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Research article

Human health risk assessment of $PM_{2.5}$-bound heavy metal of anthropogenic sources in the Khon Kaen Province of Northeast Thailand

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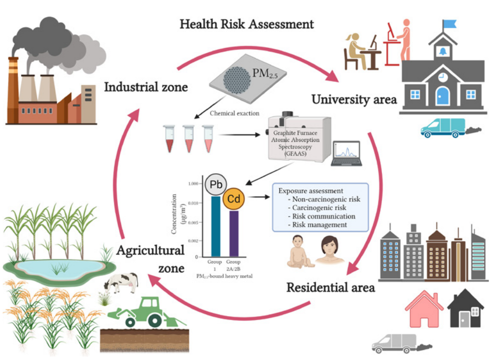
GRAPHICAL ABSTRACT



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ABSTRACT

The study aimed to assess the human health risk of $PM_{2.5}$-bound heavy metals from anthropogenic sources in Khon Kaen Province, Thailand between December 2020 and February 2021. According to the findings, the geometric mean concentration of $PM_{2.5}$ in the university area, residential area, industrial zone, and the agricultural zone was 32.78 μg/m^3, 50.25 μg/m^3, 44.48 μg/m^3, and 29.53 μg/m^3, respectively. The results showed that the estimated human health risk assessment, in terms of non-carcinogenic risks among children and adults in an urban area (residential and university), industrial zone, and the agricultural area, was of hazard index (HI) value of >1.0 indicating a greater chance of chronic effects occurring. This study showed that exposure to $PM_{2.5}$-bound heavy metal may increase the likelihood that lasting effects will result in a very high carcinogenic risk (CR) in children in residential areas, and an industrial zone with total carcinogenic risk (TCR) values of 0.23×10^{1}, and 0.12×10^{1}, respectively while resulting in a high TCR of 3.34×10^{-2} and 4.11×10^{-2}

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within the university areas and agricultural zone, respectively. In addition, health risk assessments among adults demonstrate high TCR values of 4.40×10^{-1} (residential area), 2.28×10^{-1} (industrial zone), and 7.70×10^{-3} (agricultural zone), thus indicating a potential health risk to adults living in these areas while the university area was very low effects on carcinogenic risk (CR ≤ 10^{-8}) for adults. Therefore, lowering the risk of exposure to $PM_{2.5}$ via the respiratory tract, for example, wearing a mask outside is a very effective self-defense strategy for people within and around the study site. This data study strongly supports the implementation of the air pollutant emission source reduction measures control and health surveillance.

1. Introduction

Air pollution is one of the most serious environmental problems to be addressed locally, regionally, and globally. It is endangering human health and life, as well as having a variety of indirect effects on society and the economy, such as weakening a country's economic growth potential by reducing people's working hours and lowering agricultural production (Narita et al., 2019). In 2017, Exposure to $PM_{2.5}$ resulted in 4.58 million deaths and 142.52 million years of disability worldwide, with ambient $PM_{2.5}$ accounting for 64.2 percent of fatalities and 58.3 percent of disability-adjusted life years (Bu et al., 2021). Chronic exposure to $PM_{2.5}$ has been linked to respiratory diseases, heart disease, stroke, brain, and lung cancer (Cao et al., 2011; Du et al., 2015; Fold et al., 2020; Sakunkoo et al., 2022; Pope Iii et al., 2002). PM_{10} and $PM_{2.5}$ majority include road traffic, biomass and waste open burning and industrial area (Fang et al., 2013; Li et al., 2021a; Tobler et al., 2020; Wang et al., 2020), construction of buildings and demolition (Yan et al., 2019), crop residue burning (Yang et al., 2008), and informal electric-waste recycling site (Zheng et al., 2016). PM contains a mixture of a chemical component, which can be produced from primary sources or formed by complex atmospheric reactions. (Wang et al., 2020; Zhang et al., 2015). $PM_{2.5}$ during 2016-2019 in Bangkok, 24-h Thailand Ambient Air Quality Standards (TNAAQs) a dose of 50 μg/m^3 was exceeded for about 50 days/years and a maximum daily concentration of more than 100 μg/m^3 was observed during the dry season. (November to April) (Fold et al., 2020). Previous conducted study PM episode, Ji County in Tianjin, China by Zhou et al. (2015) has reported health impact with a relative risk (RR) were 1.013 (CI 1.0074–1.019) for all-cause mortality per 10 μg/m^3 increase in $PM_{2.5}$. Therefore, chronic exposure to $PM_{2.5}$ is known to produce excessive exposure to the alveolar angiotensin-converting enzyme 2 (ACE-2) receptor, which may promote infection or toxicity in those exposed to air pollution (Domingo et al., 2020), especially, as long-term exposure to ambient $PM_{2.5}$ is linked to increased morbidity and premature death in epidemiological studies (Pope and Dockery, 2006; Tobler et al., 2020; Zhu et al., 2019). Dockery and Pope (1994) reported that the increase in mortality and morbidity in urban areas may be associated with exposure to particulate matter (PM). The potential for adverse health effects from PM varies in particle size, composition, toxicity, and degree of exposure. In addition, the hospitalization rate for chronic obstructive pulmonary disease (COPD) increased by 6.88%, and the total number of hospital, patient, and asthma emergency rooms increased by 0.67%, 0.65%, and 0.49%, respectively of 10 μg/m^3 for $PM_{2.5}$ concentration (Duan et al., 2020; Xie et al., 2019).

