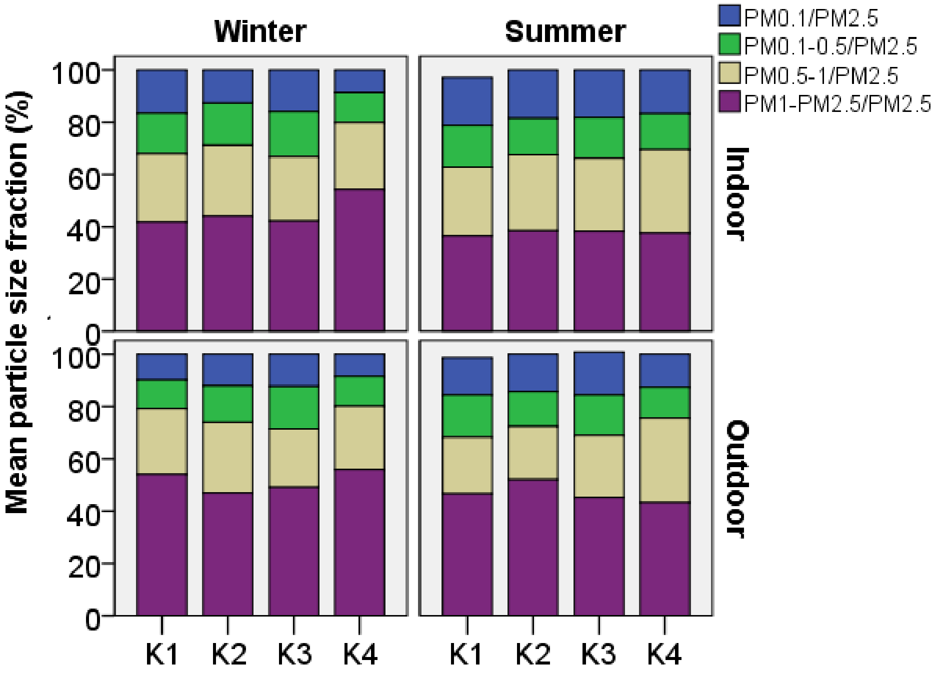


Fig 4. 10. Mass-size distribution of indoor and outdoor airborne particles at four houses (case 2).

3.4. Deposited doses of PM in the Human respiratory tract (HRT)

The average deposition fractions (DF) of $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$ and PM_{10} in the HA, tracheobronchial (TB), and alveolar (AL) regions of HRT for infants (0-1y), toddlers (1-3 y) and preschool children (3-6 y), children (6-11y), adolescents (11-21y), and adults (>21y) are presented in Table 4.7. The distribution of deposition fractions of different particle sizes in lobes of the lung are given in Table 4.8 and 4.9.

Different DFs were obtained were due to differences in particle properties and airway morphometry according to age. The assessment of particle deposition of size-segregated PM fractions in specific regions of HRT (ig. HA, TB, AL) is an essential step for understanding the doses deposited in the different regions. The total DF was found to be higher for PM_{10} than it was for $PM_{2.5}$, PM_1, $PM_{0.5}$ and $PM_{0.1}$. It is worthy to underline that, the highest DFs for PM_{10} and $PM_{2.5}$ were obtained at the adults (above 21y), whereas this group had the lowest DFs for $PM_{0.1}$, $PM_{0.5}$ and PM_1 (Table 4.7). Among the different age categories, the DFs of $PM_{0.5}$ and PM_1 were highest at the children group (6-11y)**,** while the highest DF of $PM_{0.1}$ was recorded at infants (0-1y). These findings were partially consistent with the study of Manojkumar et al. (2019), who reported that the adults (21 y) had the highest DFs for PM_{10} and $PM_{2.5}$, and that the lowest DF was observed at the toddlers (28 months). Sánchez-Soberón et al.(2015) and Sarigiannis et al.(2015) concluded that coarse particles accounted for the highest DFs compared to fine particles considering to different pattern activities. For the different lung lobes, the total DF was highest for $PM_{0.1}$, followed by $PM_{2.5}$, PM_{10}, PM_1 and $PM_{0.5}$ according to age categories, except case for the adults (>21y) at PM_{10}.

The distribution of deposited doses in HRT estimated by the MPPD model is illustrated in Fig. 11, which shows that the deposited dose (EDI) increased significantly with increases in age and particle sizes. Generally, the average EDI of $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$, and PM_{10} in the HRT (sum of EDI of HA, TB and AL) varied from 6.7 to 263.9 µg/h, 10.8 to 278.8 µg/h, 22.8 to 805.2 µg/h, 62.1 to 3362.1 µg/h and 114.7 to 5745.7 µg/h, respectively. The distribution of EDIs in different regions of the HRT (i.e., the HA, TB and AL regions) varied according to particle size and age. In general, the distribution of the EDI values were distributed as from 8.3 to 25.5% in the HA region, 13.3 to 23.4% in the TB region, and 66.9 to 84% in the AL region for $PM_{0.1}$; 21.5 to 56.9% in the HA region, 8.6 to 12.2% in the TB region, and 33 to 61.1% in the AL region for $PM_{0.5}$; 36.1 to 59.5% in the HA region, 7.1 to 11.1% in the TB, and 31.3 to 52.7% in the AL region for PM_1; 35.7 to 53.7% in the HA region, 6.1 to 18.7% in the TB region and 40.1 to 45.5% in the AL region for $PM_{2.5}$; 55 to 94.9% in the HA region, 4.2 to 44.3% in the TB region, and 0.6 to 8.4% in the AL region for PM_{10}. The highest deposition of PM_{10} was observed in the HA region, whereas the majority of $PM_{0.1}$ was deposited in the AL region, followed by the TB and HA regions. The relatively greater deposition of $PM_{0.1}$ in the AL region was due to the flow path of PM in the HRT, which is governed primarily by Brownian motion, thereby leading to preferential deposition in the AL region (Adachi, 2018; Chatoutsidou et al., 2015; Sharma and Balasubramanian, 2018). Similarly, dominant deposition of PM_{10} in the head region was reported in previously (Madureira et al., 2020; Manojkumar et al., 2019; Vu et al., 2017). Regardless of age categories, the average deposition rate of $PM_{0.5}$ and $PM_{2.5}$ in this study was relatively similar in the TB region (11.2%, 10.6%) and in the HA region (46.7%, 45.7%), whereas it differed in the AL region (42.1%, 49.5%), respectively The deposition rate of PM_{10} in different regions was quite different from other particles, which distributed as 73.6% in the HA region, 28% in the TB region and 4% in the AL region, respectively. These findings were consistent with study of Manojkumar et al.(2019), who reported that PM_{10} was highly deposited in the HA (73%) and TB (23%) regions, followed by the lowest deposition in the AL region (4%); the deposition fraction of $PM_{2.5}$ and PM_1 were 45% and 50% in the HA region; 9% and 9% in the TB region; and 45% and 40% in the AL region, respectively. Gupta and Elumalai.(2017) added that the deposited doses of PM_1, $PM_{2.5}$, and PM_{10} among inhabitants performing outdoor physical exercises distributed as 69%, 80% and 96% in the HA region, 7%, 7%, and 2% in the TB region, and 24%, 13% and 2% in the AL region, respectively.

Furthermore, deposited doses (EDIs) for PM were estimated in the five lung lobes by the MPPD model illustrated in Fig. 12. Five lung lobes included left upper (LU), left lower (LL), Right upper (RU), Right middle (RM), and Right lower (RL). Regardless of age categories, the average EDI of $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$, and PM_{10} in the lobes of the lung (Sum EDI of LU, LL, RU, RM, and RL) ranged from 6.6 to 231.9 µg/h, 5.8 to 211.8 µg/h, 11.3 to 500.4 µg/h, 39.4 to 1531.7 µg/h, and 43.6 to 208.9 µg/h, respectively among three houses. Among lung lobes, the EDIs at left lower (LL) and right lower lobes (RL) were significantly higher than those at upper and middle lobes (LU, RU and RM), which was found in all particles. These results mean that greater deposition fractions were observed in the lower lobes, and the lowest deposition for all particles was observed in the right middle lobe. The left lower lobes had higher EDIs than the right lower lobes, except for infants (0-1y) and preschool children (3-6y). Although the EDIs of $PM_{0.1}$ and $PM_{0.5}$ were lowest among investigated PM fractions, EDI proportions of $PM_{0.1}$ deposited the greatest in lung lobes, whereas the lowest proportions of EDI of PM_{10} were observed to deposit in lung lobe regions. This can be explained that majority of $PM_{0.1}$ was deposited in AL, which lead the highest deposition in lung lobes. Most of PM_{10} was settled in head airways, this resulted in lowest settlement in lung lobes. The deposited doses (EDI) in the upper lobe were 1.5 to 4 times lower than those of lower lobe, which were 5 to 20 times higher than those in right middle lobe. This variation in PM deposition was associated with lobar volume. Since the lower lobes have a greater volume, which support for the deposition of PM resulting in the high PM deposition. Meanwhile, in the middle lobe, which has a smaller volume, the extent of deposition was lower (Asgharian et al., 2006; Manojkumar et al., 2019).

Table 4. 7. Deposition fractions of PM in different human respiratory tract (HRT) regions and age categories

	$PM_{0.1}$			$PM_{0.5}$			PM_{1}			$PM_{2.5}$			PM_{10}		
Age	Head	TB	AL	Head	TB	AL	Head	TB	AL	Head	TB	AL	Head	TB	AL
0-1 y	0.08	0.09	0.42	0.19	0.04	0.18	0.22	0.04	0.18	0.23	0.12	0.30	0.51	0.41	0.01
1-3 y	0.09	0.08	0.30	0.21	0.04	0.12	0.25	0.04	0.13	0.28	0.06	0.25	0.59	0.28	0.03
3-6 y	0.09	0.07	0.33	0.21	0.03	0.13	0.25	0.03	0.15	0.28	0.05	0.28	0.60	0.26	0.07
6-11y	0.08	0.08	0.42	0.22	0.04	0.17	0.25	0.04	0.21	0.27	0.06	0.40	0.63	0.30	0.04
11-21 y	0.06	0.08	0.32	0.16	0.04	0.13	0.20	0.04	0.16	0.26	0.06	0.33	0.68	0.21	0.06
>21y	0.04	0.08	0.32	0.05	0.04	0.14	0.12	0.04	0.18	0.41	0.05	0.31	0.95	0.04	0.01

Table 4. 8. Deposition fractions of PM in different lung lobes and age categories

	$PM_{0.1}$					$PM_{0.5}$					PM_{1}				
	LU	LL	RU	RM	RL	LU	LL	RU	RM	RL	LU	LL	RU	RM	RL
0-1 y	0.05	0.18	0.04	0.04	0.19	0.02	0.08	0.02	0.02	0.08	0.02	0.08	0.02	0.02	0.08
1-3 y	0.08	0.13	0.05	0.02	0.10	0.03	0.05	0.02	0.01	0.04	0.03	0.05	0.02	0.01	0.04
3-6 y	0.05	0.13	0.04	0.03	0.14	0.02	0.05	0.02	0.01	0.06	0.02	0.06	0.02	0.01	0.06
6-11y	0.07	0.15	0.10	0.03	0.13	0.03	0.07	0.04	0.01	0.05	0.04	0.08	0.05	0.02	0.06
11-21 y	0.06	0.13	0.08	0.03	0.10	0.02	0.05	0.03	0.01	0.04	0.03	0.06	0.04	0.01	0.05
>21y	0.05	0.15	0.05	0.03	0.10	0.02	0.07	0.02	0.01	0.05	0.03	0.08	0.03	0.02	0.06

Table 4. 9. Deposition fractions of PM in different lung lobes and age categories (Cont)

	$PM_{2.5}$					PM_{10}				
	LU	LL	RU	RM	RL	LU	LL	RU	RM	RL
0-1 y	0.04	0.15	0.03	0.03	0.17	0.03	0.13	0.03	0.01	0.14
1-3 y	0.06	0.10	0.04	0.02	0.08	0.06	0.10	0.04	0.02	0.08
3-6 y	0.05	0.10	0.03	0.03	0.12	0.03	0.09	0.03	0.02	0.10
6-11y	0.07	0.14	0.09	0.03	0.12	0.04	0.09	0.06	0.02	0.08
11-21 y	0.06	0.13	0.08	0.03	0.10	0.04	0.08	0.05	0.02	0.06
>21y	0.05	0.13	0.04	0.03	0.09	0.01	0.02	0.01	0.00	0.00

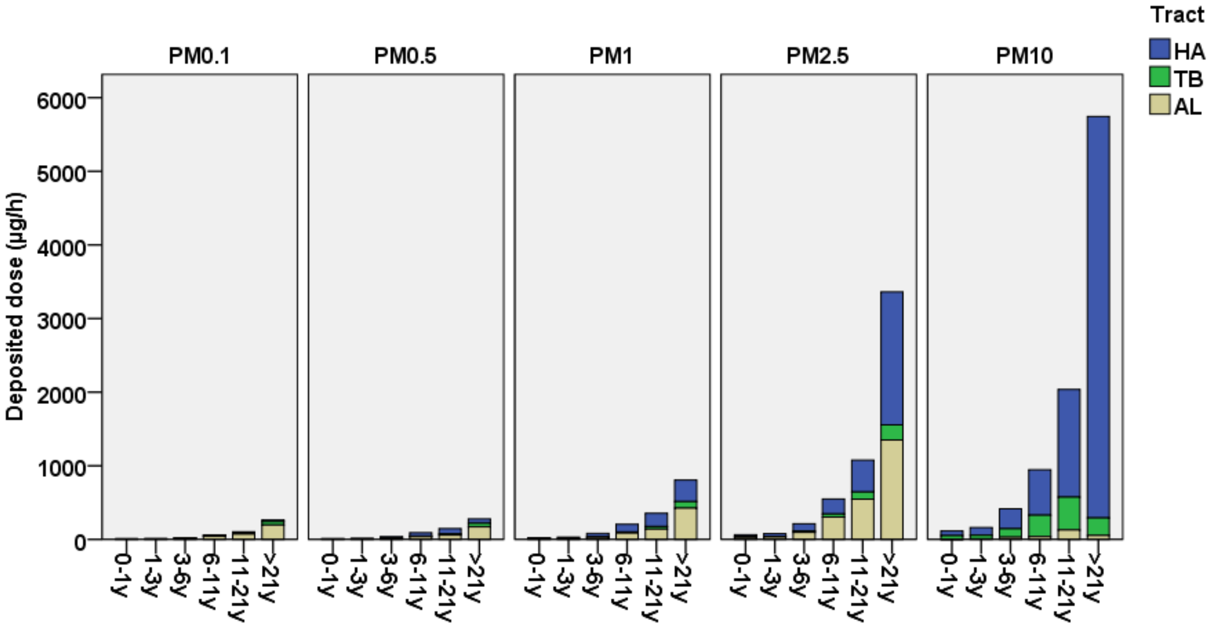


Fig 4. 11. The distribution of deposited doses at different HRT according to ages and particle sizes

HA: head airways; TB: Tracheobronchial regions; AL: Pulmonary (Alveolar) region

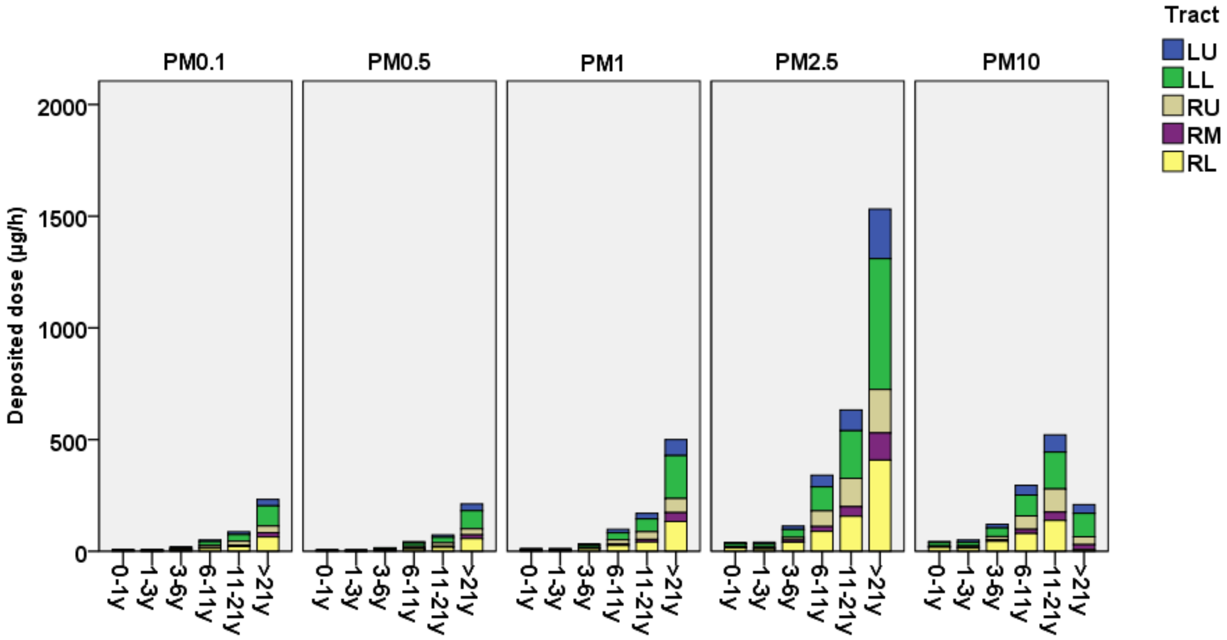


Fig 4. 12. Distribution of deposited doses at different specific lobes according to ages and particle sizes

4. CONCLUSION

Here we present one of the first studies focused on determining the seasonal variation in the mass concentration of size-fractionated PM, the relationships between indoor and outdoor PM, and the particle size distribution and deposited dose estimated in urban residential houses in Hanoi, Vietnam. The findings indicated that the average concentrations of indoor PM_{10} and $PM_{2.5}$ were 1.5- to 3.7-fold higher than WHO recommended values in summer and winter, exposing the residents of these areas to serious health risks. Indoor PM_1, $PM_{2.5}$, and PM_{10} levels were considerately higher in winter, but the change in the concentrations of indoor $PM_{0.1}$ and $PM_{0.5}$ was found to be negligible between the two seasons. Seasonal variation in PM concentrations was associated with meteorological factors, with wind speed being moderately correlated with the concentrations of all size fractions, except for $PM_{0.1}$. The particles followed unimodal distribution, with the concentrations of super-micron-sized PM having the highest concentrations, and the concentrations of coarse particles ($PM_{>10}$) and fine particles ($PM_{<0.5}$) being the lowest. In addition to indoor sources, Outdoor sources contribute primarily to indoor PM fractions. The total DFs in human respiratory tract of PM_{10} were higher than those of $PM_{2.5}$, PM_1, $PM_{0.5}$ and $PM_{0.1}$. However, the total DF in lung lobes was highest for $PM_{0.1}$, followed by $PM_{2.5}$, PM_{10}, PM_1 and $PM_{0.5}$ for all age categories (except for PM_{10} in the adults (>21y). Deposited doses for PM_{10} were mainly distributed in the HA region and $PM_{0.1}$ occurred predominantly in the AL region. The deposited doses increased with ages and PM sizes. In terms of lobar deposition distribution, the deposited doses of PM were highest in the left and right lower lobes, and the lowest deposited dose was in the right middle lobe. It is worthy to underline that, among PM fractions, the highest proportions of deposited doses of NP ($PM_{0.1}$) were observed in the lung lobes in, whereas the smallest proportion of deposited dose in lung lobes was observed for coarse particles (PM_{10}). Total lobar deposited doses were higher in the left lung than in the right lung. Human exposure to NP, fine particles, and coarse particles is emerging as an important health issue that poses significant health challenges in indoor environments.

CHAPTER 5

INDOOR $PM_{0.1}$ AND $PM_{2.5}$ IN HANOI: CHARACTERIZATION, SOURCE IDENTIFICATION AND HEALTH RISK ASSESSMENT

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 globally (GBD, 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 increased attention from researchers in recent decades. Numerous 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, particularly in Southeast Asian countries. To the best of our knowledge, few studies on indoor PM in this region are available in open databases; those that are available include 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) reported on the chemical characterization of $PM_{2.5}$ and other particle size categories 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 its seasonal variation, source identification, and health risk assessment in this region.

Indoor air quality is affected by many factors, including 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, 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 experienced high increases in the concentrations of outdoor PM, especially $PM_{0.1}$ and $PM_{2.5}$, compared to other cities in the 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 an increase in hospital admissions for respiratory ailments was reported 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.

Very few studies have been conducted on indoor PM in Hanoi to date. Studies that have been published in open databases include the following: 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) examined 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 that study. Two other studies were conducted 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., 2020b). However, these studies focused mainly 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 quantifying the chemical distribution of PM in respiratory tract regions or on 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 for evaluating health risks, as doing so can avoid adverse health impacts. Therefore, this study was designed to address this gap. Specifically, 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. METHODOLOGY

2.1. Description of the sampling area

The sampling was conducted in residential dwellings located in Hanoi, which is the second-largest city in Vietnam with a population of more than 7.5 million people and a density of up to 40,300 inhabitants/km^2 in the central districts (GSO, 2018). The climate of this region is characterized by two monsoon seasons; the northeastern monsoon in winter (November to March) and the southeastern monsoon in summer (May to September) (Phung et al., 2021, Vo et al., 2020a). Three buildings (K1, K2 and K3) in the city were selected as being representative of residential dwellings. K1, representing an urban periphery dwelling, is located in the Long Bien inner district, Northeastern Hanoi, and is strongly impacted by heavy traffic activities (upper ring road #2) and industrial activities (Sai Dong industrial zone B). K2 is considered to be typical of a roadside house, located adjacent to roads with heavy traffic densities (upper ring road #3 and lower road systems) in Southern Hanoi. K3 represents an urban dwelling in an area with a booming population and commercial areas in southeastern Hanoi. The direct distance between sites is roughly 5 to 10 km. The characteristics of three selected residential dwellings in this study are summarized in Table 5.1. The sampling sites is shown in Fig. 5.1.

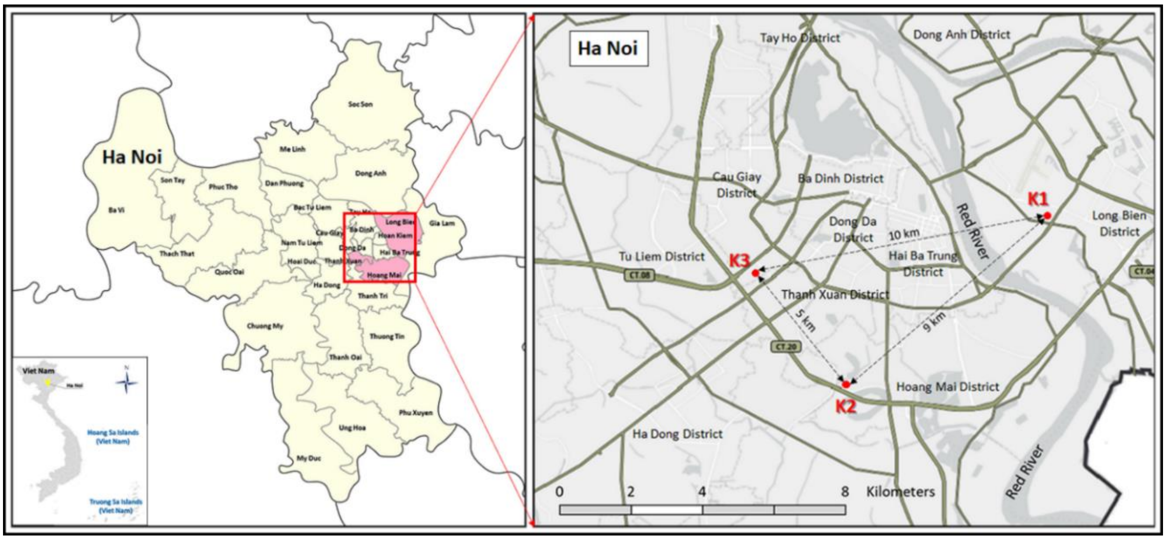


Fig 5. 1. Locations of three dwellings examined in this study in Hanoi.

Table 5. 1. Main characteristics of the three dwellings examined in this study

No.	Location	Description
K1	21°01'21.0"N 105°53'34.1"E Dam Quang Trung, Long Bien District, Hanoi	Located in the center of an urban peripheral area in Hanoi, far 50 m from upper belt road #2 and 50 m from an embankment road with a high vehicle density, 5 km from Sai Dong industrial zone. This industrial zone has a wide variety of industrial sectors, e.g., electronics, computer accessories, mobile phones, electric appliances, and non-ferrous metal and precious metal processors. The house is surrounded by a residential community, and was built in 1990 and renovated in 2005 using brick and cement. The house has two floors and a total ground area of 100 m^2. The house has ceramic floor tiles, wooden/aluminum framed windows, wooden doors, and natural and mechanical ventilation systems. Coal briquettes are used for cooking by the four occupants who sell breakfast in the morning
K2	20°58'10.9"N 105°49'41.8"E Linh Dam, Hoang Mai District, Hanoi	Located at the center of a typical urban area, approximately 5 m from upper belt road #3 and lower road system (Nguyen Huu Tho road) with a high traffic density (heavy and light vehicles). Located 500 m from the Linh Dam Detention Basin. Surrounded by high residential buildings with a high population density **and** numerous food restaurants. The structure was built in 2000 with brick and cement, and is a 2nd-floor apartment with an area of 80 m^2. The house has laminated floors, wooden windows and doors, and natural and mechanical ventilation systems. Electricity is used regularly for cooking by the two occupants.
K3	21°00'20.3"N 105°48'07.3"E Trung Hoa, Nhan Chinh Thanh Xuan District, Hanoi	Located in the center of a typical urban area, near a junction of three streets within a 100 m radius. The streets have high densities of light vehicles. Surrounded by many high residential and commercial buildings. The house was built in 2006 using brick and cement. It is a 2nd-floor apartment with an area of 120 m^2. The house has laminated floors, glass windows and doors, and natural and mechanical ventilation systems. Gas is used regularly for cooking by the five occupants.

2.2. Sample collection

Sampling at the three dwellings was conducted in two seasons, one during the winter (from November 2018 to January 2019) and the other during the summer (from April to June 2019). These sampling seasons were selected to cover the periods of the lowest and highest PM concentrations in Hanoi in the year (Phung et al., 2021, Nguyen et al., 2018). At sampling site, each sampling batch lasted for 14 consecutive days. During each sampling season, batches were sampled consecutively, from one site to the next. At each dwelling, daily samples of $PM_{2.5}$ and $PM_{0.1}$ in the indoor and outdoor air were simultaneously collected. During sampling, the windows/doors were kept closed, and only opened when entering/leaving the house and during cleaning. Daily activities at the three dwellings were performed as usual during the sampling periods. The indoor samplers were placed in the bedrooms at K1 and K2 and in the master room at K3, all of which are on the 2nd floor of the dwellings. The sampling of outdoor particles was performed in the corridors of the dwellings. Both indoor and outdoor inlets were placed approximately 1.5 m above the floor to simulate the human breathing zone. The sampling instruments were placed at least 1 m away from any obstacles (windows, doors, ventilation inlets and walls). Samples of indoor and outdoor $PM_{0.1}$ were collected using quartz filters (55 mm in diameter) by two identical Nano Sampler II devices (Model 3182, KINOMAX, Japan) using a constant flowrate of 40,0 L/min. Simultaneously, samples of indoor and outdoor $PM_{2.5}$ were also collected using 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 flowrate of 16.7 L/min. The collection of field blanks was also taken. Before sampling, all samplers were calibrated to obtain recommended flowrates using a mass flow meter (4040, TSI Incorporated). Quartz filters were pre-baked at 900°C for 4 h to remove possible contaminants. A total of 320 samples was collected.

2.3. Mass and chemical analysis

The collected filters, real and blank samples, were equilibrated in a desiccator in a room, where the relative humidity and temperature were maintained at 30 to 40% and 25 ± 5°C, respectively, for 48 hours 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 weighing samples. After weighing, the quartz filters were subsequently stored in a refrigerator at 4°C for further analysis. A haft of each quartz filter was digested in a concentrated acid mixture ($1HNO_3$: 3HCl, v/v) in a Mars 6 microwave for 30 minutes as specified by the US EPA IO-3.1 method. Solutions after digestion were analyzed for 10 trace elements (TEs) (Cr, Mn, Co, Cu, Ni, and Zn, As, Cd, Sn, and Pb) by inductively coupled plasma mass spectrometry, ICP-MS (X-series 2; Thermo Fisher Scientific, Massachusetts, USA) at 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 with relative standard deviations of 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). Recovery tests for TEs were also conducted using spiked samples with the recoveries of all the TEs being 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 a blank sample and a laboratory control sample.

2.4. Determination of infiltration factor *(F_{inf})*

The infiltration factor (F_{inf}) is defined as the fraction of outdoor PM and 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 or the chemical components in the indoor and outdoor environments, respectively. C_{ig} is the concentration of PM or the components generated from indoor sources. C_{ig} intensity (%C_{ig}) is the fraction of indoor-generated PM, which is calculated using Eq. (2)

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

2.5. Source identification

2.5.1. Enrichment factor

Enrichment factor (EF) is used to determine whether atmospheric aerosol elements are derived from indoor or outdoor sources (Kulshrestha et al., 2014, Wang et al., 2019). Outdoor PM concentration is taken as a reference value in this term, and all atmospheric particulate matter is assumed to be derived from outdoor sources. EFs can be calculated using the following formula:

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

where C is the concentration of the elements in the particle phase. The elements with EFs close to 1 regarding as "non-enriched" elements and outdoor sources are considered as the main source. Conversely, if the EF for elements is greater than 1, this suggests that the elements are "enriched" and major sources other than outdoor sources are considered to be the primary source.

2.5.2. Principle Component Analysis (PCA)

Principle 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 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 Eigenvalues of the correlation matrix are greater than 1 (David et al., 2019; Kulshrestha et al., 2014, Wang et al., 2019). In this study, factor loadings greater than or equal to 0.7 are employed for source apportionment.

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 consisting of three main parts, i.e., the head airways (HA) region, the tracheobronchial (TB) region, and pulmonary/alveolar (AL) region, and then to estimate the internal dose of TEs. In our study, we used a simplified equation based on the ICRP model with Hind's parameterization for calculating the deposited dose of inhaled TEs in PM in the human respiratory tract (HRT) (Bair,1994; Gao et al., 2017; Hinds., 1999). Major mechanisms of PM deposition across the HRT include diffusion, sedimentation, and impaction. The deposition mechanisms of PM are differentiated in each HRT region considering PM size, HRT anatomy, and physiology of respiratory systems (Bair, 1994; Gao et al., 2017). The deposited doses from inhaling TEs in $PM_{0.1}$ and $PM_{2.5}$ in the HRT are the sums of the deposited doses in the three regions, which are estimated following the US EPA model (USEPA, 1989, US-EPA, 2009):

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

where, EDI is the total deposited dose of TEs (ng/day); DF_i is the particle deposition fraction of the *i* region in human respiratory tract (HA, TB, AL); C_j is the concentration of TEs of the *j* trace element (ng/m^3); *j*=1-n, in which n is the number of TEs; IR is the inhalation rate (m^3/day); ET is the exposure time (hours/day); EF is the exposure frequency (days/year); ED is the exposure duration (years); and AT is the average lifetime (days). DF_i is a function of the aerodynamic diameter (dp) (Chalvatzaki and Lazaridis, 2015) and is estimated using Eqs. (5-8), presented in Table 5.2

2.6.2. Estimation of non-carcinogenic and carcinogenic risks

The assessment of non-carcinogenic and carcinogenic health risks was performed for seven age categories in winter and summer following the US EPA model, which has been widely used to assess the health risk in earlier studies (Sharma and Balasubramanian, 2018; Wang et al., 2018; Oh et al., 2020).

Regarding to non-carcinogenic risk (non-car risk), the Hazard Quotient (HQ) for individual trace elements and the Hazard Index (HI) was the sum of HQ, which was calculated using Eq. (9) and Eq. (10):

$$HQ = \frac{EDI_j}{BW \times RfD_j} \quad (9)$$

$$HI = \sum_{j=i}^{n} HQ \quad (10)$$

Regarding to carcinogenic risk (car-risk), the increased lifetime cancer risk (ILCR) was quantified as the incremental probability of an individual developing cancer over a lifetime due to exposure to potential carcinogenic substances, and is estimated using the Eq (11):

$$ILCR = \sum_{j=1}^{n} ILCR_j = \frac{EDI_j \times CSF_j}{BW} \quad (11)$$

where BW is body weight (kg) and RfD_j is the chronic reference dose (mg/kg.day) for element j. CSF is the cancer slope factor for a specific element (kg.day/mg). In the estimation of EDI and ILCR, the values of parameters (Cj, ET, BW) were determined by distributing 500 questionnaires to residents of Hanoi; IR, EF, ED, AT, CSF were based on the exposure handbook published by the US EPA. Details of data sources for the calculations are shown in Table 5.3 and Table 5.1 in the appendix. To protect the public health**.** 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 value $\leq 10^{-6}$ indicates “zero risk” or no adverse risk, the acceptable ILCR value for car risk is within $10^{-6} < \text{ILCR} \leq 10^{-4}$. In contrast, an ICLR value greater than 10^{-4} is considered unacceptable (US-EPA, 1989, US-EPA, 2009).

2.7. Questionnaire method

A total of 500 questionnaires were distributed to households in Hanoi to gather basic information about families and their daily activities during sampling campaign. The collected data included exposure parameters (BW, age, ET in indoor air and outdoor air, daily indoor and outdoor activities), which are summarized in Table 5.3 and used for the health risk assessments. At four of the surveyed houses, the questionnaires included additional information about the characteristics of the houses (ventilation system, flooring, walls, structure of the windows, and building age) and daily indoor activities (cooking, cleaning, smoking, incense burning, etc.), type of stove, number of occupants, etc., through a daily_report were carried during sampling period.

2.8. Statistical analysis

In this study, the statistical analysis was carried out using IBM SPSS Statistics 20 software. Normal distribution was tested by Shapiro-Wilk *t*-tests and linear regression analysis. Mann-Whitney and Kruskal-Wallis tests were used for nonparametric tests. All results were considered to be statistically significant if $p<0.05$. Monte Carlo simulation was used to perform a probabilistic risk assessment. This probability-based method can generate output as a cumulative distribution function from the random input variables instead of using fixed single values that can minimize the uncertainties of expected outputs. Cumulative distribution functions were determined by Monte Carlo simulation with 100,000 iterations, run via the Oracle Crystal Ball software program in the EXCEL (ver. 11.1.2.4.850, Oracle, Inc., USA).

Table 5. 2. Deposition fraction estimation

Parameter	Equation	
IF (Inhalation fraction)	$\mathrm{IF} = 1 - 0.5\,[1 - \frac{1}{1+0.00076\mathrm{D}_\mathrm{p}^{2.3}}]$	(5)
$\mathrm{DF}_{\mathrm{HA}}$ (Deposition fraction of head airways)	$\mathrm{DF}_{\mathrm{HA}} = \mathrm{IF}\,[\frac{1}{1+\exp\,(6.84+1.183\ln\mathrm{Dp}} + \frac{1}{1+\exp\,(0.924-1.885\ln\mathrm{Dp})}]$	(6)
$\mathrm{DF}_{\mathrm{TB}}$ (Deposition fraction of trachea-bronchia region)	$\mathrm{DF}_{\mathrm{TB}} = \frac{0.00352}{\mathrm{D}_\mathrm{p}}[\exp\left(-0.234(\ln\mathrm{D}_\mathrm{p}+3.40)^2\right) + 63.9\exp\left(-0.819(\ln\mathrm{D}_\mathrm{p}-1.61)^2\right)]$	(7)
$\mathrm{DF}_{\mathrm{AL}}$ (Deposition fraction of alveolar region)	$\mathrm{DF}_{\mathrm{AL}} = \frac{0.0155}{\mathrm{D}_\mathrm{p}}[\exp\left(-0.416\left(\ln\mathrm{D}_\mathrm{p}+2.84\right)^2\right) + 19.11\exp\left(-0.482(\ln\mathrm{D}_\mathrm{p}-1.362)^2\right)]$	(8)

Table 5. 3. Parameters used for the average daily exposure assessment

	0-1 y	1-3 y	3-6 y	6-11 y	11-21 y	21-60 y	>60 y
Inhalation rate-(IR) (m^3/day) [a,c]	5.3	8.9	10.1	12.9	15.4	18.6	14.3
Indoor exposure time (ET) (h/d) [b]	22.5	19.5	14.5	12.4	12.3	14.4	18.5
Outdoor exposure time (ET) (h/d) [b]	1.5	1.5	1.5	2.5	3.0	3.0	2.5
Exposure frequency-(EF) (day/year) [c]	365	365	365	365	365	365	365
Exposure duration (ED) (year)[c]	1	1	3	6	11	21	60
Body weight (BW) (kg)[b]	9.1	10.6	14.5	26.3	46.7	55.4	56.5
Average life time for carcinogenic risk (AT) (day)[c]	25550	25550	25550	25550	25550	25550	25550
Average life time for non-carcinogenic risk (AT) (day)[c]	ED*365	ED*365	ED*365	ED*365	ED*365	ED*365	ED*365

1. [a]: USEPA, “Exposure Factors Handbook ,2011 Edition (Final Report), Chapter 6, 2011
2. [b]. Questionnaires administered in this study.
3. [c] USEPA, Risk Assessment Guidance for Superfund Volume I: Human Health Evaluation Manual (Part F, Supplemental Guidance for Inhalation Risk Assessment), 2009

3. RESULTS AND DISCUSSION

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

The mass concentrations of indoor $PM_{2.5}$ and $PM_{0.1}$ in the three dwellings (K1, K2, and K3) are shown in Fig.5.2. The average concentration of indoor $PM_{2.5}$ in the three dwellings was 59.9±23.6 μg/m^3, which is approximately four times higher than WHO guidelines of 15.0 μg/m^3 (WHO, 2021). 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 comparable to that for residential apartments in China, which have similar characteristics to the houses in this study (i.e., natural ventilation and type of house) (Wang et al., 2016). However, the $PM_{2.5}$ concentrations measured in this study were higher than those in houses with mechanical ventilation in China and Canada (Kearney et al., 2014; Wang et al., 2016), as air cleaning systems can enhance indoor air quality. Meanwhile, the average concentration of indoor $PM_{0.1}$ in this study was considerably higher than that in schools in Hanoi, in which no indoor PM sources, such as cooking and burning incense, were present (Tran et al., 2020). The high concentrations of indoor $PM_{0.1}$ and $PM_{2.5}$ observed in this study may increase the disease burden of occupants, adversely affecting microvascular function, inflammation, and lung cell integrity (Karottki et al., 2015).

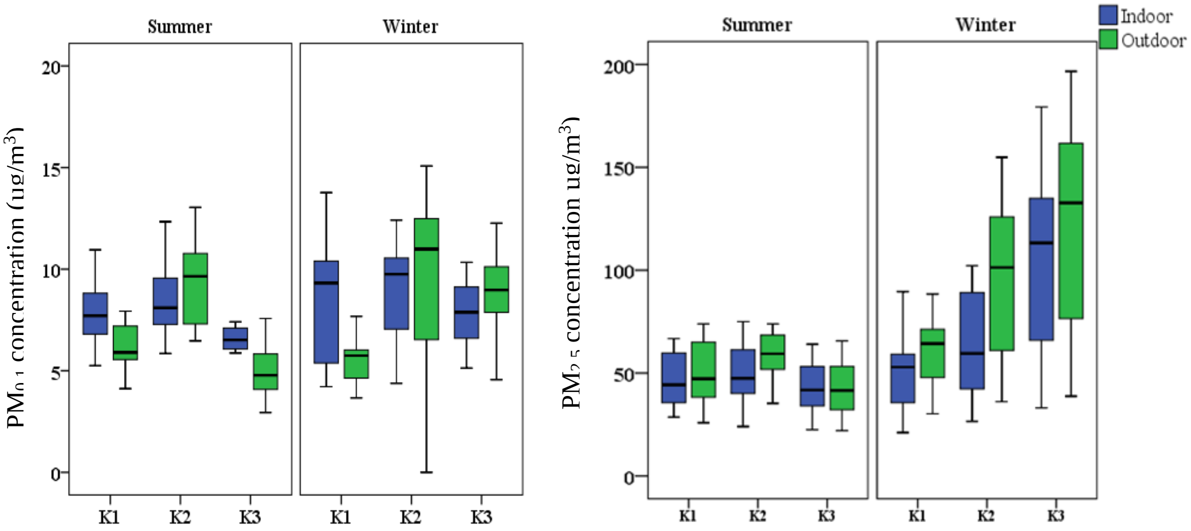


Fig 5. 2. Indoor and outdoor concentrations of $PM_{2.5}$ and $PM_{0.1}$ in two seasons

Significantly seasonal variation was observed for indoor $PM_{2.5}$, but not for indoor $PM_{0.1}$. The indoor mass concentrations of $PM_{2.5}$ were in the range of 43.4 μg/m^3 - 49.5 μg/m^3 in summer, whereas those varied from 50.6 μg/m^3 - 105.9 μg/m^3 in winter, respectively. The level of indoor $PM_{2.5}$ in winter increased 2.5-folds in comparison that in the summer. The seasonal variation of indoor $PM_{2.5}$ observed in this study was closely associated with by variations in outdoor $PM_{2.5}$, which was also reported the studies of Phung et al., 2021 and Nguyen et al., 2018. This relationship can be explained by the value of I/O (indoor/outdoor concentrations) ratio of $PM_{2.5}$, which is less than 1, suggesting that indoor $PM_{2.5}$ was strongly influenced by outdoor $PM_{2.5}$.

Seasonal trends of indoor $PM_{2.5}$ were also observed in residential houses in North-central India and Lahore in Pakistan (Massey et al., 2012, Sidra et al., 2015). Meanwhile, for indoor $PM_{0.1}$, the mass concentration remained relatively constant between seasons. The levels of indoor $PM_{0.1}$ varied from 7.2 to 8.7 µg/m^3 in summer, whereas those in winter ranged from 8.1 to 8.9 µg/m^3. The I/O ratio for $PM_{0.1}$ was slightly higher than 1, implying that the indoor $PM_{0.1}$ was, to some extent, affected by indoor sources. However, it is likely that the observed trend of indoor $PM_{0.1}$ was primarily attributed to the seasonal stability of outdoor $PM_{0.1}$ in Hanoi, as observed in this study and reported in the study of Nguyen et al. (2018).

Furthermore, the contribution of the particles was also evaluated among three houses for a better understanding of PM distribution. The ratios of indoor $PM_{0.1}/PM_{2.5}$ were 18%, 17%, and 13% at K1, K2, and K3, respectively. Meanwhile, the ratios of outdoor $PM_{0.1}/PM_{2.5}$ were 12.5%, 14%, and 11% at K1, K2, and K3, respectively. At each house, the $PM_{0.1}$ to $PM_{2.5}$ in the indoor air was slightly higher than that in the outdoor air, implying that $PM_{0.1}$ makes a larger contribution to $PM_{2.5}$ in indoor air. Morawska et al. (2017) suggested that cooking, burning incense, and cleaning activities released particles smaller than 300 nm, the majority of $PM_{0.1}$. Meanwhile, outdoor air was the primary of $PM_{2.5}$. In addition, the ratios of indoor $PM_{0.1}$/ $PM_{2.5}$ at K1 and K2 are at almost the same level, implying that the indoor PM in these houses was originated from the same types of indoor sources. However, this ratio at K3 is slightly different from those at K1 and K2, implying that indicating the different kinds of indoor sources and their contributions at K3.

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

The concentrations of ten trace elements (TEs) bound to $PM_{2.5}$ and $PM_{0.1}$ collected at residential dwellings (K1, K2, K3) are shown in Fig.5.3. The average concentrations of ∑10TEs bound to indoor $PM_{2.5}$ and $PM_{0.1}$ were 763±324 ng/m^3 and 84±22 ng/m^3 in summer; and 1511±736 ng/m^3 and 193±31 ng/m^3 in winter, respectively. In general, higher concentrations of TEs were observed in $PM_{2.5}$ and $PM_{0.1}$ at roadside house, where those increased 1.5 - 2.3 folds in $PM_{2.5}$ and 1.2 -1.3 folds for $PM_{0.1}$ in comparison with those in urban houses and the urban periphery. This point is similar to that reported by Kulshrestha et al. (2014), who also found that that the concentration of TEs in indoor $PM_{2.5}$ at the roadside houses were higher than those in urban and rural houses.

