



Chemical characterization, source apportionment, and human health risk assessment of PM_{2.5} in an urban area in Hanoi, Vietnam

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Abstract

Forty-seven ambient 24-h PM_{2.5} samples were collected during summer and fall at a typical inner-city sampling site in Hanoi, a metropolitan city in Vietnam for characterization, inorganic chemical fractionation, source apportionment, and potential health risk assessment of airborne PM_{2.5}. The average 24-h PM_{2.5} concentrations observed in this study were substantially higher than the World Health Organization (WHO) guidelines. The most abundant elements in PM_{2.5} were Al, Fe, Pb, and Zn accounting for 85.14% and 89.06% of the total obtained elements in summer and fall, respectively. Noticeably, the levels of Cd were 40.33 times in summer and 26.64 times in fall higher than WHO guidelines. The obtained results from principal component analysis (PCA) combined with enrichment factor (EF) analysis showed good agreement and suggested that the major sources of PM_{2.5} were non-exhaust traffic emission and crustal dust, exhaust traffic emission, coal combustion, and industrial source. Both the hazard index (HI) and carcinogenic risk (CR) values of all investigated elements were lower than the safe levels for adults and children in the two seasons. However, the carcinogenic risks of Cr for both adults and children were close to the acceptable values, implying potential carcinogenic risks caused by this metal. Besides, both the non-carcinogenic and carcinogenic risks for children were substantially higher than that for adults, suggesting more attention on children's health adverse effects resulting from exposure to trace metals bounded with PM_{2.5} in Hanoi should be paid.

Keywords Heavy metal · Health risk assessment · PM_{2.5} · Principal component analysis · Hanoi

Introduction

In Vietnam, elevated air pollution due to population growth, rapid urbanization, industrialization, and a large number of vehicles has been an increasingly concerning issue in recent years (Tran 2017; Nguyen et al. 2017; Bui et al. 2019). Some

recent studies conducted in Vietnam have shown that particulate matter (PM) (including PM₁₀ with an aerodynamic diameter ranging from 2.5 to 10 μm, and PM_{2.5} with an aerodynamic diameter ≤ 2.5 μm) is a common air pollutant in most urban areas, especially in the large cities such as Ho Chi Minh and Hanoi (Oanh et al. 2009; Ly et al. 2018; Hien et al. 2019). The environment report of the Vietnam Ministry of Natural Resources and Environment (MONRE) (2019) has pointed out that the concentrations of PM_{2.5} in urban areas are higher than the Vietnam national ambient air quality standard (NAAQS, 50 μg/m³ for 24-h mean) in 2019. Fine particulate matter exhibited diverse anthropogenic sources such as traffic-related emissions and industrial processes with large variations over different regions (Manoli et al. 2002; Yongyong et al. 2018). Therefore, it is necessary to apply the source apportionment techniques for source identification. Several receptor models have been developed and widely used to determine the possible sources of elements bounded with PM. The methodology of the source apportionment techniques is different, which

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requires different levels of understanding about the source types and the emission profile of the sources (Contini et al. 2010). Although chemical mass balance (CMB) has been one of the most popular receptor models, the major difficulty in its applications is the requirement of the local source profiles which are identified by experiments (Ogundele et al. 2016). On the other hand, several receptor models which applied factor analysis approaches such as principal component analysis (PCA) and positive matrix factorization (PMF) have been widely applied for source apportionment of PM. The advantage of these receptor models is the requirement of general qualitative knowledge of source emissions instead of extensive information about the source profiles (Ogundele et al. 2016).

Air pollution not only causes economic damage but also affects human health (Staif et al. 2013). The estimated economic loss due to respiratory illness in Hanoi and Ho Chi Minh City was 66.83 and 70.96 million USD per year, respectively (Ministry of Natural Resources and Environment 2013). Data from the Ministry of Health have shown that in recent years, the incidence of respiratory diseases was the highest nationwide and air pollution was one of the main causes of the increase in incidence (Ministry of Health 2012). Research by Lâm et al. (2018) showed that the incidence of allergic rhinitis in adults and asthma in people living in urban areas in Hanoi (30%) was higher than that in rural areas (10%). According to the environment report of the Vietnam Ministry of Natural Resources Environment (MONRE) (2014), the incidence of respiratory disease in children was higher than that of adults in Hanoi.

In the air, the composition of $PM_{2.5}$, which easily penetrates the human respiratory system, is toxins that affect human health. Many studies have demonstrated that PM exposure is associated with cognitive impairment, neuronal stress (oxidation stress), and neurodegeneration (Calderón-Garcidueñas et al. 2013). Some organic pollutants related to $PM_{2.5}$ can cause mutations (Sancini et al. 2014) and cancers (Harrison 2004; Pope et al. 2011; Li et al. 2016), cardiovascular diseases (Xu et al. 2016), neuropathy (Bellinger 2005), respiratory problems such as cough, asthma, and bronchitis (Cobbina et al. 2015) through various pathways: inhalation, ingestion, and digestion. In the past decades, many studies have stressed the characterization of metal and inorganic or organic components in $PM_{2.5}$ at urban sites. Research on the impact of highway traffic exhaust on human health in Switzerland figured out that adults living near highways were at greater risk of respiratory illnesses than adults living in urban areas (Hazenkamp-von Arx et al. 2011). Jeong et al. (2019) reported that traffic-related $PM_{2.5}$ sources were mainly exhausted emissions (9–19% of $PM_{2.5}$) and non-exhaust emissions including brake wear (2–6%) and resuspension of road dust (3–4%). During morning rush hours, traffic-exhausted and non-exhaust emissions contributed to 35–48% of total $PM_{2.5}$ mass. Although the contribution of brake wear and road dust sources to total $PM_{2.5}$ mass

was relatively low, non-exhaust emissions contributed a substantial fraction of trace elements, especially for Ba (74–79%), Cu (66–71%), and Mn (53–65%) in the urban atmosphere (Jeong et al. 2019). These results showed that most trace elements exhibited the highest levels in urban areas and the lowest in rural areas, and industrial emissions, coal emissions, and traffic emissions were the main sources of these elements.

Earlier studies (Hien et al. 2004; Gatari et al. 2005; Michael et al. 2006; Gatari et al. 2006; Oanh et al. 2009; Cohen et al. 2010) have reported trace elements associated with $PM_{2.5}$ and source apportionment for $PM_{2.5}$ in some locations in the inner of Hanoi. These researches were carried out at least 12 years ago. Besides, they did not investigate the health risk assessments imposed by trace elements bounded in $PM_{2.5}$. Hanoi has experienced a fast development in the recent decade through dramatic economic growth as well as high-speed urbanization with an urban expansion rate of 2.9% (Mauro 2020), which would result in obvious changes in the chemical characteristics of fine particles and their original sources. According to statistics of the MONRE, air pollution in Hanoi has significantly increased in 2019 which was unusual compared to previous years. Particularly in 2019, Hanoi has experienced at least five periods of serious air pollution (January 16–26; March 11–27; September 12–October 3; November 5–12; and December 7–16) which might have adverse health impacts for local people. Therefore, the objectives of our study are (1) to provide an up-to-date investigation on the characterization of the trace metals bounded with $PM_{2.5}$ as well as the pollution level of $PM_{2.5}$ in Hanoi; (2) to perform the source apportionment analysis to identify the major sources acting on a typical urban site in the inner city of Hanoi; and (3) to assess human health impacts due to exposure to trace elements associated with $PM_{2.5}$. We selected the National University of Civil Engineering (NUCE) as a sampling site because it has the typical characteristics of the inner-city area of Hanoi with a high population and dense key road networks with high traffic density. Besides, the NUCE site is also affected by the industry activities of several medium and large industrial zones located within a radius of smaller than 16 km. The obtained results would provide comprehensive knowledge to support local government agencies for planning the strategy to mitigate the serious air pollution and public health impact in the circumstances of fast development of Hanoi results in serious air pollution.