Furthermore, the chemical compositions of $PM_{2.5}$ include organic matter, trace elements, water-soluble inorganic ions, inorganic substance, sulfate, nitrate, and polycyclic aromatic hydrocarbons (PAHs) and others (Chen et al., 2021; Cheng et al., 2015; Fan et al., 2021; Jiang et al., 2018; Sajjadi et al., 2018; Zhang et al., 2021). In particular, a small fraction of particulate matter with aerodynamic equivalent diameter ≤2.5 μm constitutes heavy metals, and is highly toxic even at low concentrations, levels such as Cr, Cd, As, Pb, and Ni (Baensch-Baltruschat et al., 2020; Nirmalkar et al., 2021). A recent study has exhibited that heavy metals attached in $PM_{2.5}$, such as Pb, Cu, Ni, and Fe collected from high-traffic regions, have been shown in recent study to be capable of triggering a variety of respiratory and cardiovascular illnesses, even a low concentration level of metals in $PM_{2.5}$ can be harmful to human health (Kastury et al., 2018; Moryani et al., 2020). Metal elements from natural sources such as soils/crustaceans are usually found in coarse particles, while fine particles are associated with anthropogenic sources (Ny and Lee, 2011; Shaltout et al., 2019). Heavy metals can distribute and accumulated in the environmental cycle, for example, surface soil (Han et al., 2021), groundwater (Varol and Tokatlı, 2022) and atmosphere (Li et al., 2021b; Moryani et al., 2020; Pongpiachan et al., 2017; Wu et al., 2019; Xu et al., 2020; Zheng et al., 2016; Zhi et al., 2021). Previous studies shown a linkage between $PM_{2.5}$ exposure and toxic heavy metal components having negative adverse health effects (Liu et al., 2018; Zhang et al., 2015). Currently, several toxicological studies have pointed out that excessive exposure to heavy metals in $PM_{2.5}$ could threaten on human health, causing respiratory and inflammation of lung, cardiovascular disease, heart disease, and cancer (Xie et al., 2020; Xu et al., 2020; Zhang et al., 2021), specially, the heavy metals and organic pollutants adsorbed by $PM_{2.5}$ were risk factors for lung cancer (Duan et al., 2020). Oral ingestion, inhalation, and dermal contact are the main routes of exposure to airborne heavy metals in $PM_{2.5}$ (Hu et al., 2012; MohseniBandpi et al., 2018; Wang et al., 2020; Zhang et al., 2021), and are they are easily absorbed by the human body's alveolar, blood, cutaneous, and gastrointestinal systems (Cao et al., 2012; Imani, 2021). Undoubtedly, the study results show the possibility that adverse health effects are associated with particulate matter exposure bound to toxic heavy metals. To date, the chemical characterization, human health risk assessment, and source apportionment of $PM_{2.5}$-bound heavy metals have been widely studied (Chen et al., 2021; Li et al., 2021b; Zhang et al., 2021).

Data from the Pollution Control Department (PCD) air quality monitoring system showed that the provinces with the most frequent detections of air quality exceeding the standard more than 70 days per year such as Saraburi, Lampang, Khon Kaen, Phrae, Phayao, Nan, Chiang Mai, Chiang Rai, Tak and Mae Hong Son (PCD, 2020). Khon Kaen Province is one of the agriculture, urban and industrial areas that have been rapidly growing in Northeast Thailand due to urban development; rapid industrial economy expansion has resulted in the continued increase of many transport systems to meet human needs. This information, is therefore, an important highlight the researcher to choose to study in such areas. While studies on heavy metal concentrations in $PM_{2.5}$ in Thailand have been conducted in the past, they have been limited due to scarce data, especially on the country's regional aspect. Understanding the levels of particulate matter in the air have important public health consequences considering the increasing trend of air pollution. Hence, this study aims to examine the human health risk assessment of $PM_{2.5}$-bound heavy metal of anthropogenic sources in Khon Kaen Province of Northeast Thailand. The data of this study will help support policies and future control measures in reducing pollutant emission from anthropogenic activities while also communicating health risks and surveillance to develop a further plan in the short and long-term to minimize adverse effects on human health.

2. Materials and methods

2.1. Study area

Due to the topography of Khon Kaen Province (KKP), most of the plateaus and low mountains; the weather is quite hot in the summer and

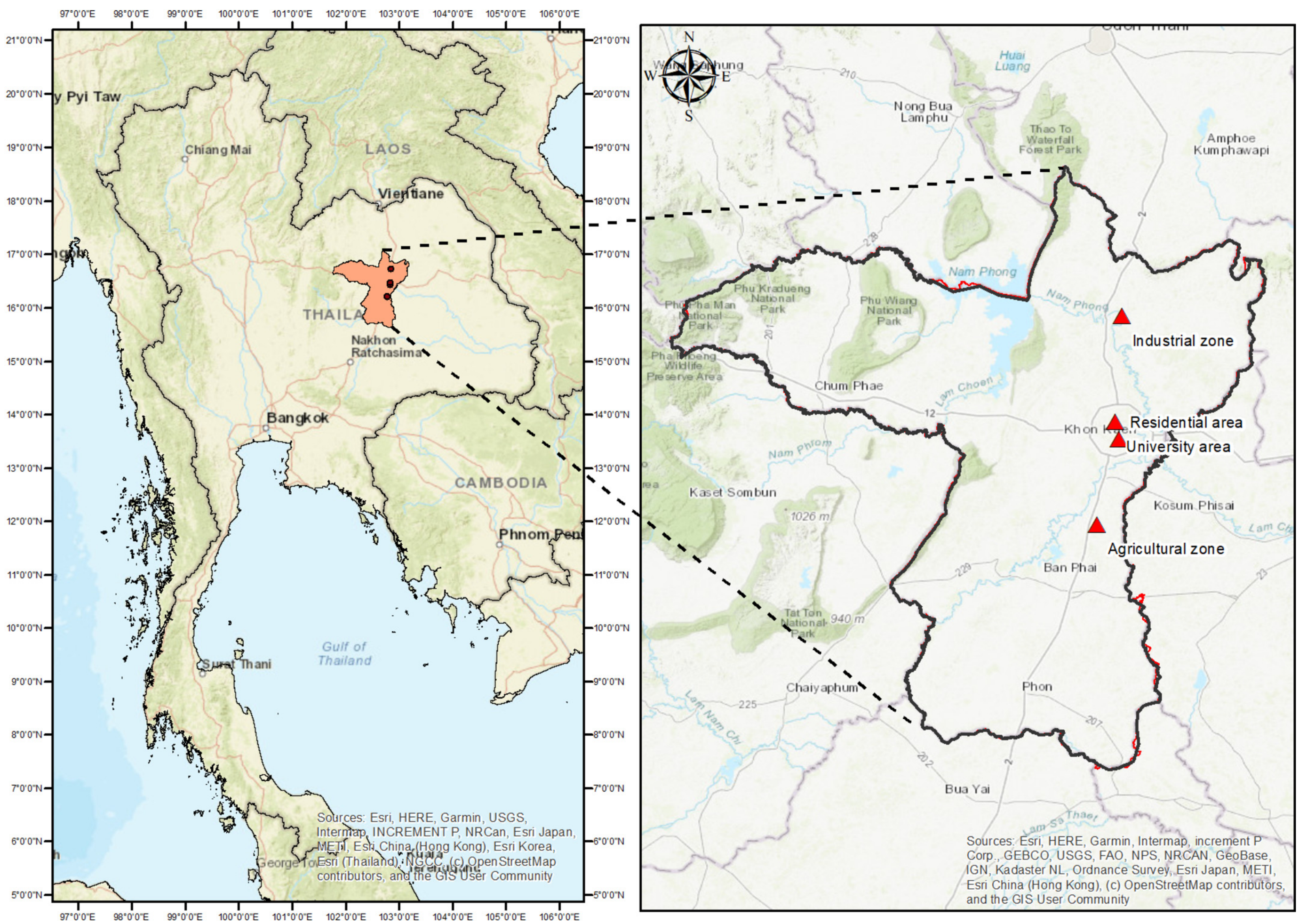

Fig. 1. Map of location of sampling site; the boundary line of this study area is Khon Kaen Province of Northeast Thailand.

