THE APPLICATION OF STREET DUST FOR MERCURY RISK ASSESSMENT OF LIVING ENVIRONMENT – A CASE STUDY OF VIETNAM

(生活環境の水銀のリスク評価のための市街路のダスト調査：

ベトナムにおけるケーススタディから)

A dissertation submitted
by

PHAN DINH Quang

to
Prefectural University of Kumamoto for the degree of Doctor of Philosophy in Environmental and Symbiotic Sciences

Graduate School of Environmental and Symbiotic Sciences
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ABSTRACT

Environmental pollution is a major problem in most developing countries. Street dust pollution is one of the issues that need to be addressed, mainly come from mines, factories, and urban areas. Street dust can affect the health of residents on a large scale due to the movement of dust by wind. Substances like Hg in dust can affect human health. As a case study, the human health effect of Hg was evaluated by focusing on dust in Quang Ninh where have coal mines and thermal power plants (TPPs), the urban areas in Hanoi, and cigarette smoke from cigarettes in Vietnam. The following attempts can be understood by looking at human health by Hg in street dust.

Hg in the street dust near coal mines (Duong Huy area) and TPPs in Quang Ninh province, where there are large coal mines and three TPPs were evaluated. Hg polluted in street dust from Duong Huy area and TPPs located near the coal mines were the main sources of Hg emission probably results from coal mines and combustion activities. Besides, the high concentration of Hg in street dust from sites Q17 (525.44 ng/g) may come from the close to the non-ferrous metal productions name Quang Ninh Metallic Color Company. The geoaccumulation index (Igeo) showed that street dust in Duong Huy area and TPPs were contaminated by Hg. About 50 % samples from the densely populated sites (Quang Hanh and Cam Pha city) were also contaminated by Hg. The contamination of Hg in street dust of Duong Huy areas resulted from anthropogenic activities based on the enrichment factor (EnF). Although, the human health risk assessment showed that Hg in street dust near coal mines and TPPs had low human health effects (HQs and HIs < 1), adult men may experience higher human health risk due to the continuous exposure to Hg from the workplaces including coal mines and TPPs.

Based on the content of Hg in street dust from Quang Ninh province, a study on Hg in street dust from urban areas - a case study in Hanoi, Vietnam was conducted in March 2019 (spring). Street dusts were sampled from the outer areas to the city center areas of Hanoi. Hg was found at highest concentration in the central areas and significantly higher than in other areas of Hanoi. The street dusts from the central Hanoi were also polluted by Hg. The EnF indicated anthropogenic activities as the main sources of Hg in street dusts. The source of Hg from the center of Hanoi probably resulted from the honeycomb coal commonly used for cooking by restaurants and street foods vendors due to high numbers of these facilities in the area. This indicated the adult women who commonly use honeycomb coal for cooking were at higher risks of Hg due to the constant exposure. During the study, the human health risk assessment of Hg in street dust was also evaluated after a fire outbreak of the compact fluorescence lamps factory (CFL) in Hanoi on August 28^{th}, 2019. The high Hg concentration was found in street dusts near the CFL factory. The results showed that the areas located downstream of the wind direction had a trend higher than other directions. The health risk assessments for both children and adults at the site nearest the CFL showed potential to cause non-carcinogenic human health effects of Hg because HI is almost equal to 1. Besides, the human health risk of Hg in street dusts that were sampled in spring was also evaluated and showed relatively lower health risk for both children and adults in Hanoi because HQs and HIs was far lower than 1. However, adult men in Vietnam may be more at risks of Hg in cigarette smoke based on the health risk assessment of Hg in cigarette smoke that was also investigated. Based on the health risk assessment of Hg in street dusts and cigarette smoke, adult men who smoke may be at higher risk of Hg since they are exposure to both street dust and the cigarette smoke.

This is the case study on the human health risk of Hg in coal mines and combustion, and urban Vietnam. This study provides a valuable information to the Vietnam government about the status of Hg pollution and the associated human health risks. Hence, there is the need to control the Hg pollution from coal combustion and reduce Hg levels in cigarette to preserve human lives and the environment. The new point in this study were: street dust was confirmed as a good tool for environmental and human health risk assessment, the Hg emission and can release Hg into street dust, children are considered as sensitive, hence should not be exposed directly to Hg especially street dust, wind may affect the distribution of Hg in street dust, and cigarettes smoke could be a major source of Hg exposure in adult men. The street dusts could be used as a tool for pollution and human health risk assessment of Hg by other developing countries.

Keyword: Street dust, Mercury, Pollution characteristic, Coal, Compact fluorescence lamps factory (CFL), Cigarette smoke.

CHAPTER 1

General Introduction

1.1. Introduction

Mercury (Hg) is a naturally occurring metal found throughout the environment. Hg is a metal that can be passed into the environment via both natural and anthropogenic sources (Agency for Toxic Substances and Disease Registry, 1999). Hg is a known neurotoxin, the main effects impact vision, hearing, and muscle weakness in children and unborn babies (Sheehan *et al.*, 2014) (Solan and Lindow, 2014). The symptoms of Hg toxics are clearly observed in seafood consumption, including altered memory, attention, and language development in children (Grandjean and Landrigan, 2014). In humans, Hg is mainly exposed through the gastrointestinal tract, and then Hg transfer to the blood. About 5% of Hg is distributed to the tissues within a few days. Hg is a heavy metal that many studies have also suggested can be harmful to the nervous, digestive, respiratory, immune systems, and kidneys, besides causing lung damage. Adverse health effects from Hg exposure can be tremors, impaired vision, attention deficit, and developmental delay during childhood. Around 80% of the inhaled Hg vapor is absorbed in the blood through the lungs. Recent studies suggest that Hg may have no threshold below which some adverse effects do not occur (World Health Organization. Water Sanitation and Health Team, 2005). Especially, methyl mercury is known as a toxic chemical with neurological symptoms. Minamata Disease is known as one of the most significant negative consequences of MeHg poison in Japan caused by consumption of methylmercury-contaminated fish (Hachiya, 2006).

The sources of Hg may come from natural sources and anthropogenic sources. The natural sources have known include volcanoes and forest fires. Meanwhile, the main source of mercury emissions into the environment is mainly due to human activities (anthropogenic sources). The sector of anthropogenic sources that emits Hg was quantity to air in 2015 was described in Table 1. This shows that Hg is emitted from many different sources with different levels. Among them are the main sources of mercury emissions such as artisanal and small-scale gold mining (ASGM, 838 tons, 37.7%), cement production (233 tons,10.5%), stationary combustion of coal (power plant, 292 tons, 13.1%), and non-ferrous metal production (228 tons, 10.3%) (United Nations Environment, 2019).

Table 1. Quantities of mercury emitted to air from anthropogenic sources in 2015, by different sectors.

No.	Sector	Mercury emission (range), tons	Sector % of total
1	Artisanal and small-scale gold mining (ASGM)	838 (675 – 1000)	37.7
2	Biomass burning (domestic, industrial and power plant) *	51.9 (44.3 – 62.1)	2.33
3	Cement production (raw materials and fuel, excluding coal)	233 (117 – 782)	10.5
4	Cremation emissions	3.77 (3.51 – 4.02)	0.17
5	Chlor-alkali production (mercury process)	15.1 (12.2 – 18.3)	0.68
6	Non-ferrous metal production (primary Al, Cu, Pb, Zn)	228 (154 – 338)	10.3
7	Large-scale gold production	84.5 (72.3 – 97.4)	3.8
8	Mercury production	13.8 (7.9 – 19.7)	0.62
9	Oil refining	14.4 (11.5 – 17.2)	0.65
10	Pig iron and steel production (primary)	29.8 (19.1 – 76.0)	1.34
11	Stationary combustion of coal (domestic/residential, transportation)	55.8 (36.7 – 69.4)	2.51
12	Stationary combustion of gas (domestic/residential, transportation)	0.165 (0.13 – 0.22)	0.01
13	Stationary combustion of oil (domestic/residential, transportation)	2.70 (2.33 – 3.21)	0.12
14	Stationary combustion of coal (industrial)	126 (106 – 146)	5.67
15	Stationary combustion of gas (industrial)	0.123 (0.10 – 0.15)	0.01
16	Stationary combustion of oil (industrial)	1.40 (1.18 – 1.69)	0.06
17	Stationary combustion of coal (power plants)	292 (255 – 346)	13.1
18	Stationary combustion of gas (power plants)	0.349 (0.285 – 0.435)	0.02
19	Stationary combustion of oil (power plants)	2.45 (2.17 – 2.84)	0.11
20	Secondary steel production *	10.1 (7.65 – 18.1)	0.46
21	Vinyl-chloride monomer (mercury catalyst) *	58.2 (28.0 – 88.8)	2.6
22	Waste (other waste)	147 (120 – 223)	6.6
23	Waste incineration (controlled burning)	15.0 (8.9 – 32.3)	0.67
24	Total	2220 (2000 – 2820)	100

* Sectors included for the first time in the 2015 inventory.

(Source: UNEP, 2019, Global Mercury assessment 2018)

According to United Nations Environment (2019), the majority of mercury emissions occurred in Asia (49% of which 39% in East and South-east Asia,) followed by South America (18%) and sub-Saharan Africa (16%). Meanwhile, the Europe area accounted for a small percentage, only 3.5% of the global total, and lower in the Middle Eastern States (2.4%), Central America and the Caribean (2.1%), North America (1.8%), North Africa (0.9%), and Australia, New Zealand & Oceania (0.4%) (United Nations Environment, 2019). This shows that East and Southeast Asia where areas emit the most mercury into the environment in the world.

As a developing country, Vietnam is located in southeast Asia. Vietnam became a signatory to the Minamata Convention on Mercury on October 11th 2013 in Japan. After joining the Minamata Convention, Vietnam Chemicals Agency collaborated with the United Nations Industrial Development Organization (UNIDO) has developed the project named "Minamata Convention Initial Assessment in Vietnam" sponsored by the Global Environment Facility (Unido Vietnam Country Office, 2015). The output of the project showed that mercury in Vietnam could be discharged from many sources include energy consumption, fuel production, crude metal production, other material productions, production of containing mercury products, use and disposal of products containing mercury, production of metal from recycling material, waste incineration, waste burning, wastewater treatment, and cremation and burial. In fact, mercury management in Vietnam is still limited. With abundant coal resources, there are now 31 thermal power plants across the country. In addition, many factories that use coal as fuel, such as paper mills, cement factories located in the industrial zones. The amount of Hg released from these sources is difficult to control, treat and recover. Besides, the control and management of factories that use mercury in industrial and household products such as fluorescent lamps, batteries, thermometers need to have clear regulations on mercury. It is necessary to design programs to monitor mercury and prevent incidents of mercury emissions into the environment to protect human health and the environment. Furthermore, the separation of waste at the source is not done, so mercury-containing waste such as fluorescent lamps, thermometers, batteries are mixed with many other types of waste and buried in open landfills.

There are many subjects to determine the level of pollution, the sources of pollution, and the movement of mercury in the environment, thereby conducting a human health risk assessment and giving advice to cleaning of pollution environment and protect human health. Basically, the objects were commonly used for evaluation include conducting mercury observations in water, soil, sediment, or air samples. However, these objects often represent long-term, permanent pollution. In this study, street dust was used as a tool to assess the level of mercury pollution to identify emission sources as well as assess human health risk effects, in the case study in Vietnam.

CHAPTER 2

The application of street dust for mercury risk assessment of living environment – a case study in Quang Ninh Province, Vietnam

2.1. Introduction

Mercury is a toxic element from both natural sources and anthropogenic activities. The main natural sources of Hg include volcanic eruptions, fire forests, and the ocean. Anthropogenic (human-caused) emission Hg from fuels or raw materials or from uses in products or industrial processes. In which, artisanal and small-scale mining, stationary combustion of coal, and nonferrous metals production are most Hg emissions by sector (United Nations Environment, 2019). The dust containing toxic substances such as mercury can enter the air from mining deposits of ores that contain mercury. Street dust is particulate matter emission from the surrounding activities and vehicles. Street dust is characterized by a shorter residence time and is influenced by different factors such as wind, road runoff, and cleanup (Sahakyan *et al.*, 2019).

As a developing country in Southeast Asia, Vietnam has rich natural resources, especially coal mines. The coal mines in Vietnam mainly focus in the northern, due to the abundant coal capacity, many thermal power plants have built and supply electricity for the whole of Vietnam's energy demand (Figure 1). Vietnam was evaluated be a major coal producer for both domestic power plants and as a significant exporter to the Asia Pacific region (Wu, 2009). This country currently has the larger coal development program in the world, after China, India, and Indonesia (Cui *et al.*, 2019). The coal mining activities of Vietnam are mainly located in the north and underground mines. Quang Ninh province – a coastal province in northern Vietnam has the largest coal mines. Quang Ninh also has known as a province with thermal power plants because this province has coal mines available and adjacent to the sea. Thus, the potential risk of emission of mercury from coal mining and coal-burning activities in this area may affect the surrounding local environment and the human who live around. However, the data on mercury in these areas have not yet been reported. Therefore, it needs to conduct a study on mercury in coal mining and combustion and their effect on local human health in Quang Ninh province.