Materials and methods

Study area and sampling site

The sampling site in this study was located on the roof of the 2nd-floor building in the National University of Civil

Engineering (NUCE), Hanoi—the second-largest metropolitan city in Vietnam (Fig. 1). Hanoi is a populous city with a total population of 8.1 million people (General Statistic Office of Vietnam 2019), more than 600,000 automobiles, and 6,000,000 motorbikes (TDSI 2017). Hanoi City covers an area of about 3328 km² and features a warm humid subtropical climate influenced by the northeast monsoon in winter and the southeast monsoon in summer with four distinct seasons including spring (from March to May), summer (from June to August), fall (from September to November), and winter (from December to February). NUCE is situated in one of the four inner-city districts (Hai Ba Trung with a population density of 30,000 inhabitants per km²) which are considered to be the heart of the metropolitan area and characterized by a high population and traffic density with dense road networks. NUCE is located on the side of Giai Phong road, which is the key traffic route in the south area of Hanoi. Several industrial zones surrounding the sampling site including the North Thang Long industrial zone (NTL-IZ) are located in the northwest of the NUCE with a distance of about 15.6 km; the Sai Dong industrial zone (SD-IZ) is located in the northeast of NUCE with a distance of 6.5 km, and Vinh Hung industrial zone (VH-IZ) and Ngoc Hoi industrial zone (NH-IZ) are situated in the south and southeast direction of the NUCE site, respectively, with distances less than 10 km.

PM_{2.5} sampling

The 24-h integrated PM_{2.5} sampling program was conducted daily in the summer season (July), and fall season (October and November) in 2020. A total of forty-seven 24-h PM_{2.5} samples (eighteen samples collected in summer and twenty-nine samples collected in fall) were collected on quartz fiber filters (Whatman, QM-H pure quartz, size 47 mm, USA) using a low-volume sampler METONE, E-FRM-200 (USA), operated at 16.7 L/min. The quartz fiber filters were weighed twice before and after sampling. Before weighing, filters were equilibrated for 24 h in a desiccator at 25 ± 5 °C and relative humidity of 50 ± 5%. Before sampling, quartz fiber filters were preheated in an electric furnace at 900 °C for 3 h to remove carbonaceous contaminants. After collection, these samples were sealed in an aluminum foil and kept in a clean plastic bag, then transported to the laboratory and stored in a desiccator with silica gel particles. The collected quartz fiber samples were stored in a refrigerator at about 4 °C before chemical analysis.

Meteorological data

In this study, we utilized the fifth-generation atmospheric reanalysis products of the European Centre for Medium-Range

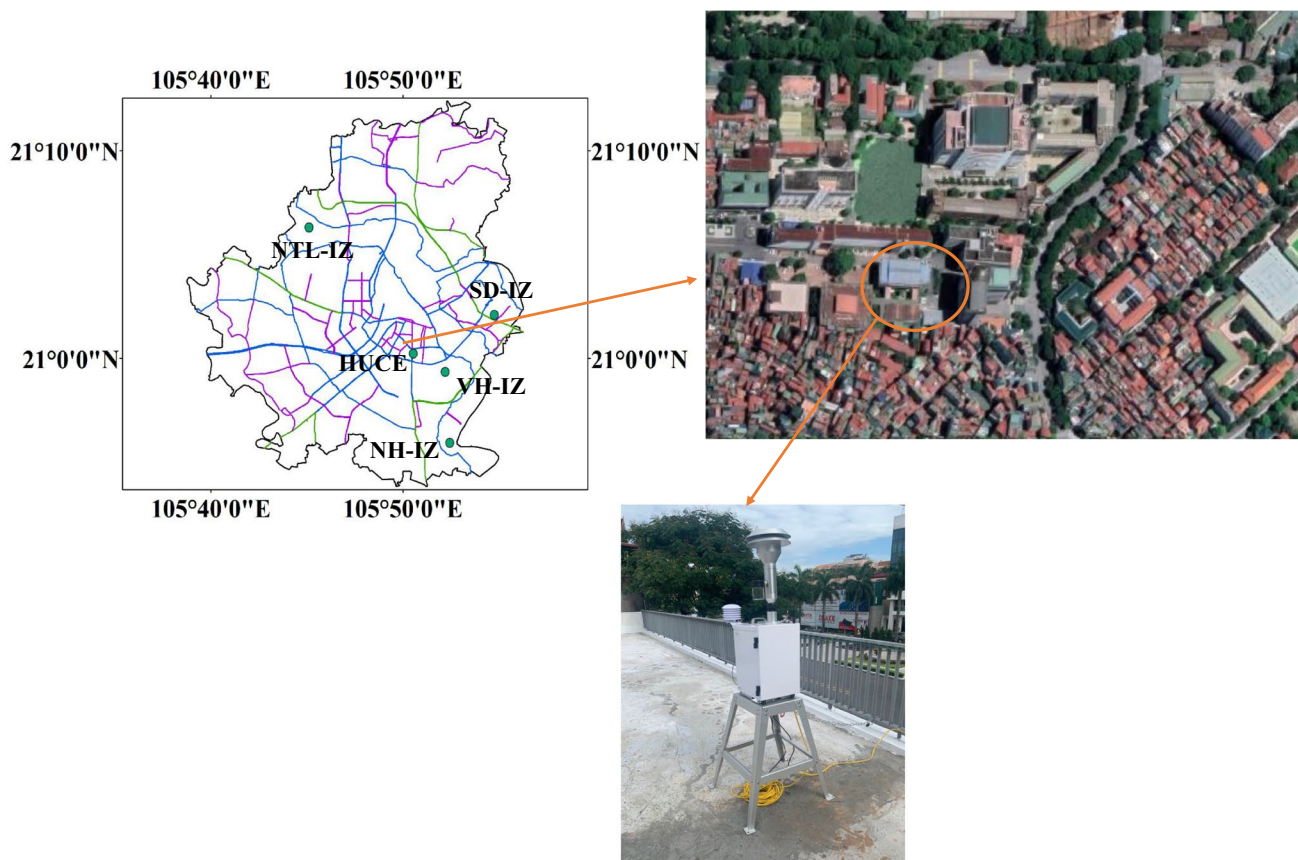


Fig. 1 PM_{2.5} sampling sites for summer and fall periods in Hanoi, Vietnam

Weather Forecasts (ECMWF) namely ERA5 to derive hourly wind speed and wind direction, dew-point temperature, and solar radiation at the sampling site. These ERA5 data were downloaded from the data climate store (<https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels?tab=overview>). The relative humidity was computed using the dew-point temperature, and temperature data based on the method described in (Alduchov and Eskridge 1996). The wind data were used to derive the pollutant rose which supported the PCA analysis to determine the pollution sources.