the weather is quite cold in winter. The average temperature throughout the year is about 26.9 °C and the average annual rainfall 1,246.8 mm. The KKP covers an area of approximately 10,885.9 km^2 and is approximately 449 km from Bangkok. Therefore, sampling site is in Khon Kaen Province, one of the largest urban development, agricultural activities, and industries in the north-eastern part. In this study, the following locations for ambient air samples were collected: The study site in Muang Khon Kaen is 16°26′ north latitude and 102°50′ east longitude (166 m above of mean sea level) is represented by urban area (includes university, and residential), and industrial zone and covering an area of 953.4 km^2 and agricultural areas in Ban Haet District (BHD) and covering an area of 205.2 km^2. The location of BHD is 16°12′ north latitude and 102°45′ east longitude at 189 m above of mean sea level (Fig. 1). The pattern of land use in KKP has shifted drastically over the past few decades. The Thai Meteorological Department (TMD, 2015, 2020) has divided the climate into three seasons: local summer (mid–February to mid–May), the rainy season (mid–May to mid–October), and the winter season (mid–October to mid–February). The KKP is the country's largest urban development region and the industry has accelerated environmental concerns. This is because the region is facing air pollution issue for nowadays; in particular, Khon Kaen is one of the provinces in north-eastern, Thailand faces a high level of seasonal air pollution every year. The community dwellers are exposed to outdoor air pollutants 24 h a day during seasonal smog periods because of their open-air housing style, and agricultural occupational hazards.

2.2. Sampling collection and analysis

The $PM_{2.5}$ samples were collected from ambient 24 h air sample collection during December 2020 and February 2021 period (dry season) using a high-volume air sampler (model ECO-HVS3000, Ecotech, Australia) was used with a flow rate of 67.8 m^3/h, at 1.50 m above ground level. After sampling, each glass fiber (GF) filter was used to adjust the filter paper before and after collecting samples by putting it in a desiccator not less than 24 h. Inside the desiccator, silica gel was placed at the bottom. These samples are then stored in the refrigerator at a temperature of −30 °C. During the previous treatment procedures, each load filter was cut and placed in a Teflon vessel, and 5 mL of HNO_3, 2 mL of HF, and 1 mL of H_2O_2 were added; later, the collected $PM_{2.5}$ was wet and digested by ultrasonic-assisted digested (model TRANSSONIC 700/H). After cooling, these solutions were diluted to 5 mL with 5% HNO_3 for use in subsequent metal analysis. In this study, seven trace elements (Pb, Cu, Cd, Fe, Mn, Al, and Zn) were measured by Graphite furnace atomic absorption spectroscopy (GFAAS) (model PinAAcle900Z, Perkin Elmer). $PM_{2.5}$ sampling and analysis methods, as well as quality assurance and quality control measures, were in line with the $PM_{2.5}$ component manual established by Chow et al. (1998) and Sugita et al. (2019).

2.3. Statistical analysis and model calculation

2.3.1. Enrichment factor (EF)

Enrichment factor (EF) was used to assess the level of heavy metal contamination in atmospheric particles and method facilitated the separation of heavy metal contribution between natural and anthropogenic origin. The EF by Zhang et al. (2021) where calculation is expressed as follows:

$$EF = \frac{\left(\frac{C^{i}_{sample}}{C^{ref}_{sample}}\right)}{\left(\frac{C^{i}_{crust}}{C^{ref}_{crust}}\right)} \quad (1)$$

where $(C_i/C_{ref})_{sample}$ and $(C_i/C_{ref})_{crust}$ denote the concentration levels of i metal and the iron metal in the sample and crust. Fe was selected as the most common reference in this study (Pongpiachan et al., 2017) and the abundance of the elements' concentration of seven metals in the continental crust (in mg/kg) were obtained from (Lide, 2004). EF values are classified into five levels based on the influence of human activities (Yongming et al., 2006), were not influenced by human activities (EF < 2), moderately influenced (EF 2–5), highly influenced (EF 5–20), highly influenced (EF 20–40), and were severely influenced (EF > 40), respectively.

2.3.2. Geo-accumulation index (I_{geo})

The geo-accumulation index (I_{geo}) was used to assess the level of heavy metal contamination by comparing the current concentration with rear levels. The calculation of I_{geo} index was done by Zhang et al. (2021) using Eq. (2)

$$I_{geo} = \log_2 \frac{C^{i}_{sample}}{1.5 \times C^{i}_{crust}} \quad (2)$$

where C^{i}_{sample} and C^{i}_{crust} represent the concentrated level of the i metal in the sample and background, respectively. The factor of 1.5 was applied as a background adjustment matrix for evolutionary and anthropogenic effects (Li et al., 2015). In this study the geological heavy metal deposition index. The geo-accumulation index; I*geo* was classified into six levels of contamination according to the study guideline of Murari et al. (2020): Level 1: ≤ 0 (Uncontaminated), Level 2: 0–1 (Uncontaminated to Moderately contaminated), Level 3: 1–2 (Moderately contaminated), Level 3: 2–3 (Moderately to strongly contaminated), Level 4: 3–4 (Strongly contaminated), Level 5: 4–5 (Strongly to extremely contaminated), and Level 6: > 5 (Extremely contaminated), respectively.

2.3.3. Human health risk assessment (HHRAs)

The subjects were divided into two groups viz., Children and Adults based on the differences in their respiratory systems and behaviors. (Dahmardeh Behrooz et al., 2021; Hu et al., 2012). Exposure assessment is a measurement process and refers to the time when people are exposed to substances in the environment or future exposure forecast. The values used for all the parameters in health risk assessment (HRA) as illustrated in Table 1.

Thus, the daily intake of the chemical through oral ingestion (CDI^{ing}_{ij}, mg/kg/day, exposure concentration through inhalation (EC^{inh}_{ij}, μg/m³), and skin absorption dose through dermal contact (DAD^{der}_{ij}, mg/kg/day) by Zhang et al., 2021 and USEPA, 2014 were calculated using the Eqs. (3)–(5) below:

$$CDI^{ing}_{ij} = \frac{C_{ij} \times IR_{ing} \times EF \times ED \times CF}{BW \times AT} \quad (3)$$

$$EC^{inh}_{ij} = \frac{C_{ij} \times IR_{inh} \times ET \times EF \times ED}{BW \times AT_n} \quad (4)$$

$$DAD^{der}_{ij} = \frac{C_{ij} \times SA \times AF \times ABS \times EF \times ED \times CF}{BW \times AT} \quad (5)$$

The corresponding hazard quotient (HQ) was calculated as shown in Eqs. (6)–(9) and the carcinogenic risk (CR) of toxic metals through the three ways were further evaluated based on Eqs. (10)–(13).