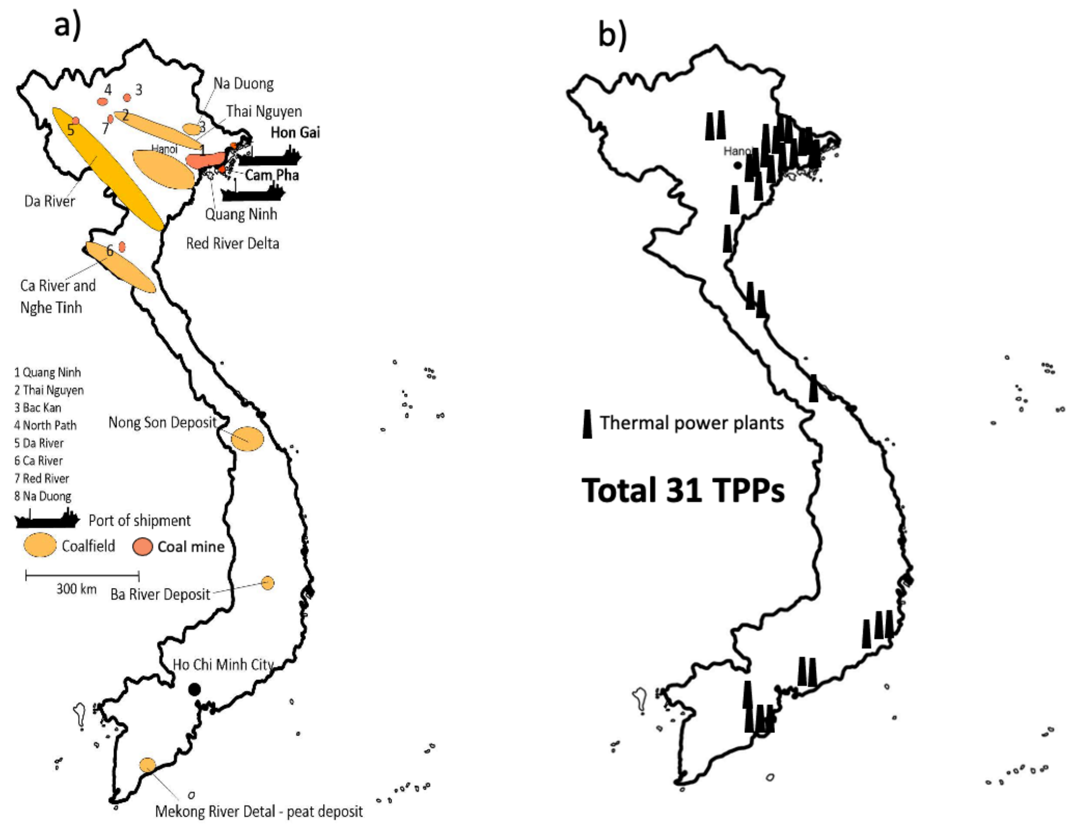

Figure 1. Vietnam'coal resources (a) and thermal power plants in Vietnam (b).

In this chapter, the street dust was used to identify the distribution of Hg in street dust in Quang Ninh, Vietnam. Then, based on these data, the pollution levels and pollution sources of Hg in street dust were discussed. Finally, the human health risk assessment of Hg in the street dust to humans was implemented. The results of this study provided a comprehended understanding of the situation and health risk of Hg in street dust in the case of Quang Ninh, Vietnam, and could be referenced to coal mining and combustion areas in other developing countries.

2.2. Materials and method

2.2.1. Sampling and pretreatment

A total of twenty-five street dust samples was collected from Quang Ninh (Cam Pha city and Ha Long city) in March 2019 and showed in Figure 2. In which, the dust samples were collected from Quang Hanh (site Q1 – Q2) and Cam Pha city (site Q4 – Q6), where rural and urban areas are, respectively. The street dust samples collected in Duong Huy area (site Q17 – Q22) are located on roads close to coal mines, where there is a frequent density of coal transport trucks. Besides, street dust samples collected close to thermal power plants include Cam Pha TPP (site Q7 – Q10), Mong Duong TPP (site Q11 – Q16), and Quang Ninh TPP (site Q23 – Q25).

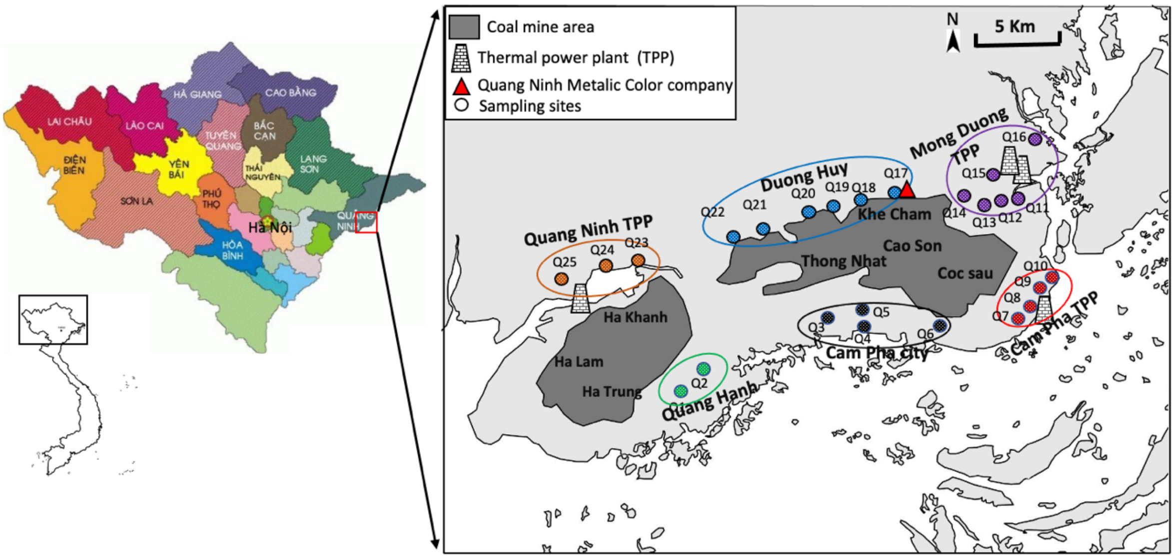

Figure 2. The dust sampling sites in Quang Ninh province.

The dust samples were collected at the edge of the street, adjacent to the asphalt. The street dusts samples were taken during clear and calm weather conditions, when there was no rain or strong wind. The sampling method used was the modified method of Tang *et al.*, 2017. Street dust samples were collected by gently sweeping an area of about 4 – 10 m^2 adjacent to the curb with a clean plastic brush and dustpan, then transferred into clean polyethylene bags. All samples were moved to the laboratory of Prefectural University of Kumamoto, kept in a clean room for two days to remove the moisture, and sieved with 150 mesh (<100 µm hole). The samples were then stored at four degrees Celsius until analysis.

2.2.2. Mercury analysis

Mercury was analyzed using the method of U.S Environmental Protection Agency, 1998, number 7473 by thermal decomposition, amalgamation, and atomic absorption spectrophotometry (U.S. EPA, 1998) was applied to measured Hg in dust samples. In which, the amount of dust sample (200 mg) was weighted using the balance in milligram value and directly measured by using pyrolysis atomic absorption spectrometry with gold amalgamation (MA 3000, NIC, Tokyo, Japan). Two calibration curves were prepared using a Hg standard solution (Wako Chemical Co., JP) for calculation of Hg concentration in samples. The low calibration curves were prepared at six points of 0.2, 0.5, 1, 5, 10, and 20 ng Hg, and the high calibration curve was prepared at six points of 10, 20, 50, 100, 200, and 500 ng of Hg. The calibration curves were described based on the real Hg concentration of analysis with the area of absorbance (ABS) from the atomic absorption spectroscopy of a mercury analyzer (MA 3000, NIC, Tokyo, JP). The correlation coefficients were 0.9999 for both calibration curves at high and low Hg concentrations. The method limit of detection (MDL) was calculated using the formula; 3SD of a blank sample (n = 20), while the method limit of quantification (MQL) was calculated using 10SD. The MDL and MQL values for the Hg analysis method in cigarette leaves were informed at 0.03 ng/g and 0.09 ng/g, respectively.

For quality control and assurance, the reagents and experiment tools were tight control and avoid cross contaminant. All reagents used for analysis were analytical grade and highest purity. All experiment tools need carefully clean before use. For Hg analysis, ceramic sample boats were sequence cleaned by dilute soap solution, tap water, and ultrapure water. Then, sample boats were kept in a furnace oven for 5 hours at 750°C to removed Hg remain before used the sample boat again. Other equipment tools were also sequence washed with dilute soap solution, tap water, ultrapure water, then kept in 5M HNO_3 solution for one day, then washed again by ultrapure water and dry in 30°C before used. The analytical validation for metals and Hg included the analysis of duplicate determinations, and the use of reagent blank. Recovery analysis was done by spiking some of the samples. The samples were spiked with 100 µL of the prepared 100 ng/mL diluted from the Hg standard solution (1000 mg/L, Wako Pure Chemical Company, Osaka, Japan). The recoveries of the spiked samples ranged from 91.92% and 105%. For certainty of the method and accuracy of the results, a certified reference material (CRM 7302-A No. A-0234, NMIJ, Japan) was used.

2.2.3. Quantification of dust pollution

The dust pollution levels of heavy metals were calculated using the geo-accumulation index (I_{geo}) (Mueller, 1969), which is commonly used for the pollution levels of metals in sediment, soil, and dust. The geo-accumulation index for Hg in street dust is expressed as formula (1):

$$I_{geo} = \mathrm{Log}_2\left(\frac{\mathrm{C_m}}{1.5\times \mathrm{B_m}}\right) \qquad (1)$$

Where C_m is the concentration of Hg in samples (µg/g), B_m is the geochemical background value (0.060 µg/g), the data from Zarcinas et al., 2005 were used as Hg of background value (B_m). 1.5 factor is the background matrix correlate factor due to natural fluctuation in the content of a given chemical element in the environment with minimum anthropogenic influence (Dytłow and Górka-Kostrubiec, 2021). Based on the I_{geo} values, the level of pollution was classified as Table 2.

Table 2. The pollution levels of Hg in street dust classified based on I_{geo}.

Igeo values	**I_{geo} classified**
$I_{geo} \leq 0$	Uncontaminated
$0 \leq I_{geo} \leq 1$	Uncontaminated to moderately contaminated
$1 \leq I_{geo} \leq 2$	Moderately contaminated
$2 \leq I_{geo} \leq 3$	Moderately to strongly contaminated
$3 \leq I_{geo} \leq 4$	Strongly contaminated
$4 \leq I_{geo} \leq 5$	Strongly to extremely contaminated
$I_{geo} \geq 5$	Extremely contaminated

The enrichment factor (EnF) is an essential value for identifying the source of emissions of metals that can be either natural or anthropogenic. The EnF was calculated using formula (2):

$$\mathrm{EnF} = \left(\frac{C_i}{C_{ref}}\right) / \left(\frac{B_i}{B_{ref}}\right) \qquad (2)$$

Where C_i is the concentration of Hg in street dust samples (µg/g), C_{ref} is the reference metal (Fe) concentration under this study (µg/g, Table S1). Bi is the target metal concentration in the background soil (0.060 µg/g, Zarcinas *et al.*, 2005). B_{ref} is the background value that was used iron for reference metal. Iron is not a matrix element, but the geochemistry of iron is similar to that of many traces elements in oxic and anoxic environments (Barbieri *et al.*, 2014). Many authors have used the average shale values of Turekian and Wedepohl (1961) as reference baselines (Ghrefat, Abu-Rukah and Rosen, 2011) (Cai and Li, 2019)(Barbieri, 2016). In this study, the B_{ref} value used Fe concentration in shale in the Earth's crust was used to calculate the background value (47200 µg/g) (Turekian and Wedepohl, 1961). The EnF values were categorized as Table 3 (Othman and Latif, 2020) (Sahakyan et al., 2019). In which, if the EnF < 1, indicated the sources of Hg come from the natural origins. If the EnF is higher than 1, indicating the sources of Hg in street dust come from anthropogenic activities. The different EnF classified of the anthropogenic sources to have more understanding of the level of pollution sources by human activities.

Table 3. The enrichment source degree of Hg in street dust classified by EnF.

EnF	Enrichment source degree
EnF<1	Natural origins
1≤EnF<2	Minimal enrichment
2≤EnF<4	Moderate minimal enrichment
4≤EnF<5	Moderately high enrichment
5≤EnF<10	Significant low enrichment
10≤EnF<20	Significant high enrichment
20<EnF<30	Significant high severe enrichment
30<EnF<40	Very high severe enrichment
EnF > 40	Extremely severe enrichment

2.2.4. Health risk assessment

The health risk assessment of both children and adults of Hg in street dust was assessed based on the model developed by the US Environmental Protection Agency (US EPA, 2001).

According to USEPA (2011), the ingestion of dust is a potential route of exposure for both adults and children to environmental chemicals. Children may ingest dust through deliberate hand-to-mouth movements, or unintentionally by eating food that has dropped. Adults may also ingest dust particles that adhere to food, cigarettes, or their hand (USEPA, 2011).

The people living in the vicinity of those areas may be exposed to Hg in street dust via ingestion, inhalation, dermal absorption, and vapor pathways. According to the Exposure Factors Handbook (USEPA, 2001), the average daily dose (ADD, mg/kg/day) for both children and adults via each exposure pathway including (1) direct ingestion of substrate particles (ADD_{ing}); (2) dermal absorption of Hg in particles adhered to exposed skin (ADD_{der}), (3) inhalation of resuspended particles through the mouth and nose (ADD_{inh}) and (4) exposure through inhalation of Hg vapor (ADD_{vap}). The average daily dose (ADD, mg/kg/day) of Hg in dust via each pathway can be estimated using equations (1–4).

$$ADD_{ing} = \frac{C_i \times IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (1)$$

$$ADD_{der} = \frac{C_i \times SA \times SL \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (2)$$

$$ADDinh = \frac{C_i \times InhR \times EF \times ED}{BW \times AT \times PEF} \quad (3)$$

$$\mathrm{ADD_{Vap}} = \frac{\mathrm{C_i \times InhR \times EF \times ED}}{\mathrm{VF \times BW \times AT}} \quad (4)$$

Where Ci (mg/kg) is the concentration of Hg in street dust, ingR (mg/kg) is ingestion rate (child = 200, adult = 100) (Berg, 1995), inhR (m^3/day) is inhalation rate (child = 7.6, adult = 20), EF (days) is exposure frequency (350), ED (years) is exposure duration (child = 6, adults = 30), SA (cm^2) is exposed skin area (child = 2100, adult = 5800), SL is skin adherence factor (child = 0.2, adult = 0.7), ABS is dermal absorption factor (0.001), PEF (m^3/kg) is particle emission factor (1.36×10^9), VF (m^3/kg) is volatilization factor (32675.6), BW (kg) is average body weight (child = 15, adult = 70), AT (days) is averaged life time for non-carcinogen (ED×365) (U.S. EPA, 2001).