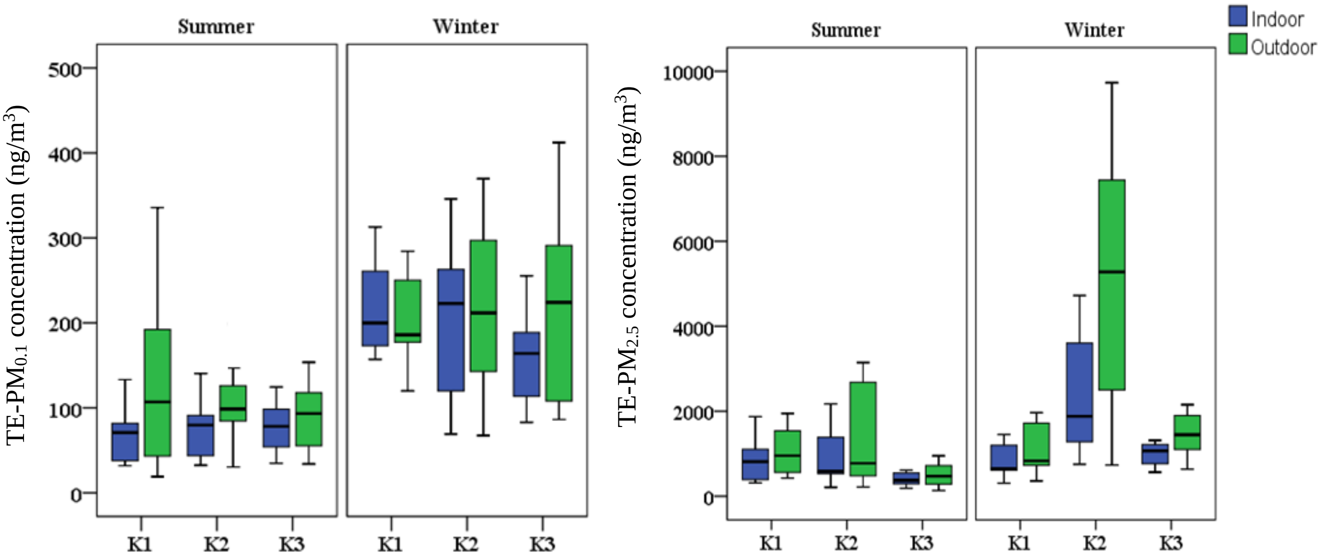


Fig 5. 3. Variation of TEs of $PM_{0.1}$ and $PM_{2.5}$ in summer and winter at three dwellings.

The mean ratios of ∑10TEs 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 is particularly apparent at K1 and K2, implying that the indoor sources of PM were similar in these houses. However, this ratio is slightly higher at K3, suggesting additional indoor sources at this site, which is consistent with the 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.

The concentrations of individual TEs in PM are shown in Tables 5.4 and 5.5. The concentrations of 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, 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 and Ni in the three houses were 1.4 to 4.6, and 1.5 to 4.5 times higher than the EU limits. The exceedances of As and 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% to 12% and 74% to 77%, respectively, whereas those of Zn, Pb, and Cr to the total concentration of TEs bound to $PM_{0.1}$ were 34% to 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 5.2 in the appendix

Table 5. 4. Concentrations of TEs bound to PM$_{2.5}$ at the three dwellings (ng/m^3) (P=1 atm, T= 25°C)

Elements	K1				K2				K3			
	Summer		Winter		Summer		Winter		Summer		Winter	
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
Cr	20.3	22.5	56.5	77.8	42.1	51.3	81.0	218	22.6	31.5	37.6	85.0
Mn	27.2	32.7	26.7	59.4	33.7	47.1	90.3	165	15.1	16.7	37.0	60.2
Co	0.5	0.5	0.9	2.1	0.5	0.8	8.2	7.8	0.7	0.7	1.1	2.3
Ni	11.6	15.9	37.4	63.4	39.7	53.7	53.3	144	11.7	17.6	30.4	58.7
Cu	22.6	28.9	25.5	32.8	16.5	26.1	24.0	56.3	5.5	8.5	29.5	31.4
Zn	774	998	905	1190	682	1100	1930	4100	267	375	736	1080
As	8.0	8.1	14.3	20.6	10.5	12.1	20.8	45.3	6.3	7.6	16.4	17.4
Cd	2.7	2.7	3.1	6.2	2.6	4.6	4.5	11.4	0.5	1.4	2.8	3.9
Sn	7.3	8.6	10.9	19.2	5.9	7.5	12.6	25.2	5.9	5.9	9.3	12.3
Pb	115	142	97.5	156	73.3	107	133	254	61.3	55.7	101	139

Table 5. 5 . Concentrations of TEs bound to PM$_{0.1}$ at the three dwellings (ng/m^3) (P=1 atm, T=25°C)

Elements	K1				K2				K3			
	Summer		Winter		Summer		Winter		Summer		Winter	
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
Cr	14.8	16.8	49.5	35.8	21.1	29.8	37.9	37.9	12.8	16.5	15.9	55.4
Mn	4.9	9.5	7.8	13.7	6.1	11.7	15.3	15.6	4.1	4.1	9.5	16.3
Co	0.3	0.4	0.5	0.8	0.3	0.4	0.6	0.8	0.2	0.1	0.7	0.8
Ni	5.7	4.5	5.9	10.3	11.9	10.8	7.2	7.9	3.9	5.8	11.3	17.1
Cu	2.2	2.4	5.1	5.1	2.9	4.5	4.8	4.7	1.4	1.1	5.5	6.1
Zn	26.7	72.2	116	109	31.0	37.5	104	108	45.4	49.7	84.8	82.6
As	1.2	1.5	5.5	8.4	2.4	2.2	6.7	7.6	1.1	0.9	3.8	4.0
Cd	0.2	0.3	0.5	0.5	0.3	0.3	0.6	0.6	0.1	0.1	0.5	0.6
Sn	1.1	1.6	5.2	3.7	1.7	1.8	4.1	4.5	0.7	0.5	3.2	3.3
Pb	9.6	13.7	20.1	15.5	9.6	10.9	23.7	26.3	6.9	7.8	17.7	20.7

3.3. Infiltration factor *(F_{inf})* and indoor generated particles *(C_{ig})*

3.3.1. F_{inf} and C_{ig} 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. 5.2 in the appendix. In summer, the average F_{inf} was 1.0 and 0.9 for $PM_{0.1}$ and $PM_{2.5}$, respectively, whereas in winter, the average F_{inf} was 1.0 and 0.8 for $PM_{0.1}$ and $PM_{2.5}$, respectively. The results 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 even when the doors and windows are closed (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 why the F_{inf} value of $PM_{2.5}$ was slightly lower than that of $PM_{0.1}$.

The contribution of indoor-generated PM to indoor PM *(%Cig)* in this study varied widely among houses in seasons, reflecting the complex and dynamic features in indoor environment. However, based on the values of *%Cig* obtained; it can be roughly estimated that about 2% to 21% of indoor $PM_{2.5}$ in the three houses originated from indoor sources. This highlights the importance of outdoor sources of $PM_{2.5}$ in the indoor environment. It was estimated that 18% and 54% of indoor $PM_{0.1}$ in 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, the $\%C_{ig}$ of $PM_{0.1}$ was negative at K1, which might be attributed to the decomposition of some species of $PM_{0.1}$ (Sangiorgi et al., 2013, Vu et al., 2017).

3.3.2. Finf and Cig for TEs concentration

The mean F_{inf} values of $\sum$10TEs 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% to 41% and 3% to 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 5.3 in the appendix. In general, As, Zn, and Co appeared to have the highest C_{ig} intensities ($\%C_{ig,\mathrm{As}}$=71% in $PM_{0.1}$ in winter at K1; $\%C_{ig,\mathrm{Co}}$=56% in $PM_{2.5}$ in summer at K3; $\%C_{ig,\mathrm{Zn}}$=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. Estimation of EFs

The EFs of TEs in $PM_{2.5}$ and $PM_{0.1}$ in the three houses in summer and winter are shown in Fig. 5.4. 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 Ni 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.

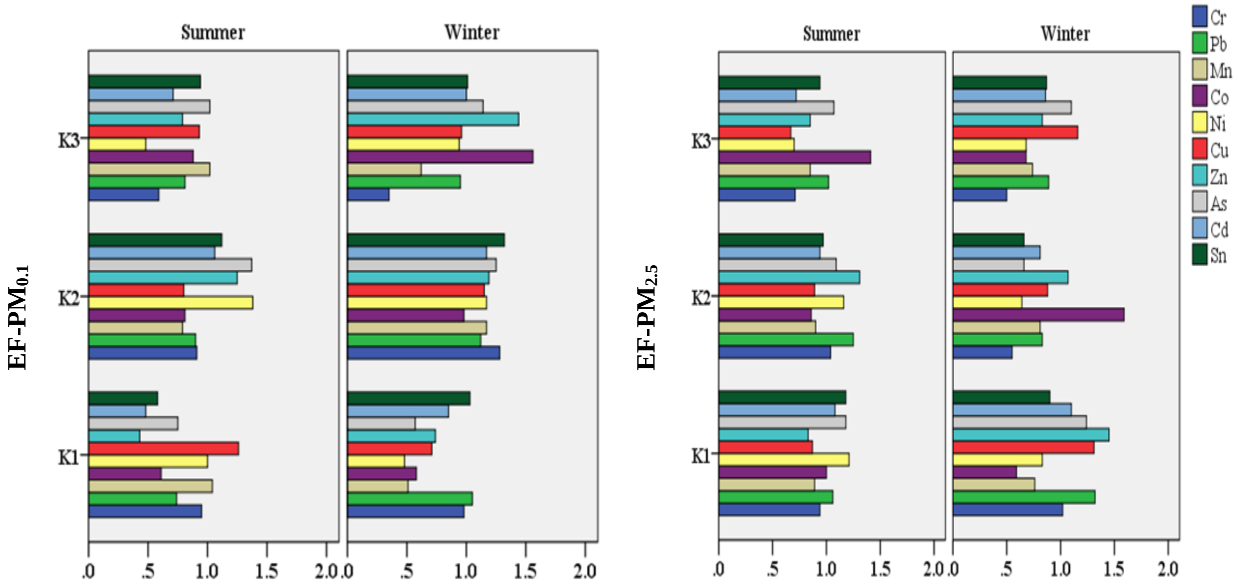


Fig 5. 4. Enrichment factors of TEs in $PM_{0.1}$ and $PM_{2.5}$ at the three dwellings in summer and winter.

3.4.2. Principle component analysis (PCA)

The results of PCA for source identification at the three investigated houses and source distribution are summarized in Fig. 5.5.

Urban periphery house (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 PM (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 the 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-exhausts (Zn), which accounted for 15%. These sources are considered to be 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 observed 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 example, 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, factor 2 accounted for 22% of indoor $PM_{2.5}$ and 17% of indoor $PM_{0.1}$, 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, in winter, outdoor sources accounted for 55% and 80% of indoor $PM_{0.1}$ and indoor $PM_{2.5}$, respectively. In summer, outdoor sources accounted for 57% and 58% of indoor $PM_{0.1}$ and indoor $PM_{2.5}$, respectively. 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, including some focused on 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 to be markers of industrial sources in Hanoi, accounting for approximately 6% of ambient $PM_{0.1}$ and $PM_{2.5}$ 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 an 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 house (K2): In winter, two PCA factors explained 83% and 80% of the variation observed 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 combustion (As, Pb and Cd), and vehicle non-exhaust emissions from brakes 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 showed that outdoor sources are the main contributors to indoor PM at roadside house. In winter, outdoor sources contributed to 64% and 52% of indoor $PM_{0.1}$ and $PM_{2.5}$, respectively, and 64% and 63% of the corresponding PM in summer. In winter, indoor sources contributed to 19% and 28% of indoor $PM_{0.1}$ and $PM_{2.5}$, 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 have been reported to 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 close proximity of traffic roads that are used extensively by both light- and heavy-duty vehicles (e.g., upper ring road #3 and the nearby Nguyen Huu Tho road). This house is also affected by heavy traffic along Giai Phong Road and the Nuoc Ngam and Giap Bat bus stations. In addition, the combustion of coal briquettes occurs routinely at nearby eateries and small restaurants.

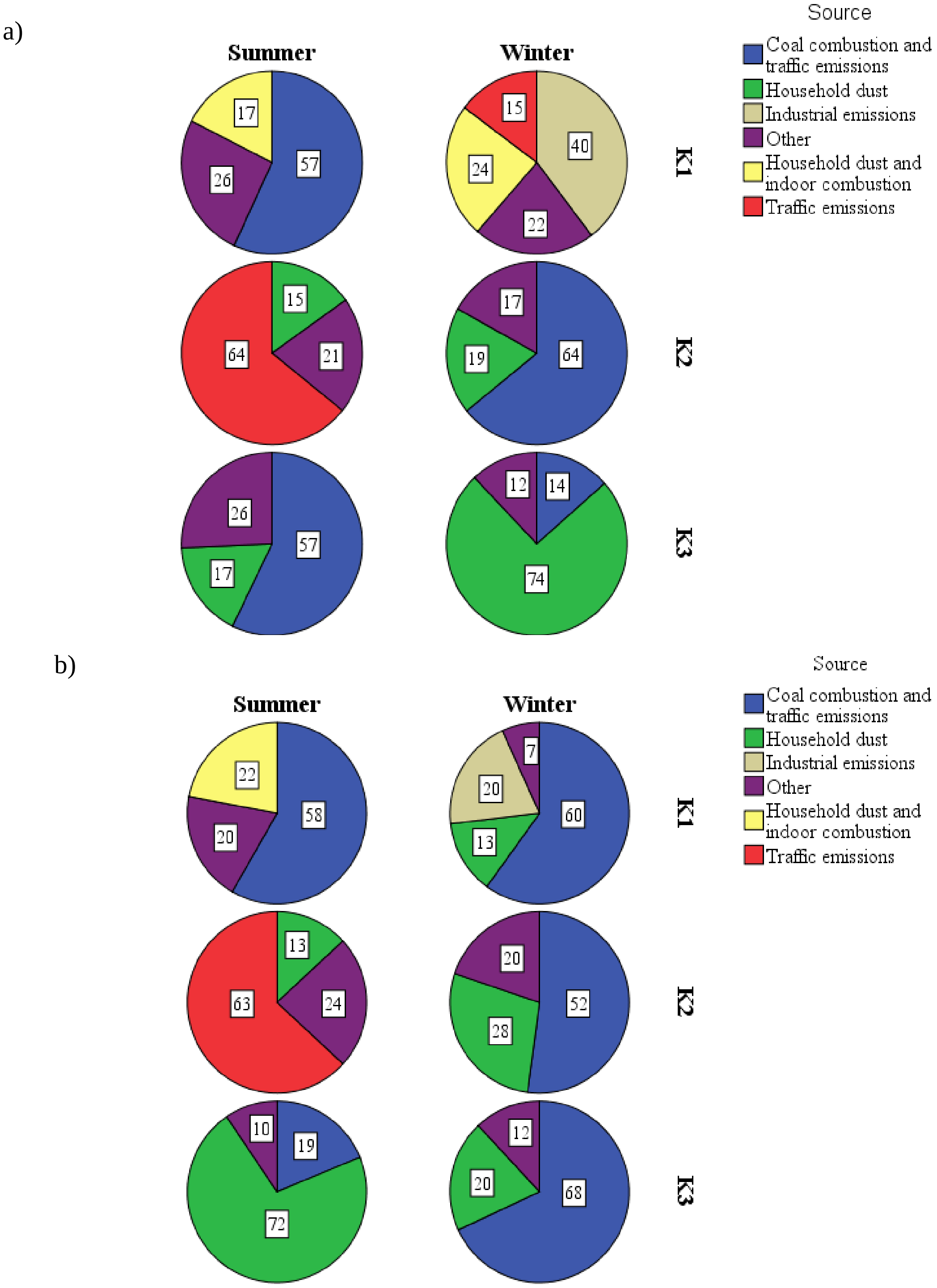


Fig 5. 5. Source distribution of trace elements in particulate matter (PM) at the three dwellings

a) $PM_{0.1}$; b) $PM_{2.5}$

Urban house (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 frequently took place 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, the contribution of these sources 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 deposited doses in human respiratory tract (HRT)

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 ($PM_{0.1}$ and $PM_{2.5}$) according to different age categories of inhabitants are shown in Tables 5.5 and 5.6, 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 to 22 500 ng/d and 4.8 to 1880 ng/d in $PM_{2.5}$, respectively, and 63.1 to 458 ng/d and 0.2 to 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 of Gao et al. (2017), who reported that 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 to 32 times higher than that in student offices, laboratories, and outdoor environments. However, our results contrasted with those of Sharma and Balasubramanian (2018) who reported that a higher EDI of TEs was found for outdoor air and that EDI increased as the haze intensity increased.

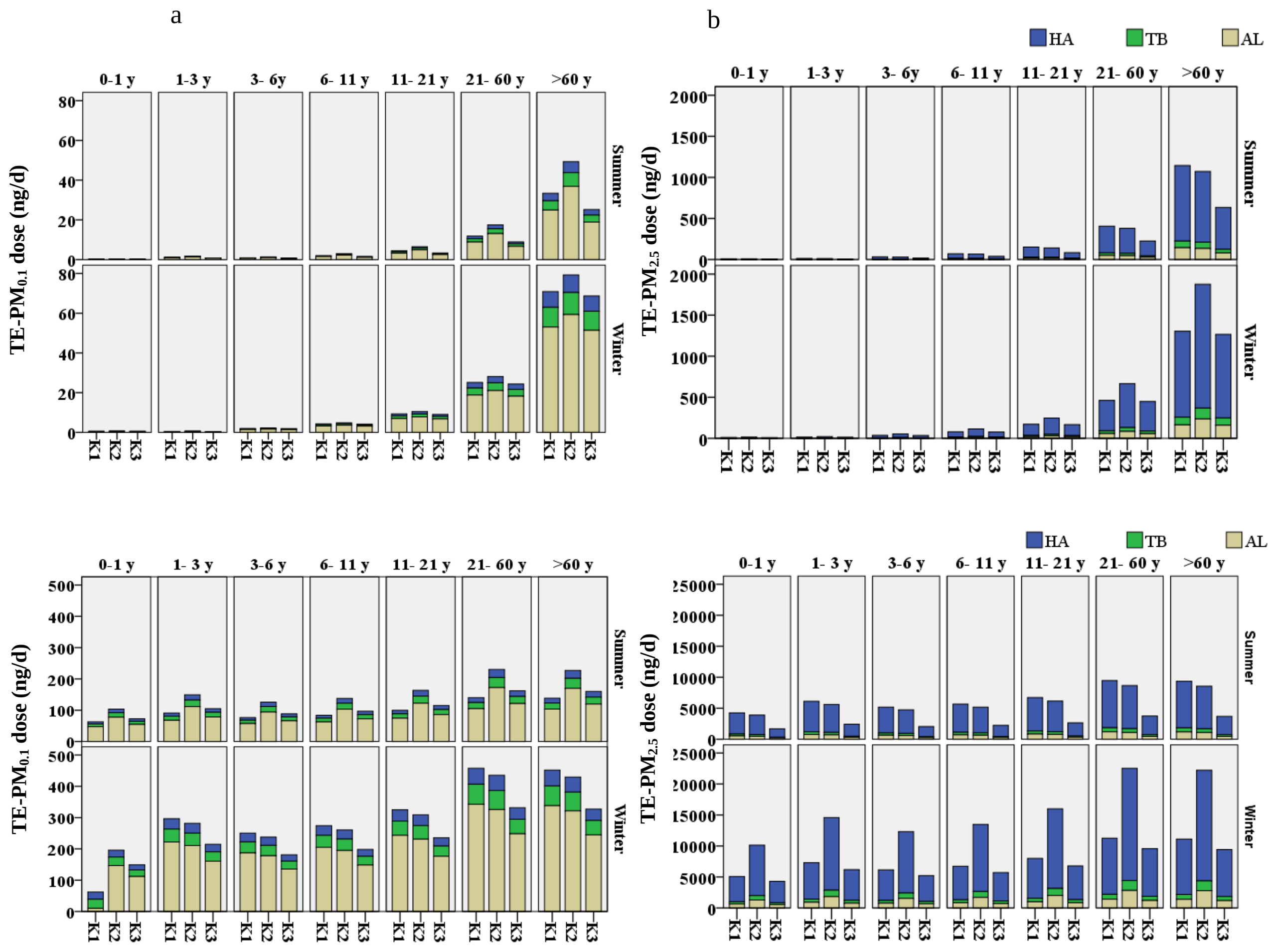


Fig 5. 6. Deposited doses of carcinogenic trace elements (TEs) (a) and non-carcinogenic TEs (b) in different respiratory tract regions (HA: head airways; TB: tracheobronchial; AL: alveolar/pulmonary)

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 the occupants of the three houses, and are presented in Fig. 5.6. In general, the EDI values for PM$_{2.5}$ were distributed as follows: 80% in the HA region, 7% in the TB region, and 13% in the AL; however, for PM$_{0.1}$, they were distributed as 11% in the HA region, 14% in the TB region, and 75% in the AL region. 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 the HA region, whereas the majority of TEs-PM$_{0.1}$ was deposited in the AL region, followed by the TB and HA regions. The greater deposition of PM$_{0.1}$ in the AL region was due to the flow path in the HRT, which is primarily governed by Brownian motion, thereby leading to preferential deposition in the AL region (Adachi, 2018; Chatoutsidou et al., 2015; Sharma and Balasubramanian, 2018). Similar results have been reported in previous studies. For example, Vu et al. (2017) reported that PM$_{0.1}$ is deposited predominantly in the AL region, followed by the TB region. Madureira et al. (2020) reported that PM$_{10}$ is primarily deposited in the HA region (86–97%), followed by PM$_{2.5}$ (60–75%) and

$PM_{0.1}$ (31–40%), whereas $PM_{0.1}$ deposition mainly occurs in the AL region. Thus, the EDI in the HA region decreased as the particle size decreased, whereas that in the TB and AL regions increased as particle size decreased. Although the total EDI of TEs-$PM_{0.1}$ was significantly lower than that in $PM_{2.5}$ (proportion 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 into the AL region, which causes severe damage to human health.

The overall results in Tables 5.6 and 5.7 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 to 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 EDIs during the exposure to TEs in both particle size categories. 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 exhibit a high tendency to deposit particles in the lower respiratory tract owing to them having less efficient uptake of particles in the HA region. Therefore, this should be considered when developing public health measures focusing on protecting the health of children and elderly adults. Our findings were similar to those of Madureira et al. (2020), who showed that the EDI is higher in mothers than in infants (0–3 months) for both $PM_{0.1}$ 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 illustrate the importance of protecting human health because people spend almost 90% of their lives indoors.

3.6. Estimation of health risk

3.6.1. Non-carcinogenic risk

Assessment of non-car risks for TEs bound to $PM_{0.1}$ and $PM_{2.5}$ in the three residential dwellings was conducted in seven age categories. The HI was determined to estimate the non-car risk associated with TE inhalation according to Eq. (10), and the findings are presented in Fig. 5.7. 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 their lower body weight (BW). It is estimated that the HI values for these age groups are twice as high as those of adults, thereby implying that there is a greater potential non-car risk for infants and toddlers.

Table 5. 6. Total deposited dose of carcinogenic TEs bound to PM in different age categories at the three sites

	Total deposited dose of carcinogenic TEs (ng/d)											
	K1				K2				K3			
	$PM_{2.5}$		$PM_{0.1}$		$PM_{2.5}$		$PM_{0.1}$		$PM_{2.5}$		$PM_{0.1}$	
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
SUMMER												
0-1 y	8.7	0.7	0.3	0.0	8.2	0.8	0.4	0.0	4.8	0.3	0.2	0.0
1-3 y	12.5	1.2	0.5	0.0	11.8	1.3	0.6	0.1	6.9	0.6	0.3	0.0
3-6 y	31.7	3.9	0.9	0.1	29.7	4.3	1.4	0.2	17.6	1.9	0.7	0.1
6-11 y	69.3	17.0	2.0	0.5	65.0	18.3	2.9	0.6	38.5	8.2	1.5	0.4
11-21 y	151	44.7	4.4	1.3	141	47.9	6.5	1.7	83.7	21.4	3.3	1.0
21-60 y	406	103	11.8	3.0	380	110	17.5	3.8	225	49.2	8.9	2.3
>60 y	1140	187	33.4	5.5	1070	201	49.3	7.0	635	89.9	25.2	4.2
WINTER												
0-1 y	9.9	1.1	0.5	0.1	14.3	2.0	0.6	0.1	9.6	0.9	0.5	0.1
1-3 y	14.3	1.8	0.5	0.1	20.6	3.4	0.9	0.1	13.9	1.6	0.4	0.1
3-6 y	36.1	6.0	1.9	0.2	51.9	11.5	2.2	0.3	35.0	5.4	1.9	0.3
6-11 y	79.1	25.7	4.3	0.9	113	48.8	4.8	1.1	76.7	23.0	4.2	1.2
11-21 y	172	67.4	9.4	2.3	247	128	10.5	2.8	167	60.5	9.1	3.1
21-60 y	463	155	25.2	5.4	665	294	28.1	6.4	448	138	24.4	7.2
>60 y	1300	282	70.9	9.8	1880	538	79.2	11.6	1270	254	68.7	13.1

Table 5. 7. Total deposited dose of non-carcinogenic TEs bound to PM in different age categories at the three sites

	Total deposited dose of non-carcinogenic TEs (ng/d)											
	K1				K2				K3			
	$PM_{2.5}$		$PM_{0.1}$		$PM_{2.5}$		$PM_{0.1}$		$PM_{2.5}$		$PM_{0.1}$	
	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor	Indoor	Outdoor
SUMMER												
0-1 y	4260	361	63.1	7.8	3900	404	104	7.4	1680	148	72.9	5.5
1-3 y	6130	599	90.8	12.9	5610	671	149	12.2	2420	246	105	9.2
3-6 y	5170	680	76.7	14.7	4740	762	125	13.9	2040	279	88.6	10.4
6-11 y	5660	1450	83.9	31.4	5190	1630	137	29.6	2240	595	96.9	22.2
11-21 y	6720	2080	99.9	45.3	6150	2330	163	42.4	2650	852	115	31.8
21-60 y	9460	2500	140	54.1	8670	2800	230	51.0	3740	1020	162	38.2
>60 y	9340	1600	138	34.6	8550	1790	227	32.6	3690	655	160	24.5
WINTER												
0-1 y	5070	467	62.1	12.9	10100	1440	196	13.6	4300	426	149	13.6
1-3 y	7290	775	296	21.5	14600	2390	282	22.6	6190	707	215	22.5
3-6 y	6150	880	250	24.4	12300	2720	238	25.6	5230	802	181	25.6
6-11 y	6740	1880	274	52.1	13500	5790	260	54.7	5720	1710	198	54.6
11-21 y	7990	2690	325	74.6	15900	8300	309	78.3	6780	2450	235	78.2
21-60 y	11300	3240	458	89.6	22500	9980	435	94.1	9560	2950	332	94.0
>60 y	11100	2070	452	57.4	22200	6390	429	60.2	9440	1880	327	60.2

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 cumulative 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). Those studies showed that the cumulative 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 obtained for $PM_{0.1}$ and $PM_{2.5}$ for other age categories were lower than acceptable level, indicating that the exposure of these age groups to TEs bound to PM via the inhalation pathway is within the safe limit.

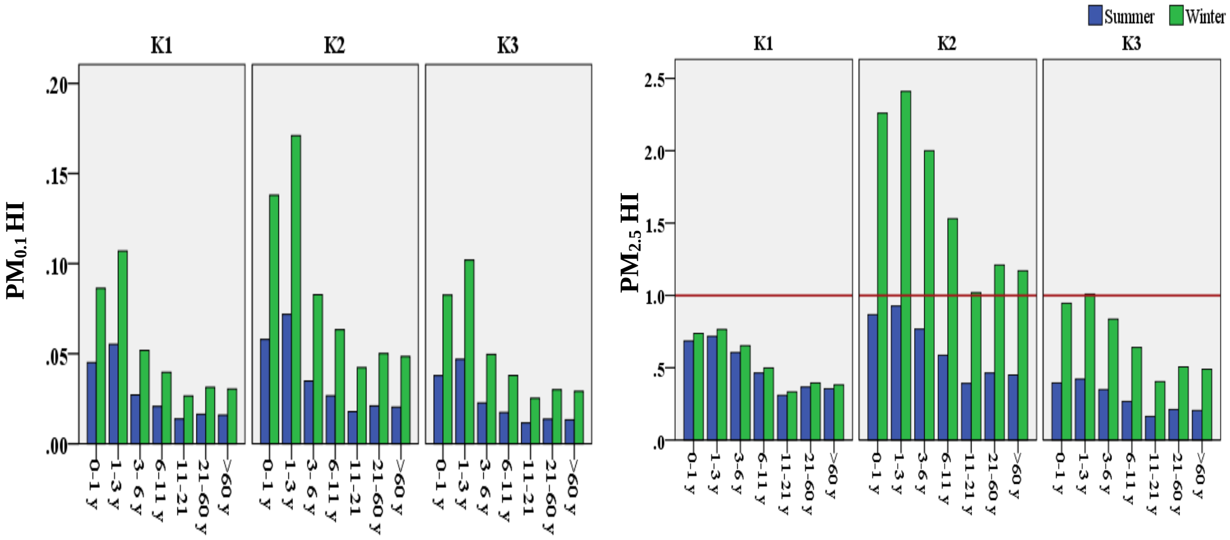


Fig 5. 7. Non-Carcinogenic risk induced by trace elements in different age groups categories

Among individual TEs, Mn was the greatest contributor to non-car TE risk, accounting for 83% to 92% for $PM_{2.5}$ and 68% to 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 reported 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 also an essential element in preventing metabolic diseases (Li, 2018).

3.6.2. Carcinogenic risk

The International Agency for Research on Cancer (IARC) has classified Cd, Cr, As, Ni, Co as carcinogenic substances due to their high toxicity raising the potential health damage. Fig 5.8 and Table 5.8 show the incremental life cancer risks (ILCR) estimated for car-TEs in $PM_{2.5}$ and $PM_{0.1}$ via inhalation pathway following seven age categories in summer and winter. In general, in the three houses, the average ILCRs in $PM_{2.5}$ were in the range of 2.3E-06 - 4.9E-05 in summer and 4.8E-06 -1.1E-04 in winter, whereas average ILCRs 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, respectively. 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 that the cumulative cancer risk for car-TEs be set at 1.0E-04 (one case per 10000 individuals for developing cancer over a human lifetime) as an acceptable level (Gao et al., 2017; US EPA, 2009). It was estimated that the ILCRs raised with the increase of age and were ordered as follows: Elderly group (>60) > (21-60 year) > (11 -21 year) > (6-11 year) > (3-6 year) > (1- 3 year) > (0-1 year). The highest ILCR was associated with the elderly group (>60 years) owing to lifetime exposure extension and was 3 to 22 times higher than other ages. Among children groups, especially in infants and toddlers, ILCRs were much lower than those of the adult groups, but the children are still growing and their organs are developing, so it is extremely necessary to protect them from chronic effects.

The cumulative risks of car TEs in $PM_{2.5}$ and $PM_{0.1}$ varied widely among the three houses. The occupants of the roadside dwelling were exposed to the highest risk, the occupants of the urban house had the lowest risk; these findings were consistent for all ages and both particle sizes and seasons. At the roadside house, elderly adults (>60 years) had a cumulative risk of 1.4E-04 in winter, which exceeded the US EPA threshold. This finding indicates that the probability of car risk is 1 to 2 people per 10000 individuals exposed to TEs in $PM_{2.5}$, which is an unacceptable level. Except for the above cases, the cumulative car risks for all age categories in our study were less than the acceptable level of 1.0E-04, thereby suggesting that the levels were safe for all ages at the three dwellings. It is well known that in addition to 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 cumulative risks were estimated for TEs in PM only. Therefore, the results of this study may have underestimated the cumulative risks because the synergistic effects of other chemical species were not taken into account (Sharma and Balasubramanian, 2018; Slezakova et al., 2014). Our findings were similar to those of a study conducted 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).

Interestingly, Cr and As were the most crucial contributors to the cumulative carcinogenic risks, which were responsible for approximately 90% of the total risks at all ages in the three houses. Similar results in previous studies also revealed that As and Cr appeared as the most carcinogenic substances in residential indoor environments in China (Wang et al., 2018), a university dormitory in China (Wang et al., 2019), and a hospital environment in Portugal

(Slezakova et al., 2014). The toxicity of chemicals depends strongly on the chemical speciation, the identification of which is vital for comprehensively evaluating the health risks. Our study estimated the human carcinogenic risk for Cr (VI) species instead of total Cr. Therefore, one-seventh of the determined total Cr concentration was used to estimate the concentration of Cr(VI) with a consumption ratio of 1:6 for Cr(III):Cr(VI) (Slezakova and Morais, 2014, US EPA, 2013). It is important to note that most of the ILCR of Cr (VI) and As bound in $PM_{2.5}$ exceeded the US EPA threshold of 1.0E-06, implying high cancer risk for all ages, whereas high cancer risk due to inhalation of Cr(VI) and As in $PM_{0.1}$ were only observed in the elderly group (>60 y). These risks may be underestimated or overestimated due to additional exposure factors, such as toxicities of other components of PM, lifestyle habits, smoking habits, diet, outdoor exposure, and uncertain outputs. These initial results of our study therefore highlighted the urgent concern about health risks associated with indoor PM exposure in an urban area in Vietnam.

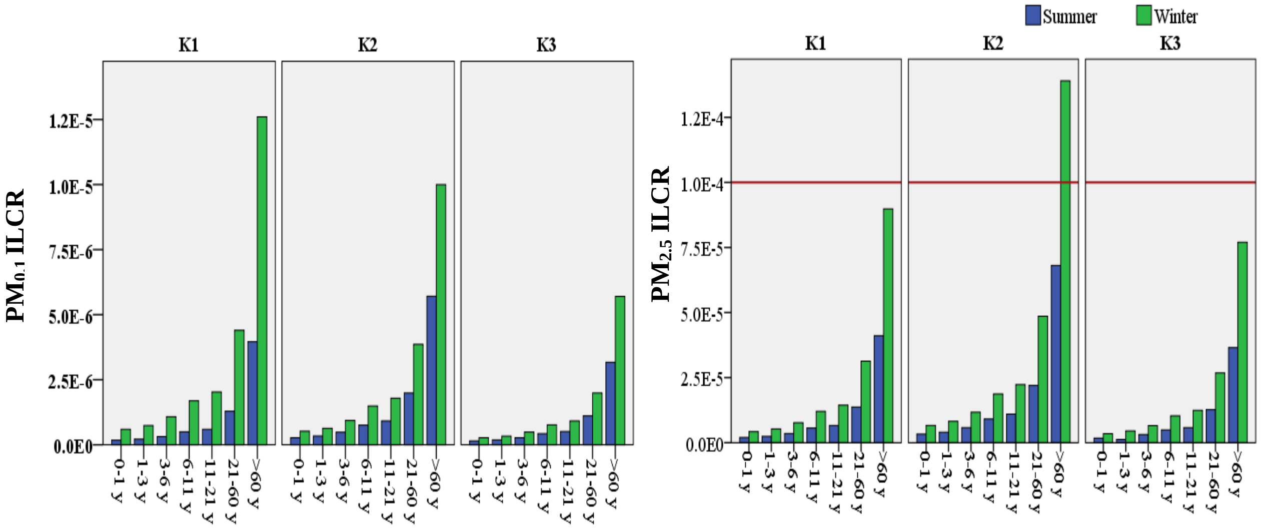


Fig 5.8. Carcinogenic risk induced by trace elements in different age groups categories

3.6.3. Sensitivity analysis and limitations

Carcinogenic risks are markedly affected by several exposure factors are used as input (C, ET, IR, EF, ED, AT, BW). Generally, the mean values of the input variables are used to estimate the single average risk value, which may result in some uncertainty. Monte Carlo simulation was used in our study to determine the probability density functions assigned to each variable. The simulation results for carcinogenic risks are summarized in Table 5.8. The probabilities of lifetime cancer risk for TEs in $PM_{0.1}$ and $PM_{2.5}$ were significantly higher in winter than in summer; for example, 90% of ILCR estimates for $PM_{0.1}$ and $PM_{2.5}$ ranged from 2.9E-07 to 9.0E-06 and 3.2E-06 to 6.8E-05 in summer, respectively, compared to the corresponding values of 6.6E-07 to 1.4E-05 and 6.5E-06 to 1.4E-04 in winter, respectively. Sensitivity analysis identified that the most influencing factor was TEs concentration, which contributed approximately 85% to 96% of the variance in ILCR (Fig. 5.9 and Fig. 5.10). The contributions of BW, EF, ED, AT, IR were negligible, ranging from approximately 1% to 6%, and BW and AT presented negative values.

4. CONCLUSIONS

A comprehensive study of indoor PM in urban areas was conducted in Hanoi in summer and winter. High levels of indoor $PM_{0.1}$ and $PM_{2.5}$ were found in the city, where the average concentration of indoor $PM_{2.5}$ is about four times higher than WHO guideline. Seasonal variation was observed for indoor $PM_{2.5}$, but not in 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, and As exceeded the respective EU limits. Such high levels have a high potential health risk and present a significant disease burden to individuals who are exposed to TEs bound to PM.

Source apportionment analysis indicated that indoor $PM_{2.5}$ was derived mainly 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 HRT varied largely with particle size. The majority of TEs bound to $PM_{2.5}$ was deposited in the HA region, whereas the dominant proportion of TEs bound to $PM_{0.1}$ was deposited in the AL region. The EDI in the HA region decreased as particle size decreased, and increased in the TB and AL regions as particle size decreased. Among the three houses investigated, the occupants of the roadside house were exposed to the highest risk. The intake of TEs in $PM_{2.5}$ at the house were sufficiently high to pose a high car risk to the occupants over 60 years old and the non-car risks were unacceptable for all ages at the roadside house in winter. Except for the above cases, 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.

Table 5. 8. ILCR values estimated by Monte Carlo simulations of cancer risk for TEs bound to PM inhalation in the three houses

Sites		$PM_{2.5}$						$PM_{0.1}$					
		Summer			Winter			Summer			Winter		
	Ages	10%	50%	90%	10%	50%	90%	10%	50%	90%	10%	50%	90%
K1	0-1 y	1.3E-06	1.8E-06	2.8E-06	2.7E-06	3.9E-06	6.1E-06	1.1E-07	1.6E-07	2.6E-07	4.4E-07	5.7E-07	7.7E-07
	1-3 y	1.5E-06	2.2E-06	3.5E-06	3.3E-06	4.9E-06	7.6E-06	1.3E-07	2.0E-07	3.2E-07	5.3E-07	7.2E-07	9.9E-05
	3- 6 y	2.2E-06	3.2E-06	5.1E-06	4.8E-06	7.2E-06	1.1E-06	1.9E-07	2.9E-07	4.6E-07	7.6E-07	1.0E-06	1.4E-06
	6 -11 y	3.5E-06	5.2E-06	8.1E-06	7.7E-06	1.1E-05	1.7E-05	3.0E-07	4.6E-07	7.3E-07	1.2E-06	1.6E-06	2.2E-06
	11-21 y	4.2E-06	6.2E-06	9.5E-06	9.3E-06	1.4E-05	2.1E-06	3.6E-07	5.6E-07	8.7E-07	1.5E-06	1.9E-06	2.6E-06
	21-60 y	8.6E-06	1.3E-05	1.9E-05	2.0E-05	2.9E-05	4.5E-05	7.9E-07	1.2E-06	1.E-06	3.2E-06	4.3E-06	5.7E-06
	>60 y	2.6E-05	3.8E-05	5.9E-05	5.8E-05	8.4E-05	1.3E-04	2.2E-06	3.5E-06	5.4E-06	9.2E-06	1.2E-05	1.6E-05
K2	0-1 y	2.2E-06	3.7E-06	4.5E-06	4.7E-06	6.4E-06	8.7E-06	1.5E-07	2.4E-07	4.2E-07	2.9E-07	4.8E-07	8.0E-07
	1-3 y	2.6E-06	3.8E-06	5.6E-06	5.4E-06	7.8E-06	1.2E-05	1.8E-07	3.1E-07	5.3E-07	3.3E-07	5.7E-07	1.0E-06
	3- 6 y	3.8E-06	5.5E-06	8.1E-06	7.7E-06	1.1E-05	1.6E-05	2.6E-07	4.4E-07	7.6E-07	4.9E-07	8.4E-07	1.5E-06
	6 -11 y	6.0E-06	8.6E-06	1.3E-05	1.2E-05	1.8E-05	2.6E-05	4.2E-07	6.8E-07	1.2E-06	7.8E-07	1.3E-06	2.3E-06
	11-21 y	7.4E-06	1.1E-05	1.5E-05	1.5E-05	2.1E-05	3.1E-05	5.1E-07	8.3E-07	1.4E-06	9.4E-07	1.6E-06	2.8E-06
	21-60 y	1.5E-05	2.1E-05	3.1E-05	3.2E-05	4.6E-05	6.7E-05	1.1E-06	1.8E-06	3.1E-06	2.0E-06	3.5E-06	6.1E-06
	>60 y	4.6E-05	6.5E-05	9.4E-05	9.2E-05	1.3E-04	1.9E-04	3.2E-06	5.2E-06	8.9E-06	5.9E-06	1.0E-05	1.7E-05
K3	0- 1 y	1.1E-07	1.6E-06	2.5E-06	2.4E-06	3.3E-06	4.6E-06	9.8E-08	1.4E-07	2.1E-07	1.6E-07	2.5E-07	4.0E-07
	1- 3 y	1.3E-06	2.0E-06	3.1E-06	3.2E-06	4.3E-06	6.1E-06	1.2E-07	1.7E-07	2.6E-07	1.9E-07	3.1E-07	5.0E-07
	3- 6 y	1.9E-06	2.9E-06	4.5E-06	4.6E-06	6.3E-06	8.8E-06	1.7E-07	2.5E-07	3.8E-07	2.9E-07	4.5E-07	7.3E-07
	6 -11 y	3.1E-06	4.6E-06	7.0E-06	7.3E-06	9.9E-06	1.4E-05	2.7E-07	4.0E-07	6.1E-07	4.5E-07	7.1E-07	1.2E-06
	11-21 y	3.7E-06	5.5E-06	8.4E-06	8.9E-06	1.2E-05	1.6E-05	3.3E-07	4.6E-07	7.2E-07	5.5E-07	8.5E-07	1.4E-06
	21-60 y	8.1E-06	1.2E-05	1.8E-05	1.9E-05	2.6E-05	3.6E-05	7.2E-07	1.1E-06	1.5E-06	1.2E-06	1.8E-06	2.9E-06
	>60 y	2.3E-05	3.4E-05	5.2E-05	5.5E-05	7.4E-05	1.0E-04	2.0E0-6	3.0E-06	4.4E-06	3.4E-06	5.3E-06	8.5E-06

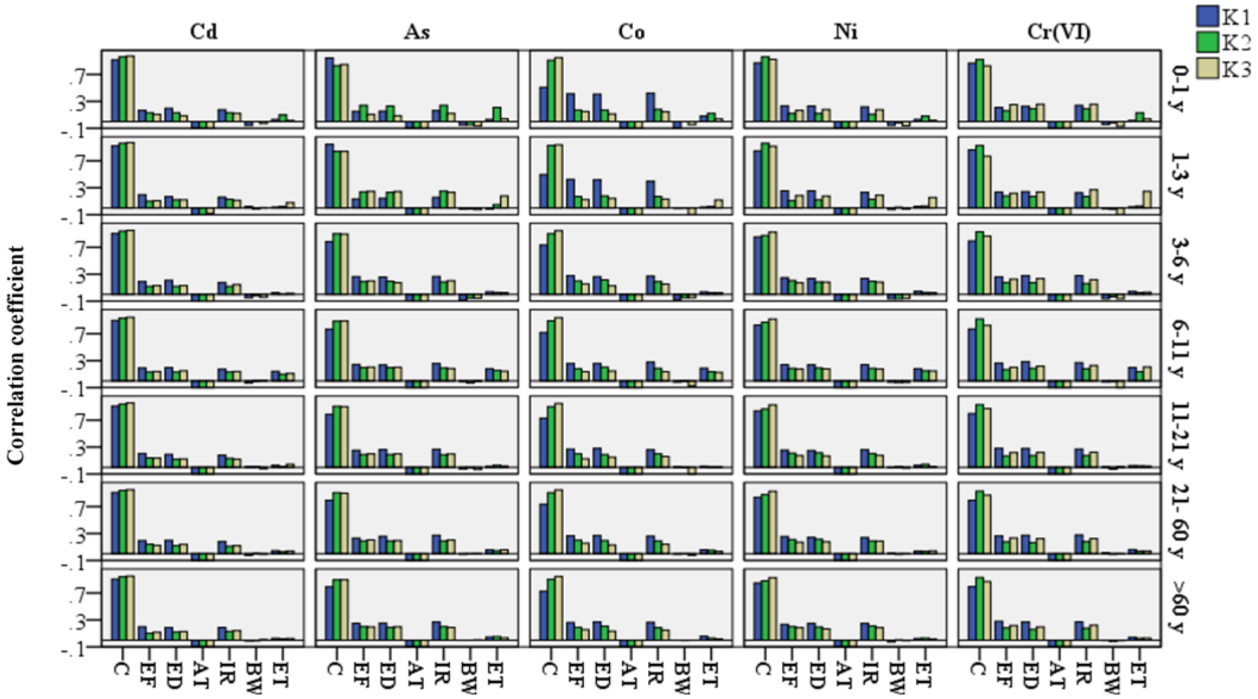


Fig 5. 9. Sensitivity analysis of $PM_{0.1}$ in the three houses

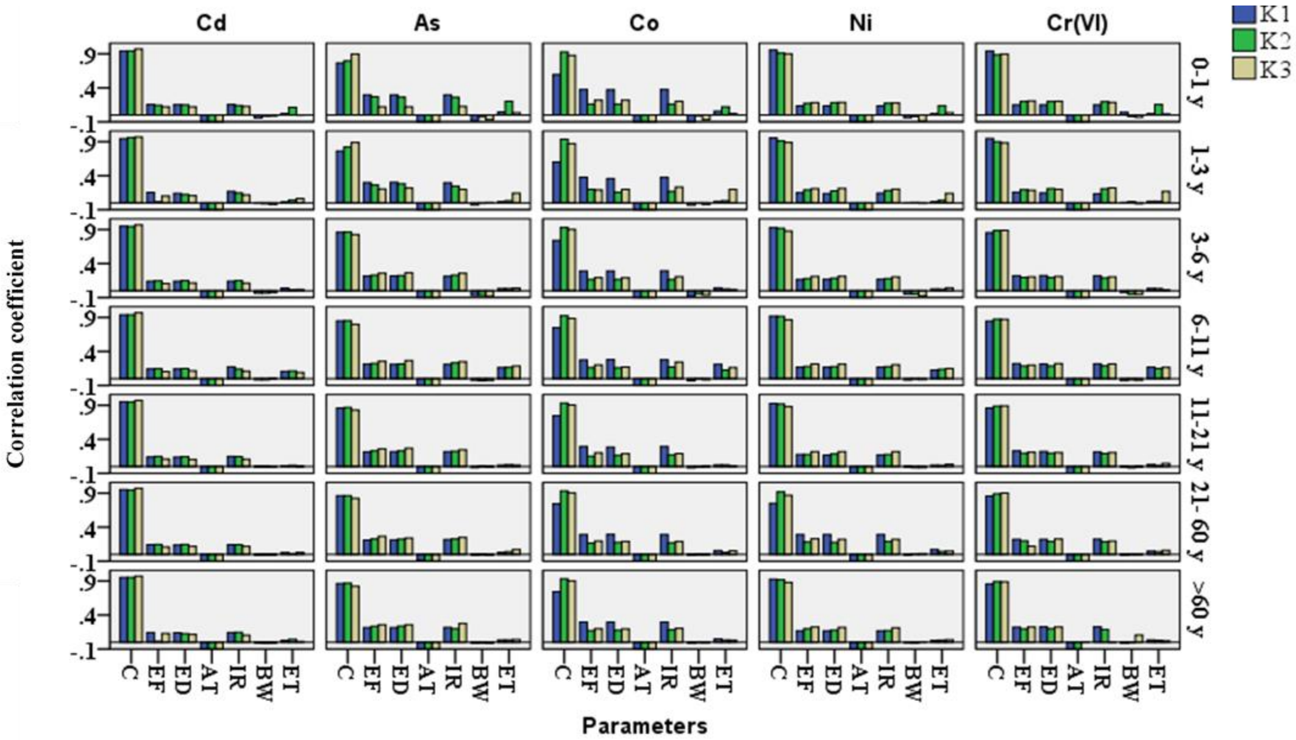


Fig 5.10. Sensitivity analysis of $PM_{2.5}$ in the three houses

CHAPTER 6

CHARACTERIZATION OF PARTICULATE POLYCYCLIC AROMATIC HYDROCARBONS IN INDOOR AND OUTDOOR AIR AND HEALTH IMPLICATION

1. INTRODUCTION

Household air pollution (HAP) has become one of biggest environmental causes of ill health in the world, with approximately 4 million premature deaths in 2016 (7.7% of global mortality) attributed to inefficiently combusted particulate matter (PM) derived from cooking, mainly in populous, low- and middle-income countries (WHO, 2016). Household air pollutants are typically derived from incomplete combustion of solid fuels, and other biomass used for cooking and heating, particularly in rural areas of developing countries (WHO, 2014). Household air pollutants contain large quantities of PM, polycyclic aromatic hydrocarbons (PAHs), Carbon monoxide (CO), and other compounds that have adverse health impacts (Sharma and Jain, 2020; Vo et al., 2020c; Yury et al., 2018). Coal briquettes and charcoal are widely used in households in Vietnam, both in the past and today. A statistical survey by the Hanoi Department of Environment and Resources (Hanoi Donre) in 2017 showed that Hanoi consumed about 528.2 tons of coal briquettes/charcoal per day and that there were approximately 55,000 coal briquette/charcoal stoves in use. Of these stoves, 63% were used for domestic cooking purposes by low-income households in the inner city (Donre, 2017). The combustion of biomass and coal by households is considered to be one of the dominant sources of indoor air pollution in the country. This combustion results in the release of numerous pollutants, such as particulate matter (PM) and other toxic chemical compounds (CO, PAHs and heavy metals) and contributes significantly to the disease burden in the region (Donre, 2017.; Vo et al., 2020c)

Numerous scientific studies have shown that exposure to PM can have severe health outcomes, including premature death due to cardiovascular diseases or lung cancer, increased morbidity incidence liked due to respiratory diseases, asthma, respiratory symptoms and other ailments (Kim et al., 2015). PM containing toxic chemicals (PAHs and heavy metals) cause more threat to human health (Kim et al., 2013; Kim et al., 2015). Some species of PAHs are potentially carcinogenic and mutagenic, and PAHs bound to fine and ultrafine particles are even more harmful and play a governing role in adverse effects onto human health (Kim et al., 2013). Lin et al. (2013) reported that the chemical composition and indoor particles can induce inflammation and endothelial dysfunction human coronary artery endothelial cells. Small particles (i.e., $PM_{0.1-1}$) increased the cytotoxicity, and naphthalene (Nap) of $PM_{0.1-1}$ was highly correlated with cytokine production and nitric oxide reduction (NO). The high incidence of lung cancer in residential houses in Hangzhou, China, has been attributed to the inhalation PAHs bound to PM (Zhu et al., 2009).