Analytical methods

PM_{2.5} mass concentration analysis

The PM_{2.5} total mass of each sample was determined gravimetrically by subtracting the initial sample filter weight from the final sampled filter weight. In this study, the glass fiber filter samples were repeatedly weighed by an analytical balance (Adam AEA-160DG, sensitivity ± 0.01 mg) until three reproducible values were obtained.

Chemical analysis

A quarter of the sample filter was treated for analysis of heavy metals by digestion method according to EPA method IO-3.1. The sample filter firstly was cut into pieces, and then digested in 5 mL of mixed acid solution (HNO₃:HCl with a ratio of 1:3) and kept on a hot plate at high temperature until a transparent solution was boiled. After complete digestion, the digested sample was heated at a low temperature until nearly dry to remove excess acid. Then, a solution was diluted in a 25-mL volumetric flask with distilled water. Samples were analyzed using an inductively coupled plasma mass spectrometer (ICP-MS, ELAN 900, PerkinElmer, USA) for 18 elements including aluminum (Al), titanium (Ti), vanadium (V), chromium (Cr), manganese (Mn), iron (Fe), Coban (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), selenium (Se), strontium (Sr), molybdenum (Mo), cadmium (Cd), antimony (Sb), barium (Ba), and lead (Pb). In the analysis, a blank sample, a duplicated sample, and a spiked sample were measured for quality control. The relative standard deviation of each element was within 10% and the analytical errors were < 10%. The detection limit for all the trace elements was 0.01 ng/m³ except for Cd (0.002 ng/m³).

Source apportionment analysis

Enrichment factor

Many works of research have used the enrichment factor (EF) analysis to specify the natural or anthropogenic sources of

metals bounded in PM_{2.5} (Bilos et al. 2001; Song et al. 2016; Cheng et al. 2018; Yongyong et al. 2018). The EF of each element was calculated according to the equation below:

$$EF_X = \left(\frac{X}{R} \right)_{PM_{2.5}} / \left(\frac{X}{R} \right)_{crust}$$

where $(X/R)_{PM_{2.5}}$ and $(X/R)_{crust}$ are the concentration of the X element and reference element R in PM_{2.5} and crust, respectively. The concentration of elements in the crust refers to their concentrations in the Earth's crust proposed by Taylor (1964). Ti, Si, Al, and Fe were generally used as reference elements of crustal materials (Kim et al. 2002; Cesari et al. 2012; Song et al. 2016). In this study, the element Ti was selected as the reference element because Ti had a small concentration range in this study. If the EF value is less than 10, it is indicated that the element is derived mainly from natural sources. The EF value between 10 and 30 suggests that the influences of anthropogenic sources on the element are small. The value of EF between 30 and 100 is considered a result of both natural and anthropogenic sources, and an EF value greater than 100 indicates that the trace element originated from anthropogenic emissions (Cesari et al. 2012; Song et al. 2016).

Principal component analysis

This study applied the principal component analysis (PCA) described in Thurston and Spengler (1985) to investigate the source contributions of PM_{2.5}. PCA is one of the most effective statistical analytical tools that minimize the dimension of original variables, and reduce the principal components for investigating the connection between samples and observed variables. Among various techniques for source apportionment, principal component analysis (PCA) based upon mass balance analysis with mass conservation has been reported to be a common and powerful tool to identify the potential sources of ambient particulate matter (Rodriguez-Espinosa et al. 2017; Ghosh et al. 2018)). Many studies worldwide applied PCA to determine possible sources of particulate matter-bound trace elements (Arruti et al. 2010; Sarti et al. 2015; Zhang et al. 2016; Bui et al. 2020; Nguyen et al. 2021; Xu et al. 2021). The SPSS 20.0 was utilized to perform PCA with the statistic significant at $p < 0.05$.

The Varimax rotation which is popular for atmospheric analysis (Thurston and Spengler 1985; Karar and Gupta 2007) has been applied to the matrix of loads. All components with eigenvalues less than 1 (before rotation) were removed in the successive analysis.

Health risk assessment

Risk assessment regarding exposure to elements bounded in PM_{2.5} was estimated to identify cancer or non-cancer risk

based on the method described in US EPA (2009). In this study, we performed the health risk assessment by exposure to contamination in PM_{2.5} through three pathways including ingestion, inhalation, and dermal contact.

According to the US EPA (1997) and US EPA (2009), the intake doses through ingestion (ADD_{ing}) (μg/kg/day), average daily intake via inhalation (EC_{inh}) (μg/kg/day), and dermal contact (DAD_{der}) (μg/kg/day) were computed using the following equations:

$$ADD_{ing} = \frac{C \times IR_{ing} \times EF \times ED \times CF}{BW \times AT}$$

$$EC_{inh} = \frac{C \times IR_{inh} \times EF \times ED}{PEF \times BW \times AT_n}$$

$$DAD_{der} = \frac{C \times SA \times AF \times ABS \times EF \times ED \times CF}{BW \times AT}$$

In these equations, *C* is the concentration of the metal in PM_{2.5} (μg/m³); IR_{ing} is the ingestion rate (100 mg/day for adults and 200 mg/day for children) (US EPA 1997; Yekken et al. 2016); EF is the exposure frequency (365 days/year); ED is the exposure duration (6 years for children and 24 years for adults) (Zhang et al. 2018) (US EPA 2001); IR_{inh} is the inhalation rate (20 m³/day for both adults and children) (US EPA 1997); PEF is the particle emission factor (1.36 × 10⁹ m³/kg for all cases) (US EPA 2001); SA is the exposed skin surface area (2800 cm² for a child and 5700 cm² for an adult) (US EPA 2001); AF is the adherence factor (0.2 mg/cm² for a child and 0.07 mg/cm² for an adult) (US EPA 2011; Cheng et al. 2018; Ghosh et al. 2018); ABS is the absorption factor (ABS = 0.001 for all trace metal except for As, ABS = 0.03 for As) (USEPA 2004; Zhang et al. 2018); BW is the average bodyweight (55 kg for Vietnamese adults and 15 kg for Vietnamese children); AT is the average time in days (for non-carcinogens, AT = ED × 365 days; for carcinogens, AT = 70 years × 365 days); AT_n is the average time in days (for non-carcinogens, AT = ED × 365 days; for carcinogens, AT = 70 years × 365 days); and CF is the conversion factor (10⁻⁶ kg/mg).

In the present study, the upper limit of the 95% confidence interval for the mean (95% UCL) was used to enhance the accuracy and rationality of the evaluation (Zhang et al. 2018). The 95% UCL is considered the reasonable maximum exposure (Kurt-Karakus 2012). The 95% UCL of the concentration of heavy metals was estimated using the following formula:

$$C_{UCL} = \bar{X} + \left[z_{\alpha} + \frac{\beta}{6\sqrt{n}} x(1 + 2xZ_{\alpha}^2) \right] x \frac{STD}{\sqrt{n}}$$

In this formula, \bar{X} is the arithmetic mean, STD stands for the standard deviation, and β is the skewness; α represents the probability of making type I error. Z_{α} presents the (1- α)th

quantile of the standard normal distribution, and *n* is the number of samples.