The hazard quotient (HQ) for the potential health effect of Hg in street dust through each pathway was calculated using the following equations (5):

$$\mathrm{HQ} = \frac{\mathrm{ADD_{ing/der/inh/vap}}}{\mathrm{RfD_{ing/der/inh/vap}}} \quad (5)$$

Where RfD: the value of reference dose (mg/kg/day, Integrated Risk Information System (RIS). RfD_{ing}: 3.00×10^{-4}, RfD_{der}: 2.00×10^{-5}, RfD_{inh}: 8.57×10^{-5}, RfD_{vap}: 8.57×10^{-5} mg/kg/day), which is taken from the US RAIS (2004). HI equals the sum of HQs for all the exposure routes and was calculated using the equation (6):

$$\mathrm{HI} = \mathrm{HQ_{ing}} + \mathrm{HQ_{der}} + \mathrm{HQ_{inh}} + \mathrm{HQ_{vap}} \quad (6)$$

If the value of HQ is less than one, it is believed that there is no significant risk of Hg in dust effects to the health of residents from this area. In contrast, if the HQ exceeds one, then there is a chance that the effects of Hg in dust properly occur, with a probability that tends to increase as the value of HQ increases (U.S. EPA, 2001). The HI greater than 1, indicates the chance that the non-carcinogenic effects of Hg in the street dusts properly occurred (USEPA, 2001).

2.3. Results and discussion

2.3.1. Hg content in street dust from Quang Ninh

In Quang Ninh province, Hg concentrations in street dusts samples were collected from each site were described in Figure 3. The highest concentration of Hg in street dust was found in site Q17 (525.44 ng/g). The site Q17 was significantly higher than other sites in that area, probably because this site was located near a non-ferrous manufacturing and processing name Quang Ninh Metallic Color Company. According to UNEP (2019), the non-ferrous metal production activity was a sector that discharged 10.3 % mercury emission to the environment (United

Nations Environment, 2019). Quang Ninh Metallic Color Company has known as a company that discharges pollutants into the environment without treatment and is opposed by the people and administratively sanctioned by the Quang Ninh provincial government. However, this company was located close to the coal mine name Khe Cham. Therefore, the high concentration of Hg in street dust in site Q17 may results of combined of the non-ferrous metal smelting activities and coal mining activities.

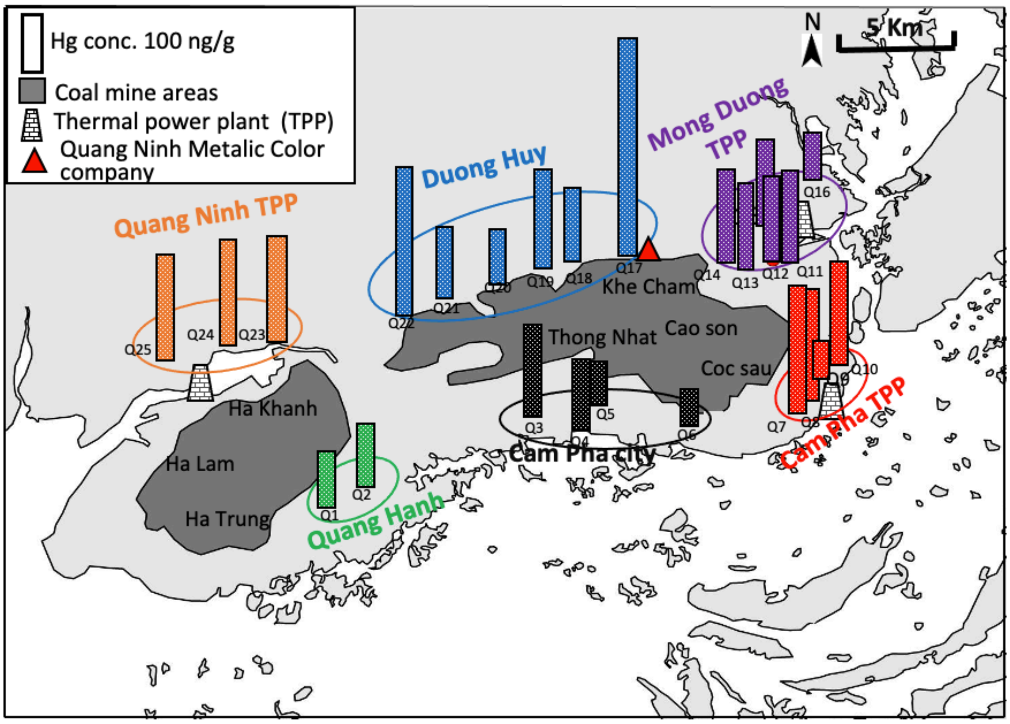

Figure 3. The concentration of Hg in street dust from each site in Quang Ninh. (Hg conc.: Hg concentration)

In the non-ferrous metal smelting process, Hg associated with sulfide ores such as copper, zinc, lead is released during extraction of the aforesaid metals (Kumari, 2011) (W. Liu *et al.*, 2019). Besides, coal has been used in non-ferrous metal smelting also released Hg during coal combustion. It had reported that in non-ferrous metal smelting processes included aluminum, copper, large scale gold, lead, and zinc, Hg was estimated that 227 tons originated in 2015 from the production of these non-ferrous metals process, and accounted for about 10.3 % of total emission (United Nations Environment, 2019). From the high detection of Hg in street dust in this study, the source of Hg was identified in the vicinity of sampling sites.

The average concentration of Hg in street dust from Quang Hanh, Cam Pha city, Duong Huy, Cam Pha TPP, Mong Duong TPP, and Quang Ninh TPP was 103.58 ±11.74 ng/g, 111.01 ±47.90 ng/g, 223.81 ±160.39 ng/g, 176.97 ±73.69 ng/g, 149.77 ±33.00 ng/g, and 196.15 ±2.71 ng/g, respectively (Figure 4). The results showed that Hg in street dust from Quang Ninh in the order of Duong Huy > TPPs > Cam Pha city > Quang Hanh. The highest concentration of Hg in Duong Huy was discussed above, which probably resulted from the non-ferrous metal production activities combines with coal mining activities discharged Hg to the environment. Meanwhile, Hg was found in street dust in the vicinity of TPPs might result from Hg emission mainly from coal combustion. Hg from TPP coal-burning moves to the high exhaust chimney with the filter treatment system. In which 49 % in fly ash, and 25 % in gaseous (Ruud, Leo H.J. and Henk te, 2002).

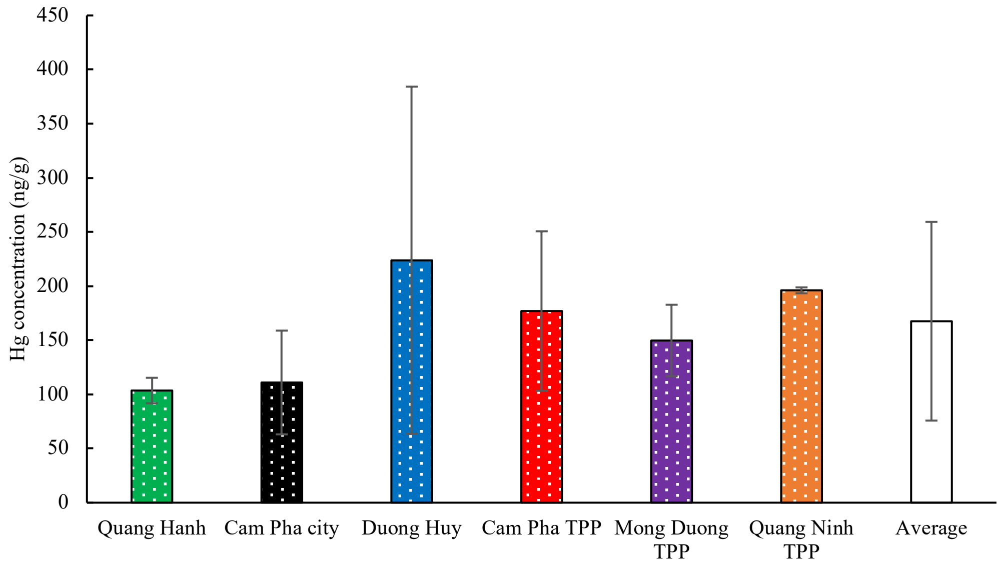

Figure 4. The average concentration of Hg in street dust from each area in Quang Ninh. Quang Hanh (site Q1, Q2); Cam Pha city: (site Q3 – Q6); Duong Huy: (site Q17 – Q22); Cam Pha TPP (site Q7 – Q10); Mong Duong TPP (site Q11 – Q16); Quang Ninh TPP (site Q23 – Q25).

Coal burning is one of the serious sources of mercury emissions, especially spreading into the atmosphere. Wang, Shen and Ma, (2000) report on coal combustion showed that 2493.8 tons of mercury were emitted into the atmosphere during the coal combustion process from 1978 to 1995 in China. The emission of Hg from coal mines may effect the vicinity of those areas. Hg was found in Cam Pha city (urban area) and Quang Hanh (rural area) might because of effected by coal mines and thermal power plants in those areas. The Hg concentration in street dust was found in Cam Pha city was a few higher than Quang Hanh, probably due to the urban activities. Another report also showed that rural areas were much lower pollution of Hg compare to coal mining areas and urban areas (Liang et al., 2017).

2.3.2. The pollution levels and enrichment sources of Hg in street dust

In Quang Ninh province, the average of Igeo values of Hg in street dust in Duong Huy (0.86) and all three TPPs include Cam Pha TPP (0.63), Mong Duong TPP (0.49), and Quang Ninh TPP (0.92) was in the range of uncontaminated to moderately contaminated, while Quang Hanh and Cam Pha city were classified uncontaminated (Figure 5).

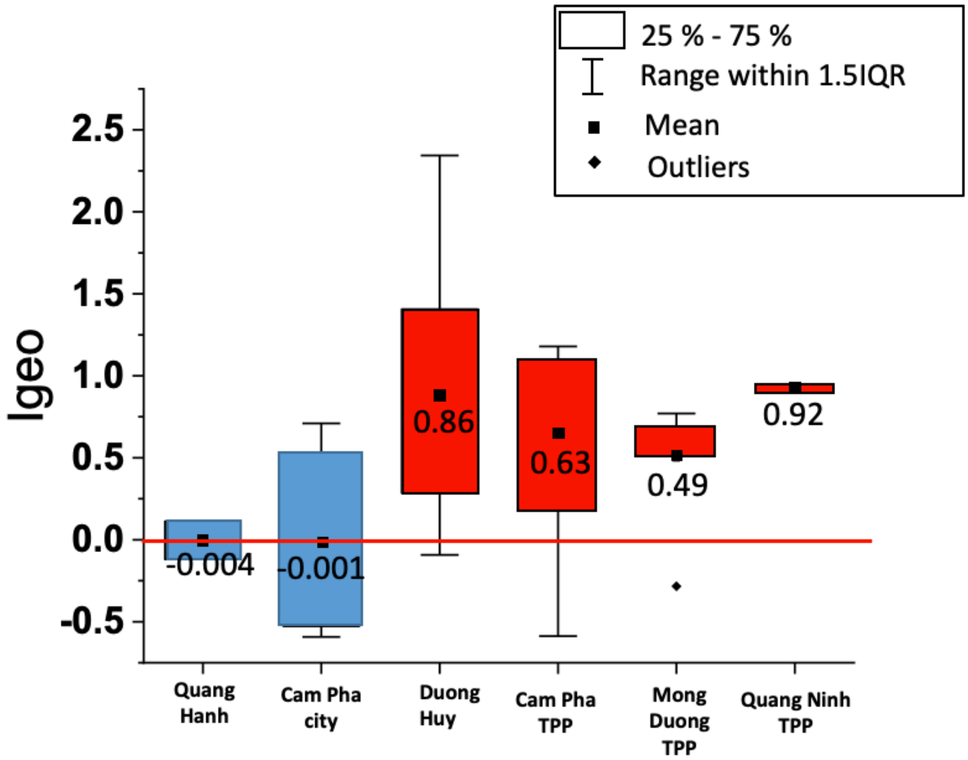

Figure 5. Pollution levels of Hg in street dust in Quang Ninh province.

Uncontaminated to moderately contaminated; Uncontaminated.

The enrichment factor (EnF) of Hg in street dust from Quang Ninh province showed that Duong Huy (23.36) was significant high severe anthropogenic sources (Figure 6). Meanwhile, the EnF of Hg in street dust from other remaining areas include Quang Hanh (12.45), Cam Pha city (11.03), Cam Pha TPP (16.85), Mong Duong TPP (14.74), and Quang Ninh TPP (18.76) were in the range of significant high enrichment by anthropogenic sources of Hg in street dust from these areas. According to our surveying, in the coal mines and TPP areas mainly adult men focus to present here for working. Therefore, adult men have more risk of Hg pollution in these areas because close to the pollution areas and frequency exposure Hg from street dusts.