$$HQ_{ing} = \frac{CDI^{ing}_{ij}}{RfD^{ing}_{j}} \quad (6)$$

$$HQ_{inh} = \frac{EC^{inh}_{ij}}{RfC^{inh}_{j} \times 1000} \quad (7)$$

$$HQ_{der} = \frac{DAD^{der}_{ij}}{RfD^{der}_{j} \times ABS} \quad (8)$$

$$\text{Hazard index } (HI_i) = \sum (HQ_{ij}) \quad (9)$$

$$CR_{ing} = LCDI^{ing}_{ij} \times SF \quad (10)$$

$$CR_{inh} = LEC^{inh}_{ij} \times IUR \quad (11)$$

$$CR_{der} = \frac{LDAD^{der}_{ij} \times RfD^{der}_{j}}{ABS} \quad (12)$$

$$\text{Carcinogen risk } (CR_i) = \sum (CR_{ij}) \quad (13)$$

RfD_o, (the oral reference doses (mg/kg/day)), RfC_i, (inhalation reference concentration (mg/m³)), ABS_{GI}, (gastrointestinal absorption factor), SF_o, (oral slope factor (mg/kg/day)) and IUR (inhalation unit risk (μg/m³). A hazard index (HI), i.e., HQ all added up, is used for assessing the chronic effects of non-carcinogenic risks. When both HQs ≤ 1.0 and HI ≤ 1.0, there is no significant risk of chronic effects. By contrast, HQs > 1 or HI > 1 indicate a possibility of the occurrence of chronic effects (Murari et al., 2020; Zheng et al., 2010). The carcinogen risk (CR) value shows the chance of developing any type of cancer by an individual due to a lifetime exposure to carcinogenic metals, divided into five categories: as very low (CR ≤ 10^{-6}), low (10^{-6} ≤ CR < 10^{-4}), moderate (10^{-4} ≤ CR < 10^{-3}), high (10^{-3} ≤ CR < 10^{-1}), and very high (CR ≥ 10^{-1}) (Roy et al., 2019; Zhang et al., 2021).

3. Results and discussion

3.1. Mass concentration of $PM_{2.5}$ in the typical anthropogenic sources

In this study, $PM_{2.5}$ sampling was done during the dry season to analyze the heavy metal composition of $PM_{2.5}$ in four human activities areas: University area, residential area, industrial zone, and agricultural zone found that in all sampling areas, the concentration of $PM_{2.5}$ in the atmosphere exceeds the standards set by the World Health Organization (WHO) (WHO, 2006), which must not exceed 25 μg/m³ and Pollution Control Department (PCD) of Thailand 50 μg/m³ for 24 h (PCD, 2020), respectively. The areas with the geometric mean concentration (μg/m³, 95% CI) of $PM_{2.5}$ were residential area at 50.25 μg/m³ (95%CI 38.97–64.78), followed by industrial zone, university area, and agriculture zone at 44.48 μg/m³ (95%CI 35.06–56.42), 32.78 μg/m³ (95%CI 26.56–40.46), and 29.53 μg/m³ (95%CI 23.62–36.91), respectively, as shown in **Table 2**. A majority of emissions were from automobiles at traffic sights with 32% and burning of biomass with 26% (Chuersuwan et al., 2008). Biomass burning emerged as the primary source of $PM_{2.5}$ mass concentrations at residential sites, for example, an issue of cross-border smog due to sampling periods (Chuersuwan et al., 2008). It is in the dry season and is influenced by the northeast monsoon that blows dust from neighboring areas and neighboring countries and meteorological factors. The mean concentration of $PM_{2.5}$ of all sampling sites that exceeded the WHO (2006), which must not exceed 25 μg/m³. While three of the four sampling sites that mean concentration were below the Pollution Control Department (PCD) of Thailand 50 μg/m³ for 24 h, while $PM_{2.5}$ concentrations in a residential area are above the PCD standard setting of Thailand.

The majority of $PM_{2.5}$ sources in Southeast Asia come from vehicular emissions, industrial pollution, and secondary aerosols as the dominating sources (Singh et al., 2017). Vehicular activities, industrial by-products, and re-suspension of crustal soil are the major factors contributed by anthropogenic activities of the emission of particulate pollutants within the environment (Hazarika and Srivastava, 2017a; Hazarika et al., 2017b; Tian et al., 2010). Additionally, Northeast Thailand is an important region for sugarcane and rice production. Due to insufficient mechanical support, approximately 60% of the crops have

Table 1. The values used for all the parameters in health risk assessment (HRA).

Factors	Notation	Unit	Value				Reference
			Children	Adults			
Ingestion rate	IR_{ing}	mg/day	200	100			(Zhang et al., 2021)
Exposure frequency	EF	days/years	365	365			(Asante-Duah, 2017)
Exposure duration	ED	year	6	24			(Han et al., 2021; Zhang et al., 2021)
Unit conversion factor	CF	kg/mg	1.0×10^{-6}	1.0×10^{-6}			
Inhalation rate	IR_{inh}	m^3/day	5	20			(Du et al., 2013)
Body weight	BW	kg	29	70			(Han et al., 2021; Zhang et al., 2021)
Average lifetime	AT	days	ED×365 (non-carcinogens)	ED×365 (non-carcinogens)			
			70×365 (Carcinogens)	70×365 (Carcinogens)			
Exposure time	ET	h/day	24	24			(Zhang et al., 2021)
Average lifetime(n)	At_n	hours	ED×365×24 (non-carcinogens)	ED×365×24 (non-carcinogens)			
			70×365×24 (Carcinogens)	70×365×24 (Carcinogens)			
Skin surface area	SA	cm^2	2800	5700			(Zhang et al., 2021)
Skin adherence factor	AF	mg/cm^2	0.2	0.07			(Zhang et al., 2021)
Dermal absorption factor	DAF						(Zhang et al., 2021)
	$DAF_{(Pb)}$		0.1	0.1			
	$DAF_{(Cd)}$		0.001	0.001			
	$DAF_{(Cu)}$		0.01	0.01			
	$DAF_{(Mn)}$		0.01	0.01			
	$DAF_{(Zn)}$		0.01	0.01			
	$DAF_{(Al)}$		0.01	0.01			
	$DAF_{(Other\ metals)}$		0.01	0.01			
Parameter	Pb	Cd	Cu	Mn	Zn	Al	
T_r	5		5		1		(Zhang et al., 2021)
RfD_{oral}	0.003500	0.000500	0.004000	0.140000	0.300000	1.000000	(Asante-Duah, 2017; Han et al., 2021; Zhang et al., 2021)
RfC_{inh}	0.003520	0.000010	0.004020	0.000050	0.301000	0.005000	(Asante-Duah, 2017; Han et al., 2021; Zhang et al., 2021)
GI_{ABS}	1		1		1		(Zhang et al., 2021)
SF_{oral}	0.008500	–	–				(Zhang et al., 2021)
Inhalation UR (IUR)	0.000012	0.001800					(Asante-Duah, 2017; Zhang et al., 2021)

Table 2. Detailed of meteorological parameter and mass concentration of $PM_{2.5}$.