Based on the classification group orders defined by the International Agency for Research on Cancer (IARC), cadmium and cadmium compounds, arsenic and inorganic arsenic compounds, nickel compounds, and chromium (VI) are the group I carcinogenic elements; lead compounds (inorganic) are group 2A; vanadium pentoxide, cobalt, and cobalt compounds are group 2B (IARC 2018); Cu, Zn, and Mn are not found in the classification group orders (IARC 2018). Therefore, in this study, Mn, Zn, V, and Cu were determined as non-carcinogenic elements, and As, Cr, Ni, Cd, Co, and Pb were identified as both non-carcinogenic and carcinogenic elements (Hu et al. 2012; Zhang et al. 2018).

The hazard quotient (HQ) was used to evaluate the non-carcinogenic risk and the carcinogenic risk (CR) impact of individual elements in PM_{2.5}. The hazard index (HI) is the sum of HQ for various substances or various exposure pathways (US EPA 2009; Zhang et al. 2018). If HI < 1, indicating adverse health effects is ignored. If HI > 1, suggesting the risk of cancer possibly occurs.

$$HQ_{ing} = \frac{ADD_{ing}}{RfD_o}$$

$$HQ_{inh} = \frac{EC_{inh}}{RfC_i \times 1000 \mu\text{g}/\text{mg}}$$

$$HQ_{der} = \frac{DAD_{der}}{RfD_{der}}$$

$$HI = \sum HQ_i$$

where RfD_o is the oral reference dose (mg/kg·day); RfC_i is inhalation reference concentrations (mg/m³); RfD_{der} is the dermal reference dose (mg/kg·day) (see Table S1).

Total cancer risk (TR) refers to the sum of CR for numerous pathways, which was calculated as the equations below (US EPA 1997, 2001; USEPA 2004; Hu et al. 2012). If CR is > 10⁻⁴, the potential risk of cancer exists seriously. If CR is between 10⁻⁶ and 10⁻⁴, the risk can be as acceptable or tolerable, and if CR is < 10⁻⁶, there can be less risk.

$$CR_{ing} = ADD_{ing} \times SF_o$$

$$CR_{inh} = EC_i \times IUR$$

$$CR_{der} = DAD_{der} \times \frac{SF_o}{ABS_{GI}}$$

$$TR = \sum CR_i$$

where SF_o is the oral slope factor ((mg/kg·day)⁻¹); IUR is the inhalation unit risk ((μg/m³)⁻¹); ABS_{GI} is the gastrointestinal absorption factor. The SF, RfD, ABS_{GI}, and IUR for selected elements were downloaded from regional screening levels (US EPA 2012) (see Table S2). The toxicity value of Cr(VI) which was much higher than Cr(III) was used to point out the worst effect of Cr. The SF and RfD values of

Cr(VI) were assumed for total Cr (Huang et al. 2014) and were used to calculate the HQ and CR for total Cr.

Results and discussion

The characteristics of heavy metals in PM_{2.5}

As shown in Supplementary Data, Fig. S1, the 24-h concentrations of PM_{2.5} were in the range from 1.720 to 82.459 µg/m³ in summer and from 4.549 to 90.681 µg/m³ in fall, respectively. The average PM_{2.5} concentrations observed in Hanoi in summer (41.028 ± 29.241 µg/m³) were higher than that measured in fall (33.643 ± 23.011 µg/m³), which would be partially attributed to the formation of secondary aerosol under the weather condition of high temperature, high RH, and intensive solar radiation (Khoder 2002; Xiu et al. 2004; Sun et al. 2006). The mean ambient temperature and solar radiation during the sampling periods in summer and fall were estimated using the ERA5 dataset (Table 1). The results showed that although there was a small difference in RH between the two seasons, the average ambient air temperature and solar radiation were much higher in summer. Thus, this weather condition could favor the secondary aerosol formation in summer, partially resulting higher concentration of PM_{2.5}. Although the PM_{2.5} concentration in both summer and fall was lower than the NAAQS (50 µg/m³ for 24-h mean), it was 2.735 times and 2.243 times higher than WHO (15 µg/m³ for 24-h mean), respectively. In addition, the percentage of sampling days that the PM_{2.5} concentration exceeded the NAAQS was still high (50% in summer and 27.586% in fall). The average PM_{2.5} concentrations in Hanoi were higher than those in Singapore, Singapore (13.02 ± 2.73 µg/m³) (Zhang et al. 2017); in Cheju Island, Korea (18 ± 13 µg/m³); in Sado island, Japan (8.9 ± 6 µg/m³) (Cohen et al. 2013); in Iasi, Romania (16.92 ± 9.07 µg/m³) (Galon-Negru et al. 2019); in Bologna (19.68 ± 0.78 µg/m³) (Sarti et al. 2015); Shangri-La, China (13.32 ± 8.23 µg/m³) (Yin et al. 2021). However, the average PM_{2.5} concentrations observed in this study in Hanoi were lower than those in Beijing, China (58.1 ± 2.2 µg/m³); in Kanpur, India (102.8 ± 20.2 µg/m³) (McNeill et al. 2020); in Shangdong, China (134 µg/m³) (Zhang et al. 2018); in Baoding City, China (155.66 µg/m³) (Liu et al. 2018b); and at two preschools in Hanoi (107–177 µg/m³) (Tran et al. 2020). Besides, the average PM_{2.5} concentrations in this study were

close to the values reported in Hong Kong (31 ± 22 µg/m³) (Cohen et al. 2013); in Tehran, Iran (41.19 µg/m³) (Kermani et al. 2016); in Mangshi (29.87 ± 20.83 µg/m³) and Mengzi, China (32.74 ± 22.56 µg/m³) (Yin et al. 2021); in Manila, Philippines (45 ± 18 µg/m³) (Cohen et al. 2013); and in Cam Hai (27 ± 12 µg/m³) and in Mong Duong (35 ± 12 µg/m³), Vietnam, during the wet season (Hang and Kim Oanh 2014).

Table 2 shows the measured concentrations of 18 trace metals in PM_{2.5}. Al, Fe, Pb, and Zn were the most abundant elements bounded with PM_{2.5} accounting for 85.144% and 89.061% of total obtained metals during summer and fall, respectively, suggesting the mass concentrations of Al, Pb, and Fe were 2.285 times, 1.788 times, and 1.096 times higher in summer compared to those in fall, respectively.

The non-crustal trace elements (As, Cd, Cr, Cu, Mn, Ni, Pb, V, and Zn) contributed to 27.886% and 48.343% of total elements in summer and fall, respectively. As shown in Table 2, the level of Mn, Pb, V, and Ni obtained in the two seasons in Hanoi was lower than the WHO guidelines and European Air Quality Directives (EU (DIRECTIVE 2004/07/EC)). The concentrations of Cd were 40.328 times in summer and 26.643 times in fall higher than the WHO guidelines (0.1 ng/m³), but were still lower than the EU directive (5 ng/m³). The concentrations of Cr observed in this study were at high levels (45.718 ± 13.250 ng/m³ during summer and 11.956 ± 4.752 ng/m³ during fall). The harmful level of Cr (VI) is much higher than that of Cr(III) (Hu et al. 2012). The concentration ratio of Cr(VI) and Cr(III) was reported to vary from 1 to 6 (Taner et al. 2013). Thus, we assumed that the concentration of Cr(VI) was at high levels compared to WHO guidelines (0.25 ng/m³), which may impose a serious risk to human health.