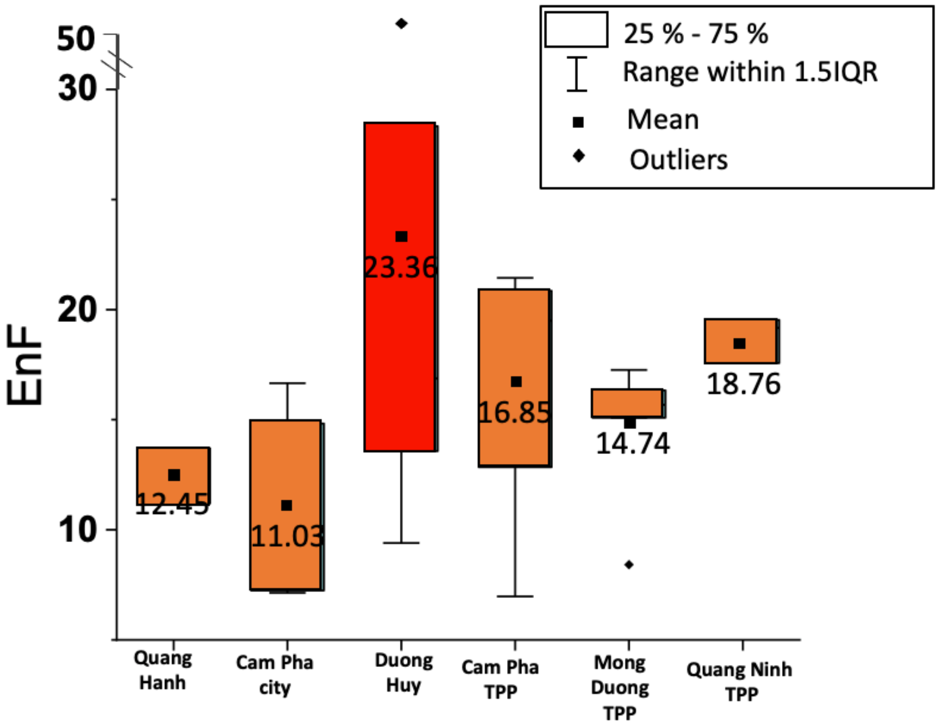

Figure 6. Enrichment factor of Hg in street dust in Quang Ninh.

Significant high severe enrichment by anthropogenic sources; Significant high enrichment by anthropogenic sources.

The street dust used the Hg could use to identify the pollution characteristic and pollution sources degree from human activities. The results showed that in coal mine areas and coal combustion areas reflected the high concentration of Hg in street dusts resulted of the degree of pollution and the level of enrichment sources from human activities.

2.3.3. The comparison of Hg in street dust in Quang Ninh and different areas

A comparison of Hg in street dust from Quang Ninh with other cities was shown in Table 4. The average of Hg in street dust in Quang Ninh was similar with Huainan – a coal energy dominant city in eastern China (Zheng et al., 2015), but much lower than Xiamen (Liang et al., 2009), Ahvaz – the capital city of Khuzestan with many oil wells in Iran (Nazarpour et al., 2018), and in Aviles – an industrial city in Spain with ferrous and non-ferrous plants (Ordóñez et al., 2003). It showed that non-ferrous and oil mining activities and are more pollution sources of Hg could find in street dust compared to coal mining and coal combustion activities. The higher concentration of Hg in street dust in site Q17 also was reflected in the case study in Quang Ninh province of the high sources of Hg from non-ferrous activities in the vicinity of the areas.

Table 4. Literature data of Hg in street dust from different areas.

Area	Description	Hg (ng/g)			Reference
		Mean	Min	Max	
Quang Ninh	Coal mines and thermal power plant	167.56 ±91.76	68.64	525.44	This study
Huainan	Coal energy dominant city in eastern China	160	20	560	(Zheng et al., 2015)
Xiamen	Scenic coastal city in southeast China with coal-fired power plants	280	34	1400	(Liang et al., 2009)
Ahvaz	Capital city of Khuzestan with may oil wells in Iran	2530	20	8750	Nazarpour et al., 2018
Aviles	Industrial city in Spain (non-ferrous factories)	2560	1200	10800	(Ordóñez et al., 2003)

In Quang Ninh province, the high concentrations of Hg in street dust were found high concentration in Duong Huy and TPPs. From these results, we also conducted to survey of the Hg concentration in several coal samples in Quang Ninh coal mines. The coal samples collected from Quang Ninh were original anthracite coal. The average concentration of Hg in coal from Quang Ninh was 592.43 ±115.63 ng/g. With about 25% Hg from coal combustion in thermal power plant's exhaust chimney go to gaseous (Ruud *et al*., 2001), the TPPs in Quang Ninh province also need to treat fly ash and gaseous for Hg reducing in the exhaust chimney system to limit Hg emission into the environment from thermal power coal-combustion.

2.3.4. Human health risk assessment of Hg in street dust from Quang Ninh

The results of human health risk assessment using hazard quotient and hazard index of Hg in street dust samples collected from Quang Ninh were shown in Table 5. In general, the HQs of Hg in street dust from these areas were in the order of $HQ_{vap} > HQ_{ing} > HQ_{der} > HQ_{inh}$ for both children and adults. These results showed that Hg in street dust might effect humans most especially through the vapor pathway. Mercury is an element that easily evaporates to become an invisible, odorless toxic vapor even at room temperature (https://www.epa.gov/mercury/basic-information-about-mercury). Therefore, vapor might be the main pathway of contact of Hg in street dust to humans, followed by ingestion, dermal contact, and inhalation. The average percentage of HQ_{vap} for children and adults accounted for 79.59 % and 93.09% of the HI value, respectively.

The HQ_{vap}, HQ_{ing}, and HQ_{inh} values of Hg in street dust for children were higher than adults; meanwhile, HQ_{der} values for adults were higher than children. For HQ_{ing}, the higher HQ of children than adults were probably due to the children constantly touching with contaminated hands. This led to higher ingestion of the street dust by children, hence the higher HQ_{ing}. For HQ_{inh}, and HQ_{vap}, the higher values of children than adults probably resulted from the higher dermal absorption, inhalation, and vaporization of Hg in street dust as again the smaller body mass of children. For HQ_{der}, due to the exposed skin area of adults is larger than children, leading to a greater chance of exposure through the skin of adults.

Table 5. Health risk of Hg in street dust to children and adults in Quang Ninh.

Area	Site	Ci (μg/g)	Children					Adults				
			HQing	HQder	HQinh	HQvap	HI	HQing	HQder	HQinh	HQvap	HI
Quang Hanh	Q1	0.09528	4.06×10^{-3}	1.28×10^{-4}	1.13×10^{-7}	1.65×10^{-2}	2.07×10^{-2}	4.35×10^{-4}	2.65×10^{-4}	6.40×10^{-8}	9.32×10^{-3}	1.00×10^{-2}
	Q2	0.11188	4.77×10^{-3}	1.50×10^{-4}	1.33×10^{-7}	1.94×10^{-2}	2.43×10^{-2}	5.11×10^{-4}	3.11×10^{-4}	7.51×10^{-8}	1.09×10^{-2}	1.18×10^{-2}
	Average	**0.10358**	$\mathbf{4.41\times10^{-3}}$	$\mathbf{1.39\times10^{-4}}$	$\mathbf{1.23\times10^{-7}}$	$\mathbf{1.80\times10^{-2}}$	$\mathbf{2.25\times10^{-2}}$	$\mathbf{4.73\times10^{-4}}$	$\mathbf{2.88\times10^{-4}}$	$\mathbf{6.96\times10^{-8}}$	$\mathbf{1.01\times10^{-2}}$	$\mathbf{1.09\times10^{-2}}$
Cam Pha city	Q3	0.16932	7.22×10^{-3}	2.27×10^{-4}	2.02×10^{-7}	2.94×10^{-2}	3.68×10^{-2}	7.73×10^{-4}	4.71×10^{-4}	1.14×10^{-7}	1.66×10^{-2}	1.78×10^{-2}
	Q4	0.13096	5.58×10^{-3}	1.76×10^{-4}	1.56×10^{-7}	2.27×10^{-2}	2.85×10^{-2}	5.98×10^{-4}	3.64×10^{-4}	8.79×10^{-8}	1.28×10^{-2}	1.38×10^{-2}
	Q5	0.07510	3.20×10^{-3}	1.01×10^{-4}	8.94×10^{-8}	1.30×10^{-2}	1.63×10^{-2}	3.43×10^{-4}	2.09×10^{-4}	5.04×10^{-8}	7.35×10^{-3}	7.90×10^{-3}
	Q6	0.06864	2.93×10^{-3}	9.21×10^{-5}	8.17×10^{-8}	1.19×10^{-2}	1.49×10^{-2}	3.13×10^{-4}	1.91×10^{-4}	4.61×10^{-8}	6.72×10^{-3}	7.22×10^{-3}
	Average	**0.11101**	$\mathbf{4.73\times10^{-3}}$	$\mathbf{1.49\times10^{-4}}$	$\mathbf{1.32\times10^{-7}}$	$\mathbf{1.93\times10^{-2}}$	$\mathbf{2.41\times10^{-2}}$	$\mathbf{5.07\times10^{-4}}$	$\mathbf{3.09\times10^{-4}}$	$\mathbf{7.45\times10^{-8}}$	$\mathbf{1.09\times10^{-2}}$	$\mathbf{1.17\times10^{-2}}$
Cam Pha TPP	Q7	0.23444	9.99×10^{-3}	3.15×10^{-4}	2.79×10^{-7}	4.07×10^{-2}	5.10×10^{-2}	1.07×10^{-3}	6.52×10^{-4}	1.57×10^{-7}	2.29×10^{-2}	2.47×10^{-2}
	Q8	0.20558	8.76×10^{-3}	2.76×10^{-4}	2.45×10^{-7}	3.57×10^{-2}	4.47×10^{-2}	9.39×10^{-4}	5.72×10^{-4}	1.38×10^{-7}	2.01×10^{-2}	2.16×10^{-2}
	Q9	0.06888	2.94×10^{-3}	9.25×10^{-5}	8.20×10^{-8}	1.20×10^{-2}	1.50×10^{-2}	3.15×10^{-4}	1.92×10^{-4}	4.63×10^{-8}	6.74×10^{-3}	7.24×10^{-3}
	Q10	0.19900	8.48×10^{-3}	2.67×10^{-4}	2.37×10^{-7}	3.45×10^{-2}	4.33×10^{-2}	9.09×10^{-4}	5.53×10^{-4}	1.34×10^{-7}	1.95×10^{-2}	2.09×10^{-2}
	Average	**0.17697**	$\mathbf{7.54\times10^{-3}}$	$\mathbf{2.38\times10^{-4}}$	$\mathbf{2.11\times10^{-7}}$	$\mathbf{3.07\times10^{-2}}$	$\mathbf{3.85\times10^{-2}}$	$\mathbf{8.08\times10^{-4}}$	$\mathbf{4.92\times10^{-4}}$	$\mathbf{1.19\times10^{-7}}$	$\mathbf{1.73\times10^{-2}}$	$\mathbf{1.86\times10^{-2}}$
Mong Duong TPP	Q11	0.16619	7.08×10^{-3}	2.23×10^{-4}	1.98×10^{-7}	2.88×10^{-2}	3.61×10^{-2}	7.59×10^{-4}	4.62×10^{-4}	1.12×10^{-7}	1.63×10^{-2}	1.75×10^{-2}
	Q12	0.16152	6.88×10^{-3}	2.17×10^{-4}	1.92×10^{-7}	2.80×10^{-2}	3.51×10^{-2}	7.38×10^{-4}	4.49×10^{-4}	1.08×10^{-7}	1.58×10^{-2}	1.70×10^{-2}
	Q13	0.16111	6.87×10^{-3}	2.16×10^{-4}	1.92×10^{-7}	2.80×10^{-2}	3.50×10^{-2}	7.36×10^{-4}	4.48×10^{-4}	1.08×10^{-7}	1.58×10^{-2}	1.69×10^{-2}
	Q14	0.17647	7.52×10^{-3}	2.37×10^{-4}	2.10×10^{-7}	3.06×10^{-2}	3.84×10^{-2}	8.06×10^{-4}	4.91×10^{-4}	1.19×10^{-7}	1.73×10^{-2}	1.86×10^{-2}
	Q15	0.14833	6.32×10^{-3}	1.99×10^{-4}	1.77×10^{-7}	2.57×10^{-2}	3.23×10^{-2}	6.77×10^{-4}	4.12×10^{-4}	9.96×10^{-8}	1.45×10^{-2}	1.56×10^{-2}
	Q16	0.08501	3.62×10^{-3}	1.14×10^{-4}	1.01×10^{-7}	1.47×10^{-2}	1.85×10^{-2}	3.88×10^{-4}	2.36×10^{-4}	5.71×10^{-8}	8.32×10^{-3}	8.94×10^{-3}
	Average	**0.14977**	$\mathbf{6.38\times10^{-3}}$	$\mathbf{2.01\times10^{-4}}$	$\mathbf{1.78\times10^{-7}}$	$\mathbf{2.60\times10^{-2}}$	$\mathbf{3.26\times10^{-2}}$	$\mathbf{6.84\times10^{-4}}$	$\mathbf{4.16\times10^{-4}}$	$\mathbf{1.01\times10^{-7}}$	$\mathbf{1.47\times10^{-2}}$	$\mathbf{1.58\times10^{-2}}$
Duong Huy	Q17	0.52544	2.24×10^{-2}	7.05×10^{-4}	6.26×10^{-7}	9.12×10^{-2}	1.14×10^{-1}	2.40×10^{-3}	1.46×10^{-3}	3.53×10^{-7}	5.14×10^{-2}	5.53×10^{-2}
	Q18	0.12618	5.38×10^{-3}	1.69×10^{-4}	1.50×10^{-7}	2.19×10^{-2}	2.74×10^{-2}	5.76×10^{-4}	3.51×10^{-4}	8.47×10^{-8}	1.23×10^{-2}	1.33×10^{-2}
	Q19	0.18592	7.92×10^{-3}	2.50×10^{-4}	2.21×10^{-7}	3.23×10^{-2}	4.04×10^{-2}	8.49×10^{-4}	5.17×10^{-4}	1.25×10^{-7}	1.82×10^{-2}	1.96×10^{-2}
	Q20	0.09706	4.14×10^{-3}	1.30×10^{-4}	1.16×10^{-7}	1.68×10^{-2}	2.11×10^{-2}	4.43×10^{-4}	2.70×10^{-4}	6.52×10^{-8}	9.50×10^{-3}	1.02×10^{-2}
	Q21	0.13398	5.71×10^{-3}	1.80×10^{-4}	1.60×10^{-7}	2.32×10^{-2}	2.91×10^{-2}	6.12×10^{-4}	3.73×10^{-4}	9.00×10^{-8}	1.31×10^{-2}	1.41×10^{-2}
	Q22	0.27427	1.17×10^{-2}	3.68×10^{-4}	3.27×10^{-7}	4.76×10^{-2}	5.96×10^{-2}	1.25×10^{-3}	7.63×10^{-4}	1.84×10^{-7}	2.68×10^{-2}	2.88×10^{-2}
	Average	**0.22381**	$\mathbf{9.54\times10^{-3}}$	$\mathbf{3.00\times10^{-4}}$	$\mathbf{2.67\times10^{-7}}$	$\mathbf{3.88\times10^{-2}}$	$\mathbf{4.87\times10^{-2}}$	$\mathbf{1.02\times10^{-3}}$	$\mathbf{6.22\times10^{-4}}$	$\mathbf{1.50\times10^{-7}}$	$\mathbf{2.19\times10^{-2}}$	$\mathbf{2.35\times10^{-2}}$
Quang Ninh TPP	Q23	0.19794	8.44×10^{-3}	2.66×10^{-4}	2.36×10^{-7}	3.43×10^{-2}	4.30×10^{-2}	9.04×10^{-4}	5.50×10^{-4}	1.33×10^{-7}	1.94×10^{-2}	2.08×10^{-2}
	Q24	0.19303	8.23×10^{-3}	2.59×10^{-4}	2.30×10^{-7}	3.35×10^{-2}	4.20×10^{-2}	8.81×10^{-4}	5.37×10^{-4}	1.30×10^{-7}	1.89×10^{-2}	2.03×10^{-2}
	Q25	0.19748	8.42×10^{-3}	2.65×10^{-4}	2.35×10^{-7}	3.43×10^{-2}	4.29×10^{-2}	9.02×10^{-4}	5.49×10^{-4}	1.33×10^{-7}	1.93×10^{-2}	2.08×10^{-2}
	Average	**0.19615**	$\mathbf{8.36\times10^{-3}}$	$\mathbf{2.63\times10^{-4}}$	$\mathbf{2.34\times10^{-7}}$	$\mathbf{3.40\times10^{-2}}$	$\mathbf{4.27\times10^{-2}}$	$\mathbf{8.96\times10^{-4}}$	$\mathbf{5.45\times10^{-4}}$	$\mathbf{1.32\times10^{-7}}$	$\mathbf{1.92\times10^{-2}}$	$\mathbf{2.06\times10^{-2}}$