The presence of particulate PAHs in the indoor environment has been linked to a mixture of sources (indoor and outdoor sources). indoor sources of PAHs have been reported to be cooking (Abdullahi et al., 2013), domestic heating (Han et al., 2015), indoor combustion (i.e., incense and candles) (Derudi et al., 2012), the evaporation of materials (Krugly et al., 2014), tobacco smoking (Zhu et al., 2009) and electronic devices (Cai et al., 2018). The penetration of particulate PAHs into buildings through windows/doors, cracks and ventilation systems also contributes significantly to indoor PAHs (Chen et al., 2017). Outdoor sources derived from vehicle emissions (Anh et al., 2019; Krugly et al., 2014; Zhu et al., 2009), industrial emissions (Anh et al., 2019) and biomass combustion (Pham et al., 2019; Sari Giannis et al., 2015).

Several publications have examined PAH bound to different-sized particles in different indoor environments globally. For example, Zhang et al. (2019) and Chen et al. (2016) examined the distribution of PAHs bound to PM in rural areas in China and found that PAH contents depend on the size of particles. Liaud et al. (2014) and Liaud et al. (2021) reported that PAHs preferentially bind to fine particles (especially PM_1). Dat and Chang (2017) also demonstrated that lighter (i.e., low molecular weight) PAHs (2 to 4 rings) bind preferentially to coarse PM, while heavier (i.e., high molecular weight) PAHs (5 to 6 rings) which are typically more toxic, mutagenic and carcinogenic PAHs, bind preferentially to fine PM ($PM_{2.5}$ and PM_1).

Numerous studies have shown that PAHs bind preferentially to fine and finer particles which penetrate deeply into pulmonary alveoli as the studies of Liaud et al. (2014) and Liaud et al. (2021) However, few publications have described the deposited dose distribution of PM-bound PAHs in the human respiratory tract (HRT), especially in the PAHs bound to ultrafine particles (particles with a diameter less than 0.1 μm) in indoor environments where the majority of our time is spent. Indeed, there is lack of open data related to ultrafine particles-bound PAHs associated with health risk assessments. Kawanaka et al.(2004) indicated that higher PAH content and increased mutagenicity were observed for ultrafine particles compared to accumulation mode particles (particles with a diameter between 0.1 and 1.8 μm) in the atmosphere, not in the indoor environment. Ultrafine particles were significant contributors to PAH deposition in the lung, despite low PM mass. In Vietnam, there is no available data on particulate PAHs in indoor environments or health risk assessments, except for publications that only investigated the concentrations of PAHs bound to $PM_{2.5}$ in the school environment (Vo et al., 2020b) Therefore, *the present study was performed to determine the content of PAHs bound to fine and ultrafine particles indoors, identify the possible sources, and estimate the contribution of indoor particles to PAH deposition in the lung in order to access human health.*

2. METHODOLOGY

2.1. Sampling methodology

Sampling was conducted in three residential dwellings (K1, K2 and K3) in Hanoi, the capital of Vietnam, which has a population of more than 7.5 million (GSO, 2018). The characteristics of the three houses were described in Table 1 and the layouts are shown in Chapter 5 and Fig. 5.1 (a,b,c) in the appendix. The direct distance between these sampling sites was approximately

5–10 km. The locations of the three dwellings are shown in Fig. 5.1 in Chapter 5. The sampling methodology was described in Sections 2.1 and 2.2 in Chapter 5.

2.2 Sample Preparation

A half of sampled filter was cut into small pieces and placed in a glass centrifuge tube (15 mL), spiked with 20 µL (10 ppb) of an internal standard (IS), and then incubated at room temperature for at least 60 mins to equilibrate. The PM was extracted from the filter by sonication using an ultrasonic probe (VCX 130 PB, Sonics, Connecticut, USA) in a 10 mL mixture of acetone/hexane (1/1, v/v) for 5 minutes. The extraction procedure was repeated three times and the final extracts were evaporated to 0.5 mL under a gentle nitrogen stream and exchanged with a hexane solvent. The extracts were then cleaned by solid-phase extraction (Bond Elut SI- Agilent). The Bond Elut SI cartridges were conditioned using a mixture of MeOH and hexane at a flowrate of 3 mL/min, before loading with the extracts. The extracts were eluted using a 12 mL mixture of hexane/dichloro-methane (1/1: v/v), concentrated under a gentle nitrogen stream until almost dry, and then refilled with 1 mL hexane. The solutions were PAH compounds by gas chromatography (GC Trace 1310, Thermo Scientific Inc., USA) including a TriPlus RSH liquid autosampler coupled with an electron impact ionization-tandem mass spectrometry (Model TSQ 8000, Thermo Scientific Inc., USA) and a Thermo DB-5MS capillary column (0.25 mm internal diameter × 30 m length × 0.25 µm film thickness, 5% methylphenyl poly-siloxane stationary phase). High-purity helium (99.999%) was used as the carrier gas and the mass spectrometer was operated in selected reaction monitoring mode (SRM). Individual PAH species were quantified including naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[a]pyrene (BaP), indeno(1,2,3-cd) pyrene (Ind), dibenz[a,h]anthracene (DahA) and benzo[g,h,i]perylene (BghiP).

2.3. Quality Assurance and Quality Control

The sampling, sample preparation, and analysis were performed according to good laboratory management practices. Laboratory blanks, sampling blanks, and international sediment exchange for tests on organic contaminants samples (proficiency testing sample: PT sample) were performed following the same procedure as mentioned in section 2.2 with real samples. PT samples were prepared to determine the recovery efficiency of investigated individual PAHs (n=3). The recovery efficiencies of individual PAHs ranged from 78% (BaA) to 107% (Acy). The recovery of ultrasonic-assisted extraction and clean-up fell within an acceptable range according to AOAC international guidelines.

The internal standards used in this study included naphthalene-D8 (Nap-D8), acenaphthene-D10 (Ace-D10), phenanthrene-D10 (Phe-D10), chrysene-D12 (Chr-D12), and perylene-D12 (Per-D12) (Code: DRE-YA08273300TO) (EPA method 8270; internal standard mixture: 2000 µg m/L in dichloromethane). Proficiency testing of samples (International Sediment Exchange for Tests on Organic Contaminants) was performed through Wageningen Evaluating Programs for Analytical Laboratories (Netherlands). All solvents, such as hexane, acetone, dichloromethane (DCM), and methanol (MeOH) were high-purity grade (Merck Inc.,Singapore). A Bond Elut SI-Solid Phase Extraction (SPE) system (Agilent Inc., United States) was used for cleaning. In addition, the sample and laboratory blanks were analyzed for every 10 samples. The of analytical signals was assessed by measuring a 2000 ng m/L standard solution at intra- and inter-day intervals, and then calculating the relative standard deviation of the analytical signals. The limit of detection (LOD) was estimated as 3 times the signal-to-noise ratio obtained from lowest matrix-matched samples. Meanwhile, the method limit of quantification (LOQ) was estimated as 10 times the signal to noise at the lowest concentration matrix-matched samples (Vu-Duc et al., 2021). LODs values ranged from 0.01 ng/m^3 to 0.05 ng/m^3, whereas LOQ values varied from 0.02 to 0.18 ng/m^3. The recovery efficiencies varied from 78% to 107%. These detailed values are shown in Table 6.1 and 6.2 in the appendix. Data from GC-MS/MS were processed by Thermo Xcalibur software (ver. 4.0, Thermo Scientific Inc.).

2.4. Source apportionment

In this study, diagnostic ratios (DR) and multivariate principal component analysis (PCA) were used for identifying the possible sources of PAHs bound to $PM_{2.5}$ and $PM_{0.1}$ by using IBM statistical software. Principle components (PCs) with eigenvalues greater than 1 were defined, and their contributions to the total variance were estimated after applying the varimax rotation. Variables with high factor loadings are considered relevant and indicate possible emission sources.

2.5. Health risk assessment

2.5.1. Exposure assessment

The deposition efficiency and deposited doses (EDI) of inhaled PAHs in the human respiratory tract (HRT) were estimated for different age categories under normal nasal breathing conditions (seated) in indoor environments using the International Commission on Radiological Protection (ICRP) model. This model is a semi-empirical model which determines the deposition fraction (DF) of particles in five regions of the airway system (anterior nasal region (ET1), main extra-thoracic region (ET2), bronchial region (BB), bronchiolar region (bb), and alveolar interstitial region (AL) (Gao et al., 2017; ICRP, 1994; Kawanaka et al., 2009; Zhang et al., 2012) based on numerical fitting of experimental data and theory. The major mechanisms of PM deposition throughout the HRT include diffusion, sedimentation, and impaction (ICRP, 1994)**.** 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 DF of particles is calculated by two kinds of deposition processes known as aerodynamic and thermodynamic transport. Each anatomical region can be represented by one or more filters (ith filters) in series.

According to ICRP (1994), synchronous thermodynamic and aerodynamic transport is significant only for particles with size range of 0.1 m (thermodynamic diameter) to 1 m aerodynamic diameter. The small particles were governed mainly by thermodynamic transport, whereas the larger particles were controlled by aerodynamic transport. The deposition efficiencies and DFs in the *i^th^* filter is given by the following equations:

$$\eta = \eta_{th}^{2} + \eta_{ae}^{2} \quad (1)$$

$$DF_i = \eta_i \emptyset_i \pi_{i=0}^{i}(1 - \eta_i) \quad (2)$$

where, η^{th} and η^{ae} are the deposition efficiencies determined by thermodynamic and aerodynamic transport; ηi is the filtration efficiency of the i^th^ filter and $\emptyset_i$ is the fraction of tidal air that reaches the i^th^ filter on inhalation. In the ICRP model, each region of the HRT is represented by an equivalent particle filter, as shown in Fig. 6.1. In terms of regional lung deposition, its efficiency (η) is expressed in terms of three parameters: *a*, *R* and *p*, which were recommended by ICPR (1994) and are shown in Table 6.1.

The total EDI of $\sum_{15}$PAHs or $\sum BaP_{(eq)}$ (toxic equivalent factor-TEF) in the form of $PM_{0.1}$ and $PM_{2.5}$ in the HRT is the sum of the EDI in the five regions, which can estimated using the following US EPA model (US EPA, 1989; US EPA, 2009) (Eq. 3):

$$EDI = \sum_{i=1}^{5} \sum_{j=1}^{n} \frac{DF_i \times C_j \times IR \times ET \times ED \times EF}{24 \times AT} \quad (3)$$

where, *EDI* is the total deposited doses of $\sum_{15}$PAHs or $\sum BaP_{(eq)}$ (ng/d); DF_i is the particle deposition fraction of region *i* in the HRT (ET1, ET2, BB, bb, and AL) or the filters *i*; C_j is the concentration of PAH species *j* (ng/m^3) or $BaP_{(eq)j}$; *j*=1-*n*, where *n* is the number of PAHs; *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. (1–2) and Table 6.1.

2.5.2. Mutagenicity and carcinogenicity of PAHs

The toxic equivalent factor (TEF) as total $BaP_{(eq)}$ ($\sum BaP_{eq}$) and mutagenic equivalent factor (MEF), which reflects the carcinogenic and mutagenic potency of an individual PAH_i to BaP have been used to evaluate the carcinogenic and mutagenic potency of PAHs, respectively (Błaszczyk et al., 2017b; Delgado-Saborit et al., 2011; Vo et al., 2020a). Calculations of the carcinogenic equivalent (TEQ) and mutagenic equivalent (MEQ) for the individual PAHs are presented in Eqs. (4–5)

$$MEQ = \sum_{j}^{n} PAH_j \times MEF_j \quad (4)$$

$$TEQ = \sum_{j}^{n} PAH_j \times TEF_j = \sum_{j}^{n} BaP_{(eq)} \quad (5)$$

where MEF_j and TEF_j are the mutagenic and toxic equivalent factors for individual PAHs. The contribution of PAHs to total carcinogenicity and mutagenicity was calculated by the carcinogenic potential and mutagenic potential. A total of 15 PAHs or 8 PAHs were employed to calculate carcinogenic potential (CP) or mutagenic potential (MP), respectively, using the following equations (6-7) (Błaszczyk et al., 2017b; Delgado-Saborit et al., 2011; Vo et al., 2020b).

$$CP_j = \frac{\frac{PAH_{ij}}{BaP} \times TEF_j}{\sum_j^n \frac{PAH_j}{BaP} \times TEF_j} \times 100\% \quad (6)$$

$$MP_j = \frac{\frac{PAH_j}{BaP} \times MEF_j}{\sum_j^n \frac{PAH_j}{BaP} \times MEF_j} \times 100\% \quad (7)$$

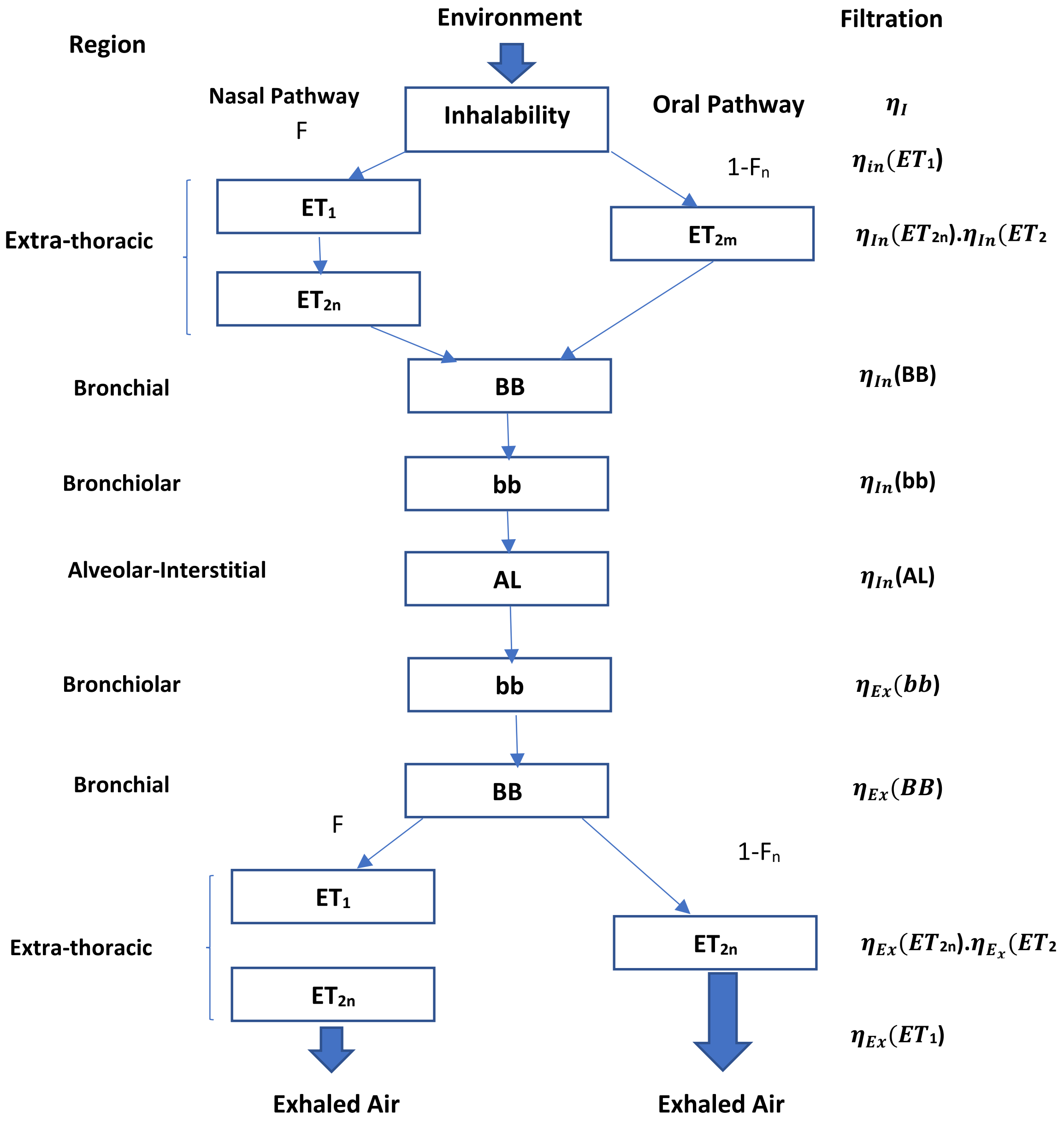


Fig.6. 1. Empirical representation of the inhalability of particles and their deposition in the extrathoracic (ET), bronchial (BB), bronchiolar (bb), and alveolar regions (AL) of the respiratory tract during continuous cyclic breathing by transport through a series of filters (ICRP, 1994)

2.5.3. Estimation of carcinogenic risk

The 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 car risk, the incremental lifetime cancer risk (ILCR) is quantified as the incremental cancer probability for an individual over their lifetime owing to potential exposure to car substances, and is calculated using:

$$ILCR = \sum_{j=1}^{n} ILCR_j = \frac{EDI_j \times CSF}{BW} \quad (8)$$

where BW is the body weight (kg), and CSF is the cancer slope factor for BaP [mg/(kg·d)]. To estimate the EDI and ILCR, variables including C_j, ET, and BW were determined from sampling and questionnaires; other variables, such as IR, EF, ED, AT, and CSF were referenced from the US EPA exposure handbook (US EPA, 2011). Details of the data sources for the calculation are shown in Table 5.3 in Chapter 5. For public health protection from PAHs, an ILCR value $\leq 10^{-6}$ 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 $>10^{-4}$ (Chen et al., 2017; Gao et al., 2017; US EPA, 2009).

2.6. Questionnaire method

A total of 500 online and offline questionnaires were distributed to the households in Hanoi to gather basic household information, e.g., daily indoor activities, characteristics of houses, and exposure parameters (body weight, age, time spent in indoor air and outdoor air). These data, which are summarized in Table 5.3 in Chapter 5, were used to estimate health risk.

2.7. Statistical analysis

Statistical analysis was performed using SPSS Statistics 20 software. The normal distribution was tested using the Shapiro–Wilk test. 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. Cumulative distribution functions were determined using a Monte Carlo simulation with 100,000 iterations run using the Oracle Crystal Ball software (Version 11.1.2.4.850, Oracle Inc., USA).

Table 6. 1. Recommended parameters for substitution in the ICRP model of regional deposition (ICRP, 1994)

Phase	Filter, j	Region	Regional deposition efficiency, η_j						Volumetric Fraction $\theta_j^{a,b}$
			Aerodynamic			Thermodynamic			
			η_{ae}= 1− exp $(-aPR^p)$			η_{th}= 1− exp $(-aPR^p)$			
			A	R	p	a	R	p	
Inhalation	1	ET_1	3.0×10^{-4}	$d_{ae}^2 V_n SF_t^3$	1	18	$D(V_n\ SF_t)^{-1/4}$	½	1
	2	ET_2	5.5×10^{-5}	$d_{ae}^2 V_n SF_t^3$	1.17	15.1	$D(V_n\ SF_t)^{-1/4}$	0.538	1
	3	BB	4.08×10^{-6}	$d_{ae}^2 V_n SF_t^{2.3}$	1.152	$22.02 SF_t^{1.24} \psi_{th}$	Dt_B	0.6391	$1-\frac{V_D(ET)}{V_T}$
	4	bb	0.1147	$(0.056+t_b^{1.5})\, d_{ae}^{t_b^{-0.25}}$	1.173	$-76.8+167SF_b^{0.65}$	Dt_b	0.5676	$1-\frac{[V_D(ET)+V_D(BB)]}{V_T}$
Exhalation	5	AL	$0.146SF_A^{0.98}$	$d_{ae}^2 t_A$	0.6495	$170+103SF_A^{2.13}$	Dt_A	0.6101	$1-\frac{[V_D(ET)+V_D(BB)+V_D(bb)]}{V_T}$
	6	bb	0.1147	$(0.056+t_b^{1.5})\, d_{ae}^{t_b^{-0.25}}$	1.173	$-76.8+167SF_b^{0.65}$	Dt_b	0.5676	$1-\frac{[V_D(ET)+V_D(BB)]}{V_T}$
	7	BB	2.04×10^{-6}	$d_{ae}^2 V_n SF_t^{2.3}$	1.152	$22.02 SF_t^{1.24} \psi_{th}$	Dt_B	0.6391	$1-\frac{V_D(ET)}{V_T}$
	8	ET_2	5.5×10^{-5}	$d_{ae}^2 V_n SF_t^3$	1.17	15.1	$D(V_n\ SF_t)^{-1/4}$	0.538	1
	9	ET_1	3.0×10^{-4}	$d_{ae}^2 V_n SF_t^3$	1	18	$D(V_n\ SF_t)^{-1/4}$	½	1

Note: (a)η_{ae}= $0.5[1-1/\exp(-aPR^p)]$ and η_{th}= $0.5[1-1/\exp(-aPR^p)]$; (b)η_{ae}= $[1-1/\exp(-aPR^p)]$ and η_{th}= $0.5[1-1/\exp(-aPR^p)$

$$\psi_{th} = 1 + 100\exp\left[-\left(\left(log_{10}\left(100+\frac{10}{d_{th}^{0.9}}\right)\right)^2\right)\right]$$

d_{ae}, d_{th} are the aerodynamic and thermodynamic diameters of a particle (μm). V_n is the total volumetric flow rate (mL/s). V_D(ET), $V_D(BB)$, $V_D(bb)$ are the anatomical and dead spaces of ET, BB and bb (L). SF_t, SF_b, SF_a are scaling parameters for different subjects; D: diffusion coefficient; t_B, t_b, t_A are the time constant for conduction of air though BB, bb and AL; ψ_{th} is an empirical correction factor for enhancement of thermodynamic deposition.

3. RESULTS AND DISCUSSION

3.1. PAH bound to $PM_{2.5}$ and $PM_{0.1}$ in indoor air

3.1.1. Concentrations of particulate PAH in indoor air

Tables 6.2 and 6.3 show a statistical summary of concentrations of the average sum of 15 PAHs ($\sum_{15}$PAHs) and individual PAHs in indoor PAHs bound to $PM_{0.1}$ and $PM_{2.5}$ during summer and winter. Indeed, the combustible and carcinogenic individual PAHs_in 15PAHs were also estimated in both fractions. Regardless of investigated houses during the whole sampling period, the $\sum_{15}$PAH concentrations ranged from 102.9 to 155.6 ng/m^3, and 25.3 to 52.9 ng/m^3 for indoor $PM_{2.5}$ and $PM_{0.1}$, respectively. The sum of the combustible PAH concentrations ($\sum$com-PAHs) ranged from 28.7 to 49.7 ng/m^3 and 10.9 to 27.3 ng/m^3 for indoor $PM_{2.5}$ and $PM_{0.1}$, respectively, whereas $\sum$the sum of carcinogenic PAH concentrations ($\sum$Car-PAHs) ranged from 16.7 to 38.9 ng/m^3; 5.5 to 18.8 ng/m^3. $\sum$com-PAHs accounted for 29.9±2.5% and 37.9±7% of $\sum_{15}$PAHs for indoor $PM_{2.5}$ and $PM_{0.1}$, respectively; while $\sum$car-PAHs were responsible for 21.3±2% and 22.7±3.5% of $PM_{2.5}$ and $PM_{0.1}$, respectively. Compared to $PM_{2.5}$, the proportions of com-PAHs/$\sum_{15}$PAH and $\sum$car-PAHs/$\sum_{15}$PAH were higher in $PM_{0.1}$. These results support the hypothesis that these percentages increased with a decrease in particle size, which was also observed by Hassanvand et al. (2015) who reported that car-PAHs accounted for approximately 35% of $\sum_{16}$PAHs in PM_{10} (0.2% to 0.4% of PM_{10} mass), 37% to 43% of $\sum_{16}$PAHs in $PM_{2.5}$ (0.5 to 0.6% of $PM_{2.5}$ mass), and 49% to 56% of $\sum_{16}$PAHs in PM_1 (0.6% to 0.8% PM_1 mass). Moreover, in this study, the content of $\sum_{15}$PAHs/PM mass was in the range of 0.1% to 0.3% and 0.3% to 0.7% for $PM_{2.5}$ and $PM_{0.1}$, respectively. Although the concentrations of total PAHs bound to $PM_{2.5}$ were greater than those bound to $PM_{0.1}$, $\sum_{15}$PAHs/PM was higher in $PM_{0.1.}$ This is implied that the PAHs bound to $PM_{0.1}$ were more enriched than those bound to larger PM ($PM_{2.5}$), which was also observed in previous studies (Lin et al., 2013; Liaud et al., 2014). Lin et al. (2013) reported that the content of $\sum$PAHs/$PM_{0.1}$ (1%) was higher than those of PAH/$PM_{1\text{-}10}$ (0.2%) and PAH/$PM_{0.1\text{-}1}$ (0.3%), whereas particles the level of PAH bound to $PM_{0.1\text{-}1}$ was higher than those of PAH bound to $PM_{0.1}$ and to $PM_{1\text{-}10.}$ It is worth to underline that $\sum$PAHs/$PM_{0.1}$ (1%) in this study was significantly higher than our results.

In addition, Liaud et al. (2014) reported that a greater concentration of $\sum$PAHs and content of PAHs (PAH/PM mass) were found in PM_1 compared to those of $\sum$PAHs and content of PAHs of TSP, PM_{10} and $PM_{2.5}$ in indoor environments in France. Thus, our study supports the hypothesis that, the smaller particles and relatively higher PAH content of $PM_{0.1}$ make them more adverse health effects. In this study, indoor $PM_{0.1}$ accounted for 15.8±3.6% of indoor $PM_{2.5}$ mass (in range of 8.6% to 18.3%), whereas indoor PAHs bound to $PM_{0.1}$ made up 35.5±7.6% of PAHs bound to $PM_{2.5}$ (in range of 26.1% to 45.9%).

Table 6. 2. Mean concentrations (ng/m^3) and standard derivation (SD) of individual PAHs_in indoor PM$_{2.5}$ and PM$_{0.1}$ at the three houses in the study in summer.

	K1				K2				K3			
	PM$_{2.5}$	SD	PM$_{0.1}$	SD	PM$_{2.5}$	SD	PM$_{0.1}$	SD	PM$_{2.5}$	SD	PM$_{0.1}$	SD
Naph	19.8	15.5	2.5	1.6	30.7	21.7	2.4	2.5	9.4	2.5	5.3	3.2
Acy	14.3	10.8	1.1	0.5	11.7	4.8	1.5	1.1	3.3	1.5	1.1	0.5
Ace	13.7	12.6	1.5	0.8	6.1	1.8	2.3	1.0	2.7	0.7	1.0	0.6
Flu	2.8	0.7	1.7	0.4	5.9	2.0	3.4	1.3	5.3	0.8	2.0	1.7
Phe	15.1	4.1	4.9	1.4	29.9	8.7	5.8	0.9	35.6	16.0	6.0	1.0
Ant	2.2	1.4	0.6	0.3	3.0	1.1	0.8	0.2	3.3	1.6	0.6	0.3
Flt	3.0	1.2	2.4	1.0	4.4	1.8	3.4	0.9	6.0	5.3	2.6	0.8
Pyr	2.4	0.6	1.7	0.4	4.0	1.1	2.9	0.6	2.2	1.3	2.2	0.8
BaA	1.5	0.9	0.8	0.3	2.4	2.0	0.7	0.2	1.7	1.4	0.9	0.3
Chr	1.4	0.5	1.2	0.5	3.5	1.9	1.1	0.5	3.2	2.9	1.1	0.4
BbF	4.6	1.9	1.9	0.5	2.9	1.5	1.0	0.3	6.4	3.6	1.4	0.8
BaP	2.3	1.4	1.6	0.5	2.5	1.6	1.0	0.5	2.3	2.5	3.2	0.4
Ind	6.2	2.7	1.3	0.9	1.8	0.4	1.0	0.3	4.1	2.6	1.2	1.5
DahA	2.0	1.3	0.7	0.4	3.6	2.2	0.6	0.0	24.0	10.3	3.5	2.1
BghiP	11.6	9.0	1.3	0.7	5.6	3.8	1.2	0.2	4.7	2.1	4.3	2.2
∑PAHs	102.9	42.5	25.3	5.3	118.1	30.4	29.2	5.5	114.2	41.6	36.3	8.5
∑Com PAHs	32.7	12.4	11.0	2.4	28.6	11.3	12.4	2.8	29.9	13.6	16.6	3.8
∑Car PAHs	18.0	6.6	7.6	2.1	16.8	7.6	5.5	1.5	41.6	20.8	11.4	4.6

Table 6. *3.* Mean concentrations (ng/m^3) and standard derivation (SD) of individual PAHs in indoor PM$_{2.5}$ and PM$_{0.1}$ at the three houses in the study in winter

	K1				K2				K3			
	PM$_{2.5}$	SD	PM$_{0.1}$	SD	PM$_{2.5}$	SD	PM$_{0.1}$	SD	PM$_{2.5}$	SD	PM$_{0.1}$	SD
Naph	45.5	37.5	4.7	2.1	40.4	12.6	3.6	1.3	13.5	4.1	4.8	1.3
Acy	20.9	14.8	8.8	2.4	4.4	2.2	1.2	0.5	10.8	4.4	2.0	1.6
Ace	10.4	9.6	1.8	0.6	7.3	2.1	2.2	0.6	7.2	4.0	0.8	0.4
Flu	5.5	3.3	2.4	1.0	12.1	2.9	5.0	1.4	16.1	6.8	6.6	2.9
Phe	20.5	8.8	7.6	2.4	36.6	13.8	8.6	2.5	32.1	17.5	10.8	3.8
Ant	2.3	1.3	1.5	0.8	3.6	1.1	1.0	0.3	4.2	2.0	1.2	0.7
Flt	4.4	2.3	3.6	1.8	4.9	1.2	3.9	1.0	3.8	2.3	1.8	1.3
Pyr	4.1	1.4	3.1	1.1	4.8	1.1	3.5	0.9	3.3	1.3	1.6	1.2
BaA	1.6	1.2	0.7	0.3	5.3	2.9	1.2	0.4	2.5	1.5	0.9	0.9
Chr	2.1	1.3	1.0	0.2	3.6	2.5	1.4	0.5	5.3	2.9	1.3	1.0
BbF	4.7	2.2	2.3	1.2	3.1	1.0	1.8	0.8	8.1	4.8	6.1	2.8
BaP	3.4	2.0	2.0	1.0	2.5	0.6	1.6	0.5	2.8	1.0	1.7	1.0
Ind	13.2	13.0	1.3	0.7	3.0	1.2	1.3	0.4	5.0	2.5	4.6	1.9
DahA	2.0	0.8	0.7	0.3	1.5	0.6	0.7	0.2	15.3	5.8	4.2	3.3
BghiP	15.0	14.4	1.6	0.7	3.0	1.2	1.5	0.5	3.9	1.8	4.5	1.6
∑PAHs	155.6	51.2	43.1	4.5	135.8	31.3	38.6	8.5	133.8	42.4	52.9	15.2
∑Com-PAHs	49.7	30.6	14.3	2.5	37.3	7.3	17.3	4.5	46.9	16.1	27.3	10.6
∑Car-PAHs	27.1	16.2	7.9	2.6	18.9	6.0	8.0	2.6	38.9	14.7	18.8	8.8

[a] Combustible PAHs includes: Flt, Pyr, BaA, Chr, BbF, BaP, BghiP and Ind

[b] Carcinogenic PAH includes: BaA, Chr, BbF, BaP, DahA, and Ind

The values of ∑PAHs_PM$_{0.1}$/∑PAHs_PM$_{2.5}$ and of mass PM$_{0.1}$/PM$_{2.5}$ in the indoor environment were greater than those in outdoor environment, respectively, implying that PAHs bound to $PM_{0.1}$ were influenced by indoor sources in the three houses. Compared with ratios reported for indoor environments in China by Zhang et al.(2012) and ambient air in Japan by Kawanaka et al.(2009), the contribution of $PM_{0.1}$ to $PM_{2.5}$ mass ($PM_{0.1}/PM_{2.5}$) and total PAH-$PM_{0.1}$ to total PAH-$PM_{2.5}$ (∑PAHs_$PM_{0.1}$/∑PAHs_$PM_{2.5}$) were higher than in this study.

Previous studies on particulate PAHs concluded that the concentrations of particulate PAHs depend on the particle sizes (Hassanvand et al.2015; Lin et al. 2013; Zhang et al. 2012). Zhang et al. (2012) reported that accumulation mode particles were composed predominantly of $\sum_{16}$PAH (68.1–76.5%), whereas ultrafine particles and coarse particles (particles with diameter larger than 1.8 μm) contributed only 3.0–4.0% and 20.4–29.2%, respectively to total PAH loads. The dominance of mass fractions of PAHs in accumulation mode particles were also reported in roadside and suburban atmosphere by Kawanaka et al. (2009), who reported that the contributions of PAHs bound to ultrafine particles to ∑PAHs concentrations in roadside and suburban outdoor environments were 10% to 15% and 4.2% to 6.9%, respectively. Accordingly, it is worth to emphasis in that study that, these contributions of PAHs bound to ultrafine particles were significantly higher than those of ultrafine particles to the total PM mass in the atmosphere at the two sites (2.3% and 1.3%), respectively. This means that, ultrafine particles contained high levels of PAHs and the contents (%) of each target PAH were significantly higher than those in accumulation mode particles and coarse particles, which was corroborated in our study, as explained above. Therefore, it seems that ultrafine particles are important carriers of PAHs and their associated toxicity.

Fig. 6.2 shows the trend in the variation of $PM_{2.5}$ and $PM_{0.1}$ and their PAHs in the three houses of this study over the course of sampling. The ranges of indoor $PM_{2.5}$ for the entire sampling period at the urban periphery house (K1), the roadside house (K2) and the urban house (K3) ranged from 28.7 to 89.6 $\mu g/m^3$, 24.4 to 102.2 $\mu g/m^3$, and 22.5 to 179.4 $\mu g/m^3$, respectively, whereas the corresponding concentrations of indoor $PM_{0.1}$ ranged from 4.2 to 11.1 $\mu g/m^3$, 4.4 to 12.4 $\mu g/m^3$, and 5.1 to 13.7 $\mu g/m^3$, respectively. Similarly, the corresponding ranges of total PAHs bound to indoor $PM_{2.5}$ were 102.9 to 155.6 ng/m^3, 118.1 to 135.8 ng/m^3, and 114.2 to 133.8 ng/m^3 at K1, K2 and K3, respectively. Accordingly, the variance of total PAHs in indoor $PM_{0.1}$ were 25.3 to 43.1 ng/m^3, 29.2 to 38.5 ng/m^3, and 36.3 to 52.9 ng/m^3 at K1, K2 and K3, respectively. As illustrated in Fig. 2 during the entire sampling period, the $PM_{2.5}$ and PAHs bound to $PM_{2.5}$ (PAH_$PM_{2.5}$) at the three houses varied significantly between winter and summer and followed a relatively similar trend. The concentrations of $PM_{2.5}$ increased in winter, which corresponded to $PM_{2.5}$ bound PAHs. However, negligible variation in the concentrations of $PM_{0.1}$ was observed between seasons in the three houses.

Regarding each house, the levels of $\sum_{15}$PAH, ∑com-PAHs and ∑car-PAHs in each house during the entire sampling period are shown in Fig.6.3. The average PAH concentrations in the urban periphery house (K1), the roadside house (K2) and the urban house (K3) were 129.2±37.3 ng/m^3, 126.9±12.5 ng/m^3 and 125.9±18.7 ng/m^3, respectively, for indoor $PM_{2.5}$, and 34.2±12.6 ng/m^3, 33.9±6.6 ng/m^3 and 44.6±11.7 ng/m^3, respectively, for indoor $PM_{0.1}$. Similarly, the levels of ∑com-PAH, ∑car-PAH bound to $PM_{2.5}$ were 41.2±12 ng/m^3 and 22.5±6.4 ng/m^3 at K1; 33±6.1 ng/m^3 and 17.8±1/5 ng/m^3 at K2; and 38.4±12 ng/m^3 and 40.3±2 ng/m^3 at K3, respectively. The results showed that the concentrations of $\sum_{15}$PAH bound to $PM_{2.5}$ did not differ significantly among three houses ($P>0.05$), whereas there were marked differences in the levels of ∑com-PAH and ∑car-PAH bound to $PM_{2.5}$ and $PM_{0.1}$ among the three houses. Interestingly, the concentrations of $\sum_{15}$PAHs, ∑Car-PAHs and ∑Com-PAHs appeared the highest in $PM_{0.1}$ at K3, where the direct influence of daily indoor activities was considered as usual during the sampling periods. Since the samples were taken in the living room, in conjunction with kitchen room. The increase in indoor $PM_{0.1}$-bound PAH at this site was attributed to burning incense, cooking activities and other indoor activities; of these activities, burning incense and cooking have been reported to generate most of the particles with a diameter less than 100 nm (cooking) and between 90 nm to 150 nm (burning incense and smoking) (Vu et al., 2017). In addition, camphor and mothballs were used in the houses, which jointly contributed to PAH generation (Chen et al., 2017; Zhu et al., 2009).

Low molecular weight (LMW)-PAHs refers to the sum of 2-4 rings and high molecular weight (HMW)-PAHs refers to sum of 5-6 rings. The concentrations of LMW-PAHs and HMW-PAHs bound to $PM_{2.5}$ and $PM_{0.1}$ in the three houses were also quantified. Regardless of the sampled houses, the mean concentrations of LMW-PAHs were in range of 72.7±25.3-122.8±31.2 ng/m^3, whereas average HMW-PAHs ranged from 16.4±7.9 to 41.5.4±29.6 ng/m^3 for $PM_{2.5}$. Accordingly, the corresponding LMW-PAHs ranged from 18.5±3.5 to 35.4±5.6 ng/m^3, while HMW-PAHs ranged from 4.9±1.1 to 21.1±8.6 ng/m^3 for $PM_{0.1}$, respectively. In spite of the higher concentrations of $PM_{2.5}$-bound HMW-PAHs and LMW-PAHs than those of $PM_{0.1}$, the contents of LMW-PAHs and HMW-PAHs in $PM_{0.1}$ (LMW-PAHs_$PM_{0.1}$/$PM_{0.1}$ mass; HMW-PAHs_$PM_{0.1}$/$PM_{0.1}$ mass) were roughly twice those of $PM_{2.5}$. This means that the smaller particles ($PM_{0.1}$) were more enriched in LMW and HMW-PAHs than bigger particles ($PM_{2.5}$) in this study. It is interesting to underline that, compared to LMW-PAHs, the concentration of HMW-PAHs was markedly lower in $PM_{2.5}$ and $PM_{0.1}$, which was also reported previously (Ali, 2019; Liaud et al., 2014; Zhu et al., 2009). The LMW-PAHs came mainly from indoor sources whereas HMW originated mostly from the penetration of outdoor emissions (traffic emissions and fuel combustion) (Ali, 2019; Liaud et al., 2014; Zhu et al., 2009). This implies that cooking activities might generate 3-4 rings PAHs abundantly during frying, boiling and streaming (Xu et al., 2020).