Sampling site	Meteorological parameter				Concentration of $PM_{2.5}$
	Aver. Temperature (°C)	Aver. Relative humidity (%)	Aver. Wind speed (m/s)	Aver. Pressure (hPa)	Geometric mean (μg/m^3, 95% CI)
University area	22.40	40	2.02	1008.8	32.78 (26.56–40.46)
Residential area	24.34	50	2.73	1015.4	50.25 (38.97–64.78)
Industrial zone	26.50	90	4.28	1006.1	44.48 (35.06–56.42)
Agricultural zone	25.32	75	3.07	1020.3	29.53 (23.62–36.91)

to be burned before harvesting (Dejchanchaiwong et al., 2020). Yunesian et al. (2019) conducted a study in $PM_{2.5}$ and PM_{10} in the metropolis of Tehran during 2016-2017, which was reportedly exposed to high levels and the associated health risks. Similarly, a study conducted in Chiang Mai, Northern Thailand has reported that the rate of chronic obstructive pulmonary disease increased by 7.2–8.9%, coronary artery disease by 8.6%, and sepsis specific causes of death were increased by 5.7–6.1%, for 10 μg/m^3 increment of $PM_{2.5}$ (Pothirat et al., 2021).

3.2. Heavy metals concentration in $PM_{2.5}$

As shown in **Table 3**, sorted from the areas with the highest concentration of $PM_{2.5}$ found that an industrial zone with heavy metal concentrations in $PM_{2.5}$ in descending proportion are as follows: manganese (Mn) < aluminum (Al) < lead (Pb) < copper (Cu) < cadmium (Cd) < iron (Fe) < Zinc (Zn), residential area (Al < Mn < Pb < Cu < Cd < Fe < Zn), university area (Mn < Al < Pb < Cu < Cd < Fe < Zn), and agricultural area (Mn < Al < Cu < Pb < Fe < (Cd = Zn), as shown in **Table 3**.

Pb, Cd, Mn, and Cu were found to be the most relevant trace metals in the period study and it has been sufficiently recognized that they are traced in traffic emissions (Rodríguez et al., 2020; Zhang et al., 2009); Mn is identified as tracers of dust or soil. The proportion of heavy metals in $PM_{2.5}$ each human activities as shown in **Fig. 2**. Rodríguez et al. (2020) described the origin of the measured metals as practical, from human-caused sources such as traffic emissions, fuel oil combustion, or

Table 3. Geometric mean concentration (GMC) of trace element in $PM_{2.5}$ during December 2020 to February 2021 in Khon Kaen Province.

Sampling site	GMC of trace element (μg/m³, 95% CI)						
	Pb	Cd	Cu	Mn	Zn	Al	Fe
University area	0.0754 (0.0228–0.2498)	0.0111 (0.0043–0.0284)	0.0338 (0.0262–0.0436)	0.2847 (0.2317–0.3500)	0.0012 (0.0004–0.0036)	0.1668 (0.1414–0.1967)	0.0047 (0.0025–0.0085)
Residential area	0.2099 (0.1345–0.3277)	0.0195 (0.0150–0.0253)	0.0573 (0.0220–0.1491)	0.4279 (0.3497–0.5235)	0.0006 (0.0004–0.0011)	0.3381 (0.2107–0.5425)	0.0059 (0.0038–0.0093)
Industrial zone	0.2729 (0.2190–0.3410)	0.1011 (0.0793–0.1290)	0.1857 (0.1674–0.2061)	2.2960 (1.9728–2.6721)	0.0044 (0.0020–0.0095)	1.7482 (1.4448–2.1154)	0.0045 (0.0034–0.0060)
Agricultural zone	0.0239 (0.0089–0.0639)	0.0035 (0.0020–0.0060)	0.1036 (0.0357–0.3009)	0.2477 (0.2149–0.2854)	0.0105 (0.0029–0.0372)	0.1760 (0.1387–0.2234)	0.0040 (0.0022–0.0072)

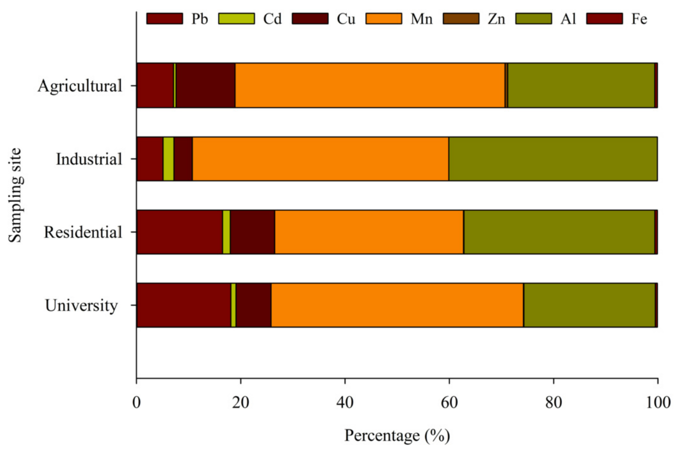

Fig. 2. The proportion of $PM_{2.5}$-bound metal in the human activities' sources during dry season in Khon Kaen Provice, Thailand.

even re-suspended particulate matter and dust storms events (Amarloei et al., 2020). For instance, some major elements connected with $PM_{2.5}$ measured in this study (Pb, Cd, Cu, or Mn), are found to be emissions from debris of wearing of tyres and old's automobile combustion systems (Adachi and Tainosho, 2004). Additionally, Kayee et al. (2020) has showed that an important source of Pb and other trace metals in the Chiang Rai aerosol in Northern Thailand were metals that were released from burning of biomass and the crustal dust formed from raging fire. Based on these comparisons, it may be decided that the $PM_{2.5}$ concentrations of metals primarily depend on the topology, meteorological factors such as wind speed, pressure, and the sampling site, which, consecutively, relies on the effects of sources of metal emissions nearby power plant, traffic transportation, and biomass and waste open burning. This study would help improve our understanding of anthropogenic activities over Khon Kaen and the significance of linking such atmospheric studies with meteorological parameters.

3.3. Environmental risk

Comprehensive anthropogenic metal risk assessments can provide valuable information for risk management at sampling sites, which are less valuable for natural-sourced metals. **Fig. 3** shows a Pb, Cd, Cu, and Mn being identified as human-caused metals with an EF greater than 10 (**Fig. 3a)**, while an *Igeo* greater than 0 was found only with Cd as shown in **Fig. 3(b)** as shown in **Fig. 3**.