The ratio of Mn and V was used to determine the sources of combustion products (Michael et al. 2006). While oil combustion showed a Mn/V ratio less than 1, coal combustion expressed a Mn/V ratio larger than 1 (Michael J et al. 2006). The average Mn/V ratio was 2.228 (from 0.955 to 6.319) in summer and 62.923 (from 4.127 to 216.678) in fall. Also, the Pb/Fe observed during summer (0.38) and fall (0.25) in Hanoi was much higher than that in Toronto, Canada (0.062) (Lee et al. 2003), Mexico City, Mexico (0.068) (Vega et al. 2004), Seattle, America (0.099) (Maykut et al. 2003). Pb/Fe reported in this study were close to the ratio found in 14 cities in China (0.39 ± 0.32) (Cao et al. 2012). The high Pb/Fe ratio observed in Hanoi indicated the ash in uncontrolled coal combustion (Cao et al.

Table 1 Mean 24-h PM_{2.5} concentration and mean of some meteorological parameters in summer and fall in Hanoi

| | PM _{2.5} (µg/m ³) | Ambient air temperature (°C) | Solar radiation (MW/m ²) | RH (%) |
|--------|--|------------------------------|--------------------------------------|----------------|
| Summer | 41.028 ± 29.241 | 31.316 ± 1.193 | 0.919 ± 0.025 | 74.910 ± 5.909 |
| Fall | 33.643 ± 23.011 | 23.390 ± 1.579 | 0.611 ± 0.078 | 74.047 ± 7.027 |

Table 2 Comparison of element components in PM_{2.5} (mean ± standard deviation) in this study with those measured at different urban sites worldwide (unit ng/m³)

| | Hanoi summer | Hanoi fall | Pre-schools in Hanoi | Cam Hai | Mong Duong | Iasi, Romania | Tehran, Iran | Iurrama, Spain | Singapore, Singapore | Seattle, USA | Beijing, China | Tianjin, China | Chongqing, China | Guangzhou, China |
|----|-------------------|-------------------|----------------------|---------|------------|---------------|---------------|----------------|----------------------|--------------|----------------|----------------|------------------|------------------|
| Al | 634.599 ± 343.330 | 277.711 ± 182.933 | 8160–9482 | 40–100 | 50–200 | 72.53 ± 47.68 | | | 128.68 ± 95.03 | 15 | 760 | | | |
| Ti | 17.388 ± 9.337 | 12.412 ± 7.811 | 547–643 | 20 | 10–20 | 0.07 ± 0.04 | | 6.31 | 2.14 ± 1.64 | 3 | | 110 ± 30 | 150 ± 20 | 60 ± 30 |
| V | 12.041 ± 3.748 | 1.548 ± 0.772 | | 7–10 | 4–8 | 0.44 ± 0.22 | | | | 3 | | | | |
| Cr | 45.718 ± 13.250 | 11.956 ± 4.752 | | 50 | 4 | 2.14 ± 1.36 | 4.24 ± 1.44 | 2.39 | 4.24 ± 1.59 | 0 | | 170 ± 60 | 180 ± 70 | 60 ± 40 |
| Mn | 25.568 ± 13.072 | 78.695 ± 54.210 | 7.63–29.3 | 20–50 | 3 | 9.90 ± 8.03 | | 2.57 | 2.66 ± 2.22 | 3 | 1170 | 1550 ± 740 | 2410 ± 750 | 780 ± 510 |
| Fe | 626.022 ± 332.425 | 571.147 ± 433.409 | 518–827 | 200 | 300–400 | 69.85 ± 66.57 | | 0.99 | 57.15 ± 47.95 | 53 | | | | |
| Co | 0.434 ± 0.157 | 0.368 ± 0.141 | 5.07–8.46 | 10 | 5–10 | 0.07 ± 0.15 | | | 0.50 ± 0.33 | | | | | |
| Ni | 4.99 ± 2.164 | 3.094 ± 1.410 | 3.57–5.69 | 20 | 10–30 | 20.53 ± 19.33 | 4.19 ± 1.38 | 1.31 | 8.80 ± 4.93 | 2 | | | | |
| Cu | 80.219 ± 64.446 | 32.214 ± 25.609 | 40.8–78.8 | 3–20 | 20–30 | 9.71 ± 5.95 | | 11.98 | 4.46 ± 2.21 | 3 | 140 | | | |
| Zn | 169.86 ± 164.703 | 608.726 ± 391.654 | 266–5606 | 100–200 | 100–200 | 29.45 ± 18.50 | 63.92 ± 19.94 | 17.98 | 107.58 ± 38.32 | 9 | | 1340 ± 500 | 920 ± 290 | 590 ± 320 |
| As | 1.806 ± 1.680 | 5.036 ± 3.062 | 1.66–4.57 | | | 6.59 ± 5.69 | 1.34 ± 0.99 | 0.16 | 0.40 ± 0.29 | 1 | | 60 ± 20 | 80 ± 30 | 40 ± 20 |
| Se | 2.422 | 1.027 ± 0.846 | 0.11–0.78 | | | 6.66 ± 3.45 | | 0.23 | | | | | | |
| Sr | 8.184 ± 4.199 | 3.407 ± 1.672 | 62.3–82.2 | 4–5 | 3–7 | 1.40 ± 1.55 | | 1.16 | 0.67 ± 0.31 | | | | | |
| Mo | 8.639 ± 10.246 | 3.247 ± 0.933 | 0.22–0.52 | 10 | 10 | 0.20 ± 0.18 | | | | | | | | |
| Cd | 4.033 ± 2.487 | 2.664 ± 1.632 | 0.35–0.72 | 0.3–2 | 2–4 | 0.29 ± 0.19 | 18.20 ± 6.13 | 0.05 | 0.14 | | | | | |
| Sb | 8.735 ± 7.240 | 3.058 ± 1.947 | 0.09–0.49 | | | 1.98 ± 0.97 | | 0.7 | | | | | | |
| Ba | 29.094 ± 11.599 | 22.882 ± 6.945 | 729–21,987 | | | | | 12.08 | 1 | | | | | |
| Pb | 184.821 ± 231.494 | 103.352 ± 37.203 | 4.38–13.4 | 100–200 | 70–100 | 2.29 | 7.9 | 4 | | 630 ± 280 | 720 ± 260 | 420 ± 260 | 184.14 ± 231.01 | 103.35 ± 37.2 |

2012), implying the coal combustion in Hanoi expressed the dominant impacts. The values of the Cu/Sb ratio reported in the literature showed a wide range. The average Cu/Sb ratio values were 2.17 for fine PM of road dust (Lin et al. 2005), 4.1 for wear particles released from brake pads (Hulskotte et al. 2014), 5.3 for brake lining and rubber from tires (Hjortenkrans et al. 2007), 12.7 for automotive brake abrasion dust (Iijima et al. 2007), and about 125 for crustal matter (Weckwerth 2001). The Cu/Sb ratios observed in this study (about 13.940 in summer and 16.215 in fall) were far lower than the value reported for crustal matter (125) and closed automotive brake abrasion dust (12.7) reported by Iijima et al. (2007), and in the range of the values for road dust (4.8 to 18.9) reported by Alves et al. (2018). Besides, the Fe/Cu ratios depend on the brake constitution, the wear for pads and discs, and the traffic conditions (Alves et al. 2018). When the share between pads and discs' wear was equivalent, the mean Fe/Cu ratio was about 10.4. When the wear from discs increased to 70% and the wear from pads was 30%, the mean Fe/Cu ratio increased to 22. More uses of brakes during the driving time would lead to a higher ratio of Fe/Cu. The average Fe/Cu ratios observed in this study were 16.097 and 27.474 in summer and fall, respectively, which is close to the value obtained from the roadside samples during the intense traffic intensity (14) reported by Alves et al. (2018). This result showed agreement with the high traffic density at sampling sites, especially during rush hour. The possible emission sources of PM_{2.5} in Hanoi were discussed in detail in "Source apportionment."