In Quang Ninh province, the average HIs of Hg in street dust collected from Duong Huy had trend higher than TPPs, followed by Cam Pha city and Quang Hanh. These results indicate that people who are frequently present in coal mine areas and in the vicinity of thermal power plants are at a higher risk of Hg exposure. According to our survey, in the areas of coal mines and thermal power plants in Quang Ninh, mainly are adult men who work in coal mines and thermal power plants. Therefore, the Hg in street dust collected from Duong Huy and TPPs areas may have more risk effect on the adult men than other groups.

The HI value in Hg street dust from site Q17 was highest in the study area for both children and adults (1.14×10^{-1} and 5.53×10^{-2} respectively), indicate this area had the highest risk to humans in the vicinity of Quang Ninh Metalic Color company. This result indicated Hg discharged from non-ferrous metal production activities probably have more health risks to human health than other areas.

The human health risk assessment of Hg in street dust in Quang Ninh were evaluated. HQ and HI values were below 1, indicates no significant risk effect by Hg to the human health in Quang Ninh province through ingestion, dermal absorption, inhalation, and vapor exposure pathways.

2.4. Conclusions

This is the first study of the Hg pollution situation in street dust from Quang Ninh. The high concentration of Hg in street dust near coal mines, TPPs, resulted in the pollution sources of Hg in these areas were identified. The high Hg concentration in site Q17 (525.44 ng/g), resulted from non-ferrous manufactory activities in the vicinity of Duong Huy areas. Street dust near coal mine areas (Igeo = 0.86) and TPPs areas (Igeo ranged 0.49 – 0.92) were classified as contaminated by Hg, which indicates adult men, may have a more health risk of Hg exposure from workplaces. The human health risk assessment showed that vaporization of Hg in street dust was most effect risk to children and adults. Although, Hg in street dust from Quang Ninh may not pose significant human health risk because HQs and HIs < 1. It was one factor that contribute risk to human if combined with other sources that have exposure Hg to human in those areas. The street dust could be used to assess the pollution level, pollution sources identified, and human health risk assessment in coal mines and combustion of coal in thermal power plants as well as non-ferrous manufacturing.

CHAPTER 3

The application of street dust for mercury in risk assessment of living environment – a case study in Hanoi city, Vietnam

3.1. Introduction

Recently, dust pollution posses a big problem in urban areas because of traffic vehicles and factories scattered around the city (Dytłow and Górka-Kostrubiec, 2021). The two primary sources of street dust are the deposition of previously suspended particles (atmospheric aerosol) and displaced urban soil. This street dust can carry pollutants, including harmful heavy metals such as cadmium (Cd), mercury (Hg), lead (Pb), zinc (Zn), and other metals (Hwang *et al.*, 2016), and effect both the environmental quality and human health. Several studies showed that these heavy metals were found in urban street dust at different levels (Zhang, Pan and Lee, 2014) (Othman and Latif, 2020) (Aguilera *et al.*, 2021). The street dust can pose potential health effects of metals upon exposure through inhalation, ingestion, and dermal contact. Residents easily come into contact with street dusts and children are the most affected due to the hand-to-mouth activities (Zheng *et al.,* 2010). Meanwhile, Hg is a ubiquitous environmental toxicant that can cause various adverse health effects in humans, especially children (Meza-Figueroa *et al.,* 2007).

Hanoi is the capital of Vietnam with more than 10 million population. Dust pollution in Hanoi is mainly caused by traffic, construction, and daily human living activities. According to the Vietnam Ministry of Natural Resources and Environment, the air quality index (AQI) in Hanoi was moderate and unhealthy for the sensitive group for many months, mostly in winter and spring.

There was an incident in Hanoi city when Rang Dong warehouse compact fluorescence light (CFL) factory was fired on August 28th, 2019. This incident might discharged a high amount of Hg into the environment. It was problematic that the pollution levels of Hg increased around the surrounding area. The monitoring of Hg around the accidental area is needed to assess their health impact on community residents. This accident occurred near the sampling site in spring 2019. In autumn 2019, the street dusts from the same site were sampled and the concentration of Hg were determined. Based on the concentrations, the pollution levels and health risk of Hg to both children and adults through ingestion, inhalation, dermal, and vapor routes were evaluated using hazard quotient (HQ) and hazard index (HI).

The aim of this chapter was, (1) used the street dust to conduct the distribution of Hg in Hanoi, Vietnam. Then, based on these data (2) the pollution levels and pollution sources of Hg in street dust were discussed. Finally, (3) the human health risks assessment of Hg in the street dust were implemented. The results of this study might provide the comprehended understanding the situation and health risk of Hg in street dust in the case of Hanoi city and could be referenced to other developing countries.

3.2. Materials and method

A total of twenty-nine street dust samples were collected in March 2019 in Hanoi city. The sampling site locations (site H1 – H29) are shown in Figure 7. Outer Road 1 (OR1): H1 – H4; Outer Road 2 (OR2): H5 – H10; Ring Road (RR1): site H26 – H29; Ring Road 2 (RR2): H19 – H25; Ring Road 3 (RR3): H11 – H18. The RR1, RR2, and RR3 areas are planned by the Hanoi government. In which, RR1 is the core central area of the city, known as the old town. This place is the most crowded and bustling place in Hanoi in terms of population density. Many restaurants, hotels, tourists, and business activities are also mainly concentrated in the RR1 area. Ring road 2 (RR2) is a closed inner-city traffic circle in Hanoi. The route has a total length of more than 43 km. This area has the problem its traffic congestion mainly by motorbikes and cars using gasoline fuel. Ring road 3 (RR3) is the heaviest traffic area. Therefore, in parallel with RR3, combine to an elevated highway that is raised above grade for its entire length with a mixture of vehicles include motorbikes, cars, trucks using both gasoline and diesel fuel. The RR3 is the combination of the normal road below and the highway above. OR1 is located outlying city area, which is the outer side of Hanoi city and contains a lot of new construction budling, construction activities in this area with higher density compared to RR3 and RR2, and RR1. OR2 is also the outer area of Hanoi but is located to the east of Hanoi city, this area near industrial complex areas. OR2 had a high-traffic road with many large diesel vehicles such as container trucks.

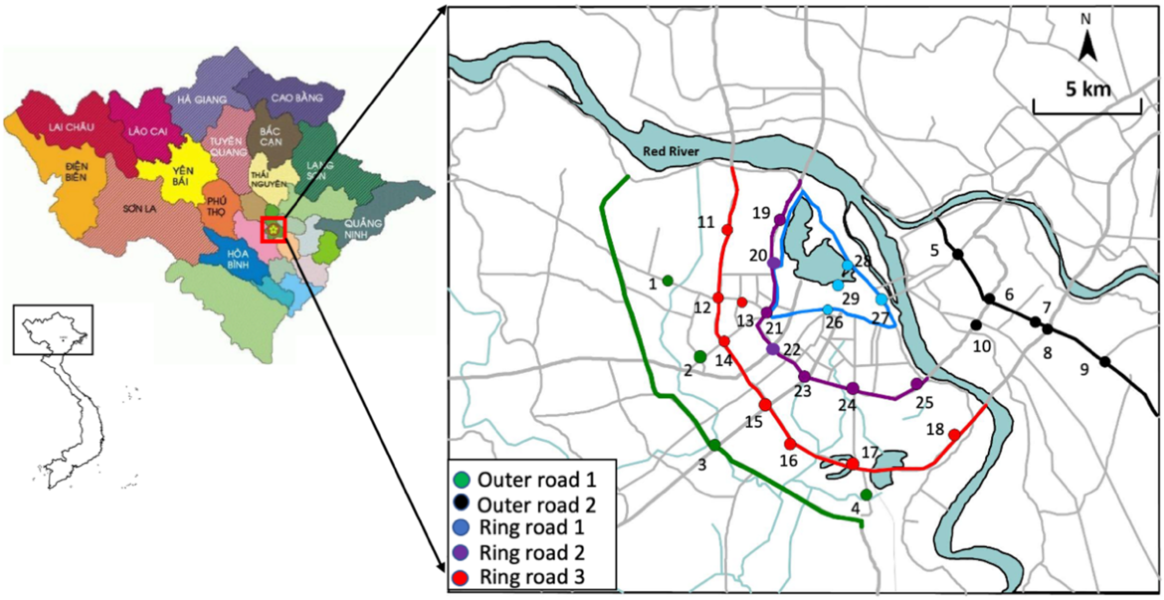

Figure 7. The dust sampling sites in Hanoi city.

The method for chemical analysis, pollution characteristic and source, and human health risk assessment through ingestion, inhalation, dermal contact, and vapor were described in sections 2.2.2, 2.2.3, and 2.2.4, respectively. The reference metal (Fe) concentrations in street dust samples for enrichment factor calculation are shown in Table S2.

3.3. Results and discussion

3.3.1. Hg content in street dust from Hanoi

During the spring of sampling, the average concentration of Hg in street dusts in each area include OR1, OR2, RR1, RR2, and RR3 are shown in Figure 8.

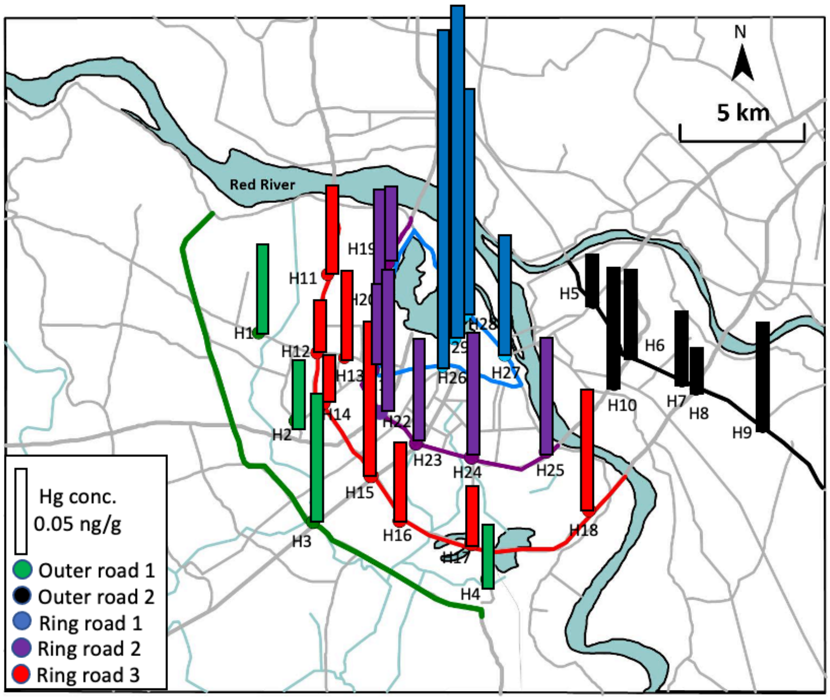

Figure 8. The concentration of Hg in street dust from each site in Hanoi during spring.
Hg conc.: Hg concentration

The average of Hg in OR1, OR2, and RR3 was 50.90 ±17.49 ng/g, 48.54 ±17.84 ng/g, 50.62 ±22.24 ng/g, respectively (Figure 9). Meanwhile, the average concentration of Hg in RR2 (64.38 ±14.26 ng/g) that was closer to the center of Hanoi was higher than outside of the city. Mercury was found at the highest concentration at RR1 with 151.42 ±61.86 ng/g on average, shown a significant higher ($p < 0.01$) than in other areas. The results showed that it probably has a high source of Hg in the center of Hanoi that leaked Hg to the environment and was found significantly higher than outside of the city. That leads the Hg in street dust from RR2 may also affect by the same source from RR1 and higher than the RR3, OR1, and OR2 in Hanoi city.

The average concentration of Hg in street dust collected in OR1, OR2, RR2, and RR3 was not significantly different ($p > 0.05$). In detail, the concentration of Hg in street dust in RR1 was 2 – 3 times higher than in those areas. This difference shows there may have anthropogenic sources of Hg in central Hanoi and need to investigate.