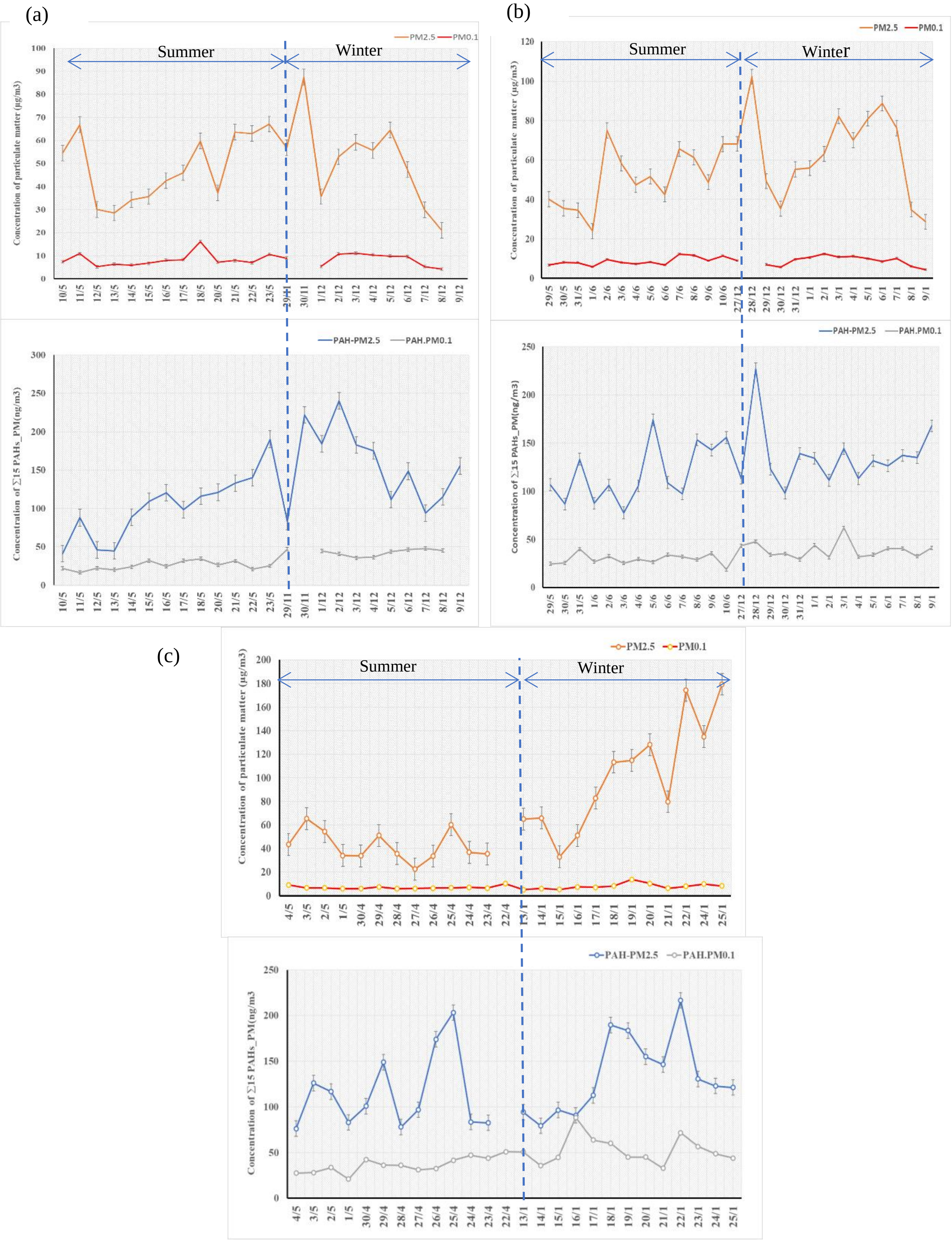


Fig.6. 2. Time series of total PAH concentrations in indoor $PM_{2.5}$ and $PM_{0.1}$ at the different houses a) K1, b), K2, and c) K3

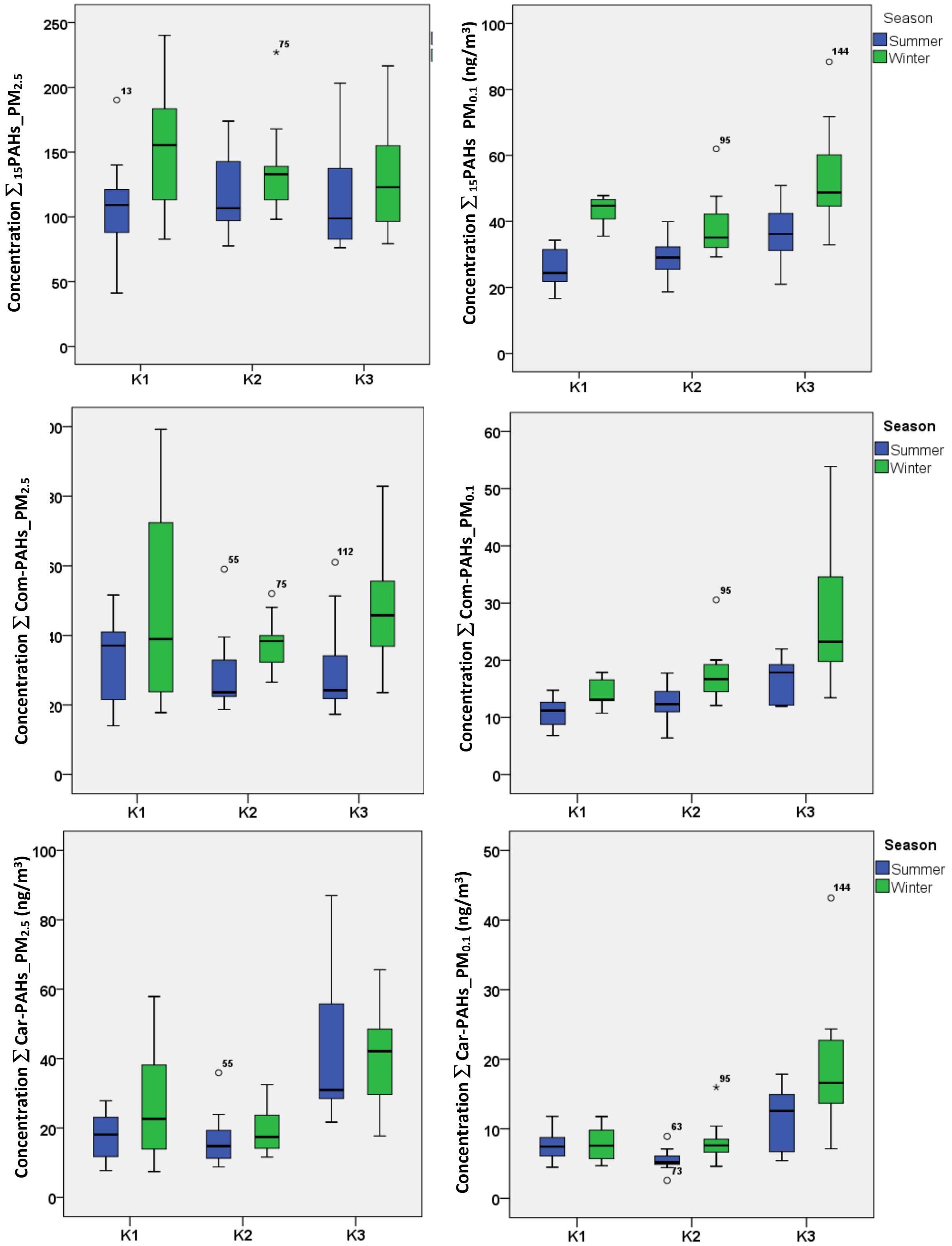


Fig.6. 3. Concentrations of ∑PAHs, ∑com-PAHs, ∑car-PAHs in summer and winter for $PM_{2.5}$ and $PM_{0.1}$ in the three houses.

In addition, high concentrations of LMW-PAHs were detected in bedrooms (at K1 and K2), which was ascribed to the use of camphor or mothballs or insect repellent, which was reported during the sampling period (Chen et al., 2017; Liaud et al., 2014; Ohura et al., 2004; Zhu et al., 2009). In comparison in three houses, the levels of LMW-PAHs in K3 were lower, whereas those of HMW-PAHs were higher than those in K1 and K2. The higher concentrations of HMW-PAHs at K3 was attributed to intensive indoor activities, such as burning incense and scented candles (Derudi et al., 2012; See and Balasubramanian, 2011), which were typical activities happing during sampling.

High levels of particulate PAHs were reported in different indoor environments in several previous studies. Compared to other Asian countries, the average $\sum_{15}$ PAHs in the residences in this study were 126.7 ± 21.2 ng/m^3 bound to $PM_{2.5}$ and 37.5±9.7 ng/m^3 bound to $PM_{0.1}$, which was considerately greater than those of ∑PAH bound to $PM_{2.5}$ in indoor environments in Korea (Kim et al., 2014), in Beijing, China (Chen et al., 2017), and residential houses in Thailand (Kanjanasiranont et al., 2021). In Korea, average $PM_{2.5}$-bound $\sum_{24}$PAH concentrations were 21.45 ng/m^3 (PC rooms); 4.13 ng/m^3 (parking lots); 8.28 ng/m^3 (underground subway stations); 7.07 ng/m^3 (terminal waiting rooms in trains); 3.79 ng/m^3 (supermarkets); 4.58 ng/m^3 (movie theaters); 2.22 ng/m^3 (childcare facilities), and 3.08 ng/m^3 (elderly care facilities) (Kim et al., 2014). Chen et al. (2017) measured $PM_{2.5}$ bound to $\sum_{16}$PAHs in a dormitory, office, residential home with average concentrations of 34.1 ng/m^3, 32.1 ng/m^3, and 39.8 ng/m^3, respectively. In Thailand, total PM_{10}-bound $\sum_{16}$PAH concentrations in residential houses were 16.2 ng/m^3 and 19.9 ng/m^3 in an industrial area and in the vicinity of an industrial area, respectively (Kanjanasiranont et al., 2021). In contrast, our results were significantly lower than those reported in India, China, and Iran. For example, Ansari et al. (2010) reported that $\sum_7$PAH bound to $PM_{2.5}$ and PM_{10}, generated by cooking in rural homes in North India, ranged from 6.2 to 12.4 μg/m^3 and 7.8 to 15.8 μg/m^3, respectively, whereas corresponding values were 1.4 μg/m^3 and 1.8 μg/m^3 when no cooking was performed. In that study, high levels were affected by the combustion of different biomass fuels during cooking in rural areas. PAHs released from the combustion of plant material (wood, leaves, twigs and crop residues) combined with cattle dung cakes doubled PAH levels in comparison to combustion of only plant material. In Iran, Hassanvand et al. (2015) found great levels of $\sum_{16}$PAHs in different particle sizes in retirement homes. The total PAH concentrations in PM_{10}, $PM_{2.5}$ and PM_1 were 317.4±80.6 ng/m^3; 281.3±80.5 ng/m^3, and 222.9±83.6 ng/m^3. The com-PAHs accounted for 41%, 43%, and 47.1% of ∑PAHs bound to PM_{10}, $PM_{2.5}$ and PM_1, respectively, whereas car-PAHs made up 55.6%, 56.3% and 60.9%, respectively. Sharma and Jain (2020) that average concentrations of particulate $\sum_{12}$PAHs were 25.7 ng/m^3 and 6139 ng/m^3 in a kitchen with traditional cookstoves (TCS) and improved cookstoves (ICS), respectively. Kitchen characteristics (enclosed, semi enclosed and open) also had a significant impact on PAHs during cooking. For example, decreases in PAH concentrations of 42% (enclosed vs. semi-enclosed), 69% (enclosed vs. open) and 46% (semi-closed vs. open) were observed in the case of TCS, whereas decreases of 67% (enclosed vs. semi-enclosed), 87% (enclosed, open) and 61% (semi-enclosed, open) were observed in the case of ICS. Thus, the reason for the disparity observed in the PAH concentrations of different kitchen configurations was likely due to poor ventilation in the kitchen. In metropolitan areas of Taipei in Taiwan, Lin et al. (2013) measured 16PAHs in household particles. The concentrations of $\sum_{16}$PAHs bound to PM_{1-10}, $PM_{0.1-1}$, and $PM_{0.1}$, were

60±3, 110±10 and 30±2 mg/m^3, respectively. That study is one of the few to investigate PAHs-bound $PM_{0.1}$ in residential areas in Asian countries, it is interesting that the results obtained for the $PM_{0.1}$-bound PAHs in the study of Lin et al. (2013) were comparable to the values we obtained for PAHs in $PM_{0.1}$ in our study.

Compared to PAH concentrations in developed countries, such as France, Italy and the US, the literature review revealed that the particulate PAH concentrations measured in residential environments in Vietnam were considerately higher. Liaud et al. (2014) measured size-segregated, PM-bound PAH (i.e., PAHs bound to TSP, PM_{10}, $PM_{2.5}$ and PM_1) in indoor environments in urban, suburban and rural areas of France. On average, total levels of particulate $\sum_{16}$PAHs found in rural sites were in the same range as those measured at the urban and suburban sites. For example, mean $\sum_{16}$PAH concentrations bound to TSP, PM_{10}, $PM_{2.5}$ and PM_1 were 1.3±0.5 ng/m^3, 0.9±0.2 ng/m^3, 0.6±0.1 ng/m^3 and 1.6±0.8 ng/m^3 in urban sites; 1.3±0.5 ng/m^3, 1.1±0 0.5 ng/m^3, 0.8±0.5 ng/m^3 and 1.8 ± 0 in suburban sites; and 1.1±0.5 ng/m^3, 0.9±0.5 ng/m^3, 0.7±0.6 ng/m^3 and 1.1±0.7 ng/m^3 in rural sites, respectively. Their findings highlighted showed that the proportion of PAHs bound to PM_1 were higher compared to PAHs bound to larger particles. Romagnoli et al. (2014) observed significant differences in $PM_{2.5}$-bound $\sum_8$PAH indifferent environmental settings in Italy, with values for schools, homes and offices, ranging from 1.84±0.3 to 8.29±6.67 ng/m^3, 5.63±2.52 to 8.36±4.16 ng/m^3, and 5.73±3.08 to 7.12±1.62 ng/m^3, respectively. Jung et al. (2011) characterized the PAH levels of different floor levels and building types between heating and non-heating seasons in New York, USA. On the 0 to 2nd, 3rd to 5th, and 6th to 32nd floors, the levels of $\sum_8PAH_{nonvolatile}$ were 2.0±2.2 ng/m^3, 2.3±6.34 ng/m^3, 1.7±2 ng/m^3, respectively, whereas $\sum_8PAH_{semivolatile}$ levels were 43.3±24.1 ng/m^3, 65.6±65.4 ng/m^3, and 45.5±25.7 ng/m^3, respectively. Naumova et al. (2002) added reported that $\sum$ PAH concentrations were 16 to 220 ng/m^3 in Los Angeles, 21 to 310 ng/m^3 in Houston, 22 to 350 ng/m^3 in Elizabeth. Indoor PM sources had a significant effect on indoor concentrations of 3-rings PAHs and a smaller effect on 4-rings PAHs, whereas outdoor sources had a dominant effect on the indoor concentrations of 5-6 rings PAHs.

3.1.2. Individual PAHs composition and size distribution

Among the 15 PAHs in the particulate phase investigated in the three houses in this study, Nap, Phe and Flt were present in the highest concentrations in indoor $PM_{2.5}$ and $PM_{0.1}$, which was in line with the previous findings of Lin et al. (2013), Liaud et al. (2021) and Zhu et al. (2009). The mean concentration of Nap and Phe ranged from 20±10.6 to 33.1±17.2 ng/m^3 and 26.8±10.5 to 29.7±8.3 ng/m^3 for $PM_{2.5}$, and 2.6±1.6 to 4.4±0.7 ng/m^3 and 4.2±0.6 to 8.9±1.7 ng/m^3 for $PM_{0.1}$, respectively. Additionally, the concentration of Flt bound to $PM_{0.1}$ was in the range of 2.1±0.5 to 3.1±1.1 ng/m^3. Previous studies found that Nap emissions in indoor environments were highly correlated with the use of mothballs, camphor and insect repellent (Chen et al., 2017; Liaud et al., 2014; Zhu et al., 2009). Phe and Flt are primarily emitted by diesel engines (Liaud et al., 2021). In addition, such high levels of Phe could be attributed to indoor emissions related to cooking and volatile materials (Krugly et al., 2014), and electronic devices (Cai et al., 2018). Although BaP was not the most abundant PAH, BaP is classified as a group 1 carcinogenic substance (US EPA, 2009). Due to the high carcinogenicity, mutagenicity and ubiquity of BaP in the atmosphere, the target annual average concentration of BaP in the atmosphere is 1 ng/m^3 (EC, 2008; Romagnoli et al., 2014). The average concentrations of BaP bound to $PM_{2.5}$ and $PM_{0.1}$ in our study were 2.6 ±0.4 ng/m^3 and 1.6±0.2 ng/m^3, respectively, which exceeded the EU guidelines for both particle sizes (EC, 2004; EC, 2008) and could potentially damage the health of the occupants of the houses in this study.

The contributions of individual PAHs to the $\sum_{15}$PAH concentrations of indoor PM are illustrated in Fig. 6.4, 6.5 and 6.6. The most abundant PAHs in $PM_{2.5}$ were Nap and Phe, which accounted for 20.6±3.9% and 22.5±2.2% of $\sum_{15}$PAH concentrations, respectively. In addition, Nap, Phe and Flt were the most enriched species in $PM_{0.1}$, which made up 10.8±0.7%, 19.1±1%, and 9.2±0.0% of $\sum_{15}$PAH concentrations, respectively. These results were generally similar to those reported in previous studies. For example, Krugly et al. (2014) reported that Nap and Phe appeared at the highest concentrations among 15 PAHs in the particulate phase in a primary school environment in Lithuania. The average concentration of Nap in that study (26.3 ng/m^3) was comparable to the results obtained in the present study. Nap levels as high as 21.7% of $\sum_{16}$PAH concentrations were also observed in urban indoor environments in Guangzhou, China (Zhang et al., 2012), but the average concentration of particulate Nap (6.2±1.6 ng/m^3) in that study was considerately lower than the mean Nap concentration of 20.6 ng/m^3 in $PM_{2.5}$ in this study. Zhu et al. (2009) Nap, Ace and Phe were among the most abundant species of the 16 PAHs measured in their study; however, these species exist almost entirely in the gas phase. Yury et al. (2018) added that Flt was the dominant PAH species in different indoor environments in China, and that it accounted for approximately 21% of the $\sum_{16}$ PAHs bound to $PM_{2.5}$. Conversely, the most abundant PAH species bound to $PM_{2.5}$ observed in indoor environments in Beijing, China, which included BbF (4.3 ng/m^3), Ind (3.4 ng/m^3), Flu (3.4 ng/m^3), BghiP (3.2 ng/m^3) and BaP (3.1 ng/m^3), accounting for 58.1% of the $\sum_{12}$PAHs. Hassanvand et al. (2015) reported that Phe, Flu, BaA, Chr and BghiP were the most abundant species of PAHs in indoor PM_{10}, $PM_{2.5}$ and PM_1 in retirement homes and school dormitories in Tehran, Iran. In addition, Liaud et al. (2021) found that Phe, BbF, BghiP and Ind were the most dominant species in PM_{10}, $PM_{2.5}$ and PM_1 in school environments in France, whereas BbF and Ace bound to PM_{10} were the major species among 16 PAHs in residential houses in Rayong, Thailand (Kanjanasiranont et al., 2021). Among individual PAHs, the most common PAHs bound to $PM_{2.5}$ were 5–6 rings PAH compounds such as, B[a]P, benzo[ghi]perylene, and indeno[1,2,3-cd]pyrene and benzo[b]fluoranthene, which was also reported by Sharma and Jain (2020) in a rural residential area in India.

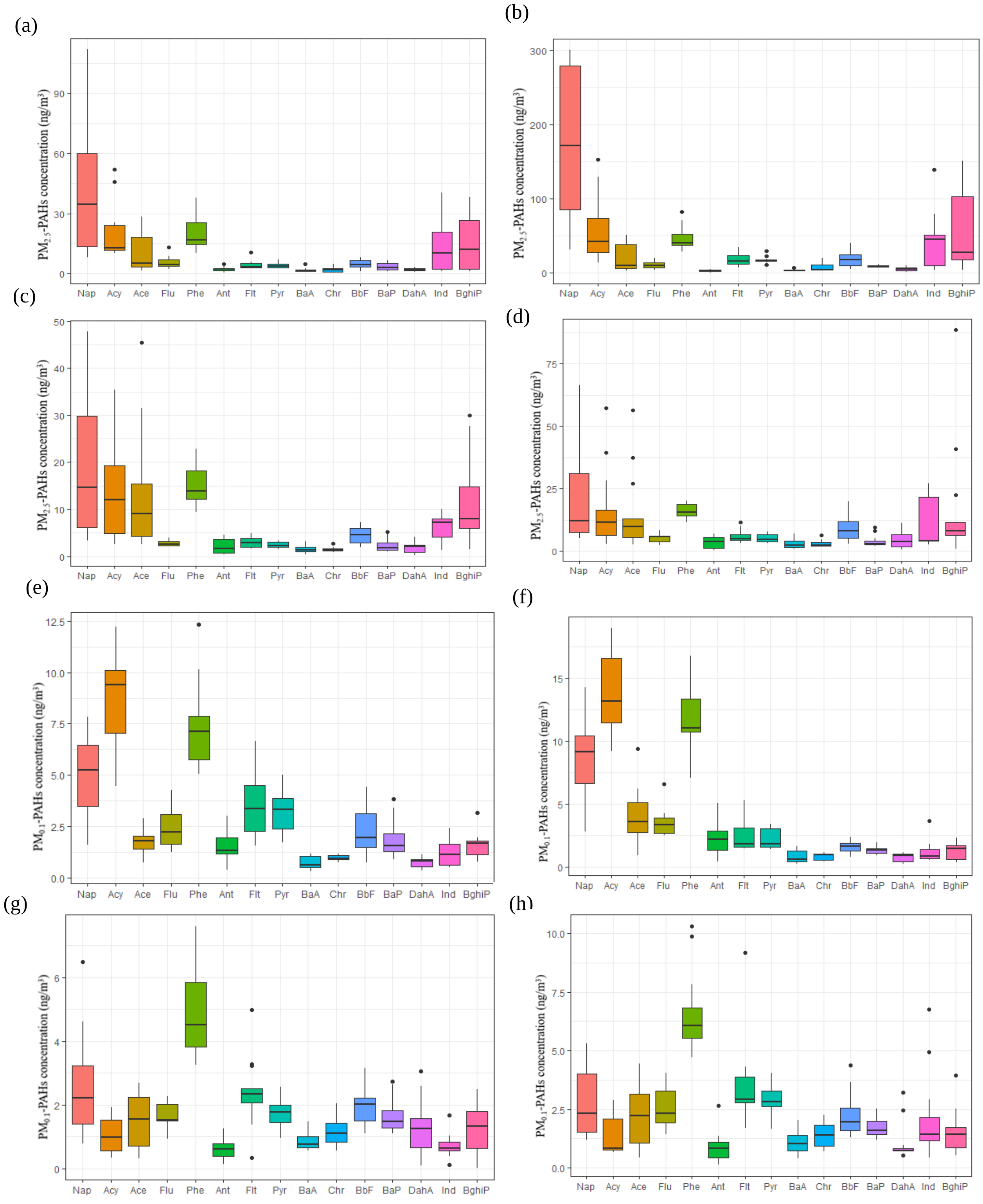


Fig.6.4. Concentrations of individual PAHs at K1: a) indoors (winter), b) outdoors (winter), c) indoors (summer), d) outdoors (summer), e) indoors (winter), f) outdoors (winter), g) indoors

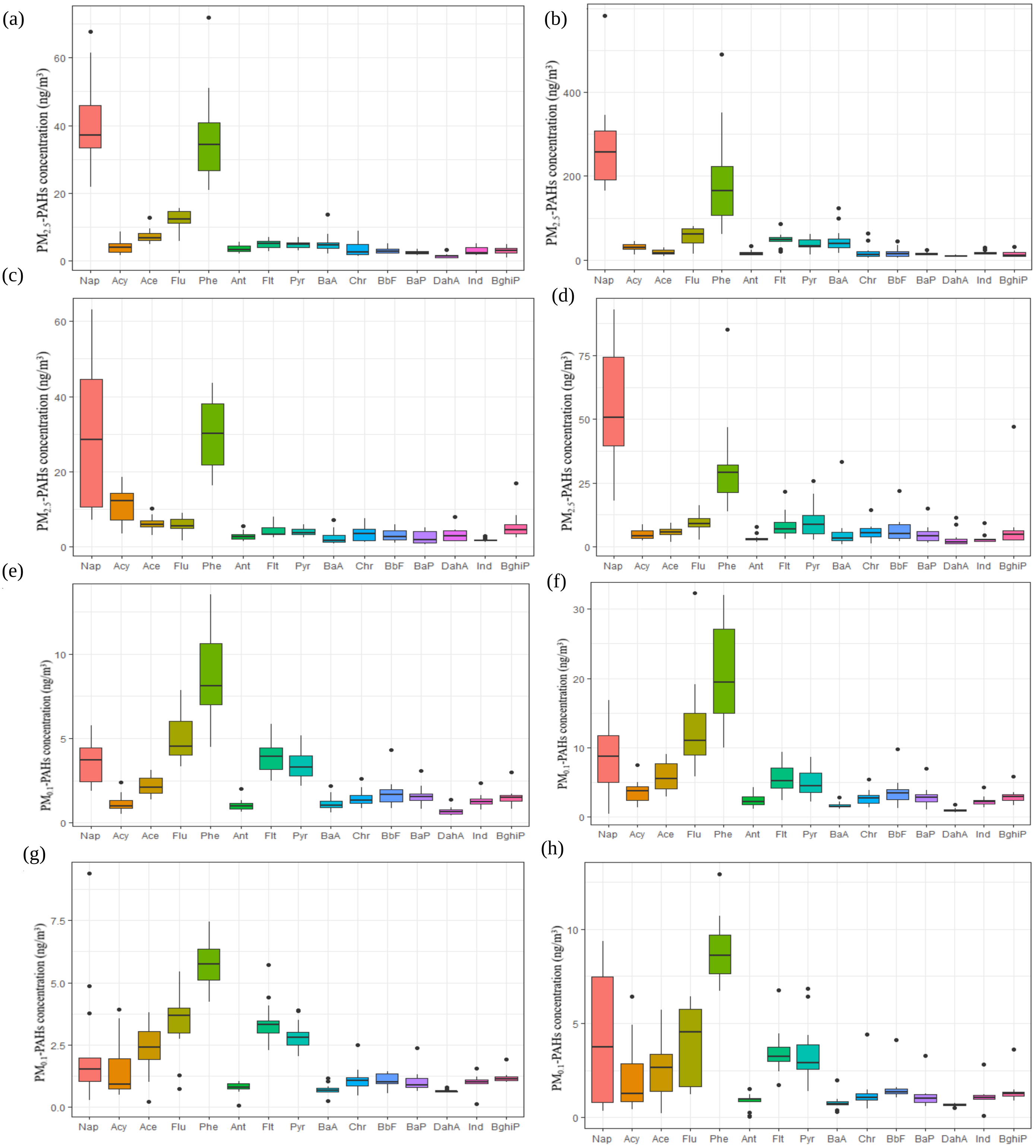


Fig.6. 5.Concentrations of individual PAHs at K2: a) indoors (winter), b) outdoors (winter), c) indoors (summer), d) outdoors. (summer), e) indoors (winter), f) outdoors (winter), g) indoors (summer), and h) outdoors (summer).

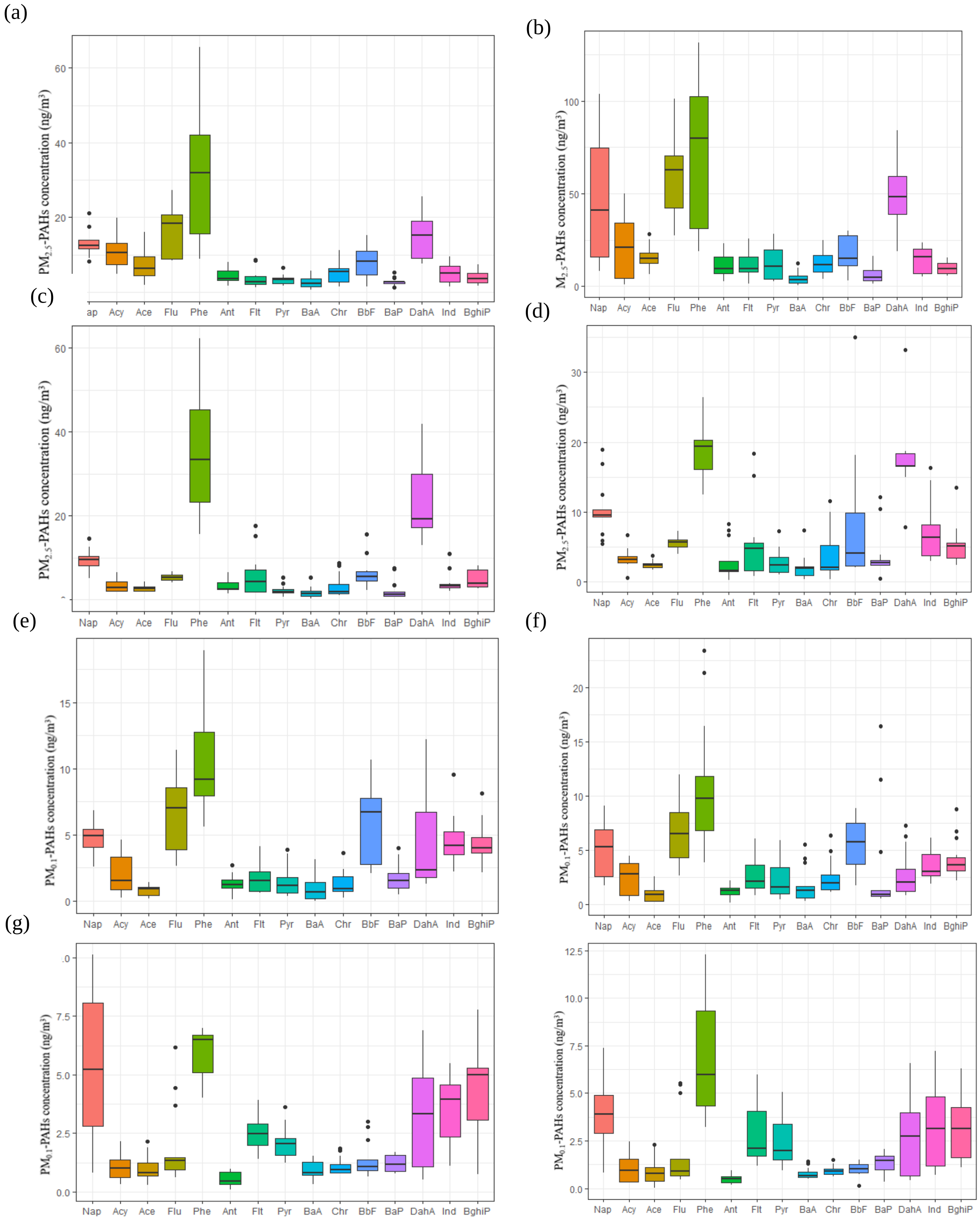


Fig.6. 6. Concentrations of individual PAHs at K3: a) indoors (winter), b) outdoors (winter), c) indoors (summer), d) outdoors (summer), e) indoors (winter), f) outdoors (winter), g) indoors (summer), and h) outdoors (summer).

(b)

PAHs can also be classified based on the number of aromatic rings, as follows: 2 rings (Nap); 3 rings (Ace, Acy, Flu, Phe, Ant; 4 rings (Flt, Pyr, BaA and Chr); 5 rings (BbF, BaP, DahA); and 6 rings (BghiP and Ind). The relative proportion of PAHs with different numbers of rings to the $\sum_{15}$PAH in indoor PM in summer and winter is shown in Fig. 6.7.

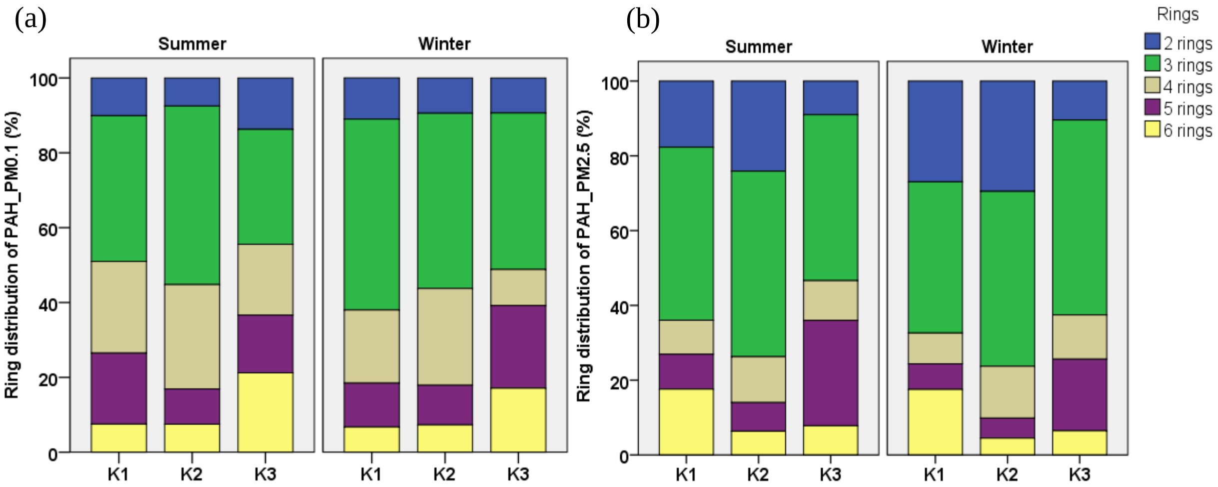


Fig.6. 7. Ring-number distribution of PAHs in the three houses: a) $PM_{0.1}$, b) $PM_{2.5}$.

The same pattern was observed for the distribution of PAH-ring number in summer and winter. Two-rings PAHs had the lowest contribution to total $\sum_{15}$PAHs (10.2±2.1 to 19.6±8.6%), whereas PAHs with 3-4 rings had a much larger contribution, ranging from 57.6±5.6 to 63.9±11.1%, and PAHs with 5-6 rings accounted for 22.8±9.4 to 26±9.9% of $\sum_{15}$PAH in the indoor environment. The lower proportion of 2-ringes PAHs is due to their higher volatility, which means that these PAHs typically exist in the gas phase. Most of the LMW-PAHs were in the gas phase due to their higher vapor pressure and higher volatility. Conversely, most of the HMW-PAHs were in the particulate phase because of their relatively low vapor pressure (Krugly et al., 2014; Zhu et al., 2009). Interestingly, the contributions of 3-4-and 5-6-rings PAHs to $\sum_{15}$PAHs bound to $PM_{0.1}$ were higher than those bound to $PM_{2.5,}$ whereas the contributions of 2- rings PAHs were higher in $PM_{2.5}$ in indoor air in both seasons. Thus, there was an increase in the contribution of HMW-PAHs (5-6 rings) and 3-4-rings PAHs to the total PAHs as PM size decreased, which was consistent with previous studies of Hassanvand et al.(2015); Kong et al.(2010); Liaud et al.(2014), and may explain the high affinity of PAHs with more rings, especially in HMW-PAHs, for ultrafine particles such as $PM_{0.1}$. Our study thus adds to the evidence that smaller particle has greater proportions of 5- and 6-rings PAHs, which are known to be more toxic than PAHs with fewer rings. Previous studies reported that PAHs with 4-6 rings condensed and were adsorbed by small particles due to their relatively large surface area (Hassanvand et al., 2015; Kong et al., 2010; Liaud et al., 2014). Conversely, the contribution of LMW-PAHs to total PAHs was higher in $PM_{2.5}$. This finding is consistent with previous studies, which reported that LMW-PAHs were more abundant in coarse fractions than in fine fractions (Kong et al.,2010; Liaud et al., 2014).

3.1.3. Seasonal variation of particulate PAHs

The results of PAH measurements carried out during summer and winter seasons in three houses are shown in Table 6.2, 6.3 and Fig. 6.3. In the houses examined in this study, the concentrations of $\sum_{15}$PAHs bound to $PM_{2.5}$ and $PM_{0.1}$ ranged from 102.9 to 114.2 ng/m^3 and 25.3 to 36.3 ng/m^3 in summer, respectively, and from 133.8 to 155.6 ng/m^3 and from 38.6 to 52.8 ng/m^3 in winter, respectively. In outdoor air, the corresponding values for $PM_{2.5}$ and $PM_{0.1}$ ranged from 101.3 to 162.8 ng/m^3 and 33.7 to 37.6 ng/m^3 in summer for $PM_{2.5}$ and $PM_{0.1}$, respectively, and from 368.4 to 801.7 ng/m^3 and 55.6 to 81.3 ng/m^3 in winter, respectively. There were significant differences between the PAHs in both particles between winter and summer. The apparent trend in the seasonal variation of $\sum_{15}$PAHs_$PM_{2.5}$ was similar to that of $PM_{2.5}$, which was observed in the studies of Nguyen et al. (2018) and Vo et al. (2022). Meanwhile, the concentration of $\sum_{15}$PAHs_$PM_{0.1}$ differed significantly between two seasons ($P<0.05$), but mass concentrations of $PM_{0.1}$ did not change in seasons in this study, which was also reported in the earlier studies of Nguyen et al.(2018) and Vo et al.(2022). The results showed that higher levels $\sum_{15}$PAH bound to $PM_{2.5}$ and $PM_{0.1}$ were observed in winter, which increased 1.3 to 1.5 times indoors and 1.8 to 4.1 times outdoors, respectively, compared to the levels observed in summer. This seasonal variations observed in this study were similar with the previous studies (Zhu et al., 2009; Chen et al., 2017; Romagnoli et al., 2014; Zhu et al., 2009). Lower temperatures and atmospheric mixing heights, as well as decreased photochemical oxidation intensity, can be explain the higher concentrations of particulate PAHs in winter in Hanoi (Nghiem et al., 2020; Ngoc et al., 2021). The combustion of biomass events during the summer-autumn harvest also affect the release of particulate PAHs in winter in Hanoi (Pham et al., 2019). The degradation of PAHs in the atmosphere is known to be associated with photochemical or thermic reactions with ozone, nitric oxides and hydroxyl radicals, especially in the warmer seasons (Ngoc et al., 2021;Pham et al., 2019). Despite differences in concentration of $\sum_{15}$PAHs_$PM_{2.5}$ and mass concentration of $PM_{2.5}$ in seasons as mentioned above, the PAH contents of indoor $PM_{2.5}$ did not differ between the two seasons, while PAH contents of indoor for $PM_{0.1}$ change. Example: Indoors, the PAH content of $PM_{2.5}$ was 0.3% in both seasons, while those of $PM_{0.1}$ were 0.4% and 0.6% in summer and winter, respectively However, outdoors, the level of particulate PAH in winter were 1.5 to 2.6 times higher than those in summer. In comparing the levels of PAH in PM between indoor and outdoor environments, the PAH levels in outdoor PM was higher than that in indoor PM. These findings imply that the particulate PAHs was dominated origins from outdoor sources for both particle sizes.

Regarding the distribution of individual particulate PAHs, there was apparent seasonal variation trend in LMW and HMW-PAHs bound to $PM_{0.1}$ and in LMW-PAHs bound to $PM_{2.5}$ in both indoor and outdoor air. Higher concentrations of LMW-PAHs and HMW-PAHs bound to $PM_{0.1}$ were observed in winter, whereas the levels of HMW-PAHs bound to $PM_{2.5}$ was stable in seasons in indoor air. Interestingly, a similar seasonal trend was observed in the contribution of LMW- and HMW-PAHs to total PAHs in $PM_{0.1}$ and $PM_{2.5}$. However, there is difference in their pattern of the contribution; the contributions of LMW-PAHs to total PAHs decreased in summer, whereas the contributions of HMW-PAHs to total PAHs increased in summer for both particle sizes. It, therefore, appears that the high temperatures in summer promoted the desorption of LMW-PAHs from the particulate phase.

Among PAHs, BaP is widely regarded as an indicator of cancer risk assessment (Chen et al., 2017; Zhu et al., 2009). The mean concentrations of BaP bound to outdoor $PM_{2.5}$ and $PM_{0.1}$ at the three houses doubled in winter, whereas the indoor particulate BaP concentrations at the houses appeared to be stable between the two seasons. The seasonal variation observed in outdoor BaP was consistent with the results of Chen et al. (2017) and Pham et al.(2019). Example, in summer, the mean concentrations of BaP in indoor $PM_{2.5}$ and $PM_{0.1}$ were 2.3±0.5 ng/m^3 and 1.5±1.1 ng/m^3, respectively, while they were 2.9±0.5 ng/m^3 and 1.8±0.1 ng/m^3 in winter, respectively. Although these BaP concentrations exceeded the annual limit of 1.0 ng/m^3 stipulated by a European commission on ambient air quality standards (EC, 2004). They were lower than those observed in residential houses in China (Chen et al., 2017), India (Ansari et al., 2010) and Iran (Hassanvand et al., 2015). However, the concentrations measured in this study were higher than those reported in European countries (Liaud et al., 2014; Romagnoli et al., 2014).

3.2 Concentration of PAH bound to PM in outdoor air

Fig. 6.8 shows the variation in mass concentrations of $PM_{2.5}$ and $PM_{0.1}$ and total their PAHs in outdoor air at the three houses investigated in this study during the sampling period. The mass concentration range of outdoor $PM_{2.5}$ for the entire sampling period in the urban periphery house (K1), roadside (K2) and urban house (K3) ranged from 25.9 to 121.6 µg/m^3, 35.3 to 154.9 µg/m^3 and 22.0 to 196.9 µg/m^3, respectively, whereas the corresponding concentrations of $PM_{0.1}$ ranged from 4.0 to 8.5 µg/m^3, 5.1 to 15.0 µg/m^3 and 2.9 to 12.3 µg/m^3, respectively. Similarly, the corresponding ranges of $\sum_{15}$PAHs bound to $PM_{2.5}$ at K1, K2 and K3 were 141.8 to 492.4 ng/m^3, 162.8 to 801.7 ng/m^3 and 101.3 to 368.4 ng/m^3, respectively. As illustrated in Fig.6.8, the $PM_{2.5}$ and $\sum_{15}$PAH_$PM_{2.5}$ followed relatively similar trends at the three houses, with the concentrations of outdoor $PM_{2.5}$ increasing together with $\sum_{15}$PAH_$PM_{2.5}$; however, the trend in outdoor $PM_{0.1}$ and $\sum_{15}$PAHs_$PM_{0.1}$ at K1 and K2 was ambiguous.

Furthermore, concentrations of the $\sum_{15}$PAH, ∑com-PAHs and ∑car-PAHs bound to outdoor $PM_{0.1}$ and $PM_{2.5}$ during the summer and winter are shown in Tables 6.4 and 6.5, respectively. Regardless sampling sites, the average $\sum_{15}$PAHs concentrations for $PM_{2.5}$ and $PM_{0.1}$ ranged from 101.3 to 801.7 ng/m^3, and 33.7 to 81.3 ng/m^3, respectively. ∑com-PAHs concentrations for outdoor $PM_{2.5}$ and $PM_{0.1}$ ranged from 39.3 to 222.3 ng/m^3 and 13.1 to 33.8 ng/m^3, respectively, whereas ∑car-PAHs ranged from 29.4 to 125.3 ng/m^3 and 6.6 to 19.6 ng/m^3, respectively. Accordingly, the average concentrations of ∑com-PAHs bound to $PM_{2.5}$ and $PM_{0.1}$ accounted for 35.2±4.7% and 41.3±10.2% of the $\sum_{15}$ PAHs, respectively; while those of

$\sum$car-PAHs were responsible for 24.6±10.5% and 23.4 ± 9.2% of the $\sum_{15}$PAHs, respectively. A higher proportion of $\sum$com-PAHs bound to $PM_{0.1}$ was obtained in comparison with that of $PM_{2.5}$, whereas the proportions of car-PAH to $\sum_{15}$PAHs were similar between the two particle sizes. The trend in the variation of the $\sum$com-PAHs contribution to total PAHs in the outdoor environment followed that observed in indoor air, which increased as particle sizes decreased. Levels of $\sum_{15}$PAHs bound to outdoor $PM_{2.5}$ and $PM_{0.1}$ were considerably higher than those in indoor PM, which was also reported in previous studies for PAHs in the particulate phase (Chen et al., 2017; Han et al., 2015; Naumova et al., 2002; Ouyang et al., 2020). For example, the average concentrations of $\sum_{15}$PAHs in outdoor $PM_{2.5}$ and $PM_{0.1}$ increased 2.7 times and 1.3 times, respectively. This implies that the concentrations of PAH bound to indoor PM were strongly influenced by outdoor PAH bound to PM.