A comparison between levels of trace elements observed in this study with other cities in the world showed that the concentrations of elements in Hanoi were higher than in Seattle in America (Chen and Lippmann 2009), and Tehran,

Iran (Kermani et al. 2016), and in other urban sites in Europe (Iasi, Romania (Galon-Negru et al. 2019); Navara, Spain (Aldabe et al. 2011); Targoviste, Romania (Radulescu et al. 2015); Singapore, Singapore (Zhang et al. 2017)). However, the concentration of elements found in Hanoi was much lower than the values recorded in Beijing and Tianjin, China (Cao et al. 2012). Generally, the concentrations of trace elements found in this study were comparable with those observed in Cam Hai and Mong Duong (Hang and Kim Oanh 2014) except for Al, Fe, Co, and Ni. The levels of Al, Fe (Co, Ni) in Hanoi were higher (lower) than in Cam Hai and Mong Duong (Hang and Kim Oanh 2014). Tran et al. (2020) reported higher (lower) levels of Al, Ti, Co, Sr, and Ba (Se, Mo, Cd, Sb, Pb), and similar ranges of remaining trace metals at two preschools in Hanoi compared to the results obtained in our study.

Source apportionment

Preliminary analysis of PM_{2.5} origin: enrichment factor

Enrichment factor results showed that group 1 included Al, Fe, Co, and Sr with EF values less than 10, which indicated that the predominant origin of those metals was natural sources (Fig. 2). Although the EF values less than 10 indicated the dominance of natural sources, the elements do not have only natural origins but also a variety of anthropogenic emissions. Group 2 included V and Ni with the EF in the range of 10 to 30, implying the small influence of anthropogenic sources. Group 3 consisted of Mn and Ba with the EF values ranging from 30 to 100, which suggested these elements were emitted from both natural and anthropogenic sources.

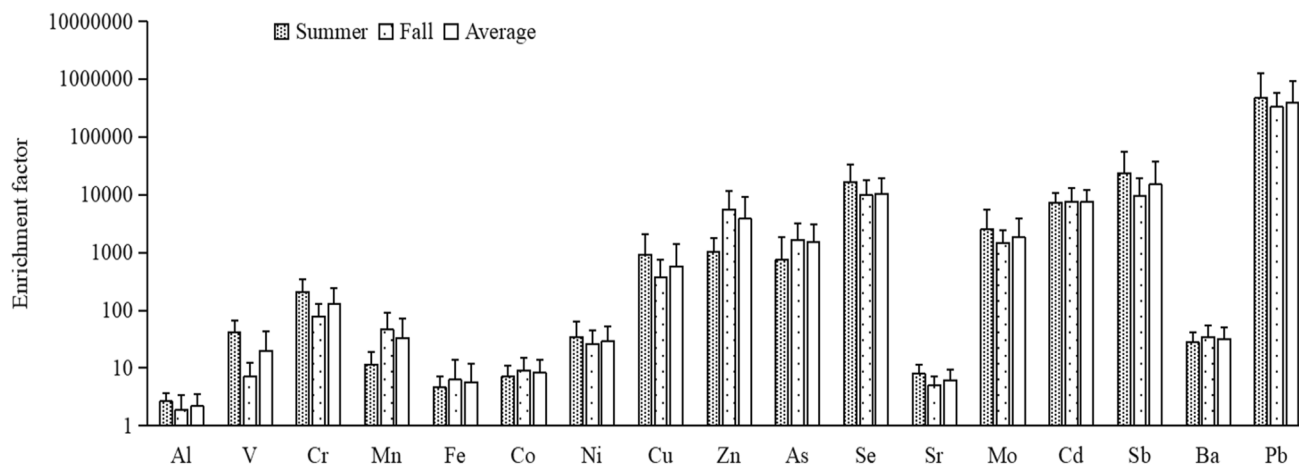


Fig. 2 Enrichment factors of elements in PM_{2.5} of urban site in Hanoi

Identification of PM_{2.5} sources: PCA analysis

In this study, we performed the PCA analysis with a total of 47 samples collected in the two seasons characterized by 16 species. As and Se had been eliminated from the analysis because these elements had more than 20% of the samples being not qualified (the levels were lower than the detection limit), which was the criteria reported by Almeida et al (2005). According to Henry (1989), the minimum number of samples (N) to retrieve a statistically stable PCA analysis is computed as follows:

$$N > 30 + 0.5x(V + 3)$$

where V is the number of species taken into the analysis. In this study, the number of samples was 47, which is larger than the requirements (40). Besides, the Kaiser–Meyer–Olkin measure of sampling adequacy obtained was 0.712, and the results of Bartlett's test showed a significant value less than 0.05, suggesting the data are eligible for PCA analysis (Hair et al. 2006).

Table 3 reports a matrix of loads (after rotation) with the variance explained by each component. Only the loads of species than 0.3 in absolute value were reported because the factor loading less than 0.3 is typically regarded as very poor (Nowak 1998). In addition, only the species with loadings larger than 0.5 were considered for PCA analysis. The first component (PC1) explained 27.154% of the total variance, and has high loadings of Al (0.800), Ti (0.857), Sr (0.758), Co (0.811), and Ba (0.726) and moderate loadings

Table 3 Results of the PCA analysis with the variance explained by the different factors (PCs)

| Species | PC1 | PC2 | PC3 | PC4 |
|------------|--------|--------|--------|--------|
| Al | 0.800 | 0.397 | | |
| Ti | 0.857 | | | |
| V | 0.394 | 0.706 | 0.345 | |
| Cr | 0.525 | 0.707 | | |
| Fe | 0.456 | | | 0.335 |
| Co | 0.811 | | | |
| Ni | | | 0.492 | 0.636 |
| Cu | | | 0.627 | 0.591 |
| Sr | 0.758 | 0.475 | | |
| Mo | | 0.325 | | 0.709 |
| Cd | 0.676 | | 0.586 | |
| Sb | | | 0.730 | |
| Ba | 0.726 | | | 0.445 |
| Pb | | | 0.761 | |
| Eigenvalue | 4.345 | 2.982 | 2.394 | 1.719 |
| % variance | 27.154 | 18.637 | 14.961 | 10.743 |