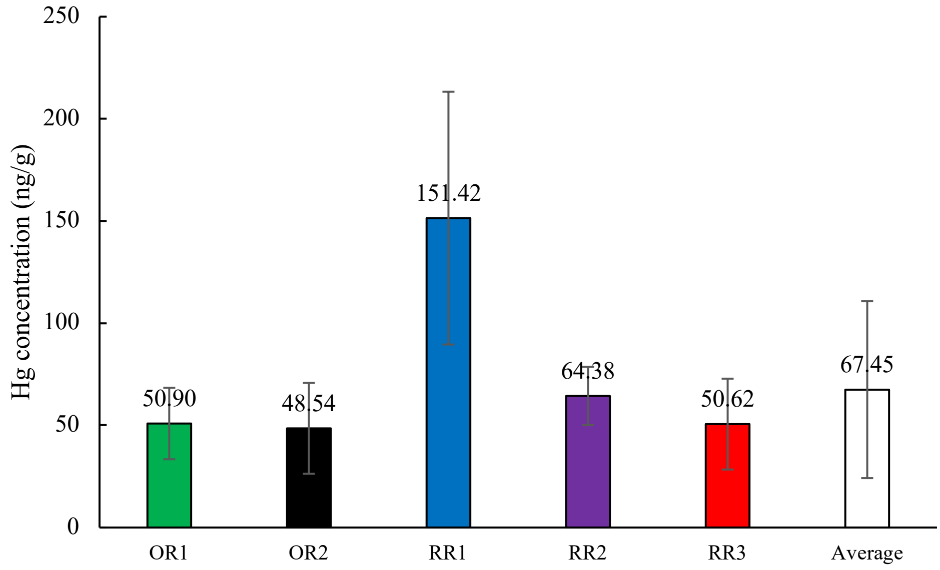

Figure 9. The average concentration of Hg in street dust from each site in Hanoi.
OR1: Outer road 1 (site H1 – H4); OR2: Outer road 2 (site H5 – H10); RR1: Ring road 1 (site H26 – H29); RR2: Ring road 2 (site H19 – H25); RR3: Ring road 3 (site H11 – H18).

The distribution of Hg in street dust from different areas in Hanoi showed that there might be sources of Hg in the RR1 that need to be a concern. Hg in street dust in OR1, OR2, and RR3 was found in similar concentrations while RR2 was a little higher than those.

3.3.2. The pollution and enrichment source characteristic of Hg in street dust

The geoaccumulation index (Igeo) of Hg in street dust from Hanoi city was showed in Figure 10. The average of Igeo of Hg in street dust from OR1, OR2, RR1, RR2, and RR3 was -1.08, -1.18, 0.43. -0.72, and -1.15, respectively. The results showed that only the average of Igeo values of Hg in street dust collected from RR1 exceeded the standard value with 75 % samples in the range of 0 ≤ Igeo ≤ 1, indicate uncontaminated to moderately contaminated. Other areas include RR2, RR3, OR1, and OR2 had Igeo values < 0, indicated uncontaminated.

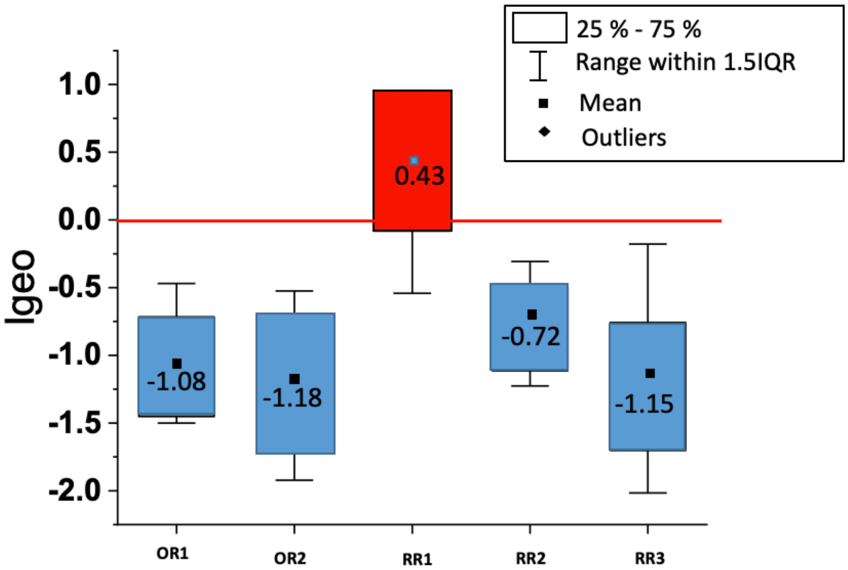

Figure 10. Pollution characteristic of Hg in street dust in Hanoi.

Uncontaminated to moderately contaminated; Uncontaminated.

In Hanoi city, the average of EnF of Hg in street dust from OR1, OR2, RR1, RR2, and RR3 was 4.47, 3.98, 11.58, 4.99, and 3.78, respectively (Figure 11). The RR1 also had highest EnF than other areas with the classification of significant high enrichment, meanwhile OR1 and OR2 were moderately high enrichment, OR2 and RR3 were significant high enrichment.

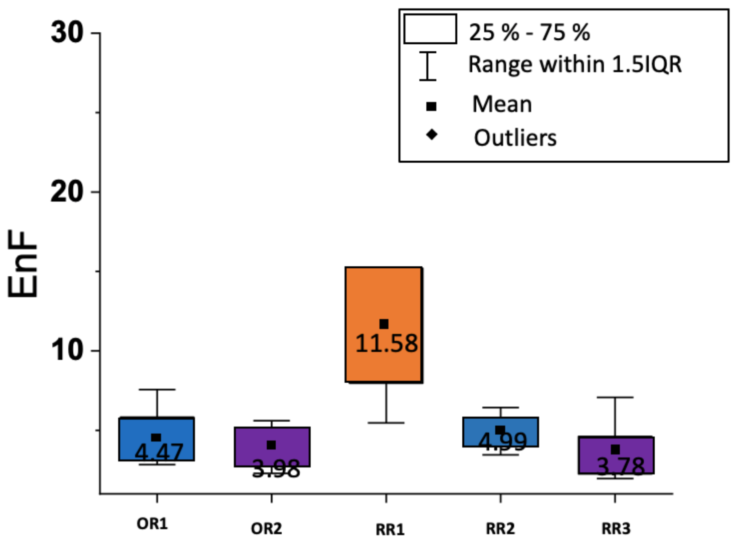

Figure 11. Enrichment factor of Hg in street dust in Hanoi.

Significant high enrichment by anthropogenic sources; Moderately high enrichment by anthropogenic sources; Moderate minimal enrichment by anthropogenic sources.

The street dust used the Hg could use to identify the pollution characteristic and pollution sources degree from human activities. The results showed that in coal mine areas and coal combustion areas, reflected the high concentration of Hg in street dusts resulted of the degree of pollution and the level of enrichment sources from human activities.

3.3.3. The comparison of Hg in street dust in Hanoi with other cities

A comparison of Hg in street dust from Hanoi with other metropolitan cities is shown in Table 6. The average of Hg in street dust in Hanoi from spring 2019 was lower than other cities from China such as Beijing (Men et al., 2018), Guangzhou (Huang et al., 2018), Shanghai (Liu et al., 2019), Huainan (Zheng et al., 2015), Xiamen (Liang et al., 2019), and Taipei, Taiwan (Zhang et al., 2014). This reflected that the source of Hg pollution cities of China might come from bigger sources than Hanoi city. According to Li et al., 2009, China had the most contaminated sites of Hg in Asia, which was nearly identical to the situation of anthropogenic Hg emission. From the comparison, Hg is a global pollution toxic substance, therefore, the Hg from China may effect the Hg pollution in other countries where close to China such as Vietnam, Korea, Japan etc. Thus, it is necessary to control Hg pollution in each country for human health protection.

Table 6. Literature data of Hg in street dust from different metropolitan cities.

Area	Description	Hg (ng/g)			Reference
		Mean	Min	Max	
Hanoi	Capital city of Vietnam	67.45 ±43.28	25.59	200.84	This study
Beijing	Capital city of China	160	40	780	(Men et al., 2018)
Guangzhou	Megacity city in southern China	235 ±245	-	-	(Huang et al., 2012)
Shanghai	Largest city in eastern China	596	210	2184	(Y. Liu *et al.*, 2019)
Taipei	Capital city of Taiwan	1590	-	-	(Zhang, Pan and Lee, 2014)

However, the high concentration of Hg in street dust was found in the RR1 area of Hanoi city, it was almost equal to Hg concentration in street dust in Beijing of China. As we have known, RR1 area has many restaurants, food streets, hotels, and tourisms. According to our surveying, in RR1, there were a lot of restaurants that used honeycomb coal for cooking because of cheap, convenient, and suitable for business works of food in restaurants. Therefore, we conduct to survey of Hg concentration in several honeycomb coal samples in Hanoi (n = 2). The honeycomb coal samples were collected from Hanoi markets. The average concentration of Hg in honeycomb coal in Hanoi market was 719.53 ±48.69 ng/g. Compared to the original anthracite coal from Quang Ninh province (592.43 ±115.63 ng/g), it showed that Hg concentration in honeycomb coal was very high, even higher than original anthracite coal from Quang Ninh coal mines. The higher concentration of Hg in honeycomb coal than original anthracite coal may result from the honeycomb coal production process. According to our investigation, in Vietnam, the honeycomb coal was made from original coal, slug, and fly ash. Sludge is used in coal production to keep the coal texture, while fly ash from thermal power plants is used to produce honeycomb coal because in fly ash, there is still unburnt carbon. It had known that 49 % of Hg from coal come to fly ash (Ruud *et al*., 2001) may result of Hg in honeycomb coal was higher than original coal that collected from Quang Ninh coal and result of Hg concentration in Hanoi probably because of honeycomb coal combustion in cooking.

Figure 12. Honeycomb coal using for cooking in Hanoi city.

Recently, there are many types of fuels that are used in cooking, such as gas, electricity, which are commonly used in Hanoi city. However, honeycomb coal is still used because of cheap, convenient, and suitable for regular cooking work, mainly in restaurants and street food eateries. In RR1 area of Hanoi city, where have known as the core of Hanoi city, have most concentrated of restaurants, hotel, and tourist. The use of honeycomb charcoal in cooking in restaurants and street food stalls here may be responsible for the higher levels of mercury detected in road dust than in other areas. Furthermore, Hg from honeycomb coal that uses in daily cooking may have more effect on women, who are common with daily cooking work (Figure 12). Therefore, we suggest the Hanoi government should consider reducing the Hg in coal for cooking to limit the risk effect of Hg on human health.

3.3.4. Hg content in street dust after the fire outbreak of the compact fluorescence lamps factory in Hanoi

On August 28th 2019, the compact fluorescence lamps (CFL) factory generates light for Rang Dong Light Source and Vacuum Flask locates in Thanh Xuan Trung ward, Thanh Xuan district, Hanoi city was incidentally fire outbreak. According to Vietnam Environment Administration, Ministry of Natural Resource and Environment, the CFL factory using Hg liquid form to produce the fluorescence lamps. It was problematic that the pollution levels of Hg and the other metals increased around the surrounding area. After the incident fire outbreak, the street dust samples in Hanoi city was conducted to collect at the same sites with March 2019. In which site H15 belongs to RR3 is very close to the CFL factory.

The average Hg in dust samples was 265.83 ±810.86 ng/g (ranged from 31.24 – 4450.39 ng/g). The highest Hg concentration in street dust was found in site H15 (4450.39 ng/g), extremely higher than in normal conditions (spring: 91.45 ng/g). Due to the high concentration of Hg in street dust in site H15, and this site is very closed to the CFL factory (only about 300 m), the source of Hg pollution after the CFL fire outbreak was identified to result from the CFL factory.

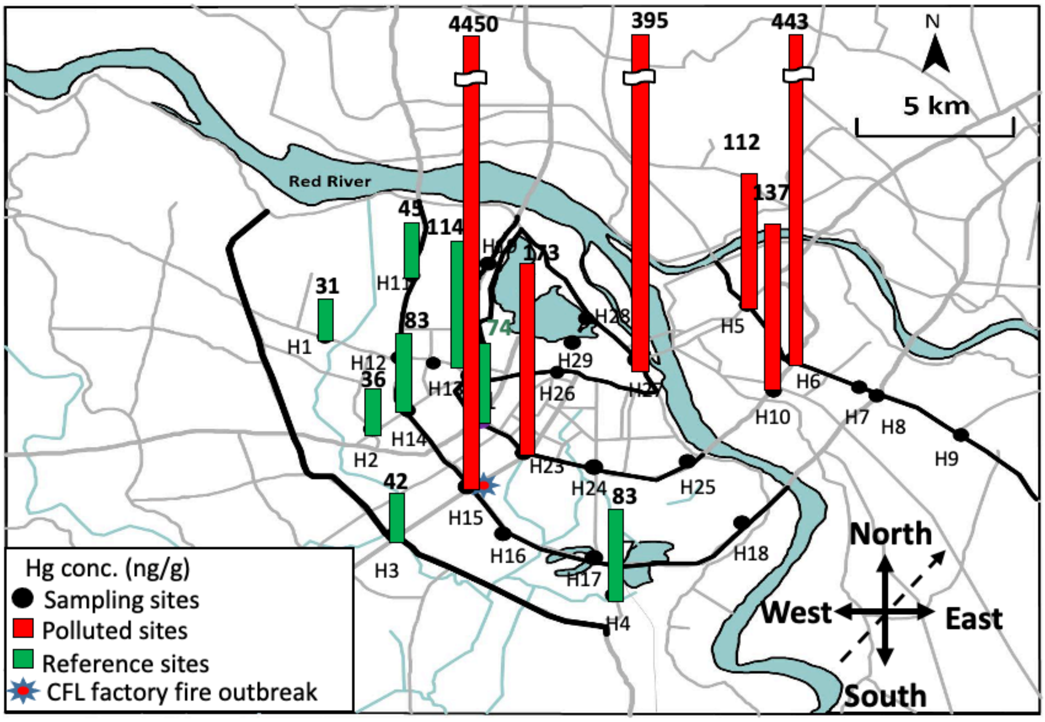

Figure 13. Hg concentration in street dust after the CFL factory fire outbreak incident.