A study conducted by Wu et al. (2019) found that augmented factors of heavy metals (Cd and Pb) in $PM_{2.5}$ were mainly from anthropogenic sources. Therefore, the variation of EF source contributions should be influenced by meteorological factors and environmental conditions (Chen et al., 2021). Furthermore, agricultural emissions of Cd and Pb in the ago-ecosystem have been shown to have a source harmful impact and can cause various long-term health issues (Pongpiachan et al., 2017). Thus, the following analysis of the human health risk assessment focuses mainly on the four metals. The findings of this study are based on the comparison of emissions from sources of human activities. However, the most important control measures must be taken into consideration, especially the air quality in which people live in the sampling area or nearby.

3.4. Human health risk assessment of heavy metal's exposure in $PM_{2.5}$

Heavy metals are considered pollutants to humans as well as the environment. Therefore, this study also focuses on two heavy metals (Cd and Pb) has been proven to be hazardous to human health (Briffa et al., 2020; Tchounwou et al., 2012). Considering that Cd is classified as a Group 1 (carcinogenic), while Pb is classified as a Group 2A (probably carcinogenic). The health risk values are overestimated as the health risk assessment was conducted using total heavy metal concentration. The appropriate heavy metal form to be used is the bioavailability form to be applied in HRA (Praveena et al., 2015; Zheng et al., 2020). Hence, these heavy metal data were used for the carcinogenic risk assessments among humans as shown in detail below

3.4.1. Non-carcinogenic risks

The risk to human health in the urban area (residential and university), industrial zone, and agricultural zone of Khon Kaen Province, Northeast Thailand through various ways of exposure merits investigations and surveillance. Furthermore, **Table 4** illustrates the six anthropogenic metals that cause health risks in children and adults. The following order was used: Pb, Cd, Cu, Mn, Zn, and Al, to study contributions in urban areas (university, and residential), and industrial zone and agricultural zone with the total HI value of >1.0 for these four sampling sites which demonstrated the total non-carcinogenic risk exceeded the safe level revealed, the potential health risk is quite high for Cd and Pb that the integrated effects of multi-metal exposure in the urban area (university, and residential), industrial, and agricultural of sampling sites, might result in severe non-carcinogenic risk. The results of this study found that HI was at a level that was at risk for chronic health effects. In line with the findings of Pongpiachan et al. (2017), metal studies in $PM_{2.5}$ in Bangkok from 17 November to 30 April 2011 found that the HI values of Cu, Zn, Cd, and Pb were at safe levels. Corresponding to the study, $PM_{2.5}$-bound heavy metal in the urban area of Kitayushu, Japan reported the total of heavy metals with a corresponding HI of 7.8 and mainly comprising ingestion exposure pathways followed by dermal contact, and inhalation for adults and among children, the route of entry into the body is through ingestion, inhalation, and dermal contact (Zhang et al., 2021). As per this study, HI_{ing}, HI_{inh}, and HI_{der}, indicated that the primary source of exposure was inhalation, followed by ingestion and contact through skin. In addition, researchers discovered that exposure to urban traffic-related air pollution had genotoxic effects on children in Malaysia's Klang Valley (Hisamuddin et al., 2020). Therefore, it is important to put in place a long-term plan to reduce the levels of Cd and Pb in urban dust, by promoting the use of electric vehicles, cycles and encouraging citizens to use public transportation (Pongpiachan et al., 2017), and conducting further research

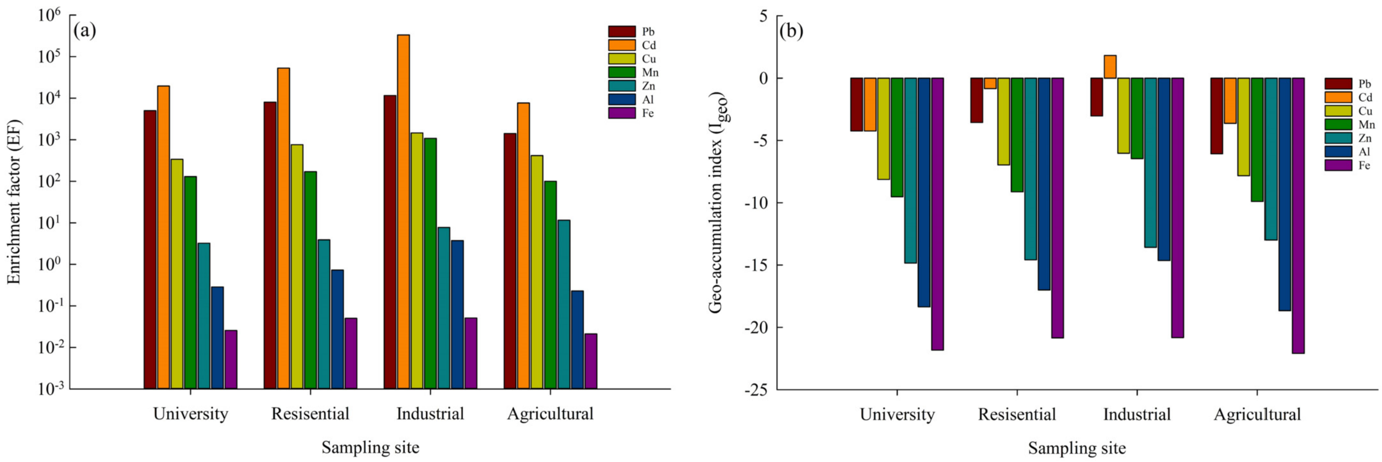

Fig. 3. Enrichment factors (a) and Geo-accumulation indexes (b) of seven metals in $PM_{2.5}$.

Table 4. Summary of estimation human health risk for children and adults due to exposure of heavy metals for non-carcinogenic risk.

Sampling site	Group	Non-carcinogenic risk*						
		CDI_{ing}	EC_{inh}	DAD_{der}	HQ_{ing}	HQ_{inh}	HQ_{der}	HI
University area	Children	3.9×10^{-6}	2.2×10^{-1}	2.4×10^{-7}	0.00	2.61	0.00	2.61
	Adults	8.2×10^{-7}	1.6×10^{-1}	7.1×10^{-8}	0.00	1.95	0.05	2.00
Residential area	Children	8.5×10^{-6}	4.7×10^{-1}	5.7×10^{-7}	0.00	10.73	0.00	10.73
	Adults	1.7×10^{-6}	3.5×10^{-1}	1.7×10^{-7}	0.00	8.02	0.00	8.02
Industrial zone	Children	3.2×10^{-5}	0.17×10^{1}	1.3×10^{-6}	0.00	21.51	0.00	21.51
	Adults	6.6×10^{-6}	0.13×10^{1}	1.6×10^{-6}	0.00	2.92	0.00	2.92
Agricultural zone	Children	2.7×10^{-6}	2.1×10^{-1}	1.1×10^{-6}	0.00	1.90	0.00	1.90
	Adults	8.1×10^{-7}	1.6×10^{-1}	3.2×10^{-7}	0.00	1.53	0.00	1.53

* Total of metal includes Pb, Cd, Cu, Mn, Zn, and Al for exposure and health risk assessment.

on the sources of air pollution and steps to control air pollution, such as open burning and waste incineration facilities.