Only the loads of species than 0.3 in absolute value were reported

of Cr (0.525) and Cd (0.676). The brake activities of traffic vehicles emitted Ba and Co (Pant and Harrison 2013). Besides, Ba and Cd were reported as the trace elements of tire wearing (Salvador 2004; Hjortenkrans et al. 2007; Pant and Harrison 2013). Al, Ti, and Sr were possibly released from abrasions of the road (Juda-Rezler et al. 2020) and dust resuspension from the road (Arruti et al. 2010). In addition, Ti and Al were generally used as sample reference elements of crustal materials (Cesari et al. 2012; Song et al. 2016; Kim et al. 2002). Furthermore, as shown in Table S3, the statistically significant correlations between these elements (the correlation coefficient ranged from 0.302 to 0.670 with one tail p -value less than 0.05) indicated that these species originated from common sources. The traffic fleet frequently moved slowly with a high occurrence of brake activities, especially during rush hours, indicating that non-exhaust traffic emission would impose a high impact at this site. Beside non-exhaust traffic emission, resuspended natural road dust and soil dust could be another source contributing to PM_{2.5} concentration. Moreover, the EF analysis showed the dominance of crustal origin of Al (2.194), Fe (5.632), and Sr (6.203), which confirmed the presence of these natural dust in PM_{2.5}. Thus, the PC1 was assigned to the non-exhaust traffic emissions (brake activities, tire wearing, abrasion of the road, road dust resuspension) and crustal dust.

The second component (PC2) appeared with high positive loadings of V (0.706), and Cr (0.707) with a variance of 18.637%. V are the components of motor oil and engine additives (Cohen et al. 2010; Pant and Harrison 2013). Besides, Cr was associated with traffic emissions and oil combustion (Tian et al. 2010; Banerjee et al. 2015). There was a large number of vehicles using gasoline such as motorbikes and cars, and diesel vehicles (car, van, bus, truck, tricycles) commuting in Hanoi (Nghiem et al. 2020). In addition, PC1 and PC2 showed a good correlation (0.687) in the component transformation matrix. Therefore, the PC2 indicated the presence of exhaust traffic emissions, which was consistent with the EF of V (20.078) and Cr (127.749).

The third component (PC3) presented high loadings of Pb (0.761) and Sb (0.730), and moderate loadings of Cu (0.627) and Cd (0.586) with a variance of 14.961%. These elements were highly enriched with the range of EF from 580.507 (Cu) to 382704.373 (Pb). In addition, Pb, Sb, Cd (Helble 2000; Gatari et al. 2005; Aldabe et al. 2011), and Cu (Aldabe et al. 2011) were emitted from coal combustion. Besides, anthracite coal used for domestic stoves was considered the popular coal emission source in Hanoi, which would be enhanced with the regional pollutants (Cohen et al. 2013; Gatari et al. 2005). Thus, PC3 suggested the small-scale coal combustion sources releasing PM_{2.5} at the NUCE sampling sites. This finding was consistent with the result reported by a previous study that investigated the source apportionment of ultrafine particles in Hanoi (Nghiem et al. 2020).

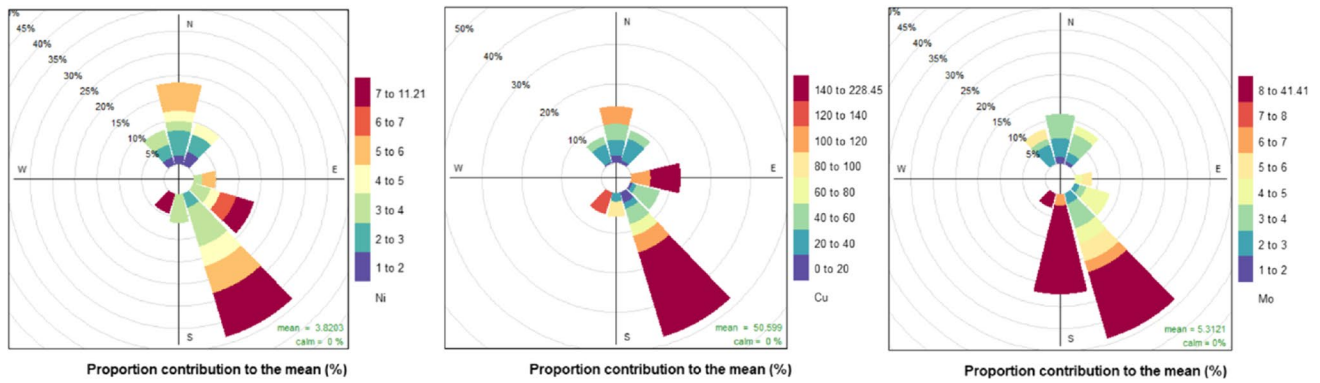


Fig. 3 Pollutant roses of some metal tracers (unit: ng/m^3) for industrial sources

The fourth component (PC4) showed high loading of Mo (0.709) and moderate loadings of Ni (0.636) and Cu (0.591) with a variance of 10.74%. Mo, Cu, and Ni were observed in emissions from industrial processes (Arruti et al. 2010; Geiger and Cooper 2010; Liu et al. 2018a). Mo was identified as the tracer of steel and ferroalloys in the manganese manufacturing industry (Arruti et al. 2010). Previous studies (Tian et al. 2010; Pant and Harrison 2013) found that Cr and Ni might be released from coal and oil combustion to provide heat for various industrial processes. Smelting furnace burning could be the possible anthropogenic source of Cu (Juda-Rezler et al. 2020). Besides, the EF of Mo (1855.410), Cu (580.507), and Ni (29.553) also expressed the influences of anthropogenic sources. Hence, PC4 suggested the impact of industry sources on $\text{PM}_{2.5}$ measured at the sampling site. On account of industry impacts on levels of Mo, Ni, and Cu, the pollutant rose of these metals was derived to clarify their industry origins as shown in Fig. 3. The pollutant rose was used widely to determine the sources of trace

metals (Arruti et al. 2010). As shown in Fig. 3, all the pollutant roses of the three metals expressed peaks pointing in the direction of Vinh Hung (VH-IZ) and Ngoc Hoi (NH-IZ) industrial zone, which is situated about 3.8 km in the southeast of the NUCE site and approximately 10 km in the south southeast of the NUCE site, respectively. Thus, this result confirmed Mo, Ni, and Cu were important metal tracers for industry sources.

Health risk assessment

Non-carcinogenic risks

Figure 4 expresses the evaluation results of non-carcinogenic risks due to trace elements through different exposure pathways in Hanoi during summer and fall. The health risk of all elements for children and adults in both summer and fall via inhalation pathway was highest, followed by ingestion and dermal contact. Therefore, the inhalation pathway was the dominant route of these trace

Fig. 4 Non-carcinogenic risks (sum HI) of elements in $\text{PM}_{2.5}$ samples of urban site in Hanoi