In the overview, from the source of Hg emission, the concentration of Hg was found higher than the normal condition (during March 2019), such as site H5 – H8, site H10, site H15, site

H23, site H24, site H26 – H28. These sites were selected as typical sites effected by the CFL factory incident as polluted sites (PO sites). Besides, other sites that had similar concentrations of Hg with the normal condition during March 2019 included site H1 – H4, site H11 – site H14, site H21, and site H22 as reference sites (RF sites) (Figure 13).

From the results, high concentrations of Hg were found in polluted sites mainly located in the northwest direction from the CFL factory. These results probably resulted from the monsoon wind blows from southwest to northwest in autumn. In the north of Vietnam, where Hanoi is located, the southwest monsoon (summer monsoon) occurs in two halves i.e., (May – Jun) and (July – October). Both halves have the wind blowing in the same southwest direction (Nguyen-Le, Matsumoto and Ngo-Duc, 2014) (https://www.vietnamdrive.com/monsoon-seasons). This led the human who lives in this direction may have more effect of Hg than other directions such as north, south, and the west sides from the CFL factory.

The most worried is the CFL factory located inside the urban area, in a case study of Hanoi city – the capital of Vietnam, where has more than eight million people with a population density of 2398 people/km^2 (General Statistics Office, 2020). When the Hg incidents happen, it was difficult to prevent and respond to the environmental effect as well as protect the local human health. Following with the southwest monsoon direction after the CFL fire outbreak, there are many kindergartens and primary schools where have the most sensitive objectives as children (Figure 14).

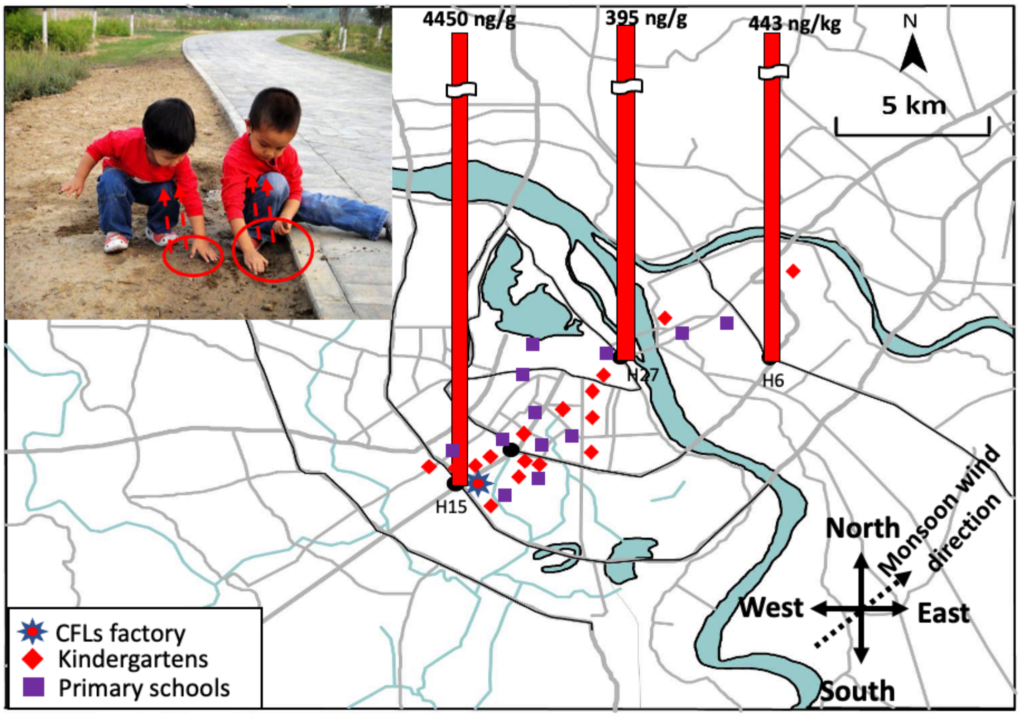

Figure 14. Kindergartens and primary schools following the monsoon wind direction.

From these results, it is shown that the authority agencies need to establish a special environmental incident control mechanism for facilities that manufacture and use Hg in the production of fluorescent lamps as well as other mercury-containing products. The relocation of the CFL factory away from densely populated areas should also be considered to protect the surrounding human health not only from mercury emission incidents such as fires and explosions but also other mercury leaks.

3.3.5. Human health risk assessment of Hg in street dust

3.3.5.1. The human health risk of Hg in street dust collected in Hanoi during March 2019

The results of human health risk assessment using hazard quotient and hazard index of Hg in street dust samples collected from Hanoi are shown in Table 7. The HQs of Hg in street dust from these areas are in the order of $HQ_{vap} > HQ_{ing} > HQ_{der} > HQ_{inh}$ for both children and adults. The HQ_{vap} values are the main effect for children and adults of the HI value. The results also found The HQ_{vap}, HQ_{ing}, and HQ_{inh} values of Hg in street dust for children were higher than adults; meanwhile, HQ_{der} values for adults were more elevated than children.

The human health risk of Hg in street dust from OR1, OR2, RR1, RR2, and RR3 showed that the HI values that summary of ingestion, dermal absorption, inhalation, and vapor pathways in RR1 was highest for both children and adults (3.29×10^{-2} and 1.59×10^{-2}, respectively), and was 2.35 to 3.11 time higher than other areas. These results indicated that humans who frequently live in RR1 might have more risk exposure and effect by Hg than other areas in Hanoi city. As the discussion above, the source of Hg in RR1 areas probably comes from the honeycomb coal using for cooking. Therefore, humans who often work with daily cooking jobs mainly adult women might have more health risk effect of Hg due to close and long-term exposure.

The human health risk assessment of Hg in street dust via ingestion was evaluated. HQ and HI values were below 1, indicates no significant human health effect by Hg in Hanoi city via ingestion, dermal absorption, inhalation, and vapor exposure pathways. However, the Hg was found in street dust could combine with other sources of Hg that need to more study in other sources such as atmospheric, food, and tobacco in these areas may increase the health effect to humans.

Table 7. Health risk assessment on children for Hg in street dusts in Hanoi city during spring.

Area	Site	Ci (µg/g)	Children					Adults				
			HQing	HQder	HQinh	HQvap	HI	HQing	HQder	HQinh	HQvap	HI
OR1	H1	0.05314	2.26×10^{-3}	7.13×10^{-5}	2.22×10^{-7}	9.22×10^{-3}	1.16×10^{-2}	2.43×10^{-4}	1.48×10^{-4}	1.25×10^{-7}	5.20×10^{-3}	5.59×10^{-3}
	H2	0.03912	1.67×10^{-3}	5.25×10^{-5}	1.63×10^{-7}	6.79×10^{-3}	8.51×10^{-3}	1.79×10^{-4}	1.09×10^{-4}	9.20×10^{-8}	3.83×10^{-3}	4.11×10^{-3}
	H3	0.07475	3.19×10^{-3}	1.00×10^{-4}	3.12×10^{-7}	1.30×10^{-2}	1.63×10^{-2}	3.41×10^{-4}	2.08×10^{-4}	1.76×10^{-7}	7.31×10^{-3}	7.86×10^{-3}
	H4	0.03659	1.56×10^{-3}	4.91×10^{-5}	1.53×10^{-7}	6.35×10^{-3}	7.96×10^{-3}	1.67×10^{-4}	1.02×10^{-4}	8.60×10^{-8}	3.58×10^{-3}	3.85×10^{-3}
	Average	**0.05090**	$\mathbf{2.17\times10^{-3}}$	$\mathbf{6.83\times10^{-5}}$	$\mathbf{2.12\times10^{-7}}$	$\mathbf{8.83\times10^{-3}}$	$\mathbf{1.11\times10^{-2}}$	$\mathbf{2.32\times10^{-4}}$	$\mathbf{1.42\times10^{-4}}$	$\mathbf{3.42\times10^{-8}}$	$\mathbf{4.98\times10^{-3}}$	$\mathbf{5.35\times10^{-3}}$
OR2	H5	0.03132	1.33×10^{-3}	4.20×10^{-5}	1.31×10^{-7}	5.43×10^{-3}	6.81×10^{-3}	1.43×10^{-4}	8.71×10^{-5}	7.36×10^{-8}	3.06×10^{-3}	3.29×10^{-3}
	H6	0.05358	2.28×10^{-3}	7.19×10^{-5}	2.23×10^{-7}	9.30×10^{-3}	1.17×10^{-2}	2.45×10^{-4}	1.49×10^{-4}	1.26×10^{-7}	5.24×10^{-3}	5.64×10^{-3}
	H7	0.04304	1.83×10^{-3}	5.78×10^{-5}	1.79×10^{-7}	7.47×10^{-3}	9.36×10^{-3}	1.97×10^{-4}	1.20×10^{-4}	1.01×10^{-7}	4.21×10^{-3}	4.53×10^{-3}
	H8	0.02730	1.16×10^{-3}	3.66×10^{-5}	1.14×10^{-7}	4.74×10^{-3}	5.94×10^{-3}	1.25×10^{-4}	7.59×10^{-5}	6.42×10^{-8}	2.67×10^{-3}	2.87×10^{-3}
	H9	0.06402	2.73×10^{-3}	8.59×10^{-5}	2.67×10^{-7}	1.11×10^{-2}	1.39×10^{-2}	2.92×10^{-4}	1.78×10^{-4}	1.50×10^{-7}	6.26×10^{-3}	6.73×10^{-3}
	H10	0.07196	3.07×10^{-3}	9.66×10^{-5}	3.00×10^{-7}	1.25×10^{-2}	1.56×10^{-2}	3.29×10^{-4}	2.00×10^{-4}	1.69×10^{-7}	7.04×10^{-3}	7.57×10^{-3}
	Average	**0.04854**	$\mathbf{2.07\times10^{-3}}$	$\mathbf{6.52\times10^{-5}}$	$\mathbf{1.83\times10^{-7}}$	$\mathbf{8.42\times10^{-3}}$	$\mathbf{1.06\times10^{-2}}$	$\mathbf{2.22\times10^{-4}}$	$\mathbf{1.35\times10^{-4}}$	$\mathbf{3.26\times10^{-8}}$	$\mathbf{4.75\times10^{-3}}$	$\mathbf{5.11\times10^{-3}}$
RR1	H26	0.20084	8.56×10^{-3}	2.70×10^{-4}	8.37×10^{-7}	3.48×10^{-2}	4.37×10^{-2}	9.17×10^{-4}	5.59×10^{-4}	4.72×10^{-7}	1.96×10^{-2}	2.11×10^{-2}
	H27	0.07111	3.03×10^{-3}	9.55×10^{-5}	2.96×10^{-7}	1.23×10^{-2}	1.55×10^{-2}	3.25×10^{-4}	1.98×10^{-4}	1.67×10^{-7}	6.96×10^{-3}	7.48×10^{-3}
	H28	0.13437	5.73×10^{-3}	1.80×10^{-4}	5.60×10^{-7}	2.33×10^{-2}	2.92×10^{-2}	6.14×10^{-4}	3.74×10^{-4}	3.16×10^{-7}	1.31×10^{-2}	1.41×10^{-2}
	H29	0.19934	8.50×10^{-3}	2.68×10^{-4}	8.31×10^{-7}	3.46×10^{-2}	4.33×10^{-2}	9.10×10^{-4}	5.54×10^{-4}	4.69×10^{-7}	1.95×10^{-2}	2.10×10^{-2}
	Average	**0.15142**	$\mathbf{6.45\times10^{-3}}$	$\mathbf{2.03\times10^{-4}}$	$\mathbf{6.31\times10^{-7}}$	$\mathbf{2.63\times10^{-2}}$	$\mathbf{3.29\times10^{-2}}$	$\mathbf{6.91\times10^{-4}}$	$\mathbf{4.21\times10^{-4}}$	$\mathbf{1.02\times10^{-7}}$	$\mathbf{1.48\times10^{-2}}$	$\mathbf{1.59\times10^{-2}}$
RR2	H19	0.04425	1.89×10^{-3}	5.94×10^{-5}	1.84×10^{-7}	7.68×10^{-3}	9.62×10^{-3}	2.02×10^{-4}	1.23×10^{-4}	1.04×10^{-7}	4.33×10^{-3}	4.65×10^{-3}
	H20	0.07326	3.12×10^{-3}	9.83×10^{-5}	3.05×10^{-7}	1.27×10^{-2}	1.59×10^{-2}	3.35×10^{-4}	2.04×10^{-4}	1.72×10^{-7}	7.17×10^{-3}	7.71×10^{-3}
	H21	0.04782	2.04×10^{-3}	6.42×10^{-5}	1.99×10^{-7}	8.30×10^{-3}	1.04×10^{-2}	2.18×10^{-4}	1.33×10^{-4}	1.12×10^{-7}	4.68×10^{-3}	5.03×10^{-3}
	H22	0.08366	3.57×10^{-3}	1.12×10^{-4}	3.49×10^{-7}	1.45×10^{-2}	1.82×10^{-2}	3.82×10^{-4}	2.33×10^{-4}	1.97×10^{-7}	8.19×10^{-3}	8.80×10^{-3}
	H23	0.06119	2.61×10^{-3}	8.21×10^{-5}	2.55×10^{-7}	1.06×10^{-2}	1.33×10^{-2}	2.79×10^{-4}	1.70×10^{-4}	1.44×10^{-7}	5.99×10^{-3}	6.44×10^{-3}
	H24	0.07267	3.10×10^{-3}	9.76×10^{-5}	3.03×10^{-7}	1.26×10^{-2}	1.58×10^{-2}	3.32×10^{-4}	2.02×10^{-4}	1.71×10^{-7}	7.11×10^{-3}	7.64×10^{-3}
	H25	0.06779	2.89×10^{-3}	9.10×10^{-5}	2.83×10^{-7}	1.18×10^{-2}	1.47×10^{-2}	3.10×10^{-4}	1.89×10^{-4}	1.59×10^{-7}	6.63×10^{-3}	7.13×10^{-3}
	Average	**0.06438**	$\mathbf{2.74\times10^{-3}}$	$\mathbf{8.64\times10^{-5}}$	$\mathbf{2.68\times10^{-7}}$	$\mathbf{1.12\times10^{-2}}$	$\mathbf{1.40\times10^{-2}}$	$\mathbf{2.94\times10^{-4}}$	$\mathbf{1.79\times10^{-4}}$	$\mathbf{4.32\times10^{-8}}$	$\mathbf{6.30\times10^{-3}}$	$\mathbf{6.77\times10^{-3}}$
RR3	H11	0.05235	2.23×10^{-3}	7.03×10^{-5}	2.18×10^{-7}	9.08×10^{-3}	1.14×10^{-2}	2.39×10^{-4}	1.46×10^{-4}	1.23×10^{-7}	5.12×10^{-3}	5.51×10^{-3}
	H12	0.02908	1.24×10^{-3}	3.90×10^{-5}	1.21×10^{-7}	5.05×10^{-3}	6.32×10^{-3}	1.33×10^{-4}	8.09×10^{-5}	6.84×10^{-8}	2.85×10^{-3}	3.06×10^{-3}
	H13	0.05240	2.23×10^{-3}	$7.03\times^{10-5}$	2.18×10^{-7}	9.09×10^{-3}	1.14×10^{-2}	2.39×10^{-4}	1.46×10^{-4}	1.23×10^{-7}	5.13×10^{-3}	5.51×10^{-3}
	H14	0.02559	1.09×10^{-3}	3.44×10^{-5}	1.07×10^{-7}	4.44×10^{-3}	5.56×10^{-3}	1.17×10^{-4}	7.12×10^{-5}	6.02×10^{-8}	2.50×10^{-3}	2.69×10^{-3}
	H15	0.09145	3.90×10^{-3}	1.23×10^{-4}	3.81×10^{-7}	1.59×10^{-2}	1.99×10^{-2}	4.18×10^{-4}	2.54×10^{-4}	2.15×10^{-7}	8.95×10^{-3}	9.62×10^{-3}
	H16	0.04777	2.04×10^{-3}	6.41×10^{-5}	1.99×10^{-7}	8.29×10^{-3}	1.04×10^{-2}	2.18×10^{-4}	1.33×10^{-4}	1.12×10^{-7}	4.67×10^{-3}	5.02×10^{-3}
	H17	0.03477	1.48×10^{-3}	4.67×10^{-5}	1.45×10^{-7}	6.03×10^{-3}	7.56×10^{-3}	1.59×10^{-4}	9.67×10^{-5}	8.17×10^{-8}	3.40×10^{-3}	3.66×10^{-3}
	H18	0.07155	3.05×10^{-3}	9.61×10^{-5}	2.98×10^{-7}	1.24×10^{-2}	1.56×10^{-2}	3.27×10^{-4}	1.99×10^{-4}	1.68×10^{-7}	7.00×10^{-3}	7.53×10^{-3}
	Average	**0.05062**	$\mathbf{2.16\times10^{-3}}$	$\mathbf{6.80\times10^{-5}}$	$\mathbf{2.21\times10^{-7}}$	$\mathbf{8.78\times10^{-3}}$	$\mathbf{1.10\times10^{-2}}$	$\mathbf{2.31\times10^{-4}}$	$\mathbf{1.41\times10^{-4}}$	$\mathbf{3.40\times10^{-8}}$	$\mathbf{4.95\times10^{-3}}$	$\mathbf{5.32\times10^{-3}}$