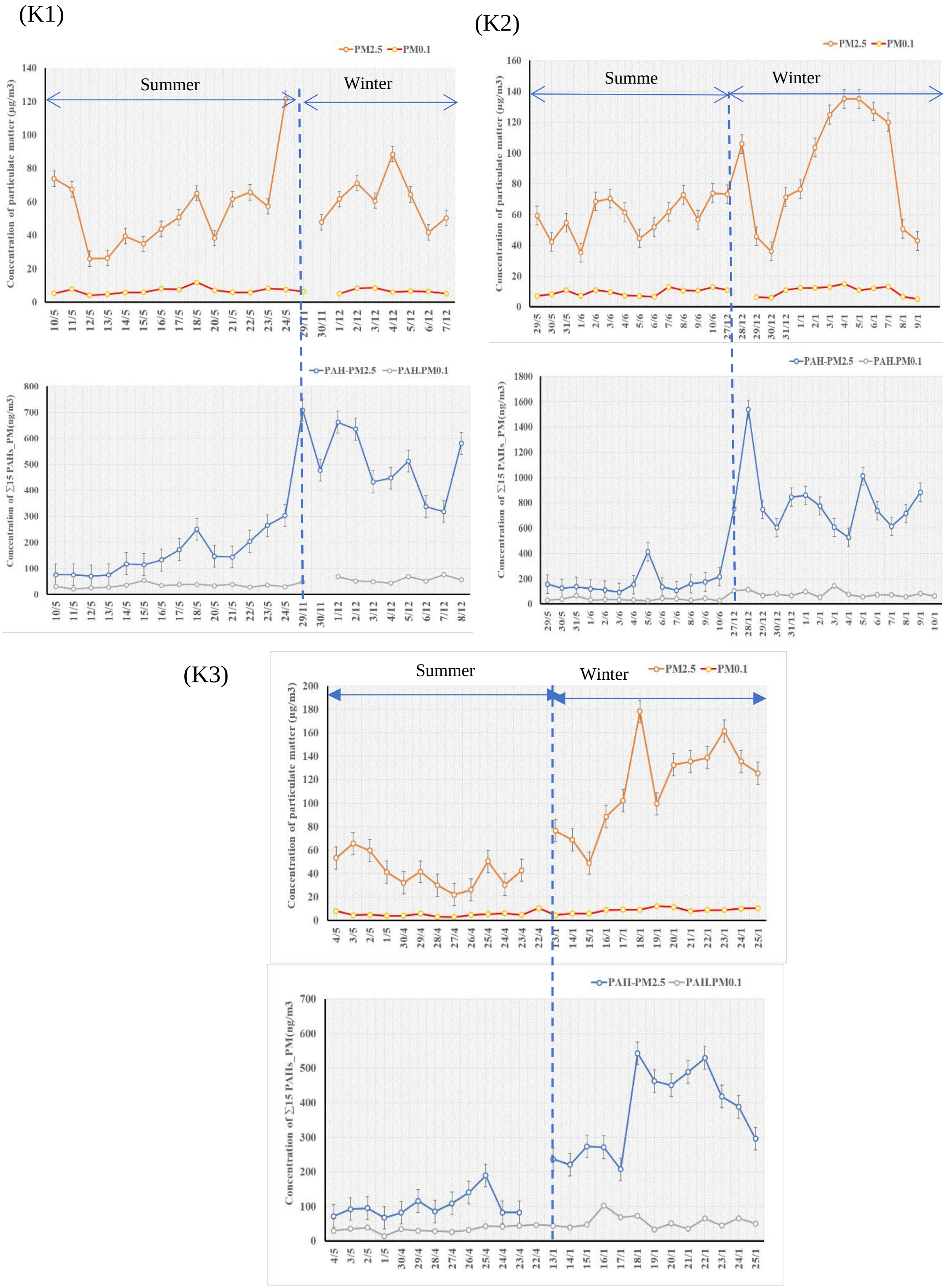


Fig.6. 8. Time series of total PAH concentrations in outdoor $PM_{2.5}$ and $PM_{0.1}$ at the different houses: a) K1, b), K2, and c) K3

Table 6. 4. Mean concentrations (ng/m^3) and standard derivation (SD) of individual PAHs in outdoor $PM_{2.5}$ and $PM_{0.1}$ in summer

	K1				K2				K3			
	$PM_{2.5}$	SD	$PM_{0.1}$	SD	$PM_{2.5}$	SD	$PM_{0.1}$	SD	$PM_{2.5}$	SD	$PM_{0.1}$	SD
Naph	22.7	20.0	2.7	1.4	52.8	24.9	4.1	3.6	10.3	4.1	4.1	1.9
Acy	16.6	15.9	1.3	0.8	4.9	2.2	2.1	1.9	3.3	1.5	1.1	0.8
Ace	15.3	15.9	2.2	1.4	5.9	2.1	2.6	1.7	2.4	0.6	1.0	0.8
Flu	5.3	1.8	2.6	0.9	9.3	3.6	4.1	2.0	5.6	1.0	1.9	2.0
Phe	16.0	3.0	6.5	1.7	31.5	18.4	8.9	1.6	18.8	4.2	6.8	2.9
Ant	3.4	2.2	0.9	0.6	3.4	1.6	0.8	0.4	2.9	2.8	0.5	0.2
Flt	5.8	2.5	3.5	1.8	8.2	5.2	3.5	1.2	5.5	5.6	2.9	1.7
Pyr	5.0	1.5	2.9	0.6	9.9	6.9	3.5	1.6	2.8	1.9	2.5	1.4
BaA	3.0	2.0	1.1	0.4	5.9	8.4	0.8	0.4	2.1	1.9	0.8	0.3
Chr	2.9	1.4	1.4	0.5	5.8	3.3	1.3	1.0	4.0	3.7	0.9	0.3
BbF	8.9	4.7	2.2	0.9	6.5	5.4	1.6	0.8	8.2	9.9	1.0	0.4
BaP	3.9	2.3	1.7	0.5	4.9	3.7	1.1	0.7	3.9	3.5	1.3	0.5
Ind	11.3	9.7	2.1	1.8	3.2	2.0	1.1	0.6	7.3	4.4	3.4	2.2
DahA	4.4	3.4	1.0	0.8	3.1	3.2	0.7	0.1	18.7	7.3	2.9	2.1
BghiP	17.1	23.8	1.5	0.9	7.6	2.0	1.4	0.7	5.3	3.0	3.1	1.5
PAHs	141.8	65.5	33.7	7.8	162.8	82.2	37.6	10.5	101.3	34.4	34.3	8.9
[a]∑Com-PAHs	57.5	34.9	15.5	4.2	53.0	41.0	14.9	6.1	39.3	20.9	15.0	4.0
[b]∑Car-PAHs	34.5	16.5	9.5	3.8	29.4	24.1	6.6	3.3	44.2	23.5	10.4	4.7

Table 6. 5. Mean concentrations (ng/m^3) and standard derivation (SD) of individual PAHs in in outdoor $PM_{2.5}$ and $PM_{0.1}$ in winter

	K1				K2				K3			
	$PM_{2.5}$	SD	$PM_{0.1}$	SD	$PM_{2.5}$	SD	$PM_{0.1}$	SD	$PM_{2.5}$	SD	$PM_{0.1}$	SD
Naph	174.5	104.4	8.5	3.4	267.2	109.2	8.6	4.7	45.4	32.7	5.0	2.4
Acy	58.6	45.5	13.9	3.5	29.4	9.5	3.5	1.7	21.6	17.7	2.4	1.7
Ace	21.4	19.2	4.1	2.6	18.3	5.9	5.8	1.9	16.1	6.3	0.9	0.7
Flu	10.6	5.3	3.6	1.3	56.4	21.0	12.8	6.7	61.7	21.5	6.7	2.9
Phe	46.7	16.6	12.1	3.0	191.6	116.1	20.8	7.8	72.9	40.6	10.9	6.1
Ant	2.5	1.6	2.3	1.4	16.3	6.7	2.4	0.8	11.5	6.3	1.2	0.7
Flt	17.7	9.3	2.4	1.3	47.4	16.9	5.5	2.1	11.7	7.1	2.6	1.6
Pyr	17.4	5.1	2.2	0.8	36.0	14.4	4.9	1.9	11.7	8.5	2.2	1.7
BaA	3.6	1.6	0.8	0.5	48.4	29.8	1.6	0.5	4.2	3.6	1.8	1.7
Chr	7.6	5.4	0.8	0.3	18.0	16.7	2.7	1.1	12.7	6.6	2.6	1.6
BbF	17.8	10.4	1.6	0.5	16.3	11.3	3.6	2.0	17.7	10.3	5.6	2.5
BaP	8.9	1.7	1.4	0.3	14.7	4.0	2.9	1.4	6.5	4.4	3.2	5.
Ind	41.8	40.4	1.3	1.0	18.7	4.9	2.2	0.7	14.0	6.8	3.5	1.4
DahA	5.0	2.9	0.8	0.3	9.2	2.0	1.0	0.3	50.5	19.0	2.8	2.2
BghiP	58.5	52.4	1.3	0.7	13.7	6.6	2.9	1.0	10.0	3.4	4.2	2.0
PAHs	492.4	141.3	57.1	11.4	801.7	248.8	81.3	25.7	368.4	121.9	55.6	19.2
[a]∑Com-PAH	166.1	89.3	13.1	2.9	222.3	49.7	33.8	13.9	138.6	36.3	29.8	15.3
[b]∑Car-PAHs	84.6	44.2	6.7	1.9	125.3	47.7	14.1	5.7	105.6	29.1	19.6	11.8

[a] Combustible PAHs includes: Flt, Pyr, BaA, Chr, BbF, BaP, BghiP and Ind.

[b] Carcinogenic PAH includes: BaA, Chr, BbF, BaP, DahA, and Ind

Moreover, the PAH content of outdoor PM varied from 0.3% to 1.1% (mean: 0.5±0.3%) and 0.4 to 1.0% (mean: 0.7±0.2%) for $PM_{2.5}$ and $PM_{0.1}$, respectively. A similar trend in the contents of PM was observed between indoors and outdoors, with the PAH contents of $PM_{0.1}$ being higher than those of $PM_{2.5}$, despite having a lower concentration of $\sum_{15}$PAHs. Accordingly, the proportion of LMW-PAHs bound to $PM_{0.1}$ was higher than $PM_{2.5}$, whereas the contents of HMW-PAHs was the same between $PM_{2.5}$ and $PM_{0.1}$.

Regarding to each house, the levels of $\sum_{15}$PAH, $\sum$com-PAHs and $\sum$car-PAHs in summer and winter are also presented in Tables 6.4 and 6.5. The average $\sum_{15}$PAHs, $\sum$com-PAHs and $\sum$car-PAHs concentrations of outdoor $PM_{2.5}$ and $PM_{0.1}$ in K1, K2 and K3 were 317.1, 482.2 and 234.8 ng/m^3, respectively, and 45.4, 59.4 and 45 ng/m^3, respectively. Accordingly, the average concentrations of $\sum$com-PAHs and $\sum$car-PAHs bound to outdoor $PM_{2.5}$ were 111.8 and 59.53 ng/m^3 (K1); 137.6 and 77.3 ng/m^3 (K2); 88.9 and 74.9 ng/m^3 (K3), respectively. Meanwhile, the corresponding values for outdoor $PM_{0.1}$ were 14.3 and 8.1 ng/m^3 (K1); 24.3 and 10.4ng/m^3 (K2); 22.4 and 15 ng/m^3 (K3), respectively. The $\sum_{15}$PAH concentrations bound to outdoor $PM_{2.5}$ and $PM_{0.1}$ in the three houses varied significantly between summer and winter. However, the concentrations of $\sum_{15}$PAHs bound to $PM_{2.5}$ and $PM_{0.1}$ were almost stable among the three houses in summer. The particulate PAH was the highest at the roadside house (K2) and the lowest particulate PAH concentration was observed at the urban house (K3). The differences in the $\sum_{15}$PAHs concentration could be due to differences in the dominant emission sources of PAH at the three houses. The highest $\sum_{15}$PAH concentrations in $PM_{2.5}$ and $PM_{0.1}$ at K2, indicate that these levels were strongly influenced by traffic sources.

Considering the contribution of individual PAHs to $\sum_{15}$PAH concentration, the most abundant species that were found outdoor air were quite similar to those found in indoor air. The greatest contributions to $\sum_{15}$PAH concentrations in PM were Nap and Phe, which was consistent with a previous study (Krugly et al., 2014). Regarding the distribution of molecular weights in the two-particle sizes, in outdoor air, the concentrations of LMW were higher than those of HMW which was similar with trend in concentrations of different molecular weight indoors. The contribution of LMW-PAHs to $\sum_{15}$PAHs bound to $PM_{2.5}$ ranged from 58.9% to 90.6% (75.2±11.5%), while that to $\sum_{15}$PAHs bound to $PM_{0.1}$ ranged from 65.6 to 88.9% (77.5±9.9%). The average contributions of HMW-PAHs to $\sum_{15}$PAHs bound to $PM_{2.5}$ and $PM_{0.1}$ were 24.8±11.5% and 22.5±9.9%, respectively. However, the contributions of HMW- and LMW-PAHs differed slightly between the two particle sizes indoors and outdoors. Specifically, in the indoor environment, slightly higher percentage of LMW was found in $PM_{2.5}$, whereas that of HMW was higher in $PM_{0.1}$. In contrast, higher percentage of LMW was observed in $PM_{0.1}$, while that of HMW was higher in $PM_{2.5}$ in the outdoor environment. It is likely explanation for differences in source emissions and the contribution of sources emissions. However, it was interesting to underline that, the contents of LMW- and HMW-PAHs bound to $PM_{0.1}$ (LMW- and HMW-PAHs/mass $PM_{0.1}$) were greater than those bound to $PM_{2.5}$ and these contents outdoors were higher than indoors. This suggested that the contents and concentrations of LMW- and HMW-PAHs bound to PM at both particles' sizes were strongly influenced from outdoor sources as main contributor, which was observed at three houses.

3.3. Relationship between indoor and outdoor PAHs bound to PM at three houses

3.3.1. Indoor and outdoor ratios (I/O)

To better understand the distribution of PAHs bound to PM, among the three houses was also evaluated. As shown in Fig. 6.9, the ratios ($\sum_{15}$PAH_PM$_{0.1}$/$\sum_{15}$PAH_PM$_{2.5}$) indoors and outdoors were estimated for the three houses in both seasons. The ratios of indoor PM$_{0.1}$/PM$_{2.5}$ at K1, K2 and K3 were 24.6%, 24.7% and 31.8% in summer and 27.7%, 28.4% and 39.5% in winter, respectively. The corresponding values of outdoor PM$_{0.1}$/PM$_{2.5}$ at K1, K2 and K3 were 23.8%, 23.1% and 33.9% in summer and 11.6%, 10.5% and 15.1% in winter, respectively. In comparison of the ratios ($\sum_{15}$PAH_PM$_{0.1}$/$\sum_{15}$PAH_PM$_{2.5}$) among three houses, the almost similar ratios indoors and outdoors were at K1 and K2, implying that PAHs bound to PM in these houses were originated from the same types of sources according to the season. However, the ratio ($\sum_{15}$PAH_PM$_{0.1}$/$\sum_{15}$PAH_PM$_{2.5}$) at K3 differs from those at K1 and K2, indicating that the kinds of indoor sources and their contributions at K3 are different from the two houses The trend in the PAHs bound to PM at the three houses was similar to that of TEs bound to PM$_{2.5}$ and PM$_{0.1}$ reported by Vo et al. (2022). In addition, the significantly higher ratio of indoor $\sum_{15}$PAH_PM$_{0.1}$ and indoor $\sum_{15}$PAH_PM$_{0.1}$ (indoor $\sum_{15}$PAH_PM$_{0.1}$/$\sum_{15}$PAH_PM$_{2.5}$) compared to ratio of outdoor $\sum_{15}$PAH_PM$_{0.1}$ and outdoor $\sum_{15}$PAH_PM$_{0.1}$ (outdoor $\sum_{15}$PAH_PM$_{0.1}$/$\sum_{15}$PAH_PM$_{2.5}$) in winter suggested that the contribution of $\sum_{15}$PAH bound to PM$_{0.1}$ to $\sum_{15}$PAH bound to PM$_{2.5}$ is greater indoors. In other words, the indoor PAHs_PM$_{0.1}$ was more greatly influenced by indoor sources than PAHs_PM$_{2.5}$. It appears that the daily activities at K3 during the sampling campaign, such as cooking; cleaning; burning incense, candles and camphor; and other indoor activities, may have had an intensive influence on the generation of PM-bound PAHs

Furthermore, the ratios of indoor and outdoor $\sum_{15}$PAHs (I/O) were estimated at the three houses in summer and winter, which described the relationship between indoor and outdoor PAH bound to PM are shown in Fig. 6.10. The red reference line in Fig. 10 shows an I/O ratio of 1. I/O ratios greater than 1 indicate that indoor sources make a significant contribution to indoor PAH, whereas in the absence of strong indoor sources, the ratios were expected to be less than 1. Fig. 10 illustrates that the I/O ratios of $\sum_{15}$PAHs_PM$_{0.1}$ at the three houses varied from 0.5 to 1.1, whereas those of PM$_{2.5}$ ranged from 0.2 to 1.1. In general, the I/O ratios of $\sum_{15}$PAHs_PM$_{0.1}$ were higher than those of $\sum_{15}$PAHs_PM$_{2.5}$ in winter at the three houses, whereas these I/O ratios of the two particle sizes were comparable in summer. Among the three houses, the I/O ratios of both particle sizes were typically less than 1 at all houses, except K3 in summer. The findings indicated that PAHs bound to indoor PM$_{0.1}$ and PM$_{2.5}$ originated mainly from outdoor sources. It should keep in notice that, the I/O ratios of PAHs bound to PM$_{2.5}$ and PM$_{0.1}$ were slightly higher than the unity (i.e., 1), was observed at K3 in summer, suggesting that, there were additional contributions from indoor sources in addition to outdoor sources, The presence of indoor sources with I/O ratios of more that 1 was also reported by Naumova et al. (2002) and Zhang et al. (2019).

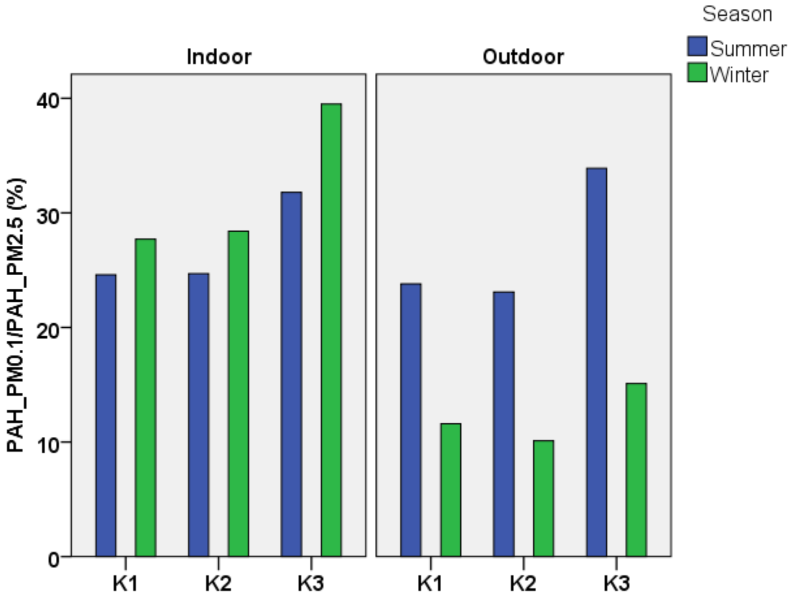


Fig.6. 9. Ratios of PAH_$PM_{0.1}$/PAH_$PM_{2.5}$ at the three houses.

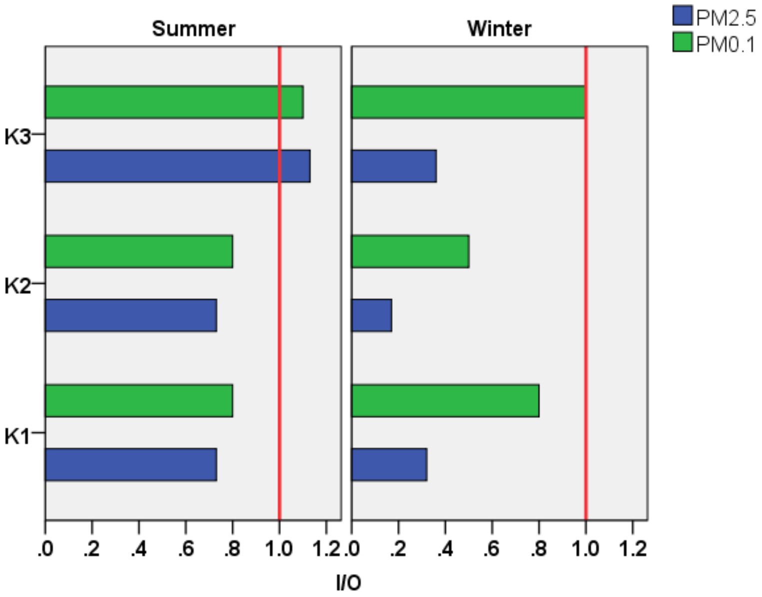


Fig.6. 10. I/O ratios at the three houses.

Furthermore, in comparisons of $\sum_{15}$PAHs I/O ratios between the two particle sizes in winter, the higher $\sum_{15}$PAHs I/O ratios observed for $PM_{0.1}$ compared to $PM_{2.5}$ in the three houses, combined with the higher ratios of indoor $\sum_{15}$PAHs_$PM_{0.1}$ and $\sum_{15}$PAHs_$PM_{2.5}$, than outdoor ratios, indicated that indoor sources appear to have had a greater influence on $\sum_{15}$PAHs_$PM_{0.1}$ than on $\sum_{15}$PAHs_$PM_{2.5}$. On average, there was significant difference in $\sum_{15}$PAHs I/O ratios of $PM_{2.5}$ in two seasons, but no such difference was observed for $PM_{0.1}$. The seasonal trend in the $\sum_{15}$PAHs I/O ratios of PAH bound to $PM_{2.5}$ observed in this study was associated with seasonal variation of outdoor $PM_{2.5}$ in Hanoi, as reported in previous studies (Nguyen et al., 2018; Vo., 2020a). However, the $\sum_{15}$PAHs I/O ratios of $PM_{0.1}$ were stable according to two seasons, which is in line with the seasonal trends in outdoor $PM_{0.1}$ reported by Nguyen al. (2018) and Vo et al. (2022).

Analysis of I/O ratios of the individual PAHs revealed several interesting findings, further considerations of the indoor sources of PAHs. The seasonal trends in the I/O ratios of HMW- and LMW-PAHs in both particle sizes were similar to those of $\sum_{15}$PAH bound to PM in both seasons. The average I/O ratios of LMW- and HMW-PAHs in $PM_{2.5}$ in summer were 0.9 and 0.9, respectively, while those were 0.3 and 0.3 in winter, respectively. Similarly, the corresponding I/O ratios in $PM_{0.1}$ in summer and winter were 0.9 and 1.0, and 0.7 and 1.0, respectively. These ratios were close the unity (≤ 1), indicating that the majority of LMW- and HMW-PAHs that were bound to indoor $PM_{2.5}$ and $PM_{0.1}$ originated from outdoor sources, which were associated with open biomass combustion, traffic activities and domestic coal combustion (Anh et al., 2019; Nghiem et al., 2020; Pham et al., 2019). Noticeably, the I/O ratios of LMW-PAHs were greater than 1 at K1 in summer, and that of HMW-PAHs was greater than 1 at K3 in summer and winter, reflecting the strong influence of indoor activities such as cooking, burning candles and incense, and combustion of coal briquettes. The prevalence of LMW-PAHs was also reported previously (Naumova et al., 2002; Zhu et al., 2009).

In brief comparison with I/O ratios reported for other indoor environments worldwide, the I/O ratios of $\sum_{15}$PAH_$PM_{2.5}$ and $\sum_{15}$PAH_$PM_{0.1}$ in our study were lower than those in residential houses in China (Naumova et al., 2002; Zhu et al., 2019). Zhu et al. (2009) reported that median I/O ratios were greater than 1 in summer, suggesting that indoor sources made significant contribution to indoor particulate PAHs. Instance, the median I/O ratio for Nap, which was associated with use of moth balls, was 2.0 in summer. However, in winter, most of the median I/O ratios of 2-4 rings PAHs were greater than were greater than 1, whereas all median I/O ratios of 5 to 6 rings PAHs less than 1. These findings indicated that indoor HMW_PAH concentrations were controlled by outdoor sources in winter in their study. Naumova et al. (2002) found that I/O ratios of individual PAHs varied considerably depending on the number of the aromatic rings of the PAHs. I/O ratios of LMW-PAHs (2-3 rings) were greater than 1, suggesting these compounds were typical dominated by indoor sources. Addition, median I/O ratios of MMW-and HMW- PAHs (4-7 rings) were lower than 1, implying the strength of outdoor sources for these PAHs. Kim et al. (2014) reported that at seven public facilities (childcare facilities, underground subway stations, internet cafés, movie theaters, terminal waiting rooms, supermarket, elderly care) in Korea, the I/O ratios of Nap and Phe were considerably higher than unity, which were indicative of strong contribution of indoor sources, such as gas utilities, smoking and burning incense. Instance, the I/O of Nap was highest at childcare facilities, followed by underground subway stations and internet cafés, whereas the I/O ratios of Phe were highest in the movie theaters and in the underground subway stations.

3.3.2. Correlations between indoor and outdoor PAH-bound PM

The indoor versus outdoor concentrations of 15 PAHs bound to PM were investigated at the three houses in summer and winter, and the results are shown in Fig. 6.11. For $PM_{2.5}$, there was an inconsistent correlation between LMW- and HMW-PAHs at urban periphery house (K1) in two seasons. A good correlation was observed between indoor LMW-PAHs and outdoor LMW-PAHs bound to $PM_{2.5}$ in summer ($R^2=0.9$), whereas indoor HMW-PAHs correlated strongly with outdoor HMW-PAHs in winter ($R^2=0.97$). These results indicated that indoor LMW-PAHs or HMW-PAHs bound $PM_{2.5}$ were strongly influenced by outdoor sources, such as transport and industrial activities, biomass burning and domestic coal combustion (Anh et al., 2019; Liaud et al., 2021; Nghiem et al., 2020; Pham et al., 2019). Liaud et al. (2021) reported that LMW-PAHs, including phenanthrene (Phe), pyrene (Pyr) and fluoranthene (Flt), were released in diesel emissions and accounted for 25.8% of all PAHs emissions in France in 2012, whereas while 63.3% of PAH emissions were induced by domestic heating achieved by combusting biomass (wood and coal) and 5.5% of PAH emissions were induced by industrial sectors. In addition, HMW-PAHs have been reported to be produced at high temperatures, such as those in gasoline engines (Zheng et al., 2018). For $PM_{0.1}$, no correlations between indoor and outdoor LMW-PAHs and HMW-PAHs were observed in winter (R^2: *0.1 to 0.2*), implying that the PAHs bound to $PM_{0.1}$ indoors were likely derived from different sources and outdoor LMW-or HMW-PAHs did not significantly explain for variability of indoor LMW-or HMW-PAHs. Meanwhile, only a moderate correlation was observed between indoor LMW-PAHs and outdoor LMW-PAHs in summer, suggesting an influence by outdoor sources. LMW-and HMW-PAHs were previously attributed to smoking, cooking, burning candles and incense in previous studies (e.g., Bootdee et al.(2016); Derudi et al. (2012); Lin and Jhuang.(2012)). Bootdee et al.(2016) reported that five-rings PAHs bound to $PM_{2.5}$ (32% to 62%) were dominant in burning incense at shrines. LMW-PAHs, such as Phe and Ant, were used as indicators of indoor sources of electronic devices (Cai et al., 2018). Moreover, BaA, Chr, BbF, BkF and BaP that have a close relationship with the combustion of natural gas and coal briquettes for cooking (Xu et al., 2020) which is common in Vietnamese kitchens, were reported at K1.

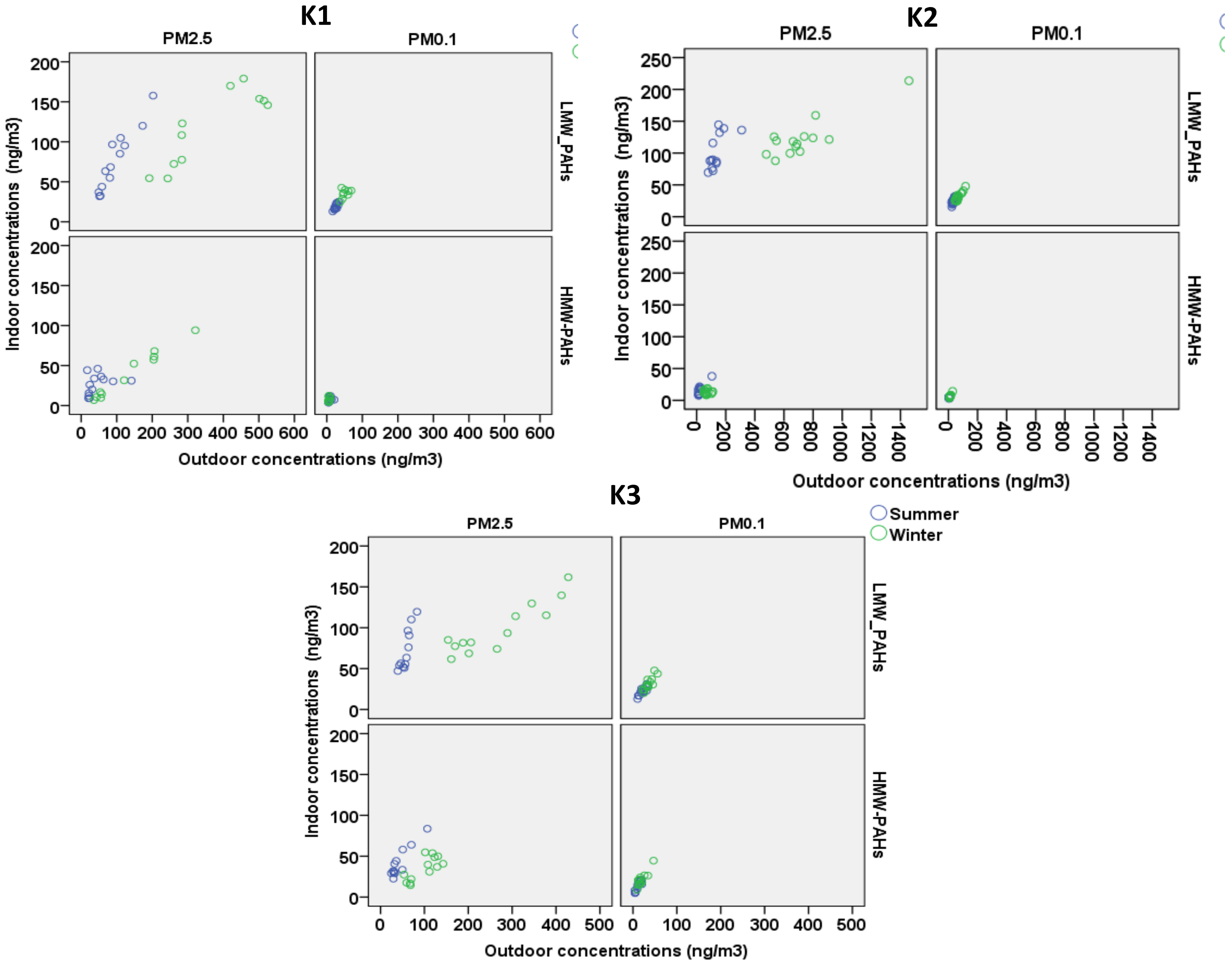


Fig.6. 11. Relationships between indoor and outdoor LMW- and HMW-PAHs at the three houses

Except for HMW-PAHs in summer, the good correlations were observed at roadside house (K2) between indoor and outdoor HMW- and LMW-PAHs in PM$_{0.1}$ in both seasons ($R^2 = 0.7$ to 0.86). These findings suggested that HMW- and LMW-PAHs bound to indoor PM$_{0.1}$ likely predominantly originated from outdoor sources. HMW- PAHs were reported to be formed at higher temperatures, such as those in gasoline engines, than those involved in most indoor combustion processes, such as cooking and smoking (Zheng et al., 2018). However, a different trend was found in the relationship between indoor and outdoor of HMW- and LMW-PAHs in PM$_{2.5}$ between two seasons. Specifically, strong correlations were observed between indoor and outdoor HMW-PAHs in summer ($R^2 = 0.76$) and LMW-PAHs in winter ($R^2 = 0.76$). In contrast, weak correlations were determined between LMW-PAHs in summer and HMW-PAHs in winter ($R^2 = 0.2$ *to* 0.43). LMW-PAHs were associated with indoor sources such as cooking and/or evaporation from indoor materials (Abdullahi et al., 2013; Liaud et al., 2021), whereas HMW-PAHs were reported to be released by domestic fuel combustion for cooking and burning incense (Abdullahi et al., 2013; Lin and Jhuang, 2012; Bootdee et al., 2016).

High correlations between indoor and outdoor HMW- and LMW-PAHs bound to $PM_{2.5}$ were determined in summer and winter at urban house (K3) ($R^2=0.8$ *to* 0.85), except for HMW-PAHs in winter. An analogous trend in the correlation between indoor and outdoor HMW- and LMW-bound $PM_{0.1}$ was observed in both seasons ($R^2=0.73$ *to* 0.83). These findings imply that most indoor particulate LMW- and HMW-PAHs in the two-particle size were controlled by outdoor sources. The moderate correlation between indoor and outdoor HMW-PAHs in winter suggested that these compounds were influenced by indoor sources, which was attributed to cooking and burning incense and scented candles, as reported previously (Bootdee et al., 2016; Derudi et al. 2012; Lin and Jhuang, 2012; Kim et al., 2014).

3.4. Source analysis

3.4.1. Diagnostic ratio (DR)

Isomeric ratios are concentrations of some PAHs that can used as a fingerprint of an emission source. The diagnostic ratios of indoor $PM_{0.1}$ and $PM_{2.5}$ in summer and winter in three houses were quantified and the results are shown in Fig.6.12. The PAH isomeric pair ratios have been extensively used to explain the characteristics of specific source, such as Flt/(Flt+Pyr) vs. BaP/BghiP and BaA/(BaA+Chr) vs. Ind/(Ind+BghiP) (Chen et al., 2017; Kong et al., 2010; Yunker et al., 2002).

Urban periphery house (K1)

The DRs of both $PM_{2.5}$ and $PM_{0.1}$ were estimated in two seasons. In summer, most Flt/(Flt+Pyr) ratios were greater than 0.5, indicating the combustion of residential fuel. These ratios are indicative of the predominance of coal combustion as the source of particulate PAHs (Chen et al., 2017). In addition, most BaP/BghiP ratios from 0.3 to 0.6, suggesting the existence of combusting a mixture of diesel and gasoline, which is typically associated with vehicle emissions (Hassanvand et al., 2015; Ouyang et al., 2020). Accordingly, the Ind/(Ind+BghiP) is a good marker for diesel sources as the DRs ranged between 0.37 to 0.7. However, most BaA/(BaA+Chr) ratios were 0.3 to 0.7, suggesting that combustion was the predominant source. In winter, the majority of the FLt/(Flt+Pyr) ratios ranged from 0.4 to 0.6, which is a good indicator of fuel oil (0.4 to 0.5) and coal and biomass combustion (DR>0.5). However, most BaP/BighP were between 0.3 to 0.5, indicating the existence of diesel vehicles, and ratios of 0.9 to 6.6 indicating the predominance of coal combustion in winter. In addition, the BaA/(BaA+Chr) ratios ranged from 0.3 to 0.6, which is indicative of fuel combustion, such as coal combustion. Ind/(Ind+BghiP) was in the range 0.35 to 0.7 are good markers for diesel source. Thus, it can be summarized that, the combustion of coal was identified as indoor sources, whereas biomass burning and diesel-source related vehicles emissions were considered as major outdoor sources for particulate PAHs in both particle sizes in two seasons at this site.

Urban roadside house (K2)

The DRs of $PM_{2.5}$ and $PM_{0.1}$ were illustrated in Fig.11 and explained as below. In summer, most Ind/Ind+BghiP ratios ranged from 0.2 to 0.5, implying the dominance of a petroleum source, whereas almost BaA/(BaA+Chr) DRs varied from 0.3 to 0.5, which is suggestive of natural gas combustion (Kong et al., 2010). In other DRs, most Flt/(Flt+Pyr) ratios fall from 0.5 to 0.6, indicating the combustion of coal. Moreover, the BaP/BghiP ratios ranged from 0.5 to 1, indicating mixed sources including gasoline combustion (0.5 to 0.6) and coal combustion (0.9 to 6.6) (Hassanvand et al., 2015; Kong et al., 2010). In winter, most BaA/(BaA+Chr) ratios range from 0.4 to 0.8, as indicator of coal combustion. Otherwise, Ind/(Ind+BghiP) ratios varied from 0.4 to 0,6, suggesting diesel combustion from vehicle exhausts (Chen et al., 2017; Yunker et al., 2002). Additionally, the majority of Flt/(Flt+Pyr) ratios varied from 0.5 to 0.6, indicating coal combustion, whereas the BaP/BghiP ratios suggested that coal combustion (0.9 to 6) and gasoline vehicles (0.5 to 0.6) were the sources of PAHs at this house (Chen et al., 2017; Kong et al., 2010). In short summary, natural gas was as identifier of indoor combustion sources, while domestic coal combustion and vehicle exhausts associated with gasoline and diesel vehicles as outdoor sources of particulate PAHs for both particle sizes in this site.

Urban house (K3)

In summer, majority of BaA/(BaA+Chr) ratios varied from 0.36 to 0.5, suggesting combustion associated with natural gas. Ind/(Ind+BghiP) ratios ranged from 0.35 to 0.6, suggesting diesel combustion related to vehicular emissions. In addition, Flt/(Flt+Pyr) ratios mainly ranged from 0.5 to 0.7, indicating a diesel source. BaP/BighP ratios ranged from 0.2 to 0.6, suggesting a mixture of sources, including diesel and gasoline vehicles. A relatively similar pattern in sources was also observed in winter, with BaA/(BaA+Chr) ratios were in range of 0.2-0.35, indicating natural gas combustion (0.35 to 0.5). Meanwhile, Ind/(Ind+BghiP) ratios ranged from 0.4 to 0.6, indicating a diesel source or coal combustion coal burning (>0.5). In addition, the Flt/(Flt+Pyr) DRs ranged from 0.5 to 0.6, as indicator of coal combustion. BaP/BghiP DRs varied from 0.2 to 1.1, implying the combustion of incense (Lin and Jhuang, 2012). It could be concluded that incense and natural gas combustion were the primary sources of PAHs indoors, while coal combustion and traffic emissions were the primary sources of PAHs outdoors at this site.

Overall, the results suggested that major sources of PAHs were domestic coal and biomass combustion, vehicular emissions, burning incense, and natural gas combustion, all of which influenced the particulate PAHs bound to $PM_{2.5}$ and $PM_{0.1}$ concentration to some extent. However, to identify the exact sources, further studies should be conducted on specific sources of PAHs at the three houses by PCA.

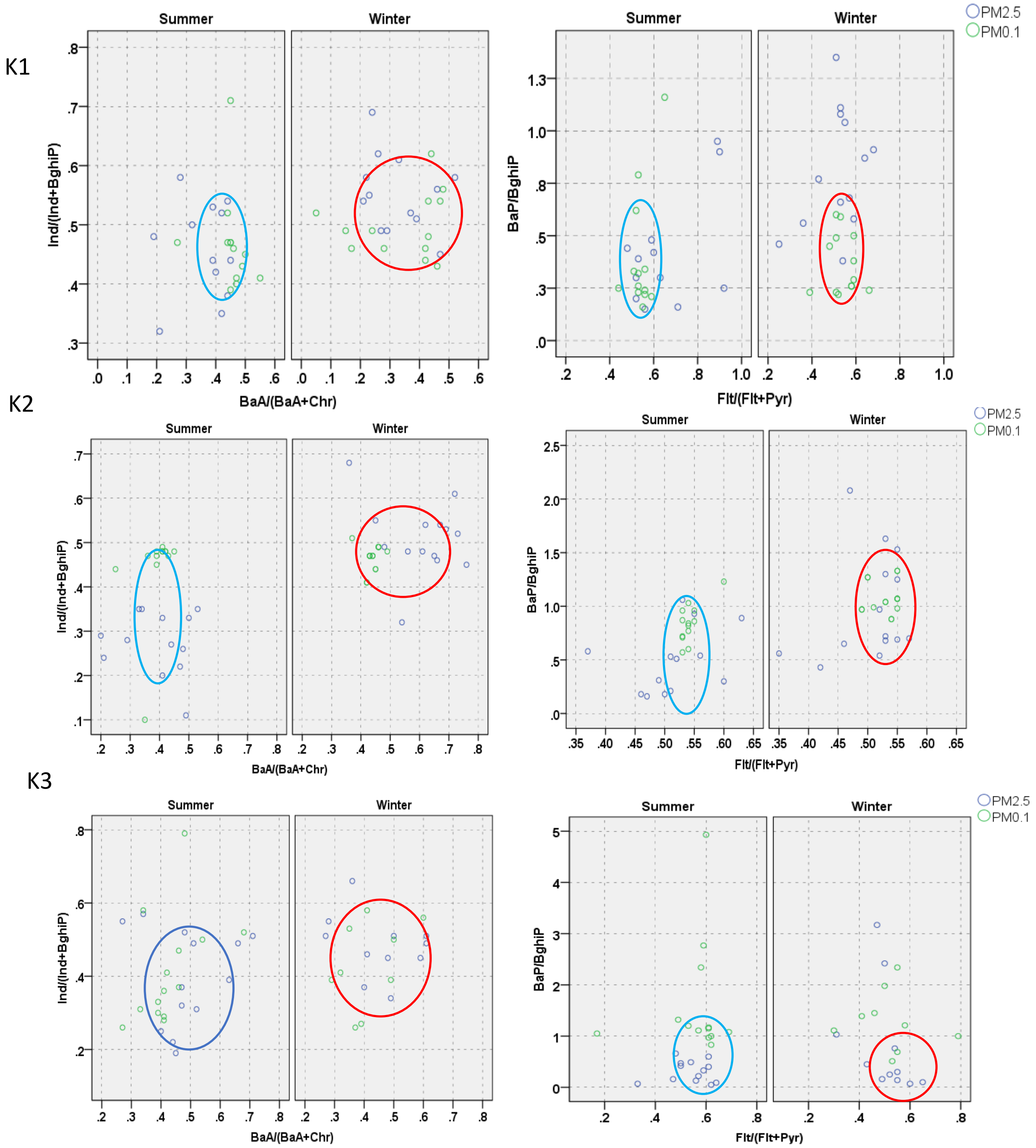


Fig.6. 12. Plots for diagnostic ratios of Flt/(Flt+Pyr) vs. BaP/BghiP and BaA/(BaA+Chr) vs. Ind/(Ind+BghiP) at the three houses in the study

3.4.2. Principal component analysis (PCA)

PCA is used to simplify the interpretations of complex systems and to reduce the number of variables. In addition to assessing DRs between PAHs bound to PM$_{2.5}$ and PM$_{0.1}$ as shown in section 3.4.1, PCA was performed to better clarify the major sources of emissions of PAHs in PM$_{2.5}$ and PM$_{0.1}$ in summer and winter at the three houses. Tables 6.6-6.11 present factor loads obtained by PCA analysis.

Urban periphery house (K1)

In summer, four factors were extracted from PCA that can explain of 82.9% and 87.8% of the source variance sour in $PM_{2.5}$ and $PM_{0.1}$, respectively. For $PM_{2.5,}$ factor 1 and factor 4 explained 47.8% of sources and were highly loaded with Chr, BbF, BaA, and Ind and BghiP, which were interpreted as indicative of vehicle emissions and coal combustion (Chen et al., 2017). Factor 2 accounted for 19.6% of biomass combustion (Pham et al., 2019; Ouyang et al., 2020). Factor 3 accounted for 15.5% of indoor sources, which were derived from indoor cooking (Abdullahi et al., 2013), mothball usage (Zhu et al., 2009), and evaporation from building material (Krugly et al., 2014). Accordingly, for $PM_{0.1}$, factors 1 and factor 3 interpreted 58.8% of sources. These factors were loaded with Phe, Pyr, BbF, BaP, BghiP Ind, which are markers of vehicle emissions and coal combustion (Chen et al., 2017; Kong et al., 2010; Ouyang et al., 2020). Factor 2 accounted for 18.1% of indoor sources of $PM_{0.1}$, which was attributed to indoor cooking and evaporation of building materials (Krugly et al., 2014). Factor 4 accounted for 10.9% of biomass combustion. In winter, PCA extracted four factors for $PM_{2.5}$ and five factors for $PM_{0.1}$, which could explain 86.8% and 93.2% of their sources, respectively. For $PM_{2.5}$, coal combustion and vehicle emissions were responsible for 60.3% of sources**,** extracted from factor 1 and 2, and 10.9% of biomass burning found in factor 4, defined as outdoor sources. Indoor sources, including indoor cooking, usage of mothballs in closets and the evaporation of building materials, made up 15.6%, which was extracted in factor 3. For $PM_{0.1}$, factor 1 and factor 2 accounted for 52.5% of coal combustion and vehicle emissions, while factors 3 and 5 can explain for 28.1% of biomass burning. Factor 4 accounted for a total of 12.7% of indoor sources, which were ascribed to indoor cooking and electronic devices, and evaporation from building materials (Cai et al., 2018; Krugly et al., 2014). In short conclusion in summer, outdoor sources, such as domestic coal and biomass combustion, as well as vehicle emissions, were responsible for 67.4% and 69.7% of $PM_{2.5}$ and $PM_{0.1}$ in summer, respectively. While, indoor sources, indoor cooking, mothballs, electronic devices, and evaporation of building materials, accounted for 15.5% and 18.1% of $PM_{2.5}$ and $PM_{0.1}$, respectively. In winter, the same source pattern was defined observed for both $PM_{2.5}$ and $PM_{0.1,}$ with 71.2% of outdoor sources and 15.6% of indoor sources were explained for $PM_{2.5}$, and 80.5% of outdoor sources and 12.7% of indoor sources were explained for $PM_{0.1}$. These results confirmed that outdoor sources (e.g., vehicular emissions, combustion of coal and biomass) were the dominant sources at this house, which is close to Ring Road #2 (upper) and an embankment road with a high density of passenger vehicles, buses, and heavy trucks. Indoor PM at this house was also affected by the combustion of coal briquettes for domestic cooking in the house itself (indoor source) and surrounding households (outdoor sources), since coal briquettes are still used in food stalls, small restaurants, and households in Hanoi (Vo et al., 2020c). Burning coal briquettes in Hanoi contributes to 17.5% of ambient $PM_{2.5}$ according to Cohen et al. (2010), 16.0% of ambient $PM_{2.5}$ according to Cao and Nguyen (2013), and 12.3% of ambient $PM_{0.1}$ according to Nghiem et al. (2020). In addition, this house was also influenced by biomass combustion during the summer-autumn crop harvests and winter-spring crop harvests during sampling. These two biomass (rice straw) combustion events occur annually in the northern areas of the city (Pham et al., 2019). Burning biomass in Hanoi is responsible for 13% of outdoor $PM_{2.5}$, according Cohen et al. (2010).