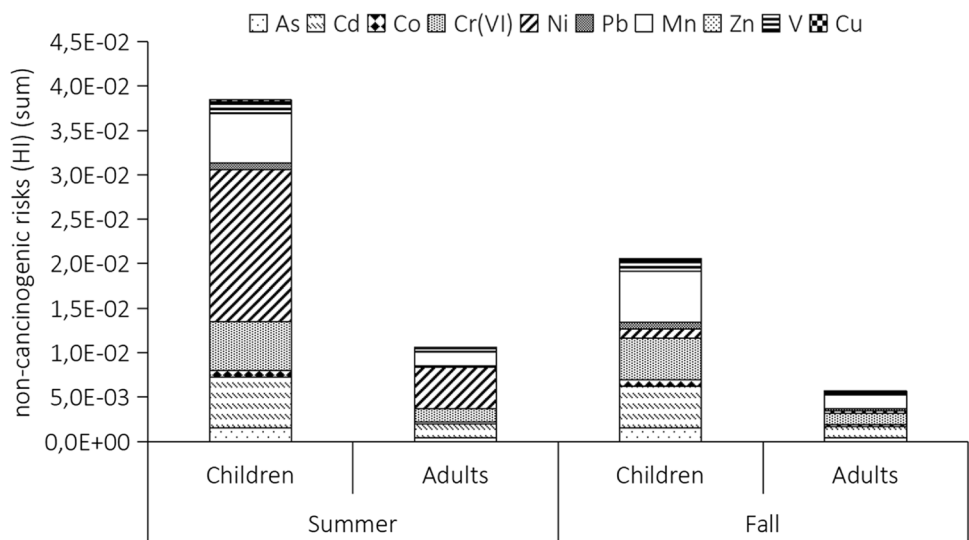
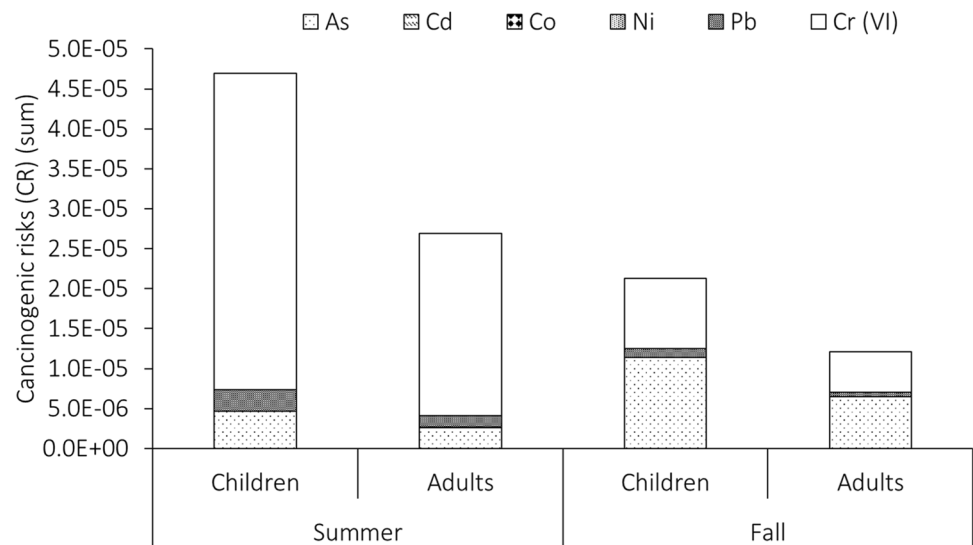


Fig. 5 Carcinogenic risks (sum CR) of elements in PM_{2.5} samples of urban site in Hanoi



elements. The summation of HI of each metal for both children and adults in the two seasons was less than the safe value (HI = 1) at all sites. This result indicated that the potential non-carcinogenic health risks imposed by these metals were tolerable.

The non-carcinogenic risk in summer was larger than that in fall for both adults and children. The summative HQ values of all the elements were less than the safe level (HI = 1), indicating that these elements did not cause non-carcinogenic effects. However, children are exposed to more potential health risks than adults. The HQ values of children were about 3.666 times higher than those of adults, suggesting the concerns for children's potential non-carcinogenic adverse effects caused by the trace elements. This result was similar to the previous studies on ambient PM_{2.5} in Shandong province, China (Zhang et al. 2018), Nanjing, China (Hu et al. 2012), Taiyuan, China (Liu et al. 2018a), and Zhejiang, China (Xu et al. 2020).

Carcinogenic risks

The carcinogenic risk induced by As, Pb, and Cr through three pathways including inhalation, ingestions, and dermal contact were estimated. Because there was a lack of SF for ingestion and dermal exposure route of Cd, Co, and Ni (Cheng et al. 2018), the carcinogenic risks of these metals were only calculated via inhalation pathways. As shown in Fig. 5, the CR of As, Pb, and Cr through the ingestion route were higher than the values via inhalation and dermal contact in both summer and fall. The CR values of Cd, Co, and Ni through the three pathways for both adults and children in the two seasons were lower than 10^{-6} , suggesting the carcinogenic risk caused by these elements could be negligible. Besides, the CR values of As, Pb, and Cr through the three pathways were in the range of 10^{-6} to 10^{-4} , implying

the carcinogenic risks of these metals were at an acceptable level. However, the CR value of Cr bounded in PM_{2.5} for both adults (1.926×10^{-5}) and children (3.342×10^{-5}) in summer was slightly smaller than 10^{-4} , suggesting the potential carcinogenic adverse effects. As shown in Fig. 5, the adverse carcinogenic risk during summer was higher than that during fall with the highest impacts of Cr. The accumulative CR values for children were 0.917 to 1.829 times higher than that for adults, implying that children were exposed to more carcinogenic adverse effects. In addition, the total CR values for children were close to the safety level in both summer and fall, implying that the carcinogenic risks due to exposure to trace metals bounded in PM_{2.5} for children in Hanoi should be paid attention to.

Conclusions

This study performed a characterization of pollution level, inorganic chemical composition, and source apportionment of PM_{2.5} measured during summer and fall at an urban site in Hanoi, Vietnam. Besides, the health risk due to exposure to trace metals bounded with PM_{2.5} was also investigated. The observed average PM_{2.5} concentrations in the two seasons considerably exceeded the guidelines of WHO with slightly higher values recorded in summer. Al, Fe, Pb, and Zn were the most abundant elements bounded with PM_{2.5} in the two seasons. The concentrations of Cd were substantially higher than the WHO guidelines. An analysis of the crustal EF and PCA was applied to identify the main sources imposed on the study area. The results obtained by these two analyses showed a good agreement. The major pollution sources were non-exhaust traffic emissions and crustal dust, exhaust traffic emission, coal combustion, and industrial activities. Although the non-carcinogenic risk in summer was higher

than that in fall, the HI values of all metals were within the safe level for both adults and children in the two seasons. The CR values of Cr were close to the acceptable levels for both adults and children, implying the possible existence of a potential carcinogenic of this metal. Besides, both the HI and CR values for children were significantly higher than that of adults, suggesting more attention to children's health adverse effects due to exposure to metals bound with PM_{2.5} in Hanoi. The results derived from this study provided a comprehensive knowledge on characterization, source apportionment, and health risk assessments of inorganic metals in PM_{2.5} in the typical urban area in the inner city of Hanoi, which would support local government agencies for planning the strategy to mitigate the serious air pollution and public health impact.

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Author contribution Bui Thi Hieu designed the structure, wrote the manuscript, processed, analyzed, and interpreted data, and prepared figures and the manuscript. Nguyen Duc Luong made contributions to the funding application, designed the structure, evaluated the results, and discussions the results. Nguyen Thi Phuong Mai contributed to the design of the structure, data analysis, and interpretation, prepared figures, and evaluated the results and discussions. Bui Quang Trung, Nguyen Van Duy, and Mac Van Dat made contributions to data samplings and analysis of the samples. All authors read and approved the final manuscript.

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Data availability All data generated or analyzed during this study are included in this published article.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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