3.4.2. Carcinogenic risks

In addition, Pb and Cd were carcinogens among two heavy metals. The CR values of Pb among children in university, residential, industrial, and agricultural were 1.00×10^{-8}, 1.71×10^{-8}, 2.22×10^{-6}, and 1.95×10^{-7}, respectively, and Cd where the CR values were 3.34×10^{-2}, 0.23×10^{1}, 0.12×10^{1}, and 4.11×10^{-2}, respectively. In this study, the risk of above the safe level i.e. $\geq 10^{-1}$, revealed a high level of carcinogenic risk for children living in the urban area (residential) and industrial zone while the results that high carcinogenic risk ($10^{-3} \leq CR < 10^{-1}$) (Roy et al., 2019; Zhang et al., 2021) for the urban area (university) and agricultural area. As compared with adults, the CR values of Pb among children in university, residential, industrial, and agricultural were 4.00×10^{-9}, 5.00×10^{-7}, 6.48×10^{-7}, and 5.70×10^{-8}, respectively, and Cd where the CR values were 8.10×10^{-8}, 4.40×10^{-1}, 2.28×10^{-1}, and 7.70×10^{-3}, respectively. However, the risk level in this study above the safe level $10^{-3} \leq CR < 10^{-1}$, revealed a higher risk of carcinogenic for adults living in urban areas (residential and industrial), and agricultural zone exempt in university area the results that very low carcinogenic risk ($CR \leq 10^{-6}$) (Roy et al., 2019; Zhang et al., 2021). In total, the combined carcinogenic risk for children and adults' (university, residential, industrial, and agricultural) was also above the acceptable limits, it was found that the Cd of carcinogenic risk obtained was high than the ceiling value of 10^{-6}, which is significant based on the USEPA standards; it becomes evident to conclude that Cd increases the lifetime cancer risk; except for adults living in university areas, the levels were very low for carcinogenic risk. In addition, they comprise the majority of exposure dose assessments among children and adults were CR_{ing}, CR_{der}, and CR_{ing} (**Table 5**). Based on the results of the health risk assessment, it is evident that in the urban area (such as university, residential), industrial zone, and rural agricultural of Khon Kaen Province, Thailand, exposure to $PM_{2.5}$ could significantly influence the environmental risks. Furthermore, children's health is at higher non-carcinogenic and carcinogenic risk than adults. Additionally, Pb, Cd, Cu, Mn, Zn, and Al were found to be in abundance. As a result, when $PM_{2.5}$-bound heavy metals are introduced into the body, particularly through the respiratory system, they encourage and induce health problems.

4. Conclusions

In this study ambient air was collected between December 2020 and February 2021 (dry season) at a sampling site in the urban area (university and residential), industrial zone, and agricultural zone of Khon Kaen Province, Northeast Thailand. The heavy metals loaded in $PM_{2.5}$ of 70% found the highest concentration in industrial areas, followed by residential, university, and agricultural areas at 15%, 9%, and 6%, respectively. In all the study areas, the EF values of heavy metals influenced by human activities in $PM_{2.5}$ were Cd, Pb, Cu, Mn, Zn, Al, and Fe, respectively, while only one I_{geo} of Cd was found in industrial zones (biomass or waste incinerated plant) of four human activity sites at the level of uncontaminated to moderately contaminated. The results of this study found that only six heavy metals in $PM_{2.5}$ showed severe non-carcinogenic risk; giving the probability of chronic effects among the children and adults. The contributions of the studied metals are in the following order: Pb, Cd, Cu, Mn, Zn, and Al in the university area, residential area and industrial zone, agricultural zone with the total HI value of >1.0. In addition, exposure to a total of heavy metals could cause high to very high carcinogenic risk for children among residential areas, and industrial zones with TCR values of 0.23×10^{1}, and 0.12×10^{1}, respectively, which resulted in a high carcinogenic risk in the 3.34×10^{-2}, and 4.11×10^{-2} of the university and agricultural zone.

Table 5. Summary of estimation human health risk for children and adults due to exposure of heavy metals for carcinogenic risk.