Urban roadside house (K2)

In summer PCA extracted four factors for $PM_{2.5}$ and three factors for $PM_{0.1}$ which could explain of 89.5% of $PM_{2.5}$ and 85.2% of $PM_{0.1}$. For $PM_{2.5}$, 68% of vehicle emissions (Flt, Pyr, BaA, Chr, BbF, BaP, DahA, BghiP) were defined in factors 1 and 2 (Chen et al., 2017; Han et al., 2015; Kong et al., 2010), whereas factor 3 and 4 accounted for 21,5% of indoor sources, including indoor cooking, evaporation from building materials and use of electronic devices (flu, Ant, Acy). In case of $PM_{0.1}$, factor 1 was responsible for 50.2% of vehicle emissions, while factor 2 accounted for 21.2% of coal combustion. Factor 3 suggested 13.8% of mothballs and insect repellents used as indoor sources. In winter, three factors for $PM_{2.5}$ and three factors for $PM_{0.1}$ were extracted with high loading of 76.9% and 88.7% variance. For $PM_{2.5}$, factor 1 included DahA, Ind and BghiP as markers of diesel and gasoline exhaust was responsible for 32.4% of vehicle emissions (Kondg et al., 2010; Li et al., 2017; Yunker et al., 2002), while factor 2 accounted for 25.7% of coal combustion (Flt, BaA and Chr), and factor 3 accounted for 18.8% of indoor sources associated with indoor cooking, evaporation from building materials, and electronic devices. A similar source pattern was also obtained for $PM_{0.1}$. Factor 1 can explain for 60.2% of vehicle emissions and factor 2 can explain for 17.6% of coal combustion. Accordingly, indoor sources derived from indoor cooking, electronic devices, evaporation from building materials accounted for 10.9%. In overall, coal combustion and vehicle emissions were considered as outdoor sources of PM at this house, which vehicle emissions contributed as the major source of $PM_{2.5}$ and $PM_{0.1}$ in both seasons. Example: vehicle emissions accounted for 68% and 50.2% for $PM_{2.5}$ and $PM_{0.1}$ in summer, respectively, whereas they were also made up 32.4% and 60.2% in winter, respectively. The dominance of vehicle emissions in this house was also confirmed by an earlier study of Vo et al. (2022), which was consistent with previous studies, i.e., that traffic activities in Hanoi contributed to 46.5% of ambient $PM_{0.1}$ (Nghiem et al., 2020) and 40.0% of ambient $PM_{2.5}$ (Cohen et al., 2010). The predominance of traffic emissions at the roadside house can be explained by the strong influence of high traffic loads in the vicinity, particularly light- and heavy-duty vehicles on Upper Ring Road #3, heavy traffic at Nguyen Huu Tho and Giai Phong roads and Nuoc Ngam and Giap Bat bus stations. However, combustion of coal briquettes occurs routinely at nearby eateries and small restaurants, which also affected this house.

Urban house (K3)

In summer, three factors can interpret 87.3% source of $PM_{2.5}$ sources, which were comprised of 50.9% of burning incense and scented candles (Phe, Flt, Chr, BbF, DahA, Ind, BghiP) extracted in factor 1(Bootdee et al., 2016; Derudi et al., 2012; Lin and Jhuang, 2012), and 23.6% of mixed sources (camphor, natural gas combustion) (Nap, Acy, Flu) (Abdullahi et al., 2013; Chen et al., 2017; Zhu et al., 2009) found in factor 2, and 12.8% of vehicle emissions and coal combustion (Pyr, BaA and BaP) factor 3. A slightly different apportionment of sources was observed for $PM_{0.1}$, for which 48.4% of vehicle emissions and domestic coal combustion were accounted for by factor 1 and 33.3% of mixed indoor sources (combustion of incense, scented candles, natural gas and use of camphor) were accounted for by factors 2 and 3. The similar sources were also found in winter for both particle sizes, although the contributions of these sources varied. for $PM_{2.5}$, factor 1 and factor 3 were responsible for 45.7% of indoor sources such as incense/candle combustion (Phe, BbF, BaP, Ind, BghiP) and 14.3% of natural gas combustion, whereas factor 2 was attributed to 24% of vehicle emissions and domestic coal combustion (Flt, BaA, Chr, DahA). The same sources were also extracted in $PM_{0.1}$, which included 57.3% of mixed indoor sources, such as the combustion of natural gas (indoor cooking), incense and candles; and 26.3% of outdoor sources, such as vehicle emissions and domestic coal combustion. It is worthy to underline that, indoor sources play a significant role in this house in which they were responsible for 74.5%, and 33.3% for $PM_{2.5}$ and $PM_{0.1}$ in summer, respectively, and 60% and 57.3% in winter, respectively. Similarly, outdoor sources accounted for 12.8%, and 48.4% of $PM_{2.5}$ and $PM_{0.1}$ in summer, respectively, and 24% and 26.3% in winter, respectively.

Table 6. 6. Factor loading of PCA for $PM_{0.1}$ and $PM_{2.5}$ at K1 in summer

Variables	$PM_{2.5}$				$PM_{0.1.}$			
	Factor 1	Factor 2	Factor 3	Factor 4	Factor 1	Factor 2	Factor 3	Factor 4
Nap			0.89					
Acy			0.75					
Ace			0.93			0.82		
Flu						0.94		
Phe					0.78			
Ant		0.8						
Flt		0.7						0.96
Pyr					0.71			
BaA	0.78							
Chr	0.83							
BbF	0.89				0.74			
BaP		0.81			0.82			
DahA								
Ind				0.89	0.76		0.96	
BghiP				0.94			0.75	
% of variance Sources	34.4	19.6	15.5	13.4	43.4	18.1	15.4	10.9
Sources	Vehicle emissions and coal combustion	Burning biomass	Indoor cooking, mothballs, evaporation from building materials	Vehicle emissions	Vehicle+ coal combustion	Indoor cooking, evaporation from building materials	Vehicle emissions	Burning

Factor loads > 0.7 are shown; Extraction method: Principal Component Analysis; Rotation method: Varimax

Table 6. 7. Factor loading of PCA for $PM_{0.1}$ and $PM_{2.5}$ at K1 in winter

Winter	$PM_{2.5}$				$PM_{0.1}$				
	Factor 1	Factor 2	Factor 3	Factor 4	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5
Nap			0.94						
Acy								0.77	
Ace			0.73						
Flu	0.91								
Phe	0.71							0.95	
Ant	0.83								
Flt				0.89					
Pyr				0.85					0.97
BaA	0.74								
Chr	0.72						0.96		
BbF		0.78			0.86				
BaP					0.95				
DahA						0.93			
Ind		0.97				0.75			
BghiP		0.95			0.77				
% of variance Sources	39.7	20.6	15.6	10.9	30.84	21.62	16.26	12.67	11.80
Sources	Coal combustion and vehicle	Vehicle emissions	Indoor cooking, mothballs, evaporation from building materials	Burning biomass	Coal combustion and vehicle emissions	Vehicle emissions	Burning biomass	Indoor cooking, evaporation from building materials	Burning biomass

Factor loads > 0.7 are shown; Extraction method: Principal Component Analysis; Rotation method: Varimax

Table 6. 8. Factor loading of PCA for $PM_{0.1}$ and $PM_{2.5}$ at K2 in summer

	$PM_{2.5}$				$PM_{0.1}$		
Summer	Factor 1	Factor 2	Factor 3	Factor 4	Factor 1	Factor 2	Factor 3
Nap							0.95
Acy				0.99			
Ace						0.84	
Flu			0.86			0.81	
Phe							
Ant			0.88			0.84	
Flt	0.96				0.95		
Pyr	0.83				0.82		
BaA	0.82					0.78	
Chr	0.79				0.85		
BbF	0.73					0.72	
BaP	0.93				0.91		
DahA		0.97					
Ind							
BghiP		0.86			0.94		
% of variance Sources	51.8	16.2	12.2	9.3	50.2	21.2	13.8
Sources	Vehicle emissions	Vehicle emissions	Indoor cooking, evaporation from building materials, electronic devices	Electronic devices	Vehicle emissions	Coal combustion	Moth balls, insect repellents

Factor loads > 0.7 are shown; Extraction method: Principal Component Analysis; Rotation method: Varimax

Table 6. 9. Factor loading of PCA for $PM_{0.1}$ and $PM_{2.5}$ at K2 in winter

Winter	$PM_{2.5}$			$PM_{0.1}$		
	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 3
Nap						0.86
Acy			0.88	0.86		
Ace						0.86
Flu			0.88	0.79		
Phe						0.74
Ant			0.85	0.79		
Flt		0.73		0.75		
Pyr				0.9		
BaA		0.94			0.87	
Chr		0.84			0.92	
BbF					0.9	
BaP					0.42	
DahA	0.73			0.78		
Ind	0.9			0.74		
BghiP	0.88					
% of variance Sources	32.4	25.7	18.8	60.2	17.6	10.9
Sources	Vehicle emissions	Coal combustion	Indoor cooking, evaporation from building materials, electronic devices	Vehicle emissions	Coal combustion	Mothballs and insect repellents

Factor loads > 0.7 are shown; Extraction method: Principal Component Analysis; Rotation method: Varimax

Table 6. 10. Factor loading of PCA for $PM_{0.1}$ and $PM_{2.5}$ at K3 in summer

	$PM_{2.5}$			$PM_{0.1}$		
Summer	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 3
Nap		0.88			0.78	
Acy		0.79				
Ace						
Flu		0.91				
Phe	0.79					0.82
Ant						
Flt	0.89					0.96
Pyr			0.74			0.76
BaA			0.95			
Chr	0.79				0.88	
BbF	0.94				0.94	
BaP			0.81	0.85		
DahA	0.98			0.85		
Ind	0.97			0.95		
BghiP	0.81			0.93		
% of variance Sources	50.9	23.6	12.8	48.4	22.8	10.5
Sources	Burning incense and candles	Camphor and natural gas (indoor cooking)	Vehicle emissions and coal combustion	Vehicle emissions and coal combustion	Burning incense, candles and camphor	Natural gas combustion (indoor cooking)

Factor loads > 0.7 are shown; Extraction method: Principal Component Analysis; Rotation method: Varimax

Table 6. 11. Factor loading of PCA for $PM_{0.1}$ and $PM_{2.5}$ at K3 in winter

Winter	$PM_{2.5}$			$PM_{0.1}$	
	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2
Nap					
Acy					0.87
Ace					0.96
Flu			0.89	0.73	
Phe	0.88				0.91
Ant			0.79		
Flt		0.84		0.84	
Pyr			0.74		
BaA		0.96		0.97	
Chr		0.77		0.98	
BbF	0.91				0.77
BaP	0.88			0.82	
DahA		0.72		0.86	
Ind	0.87			0.88	
BghiP	0.92			0.93	
% of variance Sources	45.7	24.	14.3	57.3	26.3
Sources	Burning incense and candles	Vehicle emissions and coal combustion	Natural gas combustion	Incense and candle natural gas	Vehicle emissions and coal combustion

Factor loads > 0.7 are shown; Extraction method: Principal Component Analysis; Rotation method: Varimax

3.5. Estimation of deposited doses in human respiratory tract (HRT)

The total deposited dose of $\sum BaP_{eq}$ and $\sum_{15}$PAHs were estimated using equation (3-5), which is a sum of the deposited doses in five respiratory regions ($EDI_{ET1,}$ EDI_{ET2}, EDI_{BB}, EDI_{bb}, and EDI_{AL}). The values of these deposited doses of the two particle sizes in the different age categories of inhabitants at the three houses from indoor and outdoor air in two seasons are shown in Tables 6.12 and 6.13 and Fig. 6.13 and 6.14. The age categories were classified into seven groups, namely infants (0–1y), toddlers (1–3y), preschool children (3–6y), children (6–11y), adolescents (11–21y), adults (21–60y), and elderly adults (>60 y). Regardless of the investigated houses, the average indoor $\sum$15PAHs-EDI values for $PM_{2.5}$ ranged from 7.2 to 743 ng/day in summer and 9.1 to 943 ng/day in winter, whereas those for $PM_{0.1}$ varied from 0.8 to 79.3 ng/day in summer_and 1.2 to 118 ng/day in winter, respectively. Meanwhile, outdoor $\sum_{15}$PAHs-EDIs bound to $PM_{2.5}$ ranged from 0.6 to 121 ng/day in summer and 2.4 to 497 ng/day in winter, while those bound to $PM_{0.1}$ varied from 0.1 to 12.4 ng/day in summer and 0.1 to 22.8 ng/day in winter, respectively. There were significant differences in deposited doses between indoor $\sum_{15}$PAHs and $\sum BaP_{eq}$ in three houses between seasons and particles. For example, the EDI of $\sum BaP_{(eq)}$ bound to indoor $PM_{2.5}$ decreased dramatically by 4.1 to 17.8 times in summer and 6.7 to 20.4 times in winter, respectively, and for indoor $PM_{0.1}$, values decreased by 5.1 to 14.6 times and 7.4 to 13.7 times in comparison with $\sum_{15}$PAHs_ EDIs in this study. The total deposited doses of 16PAH bound $PM_{2.5}$ (5.9 ng/h) in indoor environment in China (Zhang et al., 2012) and those of 9PAHs bound to $PM_{2.5}$ (950 pg/h, and 560 ng/h) in roadside and suburban site, respectively, in Japan (Kawanaka et al., 2009) were calculated. The deposited doses of 15 PAH bound to $PM_{2.5}$ for infants and toddlers in our study were lower than those calculated in China and Japan, but our results calculated for adult and elderly groups were significantly higher. In comparison with outdoor deposited doses between $\sum_{15}$PAHs and $\sum BaP_{eq}$, EDI of $\sum BaP_{(eq)}$ for $PM_{2.5}$ decreased in the range of 4.1 to 16.6 times in summer and from 6 to 24 times in winter, and for $PM_{0.1}$, the decrease ranged from 7.1 to 16.9 times in summer and 7.6 to 21.8 times in winter. In other context, in comparison with outdoor EDIs and indoor EDIs, the indoor $\sum_{15}$PAHs-EDIs for $PM_{2.5}$ increased 2.5 to 12.4 times in summer and 1.2 to 3.8 times in winter, whereas EDIs of $\sum_{15}$PAHs bound to $PM_{0.1}$ increased 3.5 to 12.9 times in summer and 2.8 to 10.4 times in winter. The same trend was also observed for EDIs of $\sum_{BaP(eq)}$ bound to PM between indoor and outdoor air. The indoor EDI of $\sum BaP_{(eq)}$ of $PM_{2.5}$ raised 3.7 to 13.5 times in summer and 1.2 to 4.2 times in winter, while indoor EDI of $\sum BaP_{(eq)\ bound}$ to $PM_{0.1}$ raised 4.7 to 17.2 in summer and 3.7 to 13.5 times in winter. The significantly greater EDI values indoors were attributed to longer residence time, although the outdoor concentrations of $\sum_{15}$PAHs and $\sum BaP_{(eq)}$ were higher. Among the seven age categories, EDI of $\sum_{15}$PAHs and EDI of $\sum BaP_{(eq)}$ increased significantly as age increased. The elderly adults (>60y) exposed the highest $EDI_{\sum 15PAHs}$ and $EDI_{\sum BaP(eq)}$, whereas children, including infants (0–1 y), toddlers (1–3 y), and preschool children (3–6 y), had the lowest their EDIs during exposure to $\sum_{15}$PAHs and $\sum$BaP$_{(eq)}$ in both particle sizes. This finding is consistent with the earlier finding of Vo et al. (2022). The average EDI$_{\Sigma PAH}$ and EDI$_{BaP(eq)}$ uptakes by elder groups were 103, 71.9, 31.4, 15, 10.2 and 2.8-fold higher than those by infants (0–1y), toddlers (1-3y), preschool children (3–6y), children (6–11y), adolescents (11–21y), and adults (21–60y) during chronic exposure, respectively. Although children experienced the lowest EDIs for both particle sizes, they, especially infants and toddlers, are considered to be vulnerable to the 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 show a high tendency for particle deposition in the lower respiratory tract owing to less efficient uptake of particles in head airways. Therefore, this should be considered when developing public health initiatives focusing on the protection of children and elderly adults

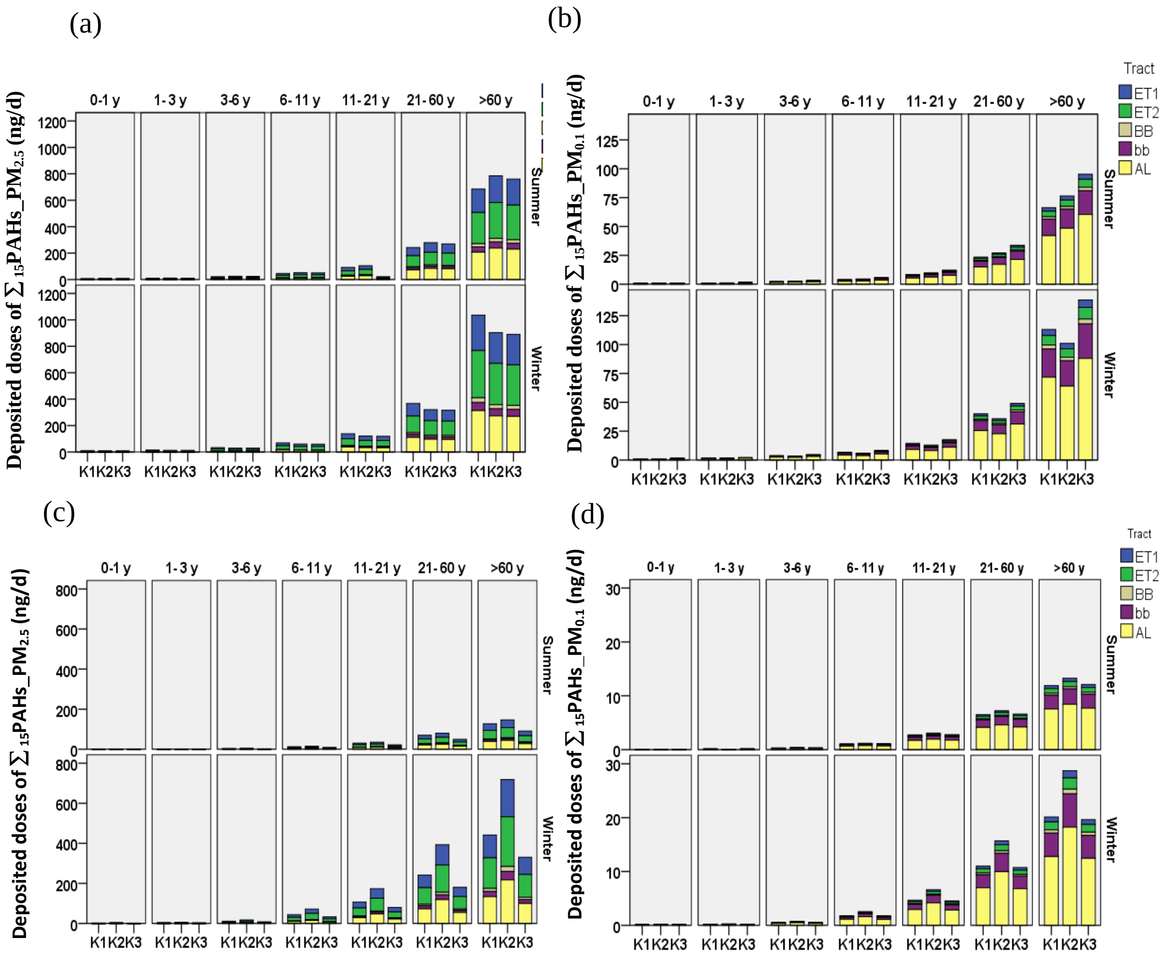


Fig.6. 13. Deposited doses of PAH-bound to PM: (a) PM$_{2.5}$ (indoor) (b) PM$_{0.1}$ (indoor); c) PM$_{2.5}$ (outdoor); d) PM$_{0.1}$ (outdoor) in different respiratory tract regions (anterior nasal region (ET1), main extra-thoracic region (ET2), bronchial region (BB), bronchiolar region (bb), and alveolar interstitial region (AL).

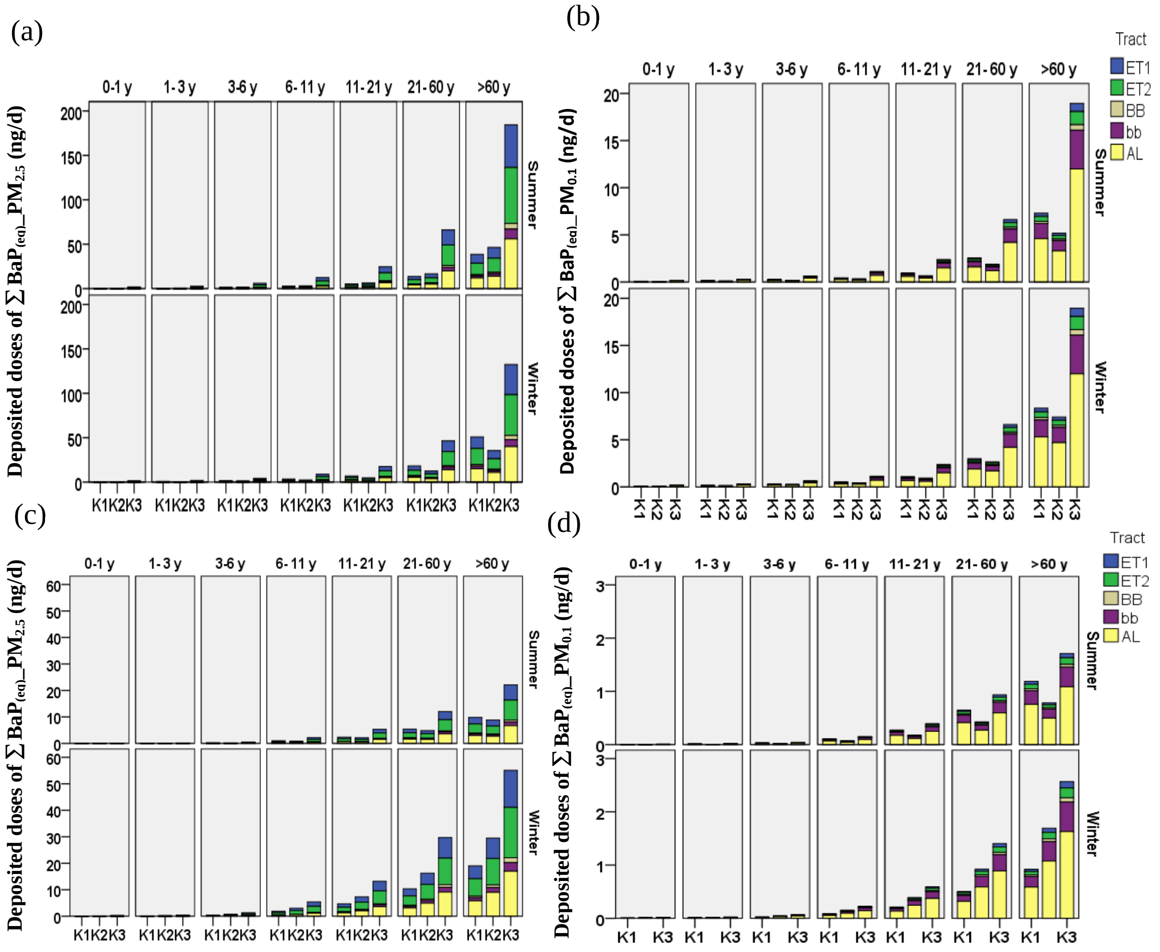


Fig.6. 14. Deposited doses of BaP(eq)-bound to PM: (a) $PM_{2.5}$ (indoor), (b) $PM_{0.1}$, (indoor), c) $PM_{2.5}$ (outdoor), d) $PM_{0.1}$ (outdoor) in different respiratory tract regions (anterior nasal region (ET1), main extra-thoracic region (ET2), bronchial region (BB), bronchiolar region (bb), and alveolar interstitial region (AL).

The $EDI_{BaP(eq)}$ for both $PM_{0.1}$ and $PM_{2.5}$ were estimated in the five respiratory tract regions (TE1, ET2, BB, bb, AL) of the occupants in the three houses, and are shown in Fig. 6.14. Deposited doses varied across the different HRT regions, which was related to PM properties (concentration and particle size) and HRT physiology (structure and morphology) (Gao et al., 2017; ICRP, 1994; Kawanaka et al., 2009)

Table 6. 12. Deposited doses of $\sum_{15}$PAHs bound to PM in different age categories at the three sites (ng/day)

	K1				K2				K3			
	Indoor		Outdoor		Indoor		Outdoor		Indoor		Outdoor	
	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$
Summer												
0-1 y	6.6	0.7	0.6	0.1	7.6	0.8	0.7	0.1	7.4	0.9	0.4	0.1
1-3 y	9.5	1.0	1.0	0.1	10.9	1.2	1.2	0.1	10.6	1.4	0.7	0.1
3-6 y	21.8	2.2	3.1	0.3	25.0	2.5	3.6	0.3	24.2	3.2	2.2	0.3
6-11 y	45.6	3.9	12.7	1.1	52.4	4.5	14.6	1.2	50.6	5.6	9.1	1.1
11-21 y	91.3	8.5	30.6	2.7	104.8	9.8	35.2	3.1	21.9	12.2	21.9	2.8
21-60 y	242.8	23.5	69.5	6.5	278.7	27.1	79.8	7.3	269.5	33.8	49.7	6.6
>60 y	684.5	66.3	127.1	11.9	785.6	76.4	145.9	13.3	759.7	95.2	90.8	12.1
Winter												
0-1 y	1.1	2.1	0.1	8.7	1.0	3.4	0.1	8.6	1.4	1.6	0.1	1.1
1-3 y	1.7	3.5	0.2	12.6	1.5	5.7	0.2	12.4	2.1	2.6	0.2	1.7
3-6 y	3.8	10.8	0.5	28.8	3.4	17.6	0.7	28.4	4.6	8.1	0.5	3.8
6-11 y	6.7	44.0	1.8	60.2	6.0	71.7	2.5	59.3	8.2	32.9	1.7	6.7
11-21 y	14.4	106.4	4.6	120.5	12.9	173.3	6.6	118.7	17.7	79.6	4.5	14.4
21-60 y	40.1	241.5	11.0	320.5	35.8	393.1	15.7	315.8	49.1	180.7	10.7	40.1
>60 y	113.0	441.4	20.1	903.5	101.0	718.7	28.7	890.3	138.6	330.3	19.6	113.0

Table 6. 13. Deposited dose of $\sum_{BaP(eq)}$ bound to PM in different age categories at the three sites (ng/day)

	K1				K2				K3			
	Indoor		Outdoor		Indoor		Outdoor		Indoor		Outdoor	
Summer	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$	$PM_{2.5}$	$PM_{0.1}$
0-1 y	0.37	0.07	0.05	0.01	0.45	0.05	0.04	0.00	1.78	0.19	0.11	0.01
1-3 y	0.53	0.11	0.08	0.01	0.65	0.08	0.07	0.01	2.56	0.28	0.17	0.01
3-6 y	1.22	0.24	0.24	0.03	1.48	0.17	0.22	0.02	5.86	0.62	0.54	0.04
6-11 y	2.56	0.43	0.98	0.11	3.10	0.31	0.88	0.07	12.26	1.11	2.19	0.15
11-21 y	5.13	0.93	2.38	0.27	6.20	0.67	2.12	0.18	24.53	2.40	5.30	0.39
21-60 y	13.63	2.60	5.39	0.65	16.48	1.86	4.82	0.43	65.23	6.67	12.03	0.94
>60 y	38.44	7.32	9.85	1.19	46.47	5.24	8.81	0.78	183.90	18.81	21.99	1.71
Winter												
0-1 y	0.49	0.08	0.09	0.00	0.34	0.07	0.14	0.01	1.28	0.19	0.26	0.01
1-3 y	0.71	0.13	0.15	0.01	0.49	0.11	0.24	0.01	1.84	0.29	0.44	0.02
3-6 y	1.62	0.28	0.47	0.02	1.12	0.24	0.73	0.04	4.21	0.63	1.34	0.07
6-11 y	3.39	0.50	1.90	0.08	2.33	0.44	2.98	0.15	8.80	1.12	5.47	0.23
11-21 y	6.79	1.07	4.58	0.21	4.67	0.94	7.21	0.39	17.61	2.40	13.22	0.59
21-60 y	18.05	2.97	10.40	0.50	12.43	2.62	16.36	0.93	46.83	6.68	30.01	1.40
>60 y	50.88	8.37	19.01	0.92	35.03	7.37	29.90	1.69	132.02	18.84	54.85	2.57

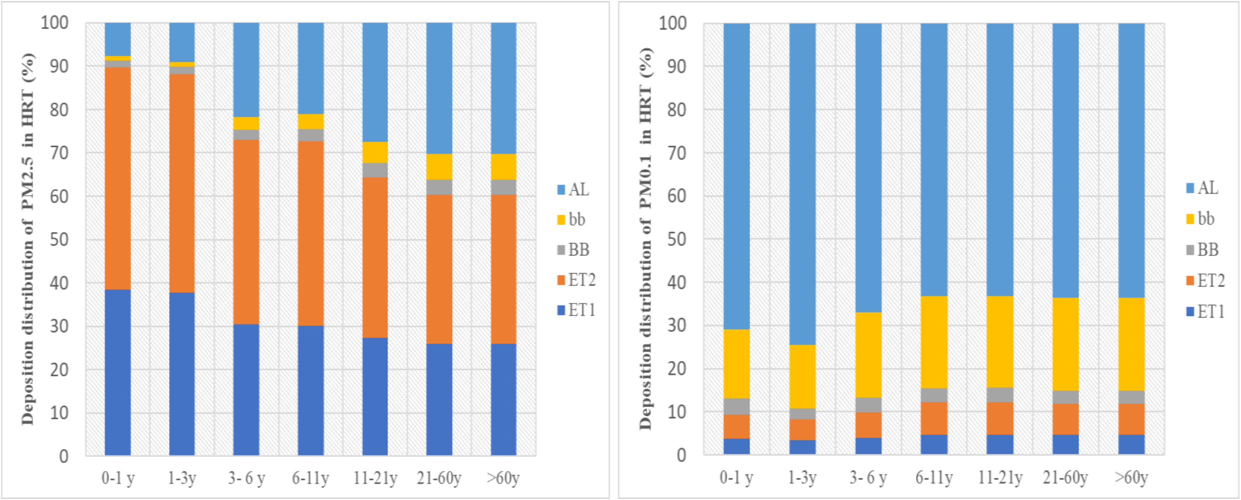


Fig.6. 15. Distribution of dose of PAH and $BaP_{(eq)}$ in the HRT among different age groups.

The distribution of doses of PAH and $BaP_{(eq)}$ in the human respiratory tract were illustrated in Fig. 6.15. In general, the values of EDI of $\sum_{15}PAH$ or $\sum BaP_{(eq)}$ bound to $PM_{2.5}$ were distributed in the range of 26% - 39% in the ET1 region, 35%-51% in the ET2 region, 2%-3% in the BB region, 1%- 6% in the bb region, and 8%-30% in the AL region, whereas those of $\sum_{15}PAH$ or $\sum BaP_{(eq)}$ bound to $PM_{0.1}$, they were distributed as in the range of 4%-5% in the ET1 region, 5%- to 8% in the ET2 region, 3%-4% in the BB region, 15%-22% in the bb region, and 63%-71% in the AL region, respectively. In other words, it can be concluded that EDI values for $PM_{2.5}$ were apportioned in the range of 60%-90% in the HA region, 3%-9% in the TB region, and 8%-30% in the AL region, whereas for $PM_{0.1}$, they were apportioned as in the range of 8%-12% in the HA region, 20%-25% in the TB region and 63%-75% in the AL region.

The proportions of EDI of $\sum_{15}PAH$ or $\sum BaP_{(eq)}$ bound to $PM_{2.5}$ in the different respiratory tract regions in our study were relatively comparable to those of $\sum$TEs bound to $PM_{2.5–1.0}$ whereas corresponding values of $\sum_{15}PAH$ or $\sum BaP_{(eq)}$ bound to $PM_{0.1}$ were similar to those of $\sum$TEs bound to $PM_{<0.25}$ in the study of Sharma and Balasubramanian (2018). These EDI proportions were also relatively consistent with an earlier study by Vo et al.(2022), who reported that EDI values for $PM_{2.5}$ were distributed as 80% in the HA region, 7% in the TB region, and 13% in the AL region, whereas those for $PM_{0.1}$ distributed as 11% in the HA region, 14% in the TB region, and 75% in the AL region during the inhalation of trace elements bound to PM. It is worthy to emphasis that the deposition of $\sum_{15}PAHs$ and $\sum BaP_{(eq)}$ bound to $PM_{2.5}$ were predominantly observed in the HA region, followed by the AL and TB regions, whereas the majority of $\sum_{15}PAHs$ and $\sum BaP_{(eq)}$ bound to $PM_{0.1}$ were deposited in the AL region, followed by the TB and HA regions. Furthermore, higher proportions of doses deposited in the HA region were observed in the infants (0-1y) and toddlers (1-3y), whereas higher proportions of doses deposited in the AL region were observed in the adults (21–60 y) and elderly adults (>60 y) during inhalation of $PM_{2.5}$. In contrast, higher doses were deposited in the AL region in infants (0-1y) and toddlers (1-3y) due to intake of $PM_{0.1}$. The differences in thoracic deposition between adults and children (3-8 y) was also reported by Sarigiannis et al. (2015), who stated observed that the $PM_{2.5}$ and PM_1 fractions, rather than the PM_{10} fraction, play a greater role in the adsorption of PAHs by the respiratory tract in younger individuals than in adults. The greater deposition of $PM_{0.1}$ in the AL region was due to the flow path of air in the HRT, which is primarily governed by Brownian motion, thereby leading to preferential deposition in AL (Adachi, 2018; Chatoutsidou et al., 2015; Sharma and Balasubramanian, 2018). Similar results have been reported in several previous studies. For example, Vu et al. (2017) reported that $PM_{0.1}$ is deposited predominantly in the AL region, followed by the TB region. Madureira et al. (2020) reported that PM_{10} is deposited primarily in the HA region (86–97%), followed by $PM_{2.5}$ (60–75%) and $PM_{0.1}$ (31–40%), whereas $PM_{0.1}$ deposition mainly occurs in the AL region. Thus, it could be underline that the EDI in the HA region decreased as the particle size decreased, whereas that in the TB and AL regions increased as the particle size decreased. The majority of $\sum_{BaP(eq)}$-$PM_{0.1}$ intrudes deeply into the AL region, severely damaging human health. Kawanaka et al.(2009) reported that as much as 18%-19% and 16%-17% of inhaled particulate PAHs in urban and suburban areas, respectively, were deposited in the alveoli, whereas Venkataraman and Raymond (1998) estimated that 23%–28% of EDIs of particulate PAHs was deposited in the pulmonary lung and 4%-5% was deposited in tracheobronchial region. Zhang et al. (2012) found that 8.6%–10.2% of $\sum_{16}$PAH was deposited in the alveoli, with accumulation particles as the largest contributor. Specially, ultrafine particles contributed 0.4–21.7% of PAHs deposited in the alveoli. The difference between these reported deposition efficiencies in the alveoli and the present results can be attributed to the particle size distributions of PAHs.

3.6. Health risk assessment

3.6.1. Mutagenicity and carcinogenicity of PAHs

In this study, the concentrations of MEQ and TEQ were estimated in three houses in two seasons for both particle sizes. The average levels of indoor TEQ bound to $PM_{2.5}$ and $PM_{0.1}$ were in the range of 5.3 to 27.6 ng/m^3 and 2.0 to 7.2 ng/m^3, respectively whereas those of outdoor values varied from 9.8 to 61.2 ng/m^3 and 2.2 to 7.3 ng/m^3, respectively. Similarly, indoor MEQ concentrations ranged from 5.6 to 13.2 ng/m^3 and 3.1 to 6.8 ng/m^3 in indoor $PM_{2.5}$ and $PM_{0.1}$, respectively, whereas those of outdoor MEQ ranged from 10.4 to 39.2 ng/m^3 and 2.4 to 7.5 ng/m^3, respectively. The values obtained for TEQ and MEQ varied considerably among the three houses and between seasons. These disparities were attributed to differences in source emissions and in the distribution of individual PAHs among the three houses in both seasons. Regardless the sampled houses, the mean concentration of indoor TEQ was 12.2±9.3 ng/m^3 and 4.2±2.4 ng/m^3 for $PM_{2.5}$ and $PM_{0.1}$, respectively, and the corresponding values for indoor MEQ was 9.6±3.3 ng/m^3 and 4.1±1.9 ng/m^3, respectively. Thus, the mean outdoor TEQ and MEQ concentrations increased more than twice for $PM_{2.5}$, while those of $PM_{0.1}$ showed a negligible difference between indoor and outdoor environments; these findings followed the trend observed in the variation of $PM_{2.5}$ and $PM_{0.1}$. The significant discrepancies were observed for the mean values of outdoor TEQ and MEQ bound to $PM_{2.5}$ and $PM_{0.1}$ between two seasons, which was attributed to seasonal variation in outdoor PM- bound $\sum_{15}$PAHs. Compared to previous studies, the mean concentrations of TEQ and MEQ bound to $PM_{2.5}$ indoors and outdoors in residential houses in this study were significantly lower than those obtained in nursery schools in Hanoi (Vo et al., 2020b), whereas our results were higher than given results in urban and rural sites in Silesian kindergartens, Poland in study of Błaszczyk et al.(2017b), who measured that indoor MEQ concentrations bound to $PM_{2.5}$ were 6.7 ng/m^3 and 6.1 ng/m^3, in urban and rural sites while outdoor MEQ concentrations were 8.4 ng/m^3 and 6.0 ng/m^3, respectively. The levels of TEQ bound to $PM_{2.5}$ were 6.4 ng/m^3 and 4.6 ng/m^3 in outdoor air, and 5.2 ng/m^3 and 4.5 ng/m^3 in indoor air in urban and rural areas, respectively. Additionally, significantly lower MEQ and TEQ concentrations were also found in indoor environments in France, Italy and Korea, which was related to lower background environment (Kim et al., 2014; Liaud et al., 2014; Romagnoli et al., 2014). Higher concentrations of MEQ and TEQ bound to $PM_{2.5}$ indoors and outdoors were measured in Tehran, which may be related to heavy vehicular traffic flow and emissions from nearby industries (Hassanvand et al., 2015), while sustainably greater TEQ and MEQ values in rural homes in North China and North India was attributed to domestic fuel combustion (Ansari et al., 2010; Zhang et al., 2019)

The health risk assessment of carcinogenic and mutagenic PAHs considers not only individual concentrations of PAH, but also the carcinogenic and mutagenic potentials of each compound. The relative mutagenic and carcinogenic contributions of individual PAH to total mutagenicity and carcinogenicity in indoor and outdoor environments are calculated using equations (6-7) as shown in Fig. 6.16 and Fig.6.17. In this study, importantly, the most abundant individual PAHs indoors and outdoors are Nap, Phe and Flt in $PM_{2.5}$ and $PM_{0.1}$, as discussed in section 3.1.2. However, the contribution of these PAH species to the total carcinogenicity of a PAH mixture is negligible due to toxicological characteristics. Fig.6.16 shows the most important PAH species that contribute to the total carcinogenicity of a PAH mixture in the two particle sizes. Regardless of the sampled houses, there was a difference in the distribution pattern of individual PAHs between $PM_{2.5}$ and $PM_{0.1}$ indoors and outdoors. The species that contributed most to the carcinogenicity of a PAH mixture for $PM_{2.5}$ in indoor air were DahA (50.7±26%), followed by BaP (31.4±16.3%), and were BaP (49.6±13.9%) followed by DahA (35.5±14.8%) for $PM_{0.1}$. A similar trend was also observed in outdoor air, where DahA (46.9±25.8%) and BaP (33.0±11.2%) in $PM_{2.5}$, and BaP (47.9±11.4%) and DahA (35.2±13.8%) in $PM_{0.1}$ were the most abundant individual PAHs that contributed to the total carcinogenicity of PAHs. These findings were relatively similar with previous studies (Błaszczyk et al., 2017b; Delgado-Saborit et al., 2011; Vo et al., 2020b). Vo et al. (2020b) concluded that DahA was the main contributor to total carcinogenicity (58.8–59.6%) and that BaP (31.6 to 32.7%) was the second most important contributor in nursery schools in Hanoi. Błaszczyk et al. (2017b) added BaP (62.5–70.0%) was the dominant contributor in Silesian kindergartens in Poland.

Delgado-Saborit et al.(2011) found that BaP (48–57%) was a dominant marker and DahA (14–33%) was the second significance to the total carcinogenicity in indoor environments. Regarding the contribution of individual PAHs to the total mutagenicity of a PAH mixture in Fig 6.17, the results showed a relatively consistent trend in individual PAHs contribution between indoor and outdoor environments. Specifically, BaP was considered to be the most dominant marker of PAHs to total mutagenicity in both particle sizes, with the contribution of BaP varying from 45.3% to 48.5% in $PM_{0.1}$ and 30% to 31% in $PM_{2.5.}$ These results implied that the contribution of BaP to total mutagenicity of PAHs in $PM_{0.1}$ was higher than that in $PM_{2.5}$, indicating that $PM_{0.1}$ is associated with more adverse health effects than $PM_{2.5}$. It can be concluded that BaP and DahA were considered as the most dominant contributions of PAH to total carcinogenicity and mutagenicity of PAH mix in both particle sizes between indoor and outdoor environment in this study. In stance, the highest contribution to TEQ was attributed to DahA for $PM_{2.5}$ and BaP for $PM_{0.1}$, respectively, whereas BaP had the highest contribution to MEQ. These results suggest that BaP and DahA are good markers for mutagenic and carcinogenic potential of environmental PAH mixtures. It should be emphasized that the MEQ and TEQ values obtained in this study were likely underestimated because not all of the PAHs were analyzed. In addition, some of the PAH derivatives (e.g., nitro-PAH) are more harmful than their parent compounds, which was not considered in this study.

3.6.2. Cancer risk assessment

3.6.2.1. Micro-environment

Table 6.18 shows the ILCRs estimated for $\sum BaP_{(eq)}$ bound to $PM_{2.5}$ and $PM_{0.1}$ via the inhalation pathway in the seven age categories in summer and winter in indoor and outdoor air. Seven age groups were used in this study: infants (0–1y), toddlers (1–3y), preschool children (3–6y), children (6–11y), adolescents (11–21y), adults (21–60y), and elderly adults (>60 y). Regardless of the sampled houses, the average ILCRs of $\sum BaP_{(eq)}$ bound to indoor $PM_{2.5}$ ranged from 3.7E-07 to 6.1E-06 in summer and 3.0E-07 to 5.0E-06 in winter, whereas those in $PM_{0.1}$ varied from 4.4E-08 to 7.1E-07 in summer and 4.8E-08 to 7.9E-07 in winter. Similarly, the ILCRs of outdoor $PM_{2.5}$ and $PM_{0.1}$ varied from 2.7E-08 to 9.2E-07 and 2.5E-09 to 8.4E-8 in summer, respectively, and 7.0E-08 to 2.4E-06 and 3.6E-09 to 1.2E-07, respectively. The ILCR was lower than in outdoor air, which was attributed to less exposure time spent outdoors, although the concentrations of $\sum_{BaP(eq)}$ was higher. Estimated ILCR increases with age, and decreases in the following order: (>60y) > (21–60y) > (11–21y) > (6–11y) > (3–6y) > (1–3y) > (0–1y). The highest ILCR was observed in elderly adults (>60y) owing to lifetime exposure extension. The ILCRs in elderly adults were 3 to 17 times, and 2 to 34 times higher than those of other ages in indoor and outdoor air, respectively. Although the ILCRs of children, especially infants and toddlers, was much lower than those of adults, children are still growing and their organ systems are developing, so it is necessary to protect this group from chronic effects. The US EPA set an ILCR threshold of 1.0E-06 for the cumulative risk for considering no adverse car risk, while levels greater than 1.0E-04 indicate a high risk for developing cancer (Chen et al., 2017; US EPA, 2009). Overall, all

ILCRs were within the US EPA threshold, implying safe level for all occupants of the three houses for both particle sizes and seasons.

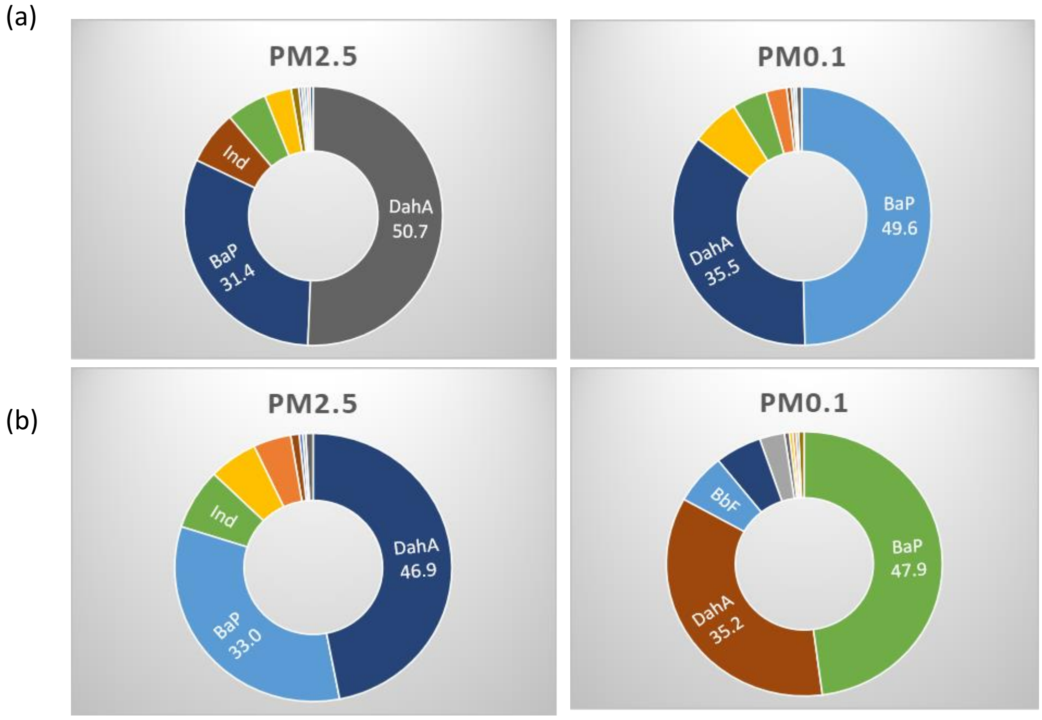


Fig.6. 17. Carcinogenic contribution of PAH in $PM_{2.5}$ and $PM_{0.1}$ in a) indoor air and b) outdoor air in the three houses.