Sampling site	Group	Metal	Carcinogenic risk						
			CDI_{ing}	EC_{inh}	DAD_{der}	CR_{ing}	CR_{inh}	CR_{der}	CR
University area	Children	Pb	7.43×10^{-9}	4.10×10^{-4}	2.08×10^{-9}	0.00	5.00×10^{-9}	2.00×10^{-9}	1.00×10^{-8}
		Cd	1.09×10^{-9}	6.02×10^{-5}	3.05×10^{-12}		3.34×10^{-2}		3.34×10^{-2}
		Cu	3.33×10^{-9}	1.84×10^{-4}	9.33×10^{-11}				
		Mn	2.80×10^{-8}	1.55×10^{-3}	7.85×10^{-10}				
		Zn	8.62×10^{-11}	4.76×10^{-6}	2.41×10^{-12}				
		Al	1.64×10^{-8}	9.07×10^{-4}	4.60×10^{-10}				
		SUM	1.00×10^{-7}	3.11×10^{-3}	3.42×10^{-9}	0.00	3.34×10^{-2}	2.00×10^{-9}	$\mathbf{3.34 \times 10^{-2}}$
	Adults	Pb	1.54×10^{-9}	3.06×10^{-4}	6.14×10^{-10}	0.00	4.00×10^{-9}	1.00×10^{-9}	4.00×10^{-9}
		Cd	2.26×10^{-10}	4.50×10^{-5}	9.01×10^{-13}		8.10×10^{-8}		8.10×10^{-8}
		Cu	6.90×10^{-10}	1.37×10^{-4}	2.75×10^{-11}				
		Mn	5.81×10^{-9}	1.16×10^{-3}	2.32×10^{-10}				
		Zn	1.78×10^{-11}	3.56×10^{-6}	7.12×10^{-13}				
		Al	3.40×10^{-9}	6.78×10^{-4}	1.36×10^{-10}				
		SUM	1.17×10^{-8}	2.32×10^{-3}	1.01×10^{-9}	0.00	8.50×10^{-8}	1.00×10^{-9}	$\mathbf{8.5 \times 10^{-8}}$
Residential area	Children	Pb	9.99×10^{-8}	4.57×10^{-3}	5.79×10^{-9}	1.00×10^{-9}	5.50×10^{-8}	1.65×10^{-6}	1.71×10^{-8}
		Cd	9.27×10^{-8}	4.23×10^{-3}	5.37×10^{-11}		$\mathbf{0.23} \times 10^{1}$		$\mathbf{0.23} \times 10^{1}$
		Cu	2.73×10^{-8}	1.25×10^{-3}	1.58×10^{-10}				
		Mn	2.04×10^{-7}	9.31×10^{-3}	1.18×10^{-9}				
		Zn	3.02×10^{-10}	1.38×10^{-5}	1.75×10^{-12}				
		Al	1.61×10^{-7}	7.35×10^{-3}	9.32×10^{-10}				
		SUM	5.85×10^{-7}	2.67×10^{-2}	8.12×10^{-9}	1.00×10^{-9}	$\mathbf{0.23} \times 10^{1}$	1.65×10^{-6}	$\mathbf{0.23} \times 10^{1}$
	Adults	Pb	1.71×10^{-8}	8.53×10^{-4}	1.71×10^{-9}	0.00	1.00×10^{-8}	4.88×10^{-7}	5.00×10^{-7}
		Cd	1.59×10^{-8}	7.92×10^{-4}	1.59×10^{-11}		4.40×10^{-1}		4.40×10^{-1}
		Cu	4.68×10^{-9}	2.33×10^{-4}	4.67×10^{-11}				
		Mn	3.50×10^{-8}	1.74×10^{-3}	3.48×10^{-10}				
		Zn	5.17×10^{-11}	2.57×10^{-6}	5.16×10^{-13}				
		Al	2.76×10^{-8}	1.37×10^{-3}	2.75×10^{-10}				
		SUM	1.00×10^{-7}	5.00×10^{-3}	2.40×10^{-9}	0.00	4.40×10^{-1}	4.88×10^{-7}	$\mathbf{4.40 \times 10^{-1}}$
Industrial zone	Children	Pb	2.69×10^{-8}	5.94×10^{-3}	7.53×10^{-9}	0.00	7.10×10^{-8}	2.15×10^{-6}	2.22×10^{-6}
		Cd	9.96×10^{-9}	2.20×10^{-3}	2.79×10^{-11}		$\mathbf{0.12} \times 10^{1}$		$\mathbf{0.12} \times 10^{1}$
		Cu	1.83×10^{-8}	4.04×10^{-3}	5.12×10^{-11}				
		Mn	2.26×10^{-7}	5.00×10^{-2}	6.33×10^{-9}				
		Zn	4.32×10^{-10}	9.55×10^{-5}	1.21×10^{-11}				
		Al	1.72×10^{-7}	3.80×10^{-2}	4.82×10^{-9}				
		SUM	4.54×10^{-7}	1.00×10^{-1}	1.88×10^{-8}	0.00	$\mathbf{0.12} \times 10^{1}$	2.15×10^{-6}	$\mathbf{0.12} \times 10^{1}$
	Adults	Pb	2.23×10^{-8}	1.11×10^{-3}	2.22×10^{-9}	0.00	1.30×10^{-8}	6.35×10^{-7}	6.48×10^{-7}
		Cd	8.25×10^{-9}	4.11×10^{-3}	8.23×10^{-12}		2.28×10^{-1}		2.28×10^{-1}
		Cu	1.52×10^{-8}	7.55×10^{-4}	1.51×10^{-10}				
		Mn	1.87×10^{-7}	9.33×10^{-3}	1.87×10^{-9}				
		Zn	3.58×10^{-10}	1.78×10^{-5}	3.57×10^{-12}				
		Al	1.43×10^{-7}	7.11×10^{-3}	1.42×10^{-9}				
		SUM	3.76×10^{-7}	1.87×10^{-2}	5.68×10^{-9}	0.00	2.28×10^{-1}	6.35×10^{-7}	$\mathbf{2.28 \times 10^{-1}}$
Agricultural zone	Children	Pb	2.35×10^{-9}	5.20×10^{-4}	6.59×10^{-10}	0.00	6.00×10^{-9}	1.88×10^{-7}	1.95×10^{-7}
		Cd	3.36×10^{-10}	7.41×10^{-5}	9.40×10^{-13}		4.11×10^{-2}		4.11×10^{-2}
		Cu	1.02×10^{-8}	2.25×10^{-3}	2.86×10^{-10}				
		Mn	2.44×10^{-8}	5.39×10^{-3}	6.83×10^{-10}				
		Zn	1.03×10^{-9}	2.28×10^{-4}	2.89×10^{-11}				
		Al	1.73×10^{-8}	3.83×10^{-3}	4.86×10^{-10}				
		SUM	5.57×10^{-8}	1.23×10^{-2}	2.14×10^{-9}	0.00	4.11×10^{-2}	1.88×10^{-7}	$\mathbf{4.11 \times 10^{-2}}$
	Adults	Pb	1.95×10^{-9}	9.71×10^{-5}	1.94×10^{-10}	0.00	1.00×10^{-9}	5.60×10^{-8}	5.70×10^{-8}
		Cd	2.78×10^{-10}	1.38×10^{-5}	2.77×10^{-11}		7.70×10^{-3}		7.70×10^{-3}
		Cu	8.46×10^{-9}	4.21×10^{-4}	8.43×10^{-10}				
		Mn	2.02×10^{-8}	1.01×10^{-3}	2.02×10^{-9}				
		Zn	8.54×10^{-10}	4.25×10^{-5}	8.52×10^{-11}				
		Al	1.44×10^{-8}	7.16×10^{-4}	1.43×10^{-9}				
		SUM	4.61×10^{-8}	2.30×10^{-3}	4.60×10^{-9}	0.00	7.70×10^{-3}	5.60×10^{-8}	$\mathbf{7.70 \times 10^{-3}}$

Adults were found to be exposed to risks that were beyond the safe level $10^{-3} \leq CR < 10^{-1}$, showing a high carcinogenic risk for adults living in the urban area (residential) and industrial zone, and agricultural zone exempt in university area the results that very low carcinogenic risk ($CR \leq 10^{-8}$). The most effective strategy to reduce the exposure risk to the $PM_{2.5}$ seems to be through the inhalation route in children around and self-protection (considerations mask-wearing outdoors), especially in urban areas, industrial zones, and biomass and waste open burning. Further investigation of appropriate process and long-term monitoring for source identification is much needed.

Declarations

Author contribution statement

Pornpun Sakunkoo: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper. **Theerachai Thonglua:** Performed the experiments; Analyzed and interpreted the data. **Sarawut Sangkham:** Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper. **Chananya Jirapornkul, Yuparat Limmongkon, Jetnapis Rayubkul, Sakesun Thongtip:** Contributed reagents, materials, analysis tools or data. **Naowarat Maneenin:** Contributed reagents, materials, analysis tools or data; Wrote the paper. **Sakda Daduang, Thanee Tessiri:** Performed the experiments; Contributed reagents, materials, analysis tools or data. **Sittichai Pimonsree:** Analyzed and interpreted the data; Wrote the paper.

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Data availability statement

The authors do not have permission to share data.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

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