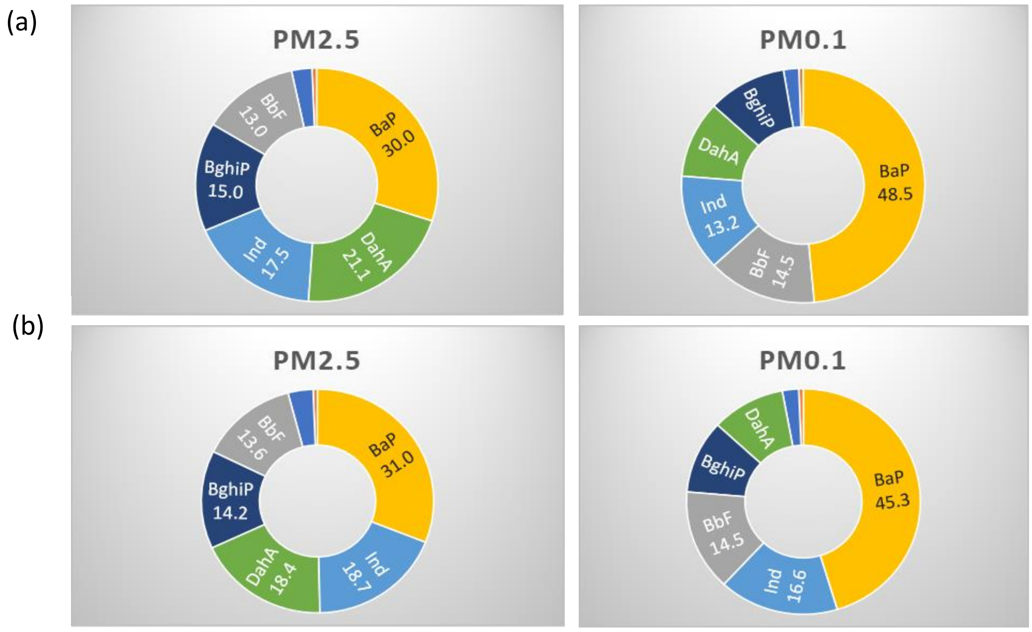


Fig.6. 16. Mutagenic contribution of PAH in $PM_{2.5}$ and $PM_{0.1}$ in a) indoor air and b) outdoor air in the three houses.

Table 6. 14. Summary of Incremental Life Cancer Risk for each age group in indoor environment

Age	Summer				Winter			
	$PM_{2.5}$		$PM_{0.1}$		$PM_{2.5}$		$PM_{0.1}$	
	Indoors	Outdoors	Indoors	Outdoors	Indoors	Outdoors	Indoors	Outdoors
0-1 y	3.7E-07	2.7E-08	4.4E-08	2.5E-09	3.0E-07	7.0E-08	4.8E-08	3.6E-09
1-3 y	4.5E-07	3.9E-08	5.8E-08	3.9E-09	3.7E-07	1.0E-07	6.3E-08	5.4E-09
3-6 y	6.0E-07	6.9E-08	4.4E-08	5.3E-09	4.8E-07	1.8E-07	5.5E-08	7.0E-09
6-11 y	8.7E-07	2.0E-07	9.1E-08	1.6E-08	7.1E-07	5.0E-07	1.0E-07	2.2E-08
11-21 y	9.8E-07	2.7E-07	1.1E-07	2.3E-08	8.0E-07	6.9E-07	1.2E-07	3.3E-08
21-60 y	2.2E-06	5.1E-07	2.6E-07	4.7E-08	1.8E-06	1.3E-06	2.8E-07	6.6E-08
>60 y	6.1E-06	9.2E-07	7.1E-07	8.4E-08	5.0E-06	2.4E-06	7.9E-07	1.2E-07

Among three investigated houses, the occupants at urban house (K3) were exposed to a higher risk than those at urban periphery house (K1) and roadside house (K2). It is quite interesting, although the highest concentration of $\sum_{15}$PAHs was measured at K2 and the lowest at K3, the ILCR of $\sum BaP_{(eq)}$ at urban house (K3) was highest in both $PM_{2.5}$ and $PM_{0.1}$ in both indoor and outdoor environments, which was attributed to the highest concentrations of $\sum BaP_{(eq)}$. The discrepancies in the PAH profiles at each house were associated with different types of source emissions as well as different contributions of sources. These findings imply that the more intensive indoor sources, such as burning incense, indoor cooking, evaporation of building materials, use of camphor etc., were attributed to elevate the concentrations $\sum_{BaP(eq)}$ at urban house (K3). Among the different age categories, elderly adults (>60 years) were also exposed the highest potential cancer risk at the three houses in the study, but were nonetheless safe. It is well known that, in addition to particle size, the chemical composition (heavy metals, PAHs, organic carbon, etc.) of PM has a substantial influence on its toxicity, which may result in synergistic effects. The cumulative risks for PAHs bound to PM were only assessed in this investigation. As a result, because the synergistic effects of other chemical species were not examined, the results of this investigation may underestimate the cumulative risks (Sharma and Balasubramanian, 2018; Slezakova et al., 2014).

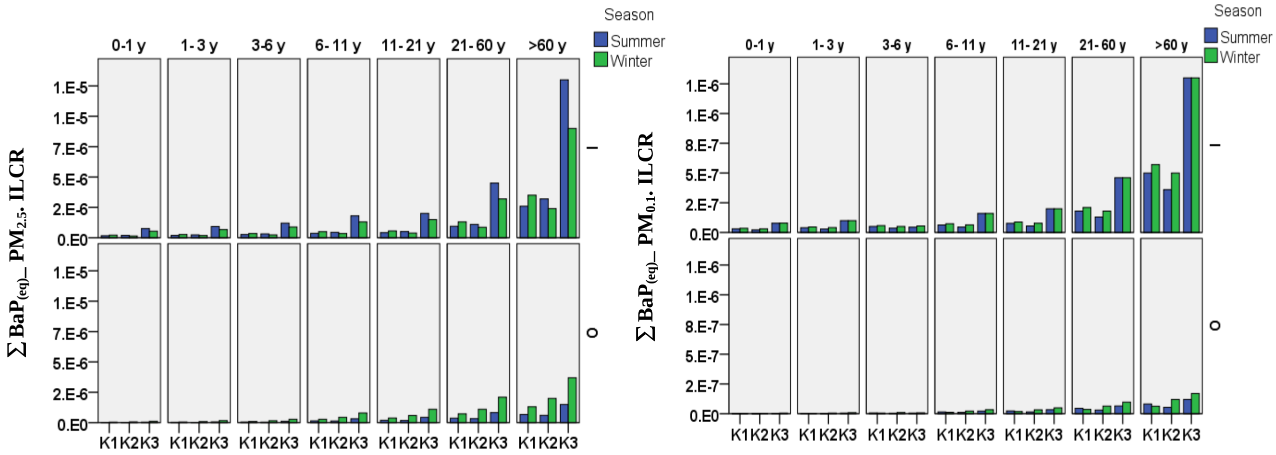


Fig.6. 18. Cancer risks for $PM_{2.5}$ and $PM_{0.1}$-bound $\sum BaP_{eq}$ in the three houses in summer and winter

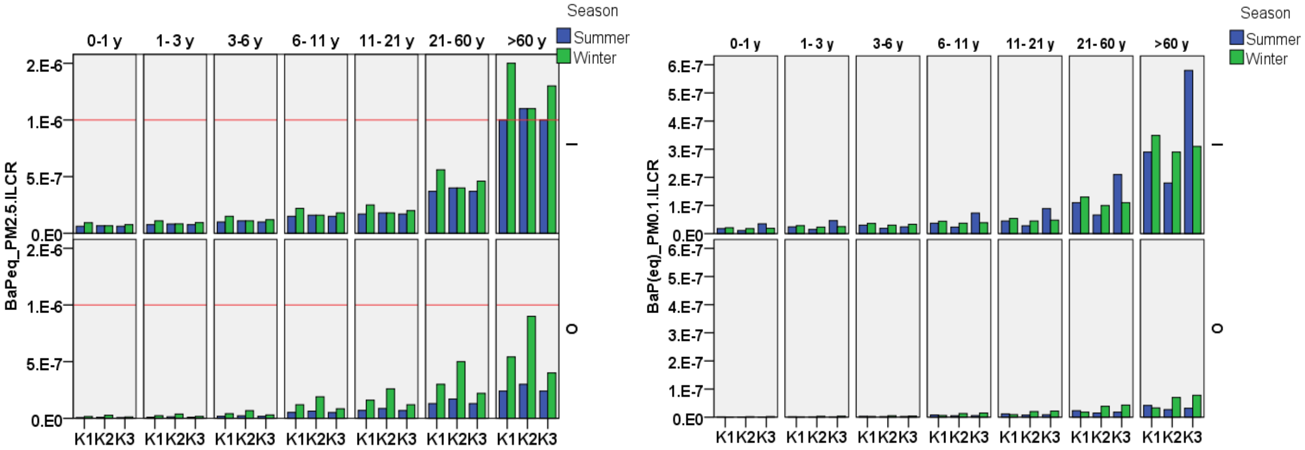


Fig.6. 19. Cancer risks for $PM_{2.5}$ and $PM_{0.1}$-bound BaP_{eq} in the three houses in summer and winter

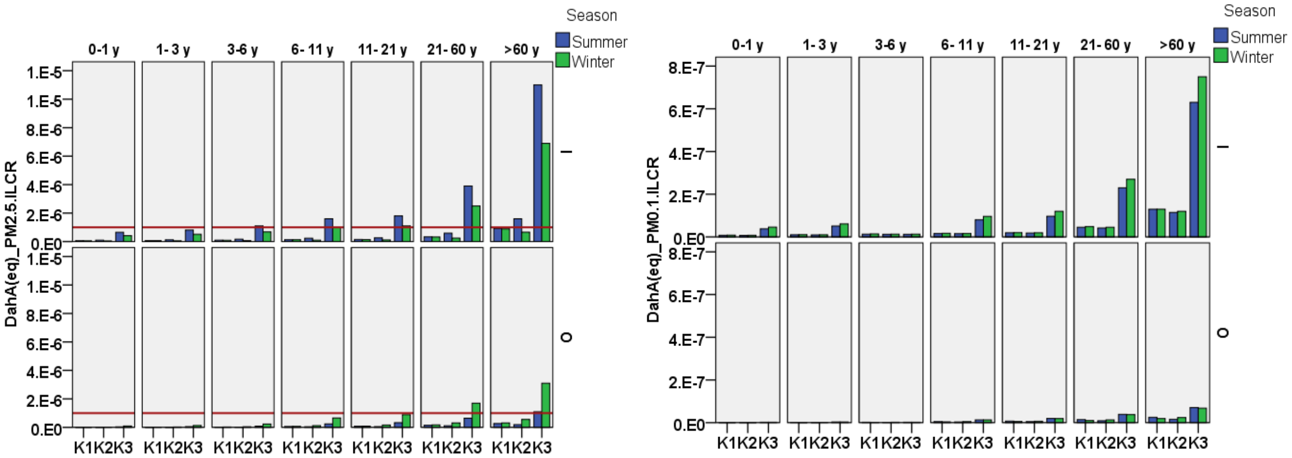


Fig.6. 20. Cancer risks for $PM_{2.5}$ and $PM_{0.1}$-bound $DahA_{eq}$ in the three houses in summer and winter

Furthermore, BaP and DahA were considered to be dominant contributors to carcinogenicity, as discussed in section 3.6.1. To assess the potential cancer risk due to exposure of the occupants to individual PAHs, the ILCRs were also estimated for BaP and DahA as individual PAHs, was illustrated Fig. 6.19 and 6.20. It should to take notice that only elderly group had ILCR values ranging from 1.1E-6 to 1.3E-06 due to intake of BaP_{eq} bound to indoor $PM_{2.5}$ at three houses. Meanwhile, ILCRs were estimated at 1.1E-06 for elderly group at K2, and 8.9E-06 for the elderly group, 3.2E-06 for adults, 1.4E-06 for adolescents, and 1.3E-06 for children at K3, which was associated with the inhalation of DahA bound to indoor $PM_{2.5.}$ Furthermore, the ILCRs of 2.1E-06 obtained for the elderly group (>60y) and 1.2E-06 obtained for adults (21-60y) at K3 in the outdoor environment These estimated ILCR values for BaP or DahA exceeded the US EPA threshold (1.0E-06) for the individual setting. These results implied the existence of a potential cancer risk due to inhalation of BaP or DahA bound to $PM_{2.5}$. Among age categories, the elderly group the most sensitive group. In addition, occupants in the other age groups, including children (6-11y), adolescents (11-21y) and adults (21-60y), were also defined as vulnerable for potential cancer risk via inhalation of DahA bound to $PM_{2.5}$ at the urban house (K3). Except for these cases, the ILCR values estimated for BaP and DahA bound to $PM_{2.5}$ and $PM_{0.1}$ as individual PAH quantitation were below the US EPA thresholds for individual setting (10^{-6}). It is worthy to emphasis that even though the cumulative cancer risk was at an acceptable level for all ages, potential cancer risk due to expose BaP and DahA bound to $PM_{2.5}$ should be considered for certain cases, as described above.

3.6.2.2. Macro-environment

It is fact that, indoor PM is strongly influenced by outdoor PM. In this study, the outdoor environment was used as a background to determine the penetration rate of PM into buildings. Therefore, outdoor air conditions are of utmost importance when controlling indoor air quality. In this study, the carcinogenic health risk associated with the inhalation of $PM_{2.5}$ and $PM_{0.1}$-bound $\sum BaP_{(eq)}$ by the urban population was estimated.

Fig. 6.21 shows the estimated lifetime excess inhalation cancer risks due to $\sum BaP_{(eq)}$ bound to outdoor PM of three inhalation unit risk (IUR_{BaP}) values recommended by different organizations: 1.1E-06 m^3/ng (CalEPA method) is recommended by the Office of Environmental Health Hazard Assessment (OEHHA) of California Environmental Protection Agency; 8.7E-05 m^3/ng is recommended by the World Health Organization (WHO method) (Bootdee et al., 2016; WHO, 2000; Yury et al., 2018), and 6.0E-07 m^3/ng is recommended by the Integrated Risk Information System (US EPA method) (Sharma and Jain, 2020). Using the CalEPA method, the average estimated excess inhalation cancer risk associated with exposure to $\sum BaP_{(eq)}$ for $PM_{2.5}$ in the outdoor environment were 1.7E-05 and 4.2E-05 in summer and winter, respectively, whereas for $PM_{0.1}$, they were 3.5E-06 and 4.9E-06, respectively. Accordingly, these results indicated that 17 and 42 cancer cases per million people could be attributed to the inhalation of $\sum_{BaP(eq}$)bound to $PM_{2.5}$ for lifetime exposure (70 years) in summer and winter, respectively, while 4, and 5 cancer cases per million people could be attributed to $PM_{0.1}$, respectively. Using the WHO method, the average excess inhalation cancer risk for $PM_{2.5}$ was 1.3E-03, and 3.4E-03 in summer and winter, respectively, while corresponding values were 3.0 E-04 and 4.3E-04 for $PM_{0.1}$, respectively. These results meant that there were 13 cases (summer), 34 cases (winter) per 10.000 inhabitants having change to develop cancer risk, when they exposed 15.1 ng/m^3 (summer), and 38.6 ng/m^3 of $\sum BaP_{(eq}$ (winter) in $PM_{2.5,}$ respectively. Meanwhile, there were 3 cases (summer) and 4 cases (in winter) per 10000 inhabitants having change to develop cancer risk, when they exposed 3.2 ng/m^3 (summer), and 4.8 ng/m^3 of $\sum BaP_{(eq)}$ (winter) in $PM_{0.1,}$ respectively. Similarly, based on Integrated Risk Information System (USEPA), the number of people at risk of developing cancer per million people due to the inhalation of $\sum BaP_{(eq)}$ included 10 in summer and 23 in winter for $PM_{2.5}$, and 2 in summer and 3 persons in winter for $PM_{0.1}$. The discrepancy in cancer cases were due to using the three inhalations unit risk for calculation by three methods as mentioned. Comparing to the US EPA threshold setting for cumulative cancer risk (one case per 10.000 inhabitants as acceptable level), the calculated ILCR values were associated with a potential cancer risk based on WHO method, while those values based on the CalEPA and US EPA methods were associated with an acceptable cancer risk. The excess cancer risk in this study was higher than values obtained in different environments in China (Chen et al., 2017; Yury et al., 2018). Yury et al. (2018) reported that ILCR due to $PM_{2.5}$-bound $\sum BaP_{(eq)}$ in the outdoor environment was equivalent to 22 cases per 10000 inhabitants can occur cancer risk according the WHO, respectively. Chen et al.(2017) calculated that the cancer cases were corresponding to 27 cases in winter and 2 cases per 10000 inhabitants in summer in the application of WHO method.

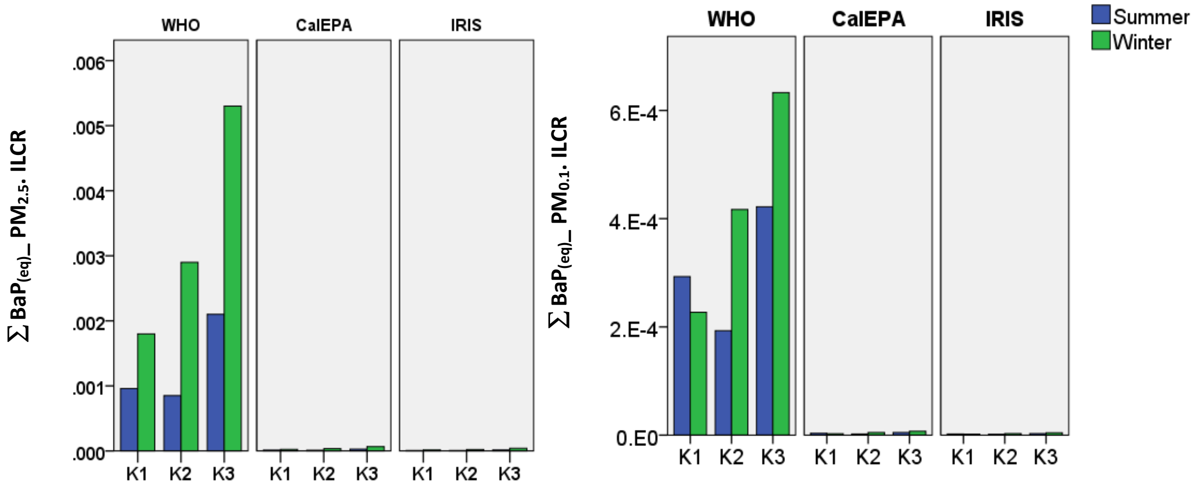


Fig.6. 21. Cancer risks for outdoor $PM_{2.5}$- and $PM_{0.1}$-bound BaP_{eq} in the three houses estimated by three methods.

3.6.2.3. Sensitivity analysis

Monte Carlo simulation was applied in our study to determine the probability density functions assigned to each input variable (C, ET, BW), instead of using the single risk value, which may cause uncertainties. The simulation results for cancer are summarized in Table 6.15 for $\sum BaP_{(eq)}$ in indoor air. The probabilities of lifetime car risk for $\sum BaP_{(eq)}$ bound to $PM_{2.5}$ in winter were lower than those in summer, with 95% of ILCR values ranging from 7.2E-07 to 1.0E-05 in summer and 5.7E-07 to 8.8E-06 in winter. The lower ILCR values $\sum BaP_{(eq)}$ bound to $PM_{2.5}$ in summer were due to decreased concentrations of $\sum BaP_{(eq)}$. The similar trend was also observed for $PM_{0.1}$, with 95% of ILCR values ranging from 5.5E-08 to 8.8E-07 in summer and 8.4E-08 to 1.5E-06 in winter. The sensitivity analysis identified that the most influential factor to ILCR variation was the $\sum BaP_{(eq)}$ concentration, which contributed to approximately 87% to 98% of the ILCR variance. BW and AT had negative values and negligible significance, ranging from 2% to 13%. These results supply useful information for creating effective mitigations for decreasing indoor PM and protecting human health, particularly in urban areas during in winter

Table 6. 15. Monte simulation results of ILCR for $\sum BaP_{(eq)}$ in summer and winter for indoor $PM_{2.5}$ and $PM_{0.1}$

	$PM_{2.5}$						$PM_{0.1}$					
	Summer			Winter			Summer			Winter		
	5%	50%	95%	5%	50%	95%	5%	50%	95%	5%	50%	95%
0-1 y	1.6E-07	3.3E-07	7.2E-07	1.3E-07	2.7E-07	5.7E-07	3.4E-08	4.3E-08	5.5E-08	2.4E-08	4.5E-08	8.4E-08
1-3 y	1.9E-07	4.1E-07	8.9E-07	1.7E-07	3.4E-07	6.7E-07	4.4E-08	5.7E-08	7.4E-08	3.1E-08	5.9E-08	1.1E-07
3-6 y	2.4E-07	5.4E-07	1.2E-06	2.3E-07	4.5E-07	8.8E-07	5.4E-08	7.2E-08	9.5E-08	3.9E-08	7.4E-08	1.4E-07
6-11 y	3.8E-07	8.2E-07	1.8E-06	3.5E-07	6.7E-07	1.3E-06	7.3E-08	9.3E-08	1.2E-07	5.2E-08	9.6E-08	1.8E-07
11-21 y	4.2E-07	9.0E-07	1.9E-06	3.9E-07	7.4E-07	1.4E-06	9.1E-08	1.1E-07	1.4E-07	6.2E-08	1.2E-07	2.1E-07
21-60 y	9.1E-07	1.9E-06	4.2E-06	7.4E-07	1.7E-06	3.7E-06	2.0E-07	2.5E-07	3.1E-07	1.4E-07	2.6E-07	4.8E-07
>60 y	2.6E-06	5.5.E-06	1.0E-05	2.4E-06	4.6E-06	8.8E-06	5.8E-07	7.2E-07	8.8E-07	4.0E-07	7.4E-07	1.5E-06

4. CONCLUSION

This study is one of the first and most comprehensive studies of indoor PM in urban areas focus on the chemical components and health risk assessment in Hanoi in summer and winter. High levels of $\sum_{15}$PAHs, $\sum$com-PAHs and $\sum$car-PAHs bound to $PM_{0.1}$ and $PM_{2.5}$ indoors and outdoors were found in Hanoi, Vietnam. The average concentrations of $\sum_{15}$PAHs, $\sum$com-PAHs and $\sum$car-PAHs were 126.7±18.1 ng/m^3, 37.5±8.9 ng/m^3 and 26.9±11 ng/m^3 for indoor $PM_{2.5}$, and 37.6±9.9 ng/m^3, 16.5±5.8 ng/m^3 and 9.9±4.8 ng/m^3 for indoor $PM_{0.1}$, respectively. The corresponding outdoor concentrations were 344.7±270.1 ng/m^3, 112.8±74.2 ng/m^3 and 70.6±40.3 ng/m^3 for $PM_{2.5}$, and 49.9±18.6 ng/m^3, 20.3±9 ng/m^3 and 11.2±5 ng/m^3 for $PM_{0.1}$, respectively. Among 15 individual PAH, Nap, Phe and Flt were present at the highest concentrations in indoor $PM_{2.5}$ and $PM_{0.1}$. The mean concentrations of Nap and Phe varied from 20±10.6 to 33.1±17.2 ng/m^3 and 26.8±10.5 to 29.7±8.3 ng/m^3 for $PM_{2.5}$, respectively, and 2.6±1.6 to 4.4±0.7 ng/m^3 and 4.2±0.6 to 8.9±1.7 ng/m^3 for $PM_{0.1}$, respectively, whereas the mean concentration of Flt bound to $PM_{0.1}$ was in the range of 2.1±0.5 to 3.1±1.1 ng/m^3. Accordingly, the most abundant PAH species found in outdoor air were quite similar to those found in indoor air. Although BaP was not as abundant, the average concentrations of BaP bound to indoor $PM_{2.5}$ and $PM_{0.1}$ in our study were 1.6 to 2.6 times higher than European guidelines for both particles (1 ng/m^3), while the corresponding outdoor corresponding values were 1.9 to 7.1 times higher than the threshold recommended by European guidelines. This means that the levels were sufficiently high to pose a threat to the health of the occupants at the three investigated houses, which could not be ignored. Furthermore, the contents of PAH bound to $PM_{0.1}$ were higher than those of $PM_{2.5}$ in both indoor and outdoor air according to the season. Specifically, proportion of LMW-PAHs to total PAHs was higher in $PM_{2.5}$, whereas that of LMW to total PAHs in $PM_{0.1}$ were greater. But the contents of LMW, HMW-PAHs bound to $PM_{0.1}$ were higher than in $PM_{2.5.}$ Significant seasonal variation was observed in the $\sum_{15}$PAHs bound to indoor and outdoor $PM_{2.5}$ and $PM_{0.1}$, with concentrations increasing by 1.3 to 1.5 times in indoor air and 1.8 to 4.1 times in outdoor air in winter.

Source apportionment analysis indicated that domestic combustion of coal and biomass and traffic emissions were the main sources of PAHs bound to both indoor $PM_{0.1}$ and $PM_{2.5}$ at the urban periphery house (K1), whereas traffic emissions were the dominant source of these compounds at the roadside house (K2). Traffic and domestic coal combustion were primary contributors to PAHs bound to indoor $PM_{0.1.}$ in summer, whereas indoor sources, such as burning incense, scented candles, natural gas and camphor accounted for the majority of PAHs bound to indoor $PM_{2.5}$ in both seasons, and PAHs bound to indoor $PM_{0.1}$ in winter at the urban house (K3). In short conclusion, the contribution of outdoor sources was significant at K1 and K2, while indoor sources were predominant at K3.

Although, the deposited doses of PAHs did not differ markedly among three houses, the deposited doses of $_{BaP(eq)}$ bound to PM and ILCR values at the urban house were significantly higher than those at other sites. The distribution of the EDI of $\sum_{15}$PAHs or $\sum BaP_{(eq)}$ in the HRT varied widely with particle size, with the majority of $\sum_{15}$PAHs or $\sum BaP_{(eq)}$ bound to $PM_{2.5}$ being deposited in the HA region, whereas the highest proportion of PAHs or $BaP_{(eq)}$ bound to $PM_{0.1}$ was deposited in the AL region. Instance, the proportion of EDI values for $PM_{2.5}$ was 60% to 90% in the HA region, 3 to 9% in the TB region, and 8% to 30% in the AL region, and for $PM_{0.1}$, apportionment was 8% to 12% in the HA region, 20% to 25% in the TB region and 63% to 75% in the AL region.

In the micro indoor environment, among the three houses investigated, the occupants at the urban house (K3) were exposed to the highest potential risk, where the intake of BaP and DahA was sufficiently high to pose a potential cancer risk to the elderly group as health risk assessment for individual PAH setting, although the cumulative cancer risk was within the acceptable limits for all of the occupants in the three houses. Except for the above cases, the cancer risk was negligible for all occupants. In the macro-environment, the estimated ILCR values fell within the high potential cancer risk category of the WHO method, while estimates based on the CalEPA and US EPA methods were classified as having an acceptable cancer risk. These findings are considered to be valuable for the development of mitigation measures to control PAH sources in the indoor air environment and can provide a scientific basis for indoor air quality management, especially for developing appropriate measures to protect human health in urban areas.

CHAPTER 7

CONCLUSIONS AND RECOMMENDATIONS

1. CONCLUSIONS

This study is one of the first to conduct a comprehensive analysis of the state of indoor pollution in Vietnam and its mitigation. Specifically, this study focused on the determination of the particle size distribution the relationship between particles of different size fractions in indoor and outdoor air, as well as on quantifying the dosages of PM in the human respiratory tract (HRT). The study analyzed the chemical compositions of $PM_{2.5}$ and $PM_{0.1}$ (trace elements (TEs) and polycyclic aromatic hydrocarbons (PAHs) to identify potential sources, evaluated the distribution of chemicals bound to PM in HRT, and finally evaluated the health risk associated with PM. The results can be summarized as follows:

1.1. Indoor pollutants in urban environments in Vietnam

Thorough comprehensive literature reviews of English and Vietnamese studies, it was shown that high concentrations of PM of various sizes (PM_{10}, $PM_{2.5}$, NPs) and BTEXs were present in various indoor urban environments in Vietnam, including schools, residential homes, offices, parking basements, and commercial areas. In comparison with World Health Organization (WHO) guidelines, indoor $PM_{2.5}$ levels in residential houses, schools, and kitchens, and indoor PM_{10} levels in schools and parking basements in high-rise buildings all exceeded acceptable limits. In addition, benzene levels exceeded WHO guidelines in residential houses and schools in urban areas. These results showed that poor IAQ had a direct influence on human health.

The origins of indoor PM and BTEXs were complex, and were attributed to both indoor and outdoor sources. Outdoor sources included vehicle and industrial emissions, and combustion of coal and biomass. Indoor sources were attributed to human activities (moving, sweeping, cleaning), cooking, smoking, and burning incense. Outdoor PM was considered to be the main contributor to indoor PM

The levels on indoor PM varied both temporally and spatially. For example, indoor PM_{10} in basement parking lots and in commercial areas at weekends were more twice those on weekdays, and indoor NP count concentrations during times of peak activity (lunch time) increased several folds compared to times when there was less activity in residential houses. Schools in urban centers had higher indoor PM concentrations (i.e., $PM_{0.5}$, PM_1, $PM_{2.5}$, PM_{10}) than schools on the urban periphery. Seasonal variation in indoor PM_{10} and $PM_{0.1}$ in offices and residential houses was not marked, but a trend in indoor $PM_{2.5}$ between seasons was observed in residential houses. Cooking activities contributed to indoor PM generation, which was dependent on type of fuel consumption. Replacing wood combustion with LPG reduced PM_1, $PM_{2.5}$, and PM_{10} in the kitchen by approximately 70%. Sweeping in offices increased indoor PM_{10} approximately ten-folds. Regarding BTEXs compounds, levels of BTEXs in new/innovated houses were higher than those in old houses, outdoor air. These compounds in new/innovated houses and underground basements were markedly higher than those in school environments. Benzene levels, used as a proxy for IAQ and human health effects, in underground parking lots, residential houses, and schools largely exceeded recommended WHO levels and pose a significant risk to human health. Clear seasonal trends were observed in outdoor BTEX levels, but not in indoor BTEX levels.

From a practical standpoint, indoor air-purifier technologies and natural ventilation are most commonly adopted to control concentrations of indoor PM. In addition, regular indoor cleaning, suitable interior decoration, and using air purifiers reduced indoor pollution and improved IAQ. Environmentally friendly transportation methods and industrial adaptations should be promoted to achieve sustainable development in urban areas, as these indirectly enhance IAQ. Mitigation efforts focusing on controlling indoor and outdoor pollutants remain insufficient due to lack of technical regulations, standards, and effective interventions on IAQ.

1.2. Size-fractionated particulate matter (PM) in residential houses in urban areas in Vietnam: Relationship of indoor and outdoor PM, mass-size distribution, and deposited dose estimates

The findings indicated that high concentrations of different PM fractions were observed in four investigated houses. Overall, the average level of indoor $PM_{0.1}$, $PM_{0.5}$, PM_1, $PM_{2.5}$, and PM_{10} ranged from 5.3 to 8.9 µg/m^3, 10.8 to 20.1 µg/m^3, 20.5 to 47.6 µg/m^3, 33.7 to 105.9 µg/m^3, and 44.7 to 135 µg/m^3 in the four houses, respectively. The average concentrations of indoor $PM_{2.5}$ and PM_{10} were 1.5- to 2-fold higher than WHO recommended values, posing a threat to the health of occupants in urban areas. There are no standards for $PM_{0.1}$, $PM_{0.5}$, and PM_1 proposed by any agencies, despite their severe risk to human health, which should be emphasized.

Different trends in the seasonal variation of particles were observed among particle sizes. The levels of indoor PM_1, $PM_{2.5}$, and PM_{10} were consideratly higher in winter, but a negligible change in concentrations of indoor $PM_{0.1}$ and $PM_{0.5}$ was observed between summer and winter. Season variation of PM concentrations was associated with changes in meteorological conditions; for example, wind speed was moderately correlated with concentrations of all size fractions, except for $PM_{0.1}$. No correlation was observed between precipitation (Pr) and PM concentrations in this study, and this was attributed to low rainfall during sampling periods.

The I/O ratios of NP particles, submicron particles ($PM_{0.1-1}$), and fine particles ($PM_{1-2.5}$) were markedly higher than those of larger particles, indicating that larger particles were more effectively infiltrated into buildings from the outside environment. In additionally, the I/O ratios of $PM_{0.1}$ and submicron particles ($PM_{0.1-1}$) were greater than 1, implying the strength of indoor sources, in addition to outdoor sources. Conversely, the I/O ratios of coarse particles were significantly less than 1, indicating that outdoor sources were the main sources of indoor PM

Except for $PM_{>10}$ and $PM_{2.5-10}$ in some circumstances, indoor and outdoor PM (i.e., $PM_{0.1}$, $PM_{0.1-0.5}$, $PM_{0.5-1}$, $PM_{1-2.5}$) were strongly correlated (R^2=0.7- to 0.96), indicating that outdoor PM can explain approximately 70% to 96% of the indoor PM variation. The infiltration factors of $PM_{0.1}$ and submicron particles ($PM_{0.1-1}$) were higher than those of larger PM sizes (i.e., $PM_{1-2.5}$, $PM_{2.5-10}$). It is likely that NP and submicron particles can easily permeate indoors, regardless of whether the window is open or closed. A wide variation in the relative contribution of indoor sources to indoor PM was observed among the four houses in this study; for example, the contribution of indoor PM ranged from 0.6% to 63% $PM_{0.1}$, 5% to 63% for $PM_{0.1-0.5}$, 5% to 48% for $PM_{0.5-1}$, 3% to 32% for $PM_{1-2.5}$, and 9% to 45% for $PM_{2.5-10}$. However, most indoor fractions came from outside sources.

Unimodal distributions of indoor particles determined the supper-micron size (1 to 2.5 µm) with highest concentration and $PM_{<0.5}$ and $PM_{>10}$ with lowest concentration, whereas greatest concentrations of outdoor particles were gained at supper-micron size (1 to 10 µm), and the lowest concentrations were also found in small sizes ($PM_{<0.5}$) and coarse size ($PM_{>10}$) as well. Fine particles (i.e., $PM_{0.5-1}$, $PM_{1-2.5}$) contributed to the predominance of coarse particles both indoors and outdoors, suggesting that these particles poses a serious threat to human health.

The total deposition fraction (DF) of PM_{10} was higher than those of $PM_{2.5}$, PM_1, $PM_{0.5}$, and $PM_{0.1}$. However, the total lobar DF was highest for $PM_{0.1}$, followed by $PM_{2.5}$, PM_{10}, PM_1, and $PM_{0.5}$ for most age (except for the adults (>21y group) for PM_{10}). The distribution of deposited doses (EDIs) in different regions of the human respiratory tract (HRT) (i.e., head airways (HA); tracheobronchial region (TB), Alveolar region (AL) varied according to particle sizes and age. In general, the EDI values were distributed as 8.3% to 25.5% in the HA region, 13.3% to 23.4% in the TB region, and 66.9% to 84% in the AL region for $PM_{0.1}$; 21.5% to 56.9% in the HA region, 8.6% to 12.2% in the TB region, and 33% to 61.1% in the AL region for $PM_{0.5}$; 36.1% to 59.5% in the HA region, 7.1% to 11.1% in the TB, and 31.3% to 52.7% in the AL region for PM_1; 35.7% to 53.7% in the HA region, 6.1% to 18.7% in the TB region, and 40.1% to 45.5% in the AL region for $PM_{2.5}$; and 55% to 94.9% in the HA region, 4.2% to 44.3% in the TB region, and 0.6% to 8.4% in the AL region for PM_{10}. Deposited doses were distributed primarily in the HA region for PM_{10} and predominantly in the AL region for $PM_{0.1}$, increasing with age and particle size. The adults (>21y) was considered to be the most vulnerable group examined in this study due to chronic effects.

In terms lobar deposition distribution, EDI was distributed in the range of 8.9% to 21.9% in the left upper region (LU), 25.6% to 30.5% in the left lower region (LL); 7.5% to 17.4% in the right upper region (RU); 5.4% to 7.2% in the right middle region (RM); and 21.2% to 35.5% in the right lower region (RL) for $PM_{0.1}$; 5.5% to 10.9% in the LU region, 13.9% to 29% in the LL region, 4.8% to 10% in the RU region (RU), 2.1% to 6% in the RM region, and 11.7% to 20.4% in the RL region for $PM_{0.5}$; 5% to 9% in the LU region, 13% to 23.8% in the LL region; 3.9% to 9.8% in the RU region; 2% to 4.9% in the RM region, and 10.9% to 18.6% in the RL region for PM_1; 6.1% to 11.6% in the LU region, 16.9% to 26.7% in the LL region; 5.2% to 15.8% in the RU region, 2.7% to 5.4% in the RM region, and 14.2% to 25.4% in the RL region for $PM_{2.5}$; and 0.7% to 6.2% in the LU region, 1.8% to 13.8% in the LL region, 0.6% to 6.8% in the RU region, 0.4% to 2.5% in the RM region, and 0.1% to 10.7% in the RL region for PM_{10}. Overall, the deposited doses of PM were greatest in the left and right lower lobes, and lowest in the right middle lobe. It is worth noting that the greatest doses of ultrafine particles were observed in the upper and lower lobes, as well as the middle lobes, while the lowest dose was observed for coarse particles as PM_{10}. The total lobar doses were higher in the left lung than in the right lung. Human exposure to NP, fine particles, and coarse particles is a recent research topic in IAQ.

1.3. Indoor $PM_{0.1}$ and $PM_{2.5}$ in Hanoi: Characterization, source identification, and health risk assessment

The obtained results showed that the average concentration of indoor $PM_{2.5}$ measured in the three houses was 59.9±23.6 µg/m^3, which is approximately four times higher than WHO guideline (15.0 µg/m^3). The average concentration of indoor $PM_{0.1}$ was 8.2±0.7 µg/m^3 and the average concentrations of TEs bound to indoor $PM_{0.1}$ and $PM_{2.5}$ varied from 66 to 216 ng/m^3 and 391 to 2356 ng/m^3, respectively. Such high concentrations of indoor $PM_{0.1}$, $PM_{2.5}$, and TE concentrations could increase the disease burden of house occupants.

Significant seasonal variation was observed for indoor $PM_{2.5}$, but not for indoor $PM_{0.1.}$ The indoor mass concentrations of $PM_{2.5}$ ranged from 43.4 to 49.5 µg/m^3 in summer, and from 50.6 to 105.9 µg/m^3 in winter. Similarly, seasonal variations were also observed in the of TEs bound to both particle sizes. Higher concentrations of TEs in $PM_{2.5}$ and $PM_{0.1}$ were observed in the roadside house, which had $PM_{2.5}$ and $PM_{0.1}$ level that were 1.5- to 2.3-folds and 1.2- to 1.3-folds higher, respectively, than the urban periphery house and urban house. Among the 10 trace elements analyzed, Zn, Pb, and Cr were the most abundant in both particle sizes in summer and winter, but the average concentrations of As and Ni in the three houses were 1.4- to 4.6, and 1.5- to 4.5-times higher than EU limits, respectively.

Almost no seasonal difference was observed in F_{inf} (Infiltration factor) of $PM_{0.1}$ and $PM_{2.5}$, as well as F_{inf} of TEs bound to the corresponding PM. The contribution of indoor-generated PM to indoor PM varied markedly among houses in summer and winter, reflecting the complex and dynamic characteristics of the indoor environment. About 2% to 21% of indoor $PM_{2.5}$ in the three houses originated from indoor sources, whereas 18% and 54% of indoor $PM_{0.1}$ in roadside and urban houses originated from indoor sources, respectively.

Indoor $PM_{2.5}$ was derived mainly from outdoor sources, whereas indoor $PM_{0.1}$ was derived from both 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 the 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).

The EDI of TEs in the HRT varied largely with particle sizes and age categories. The indoor $EDI_{non\text{-}car\text{-}TEs}$ (Deposited dose of non-carcinogenic TEs) and $EDI_{car\text{-}TEs}$ (Deposited dose of carcinogenic 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. EDI values for $PM_{2.5}$ were distributed as follows: 80% in the Head airways region (HA), 7% in the Tracheobronchial region (TB), and 13% in the Alveolar region (AL), whereas for $PM_{0.1}$, they were distributed as 11% in the HA region, 14% in the TB region, and 75% in the AL region. The EDI in the HA region decreased as particle size decreased, and increased in the TB and AL regions as the particle size decreased. In winter, the deposited doses of TEs bound to PM, HI, (Hazard index) and ILCR (Incremental Lifetime cancer risk) at the roadside dwelling were significantly higher than those at other sites.

Among the three investigated houses, the occupants at the roadside house were exposed to the highest risk, whereas the intake of TEs via $PM_{2.5}$ can lead to an increased car risk for people older than 60 years and unacceptable non-car risks for all ages at the roadside house in winter. Except for the above cases, there were negligible the effects of non-car and car risk associated with exposure to indoor $PM_{0.1}$ and $PM_{2.5}$. Therefore, the findings of this study can provide a scientific basis for IAQ management, especially for the development of appropriate measures for protecting human health in winter.

Monte Carlo simulations showed that the probabilities of lifetime cancer risk for TEs in $PM_{0.1}$ and $PM_{2.5}$ were significantly higher in winter compared with summer, with 90% of ILCR ranging from 2.9E-07 to 9.0E-06 for $PM_{0.1}$ and 3.2E-06 to6.8E-05 for $PM_{2.5}$ in summer, and 6.6E-07 to 1.4E-05 for $PM_{0.1}$ and 6.5E-06 to1.4E-04 for $PM_{2.5}$ in winter. The concentration of TEs was the most influential factor affecting ILCR variance (85% to 96%), whereas the contribution of other variables (BW, EF, ED, AT, IR) contributed the minor based on sensitivity analysis.

1.4. Characterization of particulate PAHs in indoor and outdoor air and the health implications

High levels of $\sum_{15}$PAHs (Sum of 15 PAHs), ∑com-PAHs (Sum of combustible PAHs), and ∑car-PAHs (Sum of carcinogenic PAHs) bound to $PM_{0.1}$ and $PM_{2.5}$ indoors and outdoors were found in Hanoi, Vietnam. The average concentrations of $\sum_{15}$PAHs, ∑com-PAHs, and ∑car-PAHs were 126.7±18.1 ng/m^3, 37.5±8.9 ng/m^3, and 26.9 ±11 ng/m^3 for indoor $PM_{2.5}$, and 37.6 ±9.9 ng/m^3, 16.5±5.8 ng/m^3, and 9.9±4.8 ng/m^3 for indoor $PM_{0.1}$, respectively. The corresponding outdoor concentrations were 344.7±270.1 ng/m^3, 112.8±74.2 ng/m^3, and 70.6±40.3 ng/m^3 for $PM_{2.5}$, and 49.9±18.6 ng/m^3, 20.3±9 ng/m^3, and 11.2±5 ng/m^3 for $PM_{0.1}$, respectively. Nap, Phe, and Flt appeared the highest concentrations in indoor $PM_{2.5}$ and $PM_{0.1}$, and average concentrations of BaP bound to $PM_{2.5}$ and $PM_{0.1}$ were 2.6±0.4 ng/m^3 and 1.6±0.2 ng/m^3, respectively, exceeding EU guidelines and posing a potential danger to the health of the occupants of the three houses. Significant seasonal variation was observed in $\sum_{15}$PAHs bound to $PM_{2.5}$ and $PM_{0.1}$ both indoors and outdoors, with concentrations 1.3- to 1.5-folds in indoor air, and 1.8- to 4.1-folds in outdoor air in winter.

Source apportionment analysis indicated that domestic coal, biomass combustion and traffic emissions were the main sources of PAHs bound to indoor $PM_{0.1}$ and $PM_{2.5}$ at the urban periphery dwelling (K1), whereas traffic emissions were the dominant source of these elements at the roadside dwelling (K2). Traffic emissions and domestic coal combustion were the primary contributors to PAHs bound to indoor $PM_{0.1}$ in summer, whereas indoor sources such as burning incense, candles, natural gas, and camphor accounted for most of the PAHs bound to indoor $PM_{2.5}$ in both summer and winter, and indoor $PM_{0.1}$ in winter, at the urban dwelling (K3). Outdoor sources contributed significantly to PAHs at K1 and K2, while indoor sources were the main sources at K3.

There were significant differences in deposited doses between ∑15PAHs and $\sum BaP_{eq}$ at the three houses. A significant reduction in $EDI_{\sum BaP(eq)}$ was observed compared to $EDI_{\sum 15PAHs}$, and the EDIs of both ∑Bap(eq) and 15 PAHs increased in the indoor environment, although the concentrations of $\sum BaP_{(eq)}$ and $\sum_{15}PAHs$ were greater outdoors. $EDI_{\sum PAHs}$ and $EDI_{\sum BaP(eq)}$ increased significantly with age, with elderly adults (>60y) exposed the highest $EDI_{\sum PAHs}$ and $EDI_{\sum BaP(eq)}$, whereas children, including infants (0–1 y), toddlers (1–3 y), and preschool children (3–6 y), had the lowest EDI after chronic exposure to $\sum_{15}PAHs$ and $\sum BaP_{(eq)}$ in both particle size categories. Deposited doses varied in the different HRT regions due to differences in PM properties and HRT physiology. EDI values were distributed as 26% to 39% in the (anterior nasal region (ET1), 35% to 51% in the main-extra thoracic region (ET2), 2% to 3% in the bronchial region (BB), 1% to 6% in the Bronchiolar region (bb), and 8% to 30% in the alveolar region (AL) for $PM_{2.5}$, and from 4% to 5% in the ET1 region, 5% to 8% in the ET2 region, 3% to 4% in the BB region, 15% to 22% in the bb region, and 63% to 71% in the AL region for $PM_{0.1}$. Thus, it can be concluded that the majority of the PAHs bound to $PM_{2.5}$ were deposited in the head airways region, while PAHs bound to $PM_{0.1}$ were predominantly deposited in the alveolar region.

In the micro-environment, the occupants at of the urban house were exposed to the highest risk, as the intake of BaP and DahA could potentially cause cancer in the elderly group at urban house. However, the cumulative cancer risk was within the acceptable level for all occupants at three houses. Sensitivity analysis revealed that the $\sum BaP_{eq}$ concentration contributed approximately 87% to 98% to the ILCR variance. In the macro-environment, the calculated ILCR indicated a high potential cancer risk when using WHO standards, while the results obtained based on CalEPA and US EPA standards showed that cancer risk levels were acceptable.

2. RECOMMENDATIONS

High concentrations of PM with different sizes and their chemical compositions were identified in residential houses in Vietnam's urban areas, especially emerging indoor pollutants such as fine and ultrafine particles. These particles have been shown to pose a considerable risk to public health. The following recommendations can be made to reduce the negative health effects of these particles:

2.1. For governments and authorities

- Establish IAQ standards and undertake monitoring indoor air in urban areas in Vietnam.
- Enforce increasingly stringent emission regulations for vehicle manufacturers.

- More strictly control industrial emissions through passing emissions standards, especially in highly polluting industrial sectors and enhance air quality monitoring systems to inspect emissions.

- Control the combustion of biomass and domestic coal through educating stakeholders about the disadvantages of burning biomass in fields and briquette coal in urban areas; support and encourage them to apply environmentally friendly treatment measures.

- Strengthen the human resources involved in air quality management.

- Actively promote smoking bans and increase the public's health awareness.

2.2. For stakeholders

- Optimize building design and operation.

- Develop environmentally friendly cooking methods; briquette coal should be substituted with cleaner energy sources, such as natural gas, LPG, or electricity.

- Change cooking habits, such as using exhaust hoods and promoting air circulation while cooking.

- Change lifestyles to be more environmentally friendly, such as cleaning houses on a regular basis, reducing the amount of incense burned indoors, decorating sparingly, and avoiding the use of moth balls as insect repellents.

- Since the pollutants increase in winter, filters need to be used in the mechanical ventilation equipment to clean indoor air. When the environmental quality improves, it is preferable to use natural ventilation rather than artificial ventilation.

3. LIMITATIONS AND FURTHER STUDY

3.1. Potential limitations

- The building configurations studied in the in urban areas of Hanoi were limited

- Small number of samples

- Did not consider the mobility characteristics of TEs in determining the distribution of TEs bound to PM in the HRT

- The effects of HRT morphology and PM chemical compositions on the deposition fraction in the HRT were not fully considered in the Vietnam case study. For example, for deposited doses, the hygroscopic properties and clearance were not examined

- Risk can be overestimated or underestimated depending on the assumptions of the US-EPA model

3.2. Future studies

-Studies could be extended to other indoor environments, such as offices, schools, or other working environments.

-Studies could examine indoor particle dynamics in different environments.

-Studies could employ other approaches to define the possible PM sources, such as using Factor analysis (FA), Positive Matrix Factorization (PMF) models.

-Studies could focus on the hygroscopic growth of particles and their chemical compositions, and how these characteristics affect lung deposition calculations.

-Lung deposition should be calculated for different human activities, considering the clearance process.

-Notwithstanding the limitations mentioned above, this study can provide valuable insights for developing effective measures for the control of indoor PM and health protection, especially in winter in urban areas.

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APPENDIX

Chapter 2

Table 2.1. Physical characteristics of some PAHs (Source: Kim et al., 2013; Patel et al., 2020)

	Compound name (PAHs)	Boiling point (ºC)	Melting temperature(ºC)	Vapor pressure (mmHg)	Solubility in water (mg/L)	LogK_{ow}	Toxicity (IARC)
1	Naphthalen	218	80.26	0.087	31	3.29	2B
2	Acenaphthylen	268-275	92-93	0.029	16.1	4.07	3
3	Acenaphthen	96	95	4.47×10^{-3}	3.8	3.98	3
4	Fluoren	295	116-117	3.25×10^{-4}	1.9	4.18	3
5	Phenanthren	340	100	6.8×10^{-4}	1.1	4.45	3
6	Anthracen	340-342	218	1.75×10^{-6}	0.045	4.45	3
7	Fluoranthen	375	110.8	5×10^{-6}	0.26	4.9	3
8	Pyren	393-404	156	2.5×10^{-6}	0.132	4.48	3
9	Benzo(a)anthracen	438	158	2.5×10^{-6}	0.011	5.61	2B
10	Chrysen	448	254	6.4×10^{-9}	0.0015	5.9	2B
11	Benzo(b) fluoranthen	No data	168.3	5×10^{-7}	0.0015	6.04	2B
12	Benzo(K) fluoranthen	480	215.7	9.59×10^{-11}	0.0008	6.06	2B
13	Benzo(a)pyren	495	179	5.6×10^{-9}	0.0038	6.06	1
14	Indeno(1,2,3-cd)pyren	530	163.6	10^{-11} - 10^{-16}	0.062	6.58	2B
15	Dibenz(a,h)anthracen	No data	262	10^{-10}	0.0005	6.84	2A
16	Benzo(g,h,i)perylen	550	273	1.3×10^{-10}	0.00026	6.5	3

Table 2.2. Some specific uses of PAHs (Abdel-Shafy and Mansour, 2016)

	PAHs	Usage
1	Acenaphthen	Manufacture of pigments, dyes, resins, pesticides and pharmaceuticals
2	Anthracen	Thinner for wood preservation chemicals and the manufacture of dyes and pigments.
3	Fluoranthene	Manufacture of agrochemicals, dyes and pharmaceuticals
4	Fluorene	Manufacture of pharmaceuticals, pigments, dyes, pesticides and thermoplastics.
5	Phenanthrene	Manufacture of plastics
6	Pyrene	Manufacture of pigments

Table 2.3: Properties of HM (Source: (Briffa et al., 2020))

Heavy metal	Properties	Application	Effects on humans
Cr	-Density: 7.15 g/cm^3 -21st most abundant element - Hard - Shiny and steel-grey - Reacts with most acids	Alloys; Metal ceramics; Electroplating; Leather tanning; Manufacturing of synthetic rubies; - Dye paints; Chromium salts are used to color glass green	Oral intake of Cr (VI) causes acute poison and symptoms such as gastrointestinal ulceration, nausea and vomiting, fever, diarrhea, liver damage, dizzy, dead (1-3y)
Mn	-Density: 7.3 g/cm^3 -5th most abundant metal -Essential trace element - Versatile - Exists in 6 oxidation states	Alloys; Mn (III): corrosion resistance Mn (IV): rubber additive, catalyst Mn (II): powerful oxidant Fertilizers; Fireworks; Pesticides; Cosmetics	-Hypotension; Weakness; Dystonia; -Anxiety; Sperm damage; Loss of sex drive; Pneumonia
Co	-Density: 8.86 g/cm^3 - 32th most abundant element - Ores as cobaltite; red cobalt; skutterudite - Lustrous metal - Magnetic properties	Magnets (when alloyed with aluminum and nickel) In jet turbines; Electroplating The blue color in paint, porcelain, pottery, glass and enamels Radioactive 60-cobalt used in food preservation and cancer treatment	- Skin and respiratory issues; Lung hemorrhage; Wheezing; Asthma; Pneumonia and fibrosis; Cardiac effects; Liver and kidney congestion.
Ni	-Density: 8.9 g/cm^3 -22nd most abundant metal – Minerals as iron-nickel sulphide; garnierite - Silvery metal - Resist corrosion at high temperature	Jeweler; Coins; Plating other metals to avoid corrosion; Stainless steel alloy; Welding; Armor plating; Rocket engines	Lung embolisms; Asthma; Allergic reactions; Heart disorders; Possible cancer; Respiratory failure
Cu	- Density: 8.96 g/cm^3 -26th most abundant metal - Reddish-gold color - Mineral - Good conductor of heat and electricity peacock ore - Essential element	Copper alloys such as bronze and brass; Copper wires; Plating; Coins; Pipes; Fertilizer; Preservation of wood; Preservation of fabric; Barrier cream	Diarrhea; Vomiting; Irritation of the eyes; Dizziness; Irritation caused in the mouth cavity; Hepatic and kidney disease; Wilson's disease; Dead
Zn	- Density: 7.134 g/cm^3 - 24th most abundant metal - Silvery-white metal with a blue tinge - Ores such as zinc blende and calamine - Essential element	Die-casting; Painting; Cosmetic; Soap; Deodorants; Anti-dandruff shampoo; Weapons; Electrical equipment; Plastic; Ink; Pharmaceutical; Textile; Rubber; Fluorescent lights	Nausea and vomiting; Stomach cramps; Decrease in high-density lipoprotein (HDL) cholesterol; Pancreatic complications; Fatigue; - Epigastric pain; Copper deficiency; Impaired immune function
As	-Density: 5.75 g/cm3 - 55th most abundant metal - Three allotropic forms -The minerals: Arsenopyrite, Realgar, Orpiment and Enargite -Brittle - Bright silvery-grey in color	Preservation of wood, glass production; Insecticides formulations; Doping agent in semiconductors; Pyrotechnics; Bronze production.	Gastro-intestinal system irritation; Lung irritation; Decreased production of both red blood cells and white blood cells; Heart problems; Deoxyribonucleic acid (DNA) damage; Increased chances of cancer have been suggested.
Cd	- Density: 8.69 g/cm^3 -64th most abundant metal -Found frequently in combination with zinc - Greenockite mineral - Silvery bluish tint meta	Phosphate fertilizer; Pesticides; Nickel–Cadmium batteries; Glassware pigmentation; Corrosion-resistant plating; Stabilizer in plastic production; Nuclear reactors	Kidneys primarily affected causing nephrotoxicity; reproductive system failure; Psychological disorders; Central nervous system complications; Immune system deficiencies; DNA impairment; Cancer
Pb	-Density: 11.3 g/cm3 -37th most abundant metal - Galena mineral ore -Dull silver-grey metal	Lead-acid batteries in cars; Lead crystal glass; Computer screen sheets to safeguard from radiation; In buildings for roofing; painting; Weight belts for divers; Cable sheeting; Canister for corrosive liquids	Hypertension; Miscarriages; Premature and low births; Stillbirths; Renal impairment; Brain injury; Abdominal pain,

Table 4.1. Input parameters and its values specified in MPPD model (ICRP, 1994)

Parameter	**Upper respiratory tract volume(ml)**	**Functional residual capacity (ml)**	**Total lung capacity (ml)**	**Tidal volume (ml)**	**Breathing frequency (per minute)**
0-1y	2.45	17.97	43.98	30.44	39
1-3 y	7.92	30.81	90.37	100.1	26
3-6y	9.47	48.2	125.15	121.3	24
6-11y	21.03	501.32	1143.93	278.2	17
11-21 y	30.63	987.56	1943.27	388.1	16
> 21 y	42.27	2123.75	4765	477.2	14

Table 5.1. The Reference dose (RfD) and slope cancer factor (CSF) of TEs

Trace Element s	RfD (mg/kg.day) [a,b]	CSF (kg×day/mg) [a]
Cr	3.0E-05	4.2E+01
Mn	1.4E-05	-
Co	3.0E-04	9.8E+00
Ni	5.0E-02	9.0E-01
Cu	4.0E-03	-
Zn	3.0E-01	-
As	3.0E-04	1.5E+01
Cd	1.0E-04	6.3E+00
Pb	3.5E-03	-

[a] USEPA, 2020. United environmental Protection Agency. Integrated risk information risk (IRIS)
[b] Peng Gao et al, 2017. "Exposure and health risk assessments of $PM_{2.5}$ bounded TEs during winter in university campus in Northeast China

Table 5.2. Concentrations of TEs bound to PM and comparison with other studies

PM	Environment	Season	PM (µg/m3)	ng/m3										Study
				Cr	**Mn**	**Co**	**Ni**	**Cu**	**Zn**	**As**	**Cd**	**Sn**	**Pb**	
$PM_{2.5}$	Working (China)		152.1±78.3 (72.3-273.2)	266.2±26.2 (103.1-517.4)	171.1±85.5 (62-347.9)	2.9±1.1 (1.5-5.2)	50.4±28.7 (14-101)	61.1±17.7 (33.2-98.1)	705.5±432.2 (332.2-1605.7)	33.9±8.2 (16.2-44.6)	2.7±1.4 (1.4-5)	-	190.9±178.5 (77.8-570.9)	(Li et al., 2013)
$PM_{0.6}$	Academic Campus in India		94.44 (17.5-330.7)	74.5 (15-5574)			410 (4-4500)	989 (9-532)					699 (18-18408)	(Suryawanshi et al., 2016)
$PM_{2.5}$	Road site homes		130.4±37. (83.84-184.67)	670±290 (310-1130)	200±380 (20-970)	-	430±260 (80-650)	420±170 (240-740)	1060±710 (440-2260)	-	-	-	1100±430 (650-1840)	(Kulshrestha et al., 2014)
$PM_{2.5}$	Urban site homes in India		116.9±30.7 (80.1-168.7)	640±540 (50-1350)	60±10 (49-80)	-	360±200 (90-680)	260±160 (60-500)	1450±770 (420-2450)	-	-	-	840±100 (730-980)	(Kulshrestha et al., 2014)
$PM_{2.5}$	Hospital (Portuguese)		23±10	2.14 (0.85-4.81)	2.73 (0.49-5.78)		3.02 (0.77-7.74)	-	-	80.3 (39.8-140)	-	--	11.3(3.65-20.3)	(Slezakova and Morais, 2014)
$PM_{2.5-10}$			7.4±4.1	0.625(0.1-2.07)	0.77 (0.06-3.53)		0.506 (0.07-1.96)	-	-	2.71 (0.27-99.5)	-	-	0.703 (0.39-8.75)	(Slezakova and Morais, 2014)
$PM_{2.5}$	Residential homes in Najing, China	Winter	12-91	4.12 (3.85-4.51)	18.66 (15.15-20.72)		31.71 (25.42-35.99)	6.75 (5.6-8.97)	33.53 (27.35-29.86)	6.56 (5.51-7.01)	0.11 (0.16-0.26)		11.01 (6.03-15.01)	(Wang et al., 2018)
$PM_{2.5}$	University campus in China													
	Atrium	Winter	128.8±44.7	250±70	290±90	720±220	60±30	880±190	220±70	30±30	50±50		110±80	(Gao et al., 2017)
	Laboratory		99.8±19.3	170±50	180±60	410±160	50±30	590±160	200±70	30±20	20±30		90±50	
	Empty room		78.4±17.3	180±60	190±60	480±210	50±30	610±190	190±70	20±20	20±40		90±60	
$PM_{2.5}$	Student dormitory in China	Summer	61.23 (19.62-131)	53.1±52.4	32.±21.6	0.1±0.1	4.8±8.9	20.4±11.1	190.3±118.5	11.3±10.82	2.7±2.66		69.2±59.4	(Wang et al., 2019)
$PM_{2.5}$	School environment in Hanoi, Vietnam	Winter	49.4 ± 41.0	-	5.7±5.2	2.26±1.2	1.5±1.3	18.2±14.7	119±89.1	0.5±0.4	0.2±0.2	0.1±0.1	1.5±1.6	(Tran et al., 2020)
$PM_{0.1}$			3.1 ± 1.3	-	0.07±0.1	0.07±0.1	0.07±0.1	1 ±0.7	6.82±3.66	0±0	0.1±0.1	0±0	0.1±0	
PM_1			10±2	1	4		2	11	28	2				

$PM_{2.5}$				170 ± 309	1119 ± 786		109 ± 82	1450 ± 1065	1865 ± 2506				695 ± 207	
$PM_{0.25-0.5}$	Indoor environment in India		54.1 ± 16.3	32 ± 58	241 ± 443		11 ± 8	96 ± 173	26±58				130 ± 45	(Rohra et al., 2018)
$PM_{0.5-1}$			29.9 ± 12.3	39 ± 135	245 ± 197		35 ± 40	849 ± 792	698 ± 1163				255 ± 181	
$PM_{1-2.5}$			47.6± 10.1	99 ± 161	634 ± 557		65 ± 44	504 ± 614	1141 ± 1001				310 ± 178	
$PM_{2.5}$	Residential houses in China	Heating season	129.3		28.6			25.3	144.5	32.3	3.9		85.2	(Yang et al., 2018)
		Non heating season	66.9		40.4			18.61	254.3	17.1	2.6		112.4	
$PM_{2.5}$	Residential dwellings in Hanoi, Vietnam	Winter	73.3±28.9	58.4±21.8	51.3±34.2	3.4±4.2	40.3±2.	26.4±2.8	1189.8±644.6	17.2±3.3	3.4±0.9	10.9±1.6	110.5±20.1	This study
$PM_{0.1}$			8.5±0.5	34.4±17	10.9±3.9	0.6±0.1	8.1±2.8	5.2±0.4	101.7±15.7	5.3±1.4	0.5±0.1	4.1±1.	20.5±3	
$PM_{2.5}$		Summer	46.4±2.5	28.3±11.9	25.3±9.5	0.5±0.1	21.1±16.1	14.9±8.7	547.4±270.1	8.3±2.1	1.9±1.2	6.4±0.7	83.4±28.6	
$PM_{0.1}$			8±0.75	16.2±4.3	5±1	0.2±0.1	7.1±4.3	2.2±0.7	34.4±9.8	1.5±0.7	0.2±0.1	1.2±0.5	8.7±1.5	

Table 5.3. The values of LOD and LOQs of 10 TEs

No	Compounds	LOD (ng/m^3)	LOQ (ng/m^3)
1	Cr	0.2	2.0
2	Mn	0.1	0.6
3	Co	0.1	0.5
4	Ni	0.1	0.6
5	Cu	0.1	0.5
6	Zn	0.5	5.0
7	As	0.1	0.5
8	Cd	0.1	0.6
9	Sn	0.1	0.5
10	Pb	0.1	0.5

Table 5.4. Recovery test in ICP-MS analysis in TEs (spiked method)

No	Compounds	Recovery test (%)
1	Cr	92-117
2	Mn	93-113
3	Co	96-101
4	Ni	88-107
5	Cu	92-110
6	Zn	85-120
7	As	94-108
8	Cd	95-105
9	Sn	93-104
10	Pb	96-106

Chapter 6

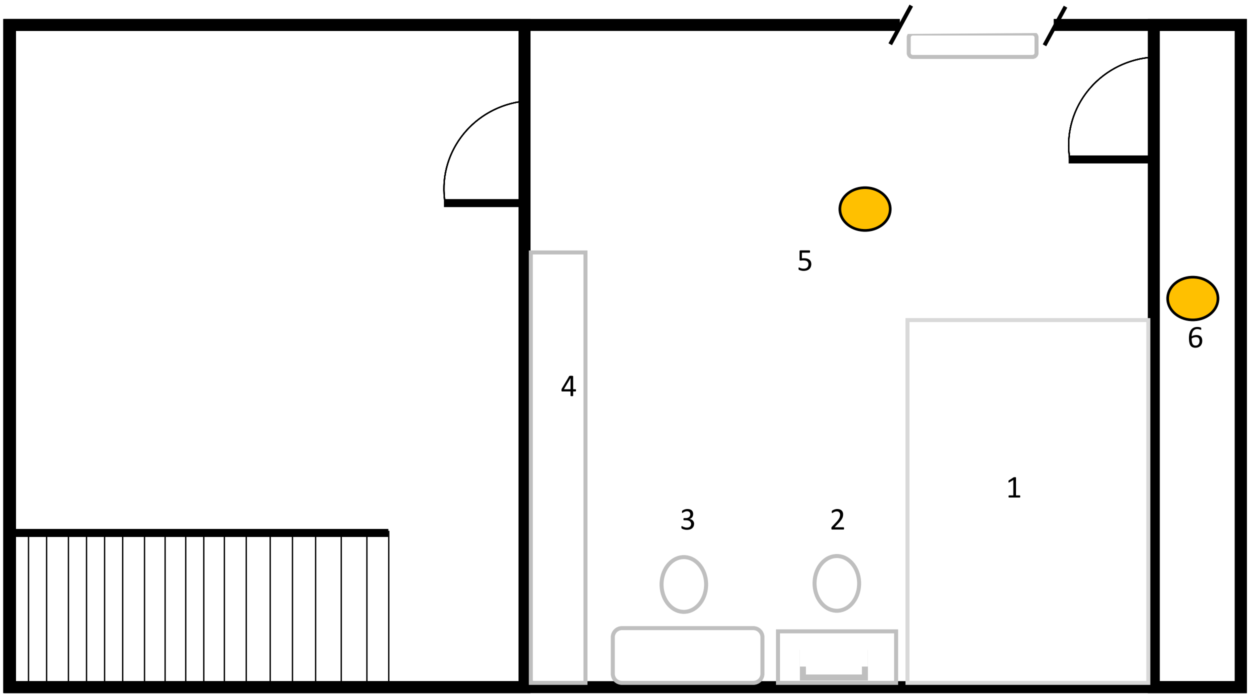
Table 6.1. The values of LOD and LOQs of 15 PAHs

No	**Parameters**	**LOD (ng/m^3)**	**LOQ (ng/m^3)**
1	Naphthalene (Nap)	0.02	0.06
2	Acenaphthylene (Acy)	0.05	0.15
3	Acenaphthene (Ace)	0.03	0.11
4	Fluorene (Flu)	0.01	0.05
5	Phenanthrene (Phe)	0.05	0.18
6	Anthracene (Ant)	0.03	0.10
7	Fluoranthene (Flt)	0.02	0.06
8	Pyrene (Pyr)	0.01	0.04
9	Benz(a)anthracene (BaA)	0.03	0.09
10	Chrysene (Chr)	0.03	0.09
11	Benzo(b) fluoranthene (BbF)	0.02	0.06
12	Benzo(a)pyrene (BaP)	0.01	0.04
13	Indeno(1,2,3-cd) pyrene (IndP)	0.01	0.02
14	Dibenz(a,h)anthracene (DahA)	0.01	0.04
15	Benzo(g,h,i)perylene (BighP)	0.01	0.02

Table 6.2. Proficiency testing sample-based recovery and stability of analytical signal in GC-MS/MS analysis of PAHs

No	**Compounds**	**Reported concentration (ng/g)**	**Recovery efficiencies (%)**
1	Naphthalene (Nap)	467	95.3
2	Acenaphthylene (Acy)	290.4	106.8
3	Acenaphthene (Ace)	185.1	80.2
4	Fluorene (Flu)	201.2	103.0
5	Phenanthrene (Phe)	2688	92.5
6	Anthracene (Ant)	663.1	93.5
7	Fluoranthene (Flt)	6753	99.9
8	Pyrene (Pyr)	5387	82.6
9	Benz(a)anthracene (BaA)	3607	78.0
10	Chrysene (Chr)	3822	80.1
11	Benzo(b) fluoranthene (BbF)	4246	88.7
12	Benzo(a)pyrene (BaP)	3345	78.0
13	Indeno(1,2,3-cd) pyrene (IndP)	2703	83.2
14	Dibenz(a,h)anthracene (DahA)	647.2	85.5
15	Benzo(g,h,i)perylene (BighP)	2703	84.9

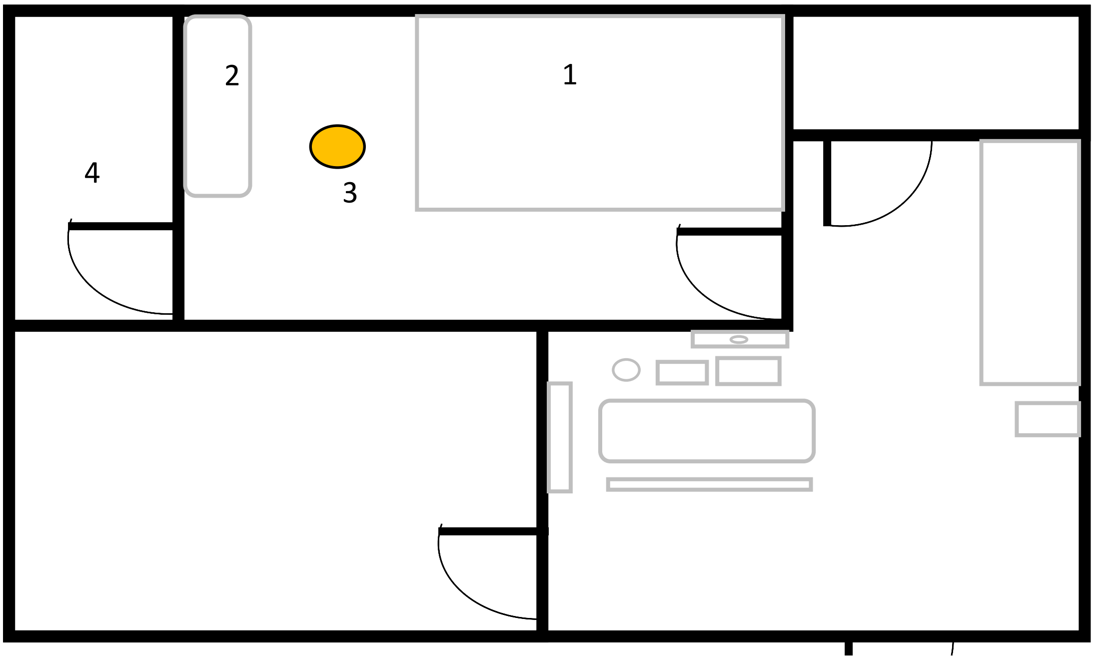
a)



1. Bed; 2. Table; 3. Computer; 4. Wardrobe; 5. Indoor inlet; 6. Outdoor inlet

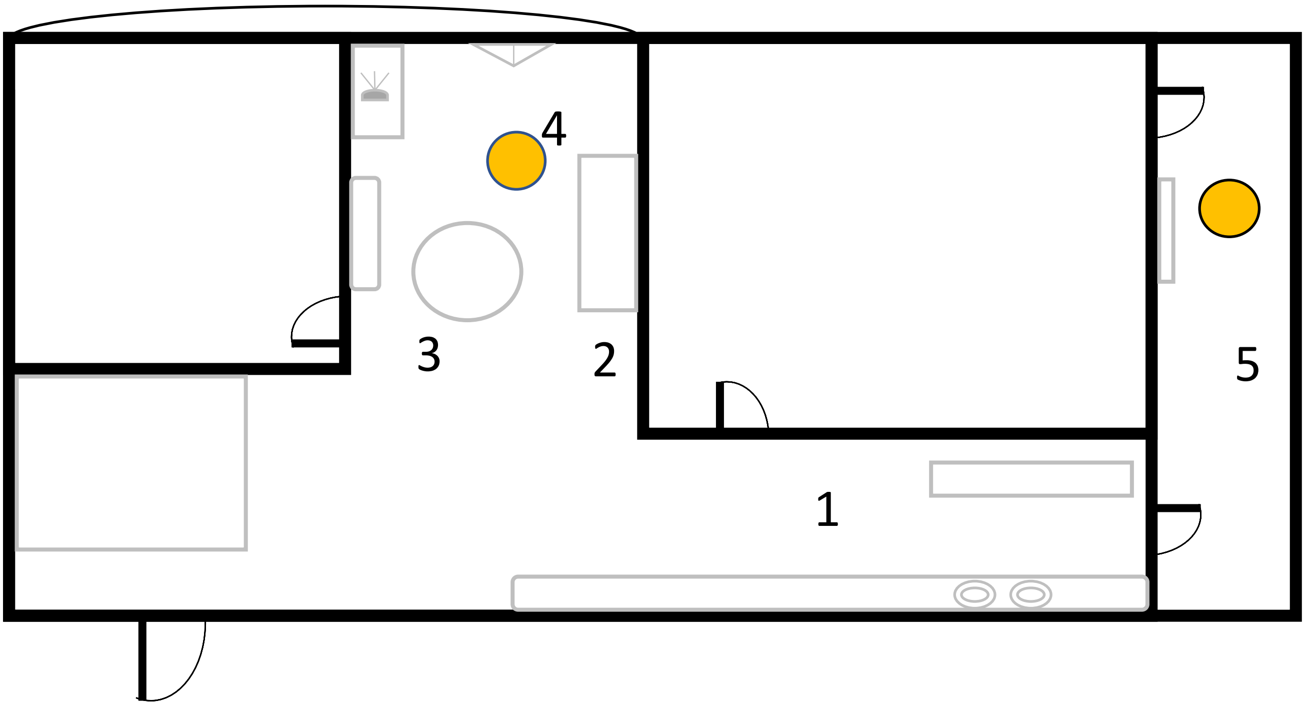
Fig. 5.1. (a). The layout of the sampling site K1

b)



1. Bed 2. Table 3. Indoor inlet 4. Outdoor inlet

Fig. 5.2 (b). The layout of the sampling site K2 c)



1. Kitchen; 2. Sofa; 3. Table; 4. Indoor inlet; 5. Outdoor inlet.

Fig. 5.1.(c). The layout of the sampling site K3

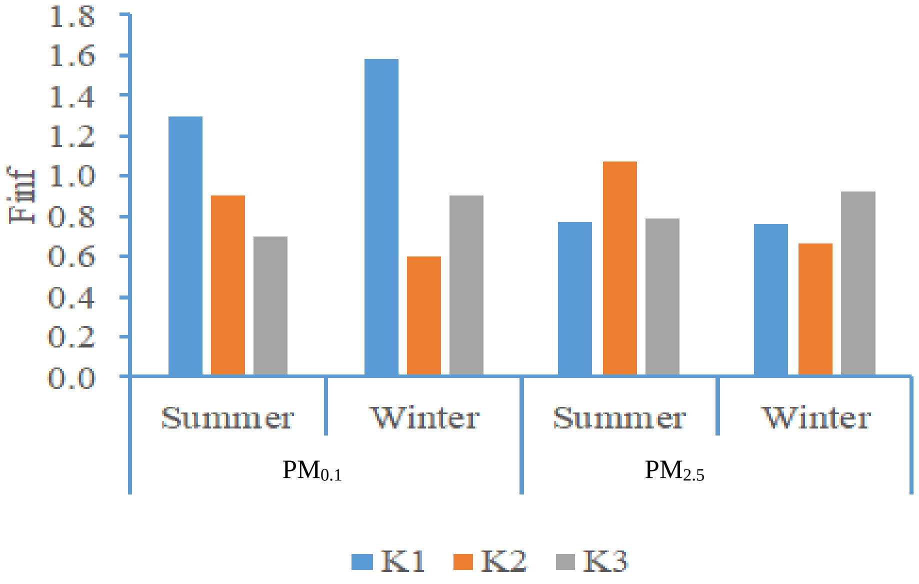


Fig 5.2. The values of F_{inf} of $PM_{2.5}$ and $PM_{0.1}$ at the three dwellings

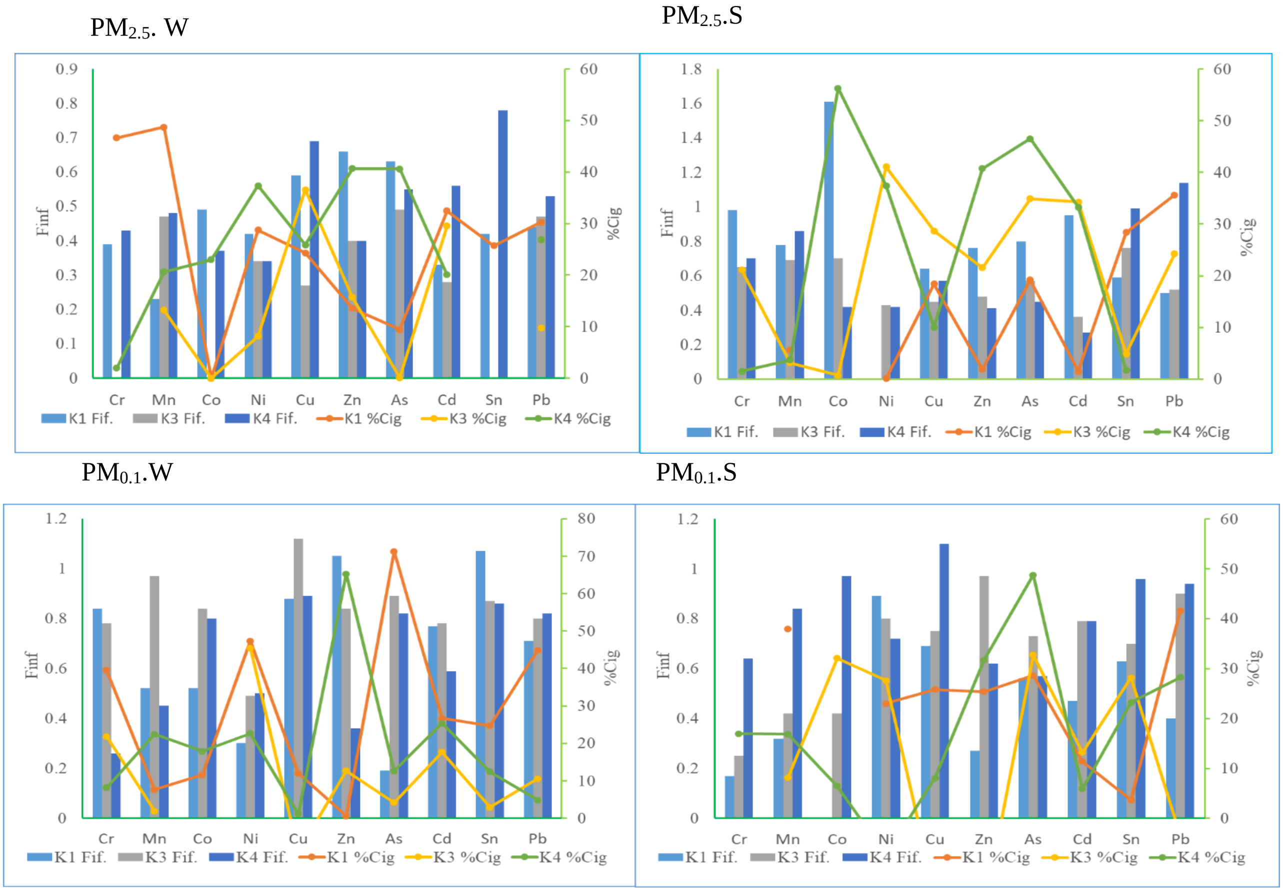


Fig.5.3. The values of F_{inf} and $\%C_{ig}$ at the three dwellings

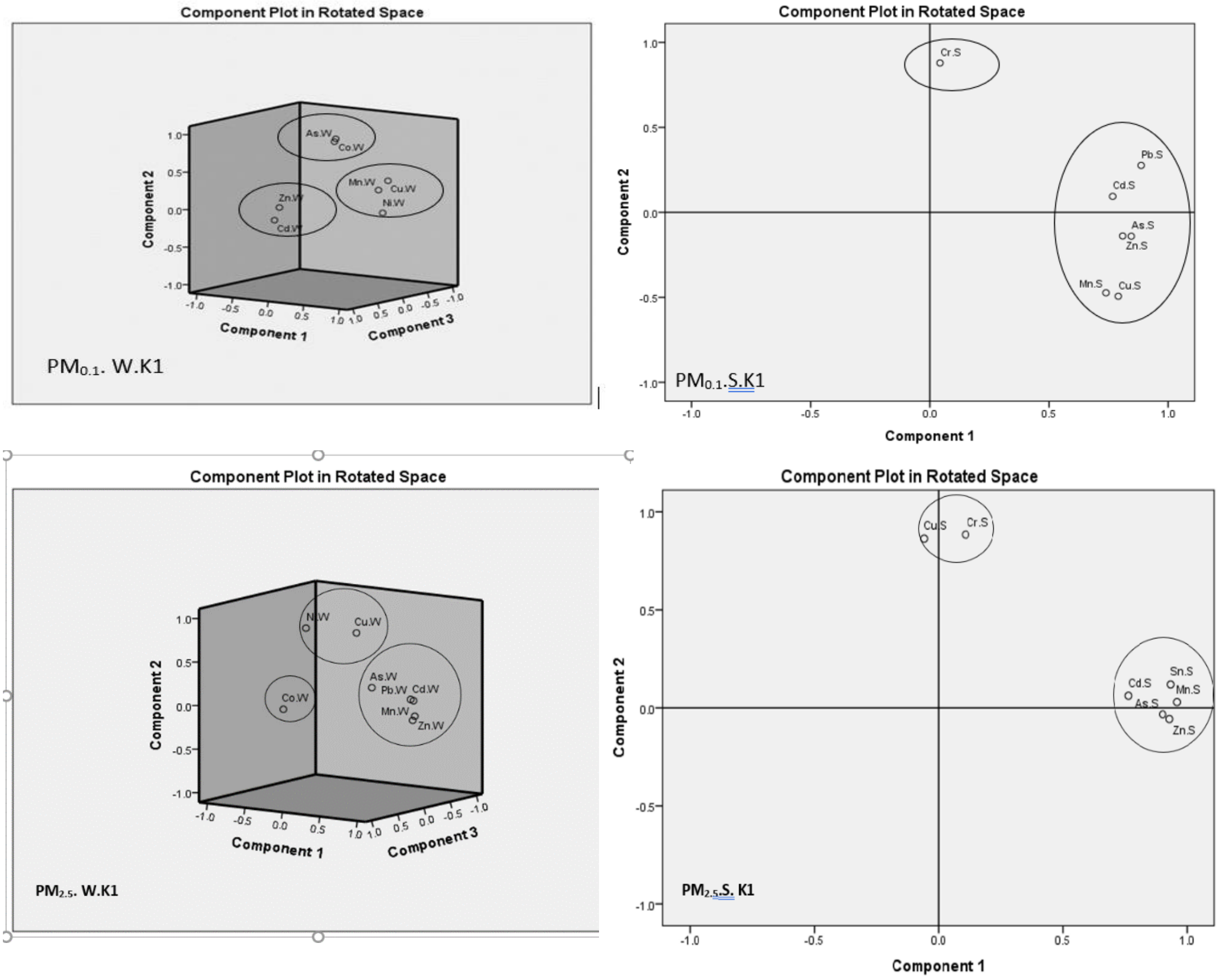


Fig. 5.4 (a). Results extracts from PCA analysis at K1

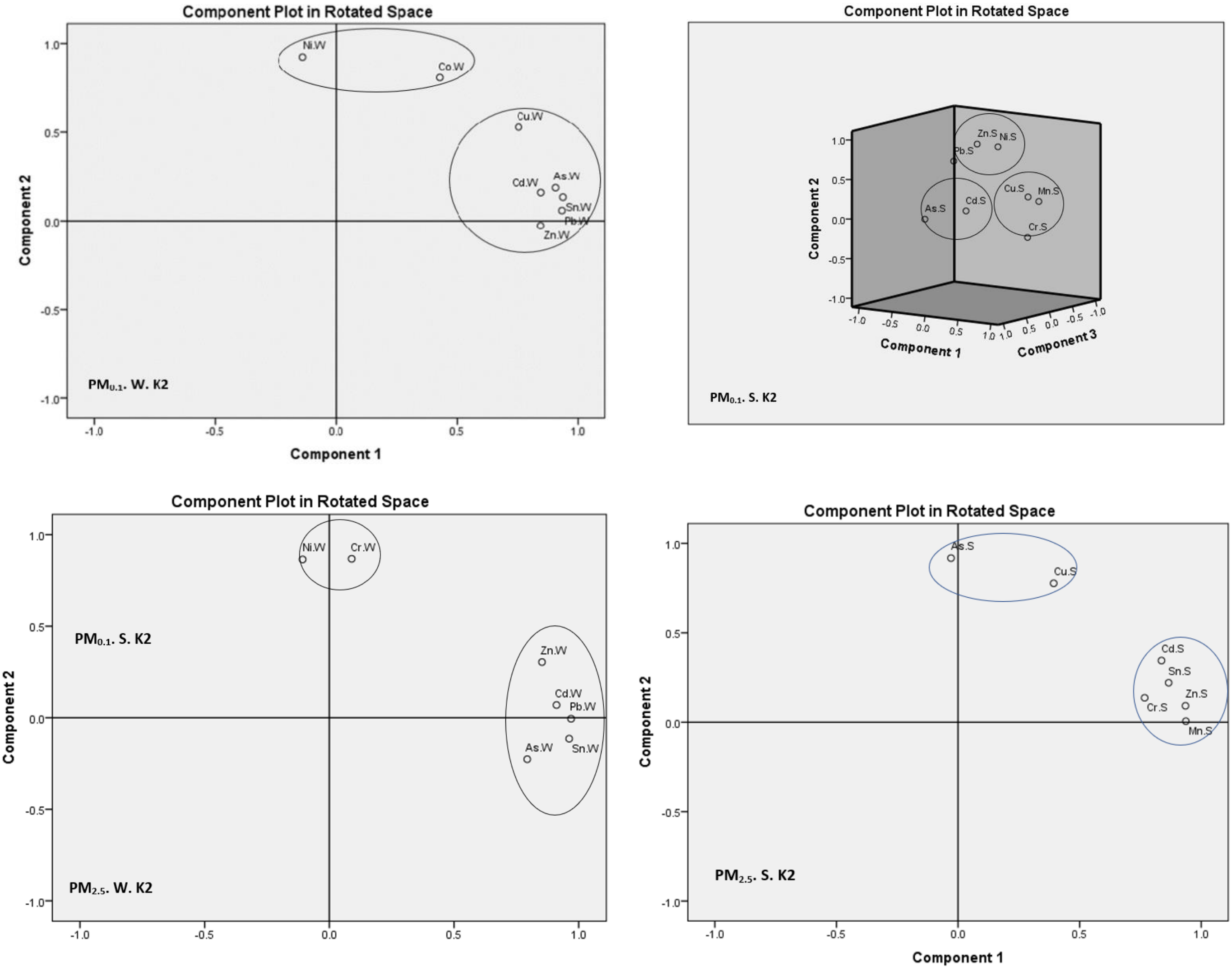


Fig. 5.4 (b). Results extracts from PCA analysis at K2

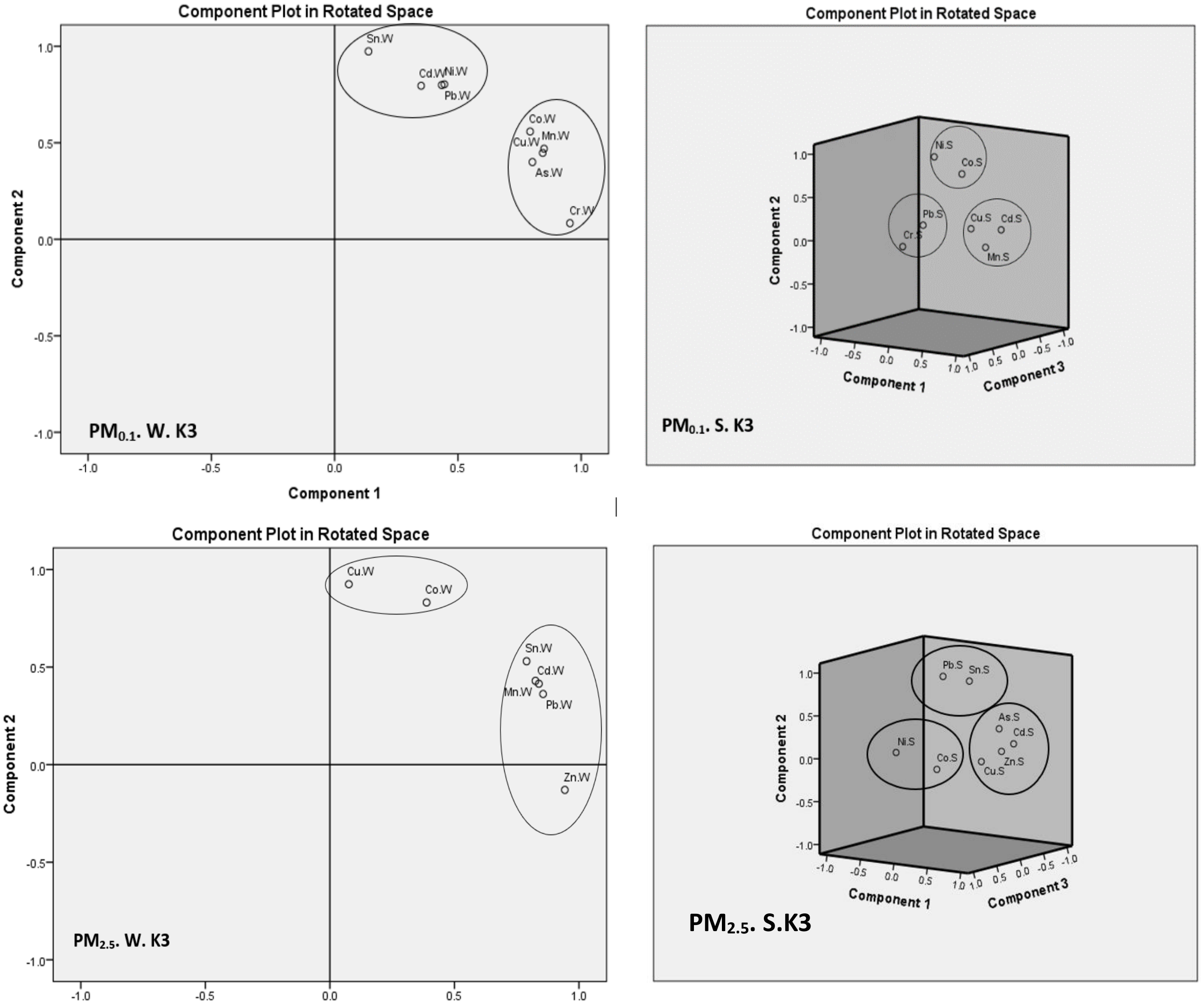


Fig. 5.4 (c). Results extracts from PCA analysis at K3