2.3.5.2. The human health risk of Hg in street dust after the CFL factory fire outbreak

The selected sites after the CFL factory fire outbreak include PO sites and RF sites for human health risk assessment. The results of the human health risk assessment of Hg in street dust from RF sites and PO sites were shown in Table 8 and Table 9, respectively.

The HI was a summation of the HQs for all the exposure routes. HI below 1 shows that there may not be any associated combined non-carcinogenic health risk of Hg while HI above 1 indicates the reverse (USEPA, 2001). Except for site H15 which had HI of 0.968 and 0.468 for children and adults, respectively, which is almost equal to 1, all other sites had HI below 1 for both children and adults. This showed that street dust from site H15 might have the potential to cause a combined non-carcinogenic human health risk to children compared to other sites. Site H15 consists of several elementary schools and kindergartens (Figure 14), which are located within the area of the CLF factory, hence the higher HI value for children. The higher HI values for children at site H15 may indicate many health problems which may be caused by exposure to Hg in street dust by inhalation, ingestion, and dermal contact. However, all other sites may not pose any combined non-carcinogenic human health risk of Hg to both children and adults since HI values were far below 1. Comparably, the PO sites had higher HI values for both children and adults than the RF sites. This showed that repeated or long-term exposure to street dust from the PO sites may have a higher potential of causing combined non-carcinogenic human health effects of Hg than street dust from the RF sites. Just like HQ, the HI values for children were higher than those for adults, an indication that children may experience more combined non-carcinogenic human health effects than adults. This may be due to the constant attachment of dust onto the skin of residents, especially children (Hu *et al.*, 2011) (Sun *et al.*, 2013) during playing hours. This result is consistent with that obtained by Meza-Figueroa *et al.*, 2007.

Table 8. Health risk assessment of Hg in street dusts for children and adults in Hanoi during autumn (RF sites).

		Average	Site H1	Site H2	Site H3	Site H4	Site H11	Site H12	Site H13	Site H14	Site H21	Site H22
	Ci	0.058812	0.03124	0.03672	0.04247	0.08299	0.04587	0.05211	0.04306	0.06504	0.11401	0.07461
Children	ADD_{ing}	7.52×10^{-7}	3.99×10^{-7}	4.69×10^{-7}	5.43×10^{-7}	1.06×10^{-6}	5.86×10^{-7}	6.66×10^{-7}	5.51×10^{-7}	8.32×10^{-7}	1.46×10^{-6}	9.54×10^{-7}
	ADD_{der}	1.58×10^{-9}	8.39×10^{-10}	9.86×10^{-10}	1.14×10^{-9}	2.23×10^{-9}	1.23×10^{-9}	1.40×10^{-9}	1.16×10^{-9}	1.75×10^{-9}	3.06×10^{-9}	2.00×10^{-9}
	ADD_{inh}	2.10×10^{-11}	1.12×10^{-11}	1.31×10^{-11}	1.52×10^{-11}	2.96×10^{-11}	1.64×10^{-11}	1.86×10^{-11}	1.54×10^{-11}	2.32×10^{-11}	4.07×10^{-11}	2.67×10^{-11}
	ADD_{vap}	8.75×10^{-7}	4.64×10^{-7}	5.46×10^{-7}	6.31×10^{-7}	1.23×10^{-6}	6.82×10^{-7}	7.75×10^{-7}	6.40×10^{-7}	9.67×10^{-7}	1.70×10^{-6}	1.11×10^{-6}
	HQ_{ing}	2.51×10^{-3}	1.33×10^{-3}	1.56×10^{-3}	1.81×10^{-3}	3.54×10^{-3}	1.95×10^{-3}	2.22×10^{-3}	1.84×10^{-3}	2.77×10^{-3}	4.86×10^{-3}	3.18×10^{-3}
	HQ_{der}	7.89×10^{-5}	4.19×10^{-5}	4.93×10^{-5}	5.70×10^{-5}	1.11×10^{-4}	6.16×10^{-5}	7.00×10^{-5}	5.78×10^{-5}	8.73×10^{-5}	1.53×10^{-4}	1.00×10^{-4}
	HQ_{inh}	2.45×10^{-7}	1.30×10^{-7}	1.53×10^{-7}	1.77×10^{-7}	3.46×10^{-7}	1.91×10^{-7}	2.17×10^{-7}	1.79×10^{-7}	2.71×10^{-7}	4.75×10^{-7}	3.11×10^{-7}
	HQ_{vap}	1.02×10^{-2}	5.42×10^{-3}	6.37×10^{-3}	7.37×10^{-3}	1.44×10^{-2}	7.96×10^{-3}	9.04×10^{-3}	7.47×10^{-3}	1.13×10^{-2}	1.98×10^{-2}	1.29×10^{-2}
	HI	1.28×10^{-2}	6.79×10^{-3}	7.99×10^{-3}	9.24×10^{-3}	1.80×10^{-2}	9.97×10^{-3}	1.13×10^{-2}	9.36×10^{-3}	1.41×10^{-2}	2.48×10^{-2}	1.62×10^{-2}
Adults	ADD_{ing}	8.06×10^{-8}	4.28×10^{-8}	5.03×10^{-8}	5.82×10^{-8}	1.14×10^{-7}	6.28×10^{-8}	7.14×10^{-8}	5.90×10^{-8}	8.91×10^{-8}	1.56×10^{-7}	1.02×10^{-7}
	ADD_{der}	3.27×10^{-9}	1.74×10^{-9}	2.04×10^{-9}	2.36×10^{-9}	4.62×10^{-9}	2.55×10^{-9}	2.90×10^{-9}	2.39×10^{-9}	3.62×10^{-9}	6.34×10^{-9}	4.15×10^{-9}
	ADD_{inh}	1.18×10^{-11}	6.29×10^{-12}	7.40×10^{-12}	8.56×10^{-12}	1.67×10^{-11}	9.24×10^{-12}	1.05×10^{-11}	8.67×10^{-12}	1.31×10^{-11}	2.30×10^{-11}	1.50×10^{-11}
	ADD_{vap}	4.93×10^{-7}	2.62×10^{-7}	3.08×10^{-7}	3.56×10^{-7}	6.96×10^{-7}	3.85×10^{-7}	4.37×10^{-7}	3.61×10^{-7}	5.45×10^{-7}	9.56×10^{-7}	6.26×10^{-7}
	HQ_{ing}	2.69×10^{-4}	1.43×10^{-4}	1.68×10^{-4}	1.94×10^{-4}	3.79×10^{-4}	2.09×10-^{4}	2.38×10^{-4}	1.97×10^{-4}	2.97×10^{-4}	5.21×10^{-4}	3.41×10^{-4}
	HQ_{der}	1.64×10^{-4}	8.69×10^{-5}	1.02×10^{-4}	1.18×10^{-4}	2.31×10^{-4}	1.28×10^{-4}	1.45×10^{-4}	1.20×10^{-4}	1.81×10^{-4}	3.17×10^{-4}	2.07×10^{-4}
	HQ_{inh}	1.38×10^{-7}	7.34×10^{-8}	8.63×10^{-8}	9.98×10^{-8}	1.95×10^{-7}	1.08×10^{-7}	1.22×10^{-7}	1.01×10^{-7}	1.53×10^{-7}	2.68×10^{-7}	1.75×10^{-7}
	HQ_{vap}	5.76×10^{-3}	3.06×10^{-3}	3.59×10^{-3}	4.16×10^{-3}	8.12×10^{-3}	4.49×10^{-3}	5.10×10^{-3}	4.21×10^{-3}	6.36×10^{-3}	1.12×10^{-2}	7.30×10^{-3}
	HI	6.19×10^{-3}	3.29×10^{-3}	3.86×10^{-3}	4.47×10^{-3}	8.73×10^{-3}	4.82×10^{-3}	5.48×10^{-3}	4.53×10^{-3}	6.84×10^{-3}	1.20×10^{-2}	7.85×10^{-3}

Table 9. Health risk assessment of Hg in streets dusts for children and adults in Hanoi during autumn (PO sites).

		Average	Site H5	Site H6	Site H7	Site H8	Site H10	Site H15	Site H23	Site H24	Site H26	Site H27	Site H28	Site H29
	Ci	0.54023	0.11197	0.44338	0.08385	0.13175	0.13787	4.45039	0.17289	0.13769	0.19295	0.39567	0.10241	0.12198
Children	ADD_{ing}	6.91×10^{-6}	1.43×10^{-6}	5.67×10^{-6}	1.07×10^{-6}	1.68×10^{-6}	1.76×10^{-6}	5.69×10^{-5}	2.21×10^{-6}	1.76×10^{-6}	2.47×10^{-6}	5.06×10^{-6}	1.31×10^{-6}	1.56×10^{-6}
	ADD_{der}	1.45×10^{-8}	3.01×10^{-9}	1.19×10^{-8}	2.25×10^{-9}	3.54×10^{-9}	3.70×10^{-9}	1.19×10^{-7}	4.64×10^{-9}	3.70×10^{-9}	5.18×10^{-9}	1.06×10^{-8}	2.75×10^{-9}	3.28×10^{-9}
	ADD_{inh}	1.93×10^{-10}	4.00×10^{-11}	1.58×10^{-10}	3.00×10^{-11}	4.71×10^{-11}	4.93×10^{-11}	1.59×10^{-9}	6.18×10^{-11}	4.92×10^{-11}	6.89×10^{-11}	1.41×10^{-10}	3.66×10^{-11}	4.36×10^{-11}
	ADD_{vap}	8.03×10^{-6}	1.66×10^{-6}	6.59×10^{-6}	1.25×10^{-6}	1.96×10^{-6}	2.05×10^{-6}	6.62×10^{-5}	2.57×10^{-6}	2.05×10^{-6}	2.87×10^{-6}	5.88×10^{-6}	1.52×10^{-6}	1.81×10^{-6}
	HQ_{ing}	2.31×10^{-2}	4.77×10^{-3}	1.89×10^{-2}	3.57×10^{-3}	5.61×10^{-3}	5.88×10^{-3}	1.90×10^{-1}	7.37×10^{-3}	5.87×10^{-3}	8.22×10^{-3}	1.69×10^{-2}	4.36×10^{-3}	5.20×10^{-3}
	HQ_{der}	7.25×10^{-4}	1.50×10^{-4}	5.95×10^{-4}	1.13×10^{-4}	1.77×10^{-4}	1.85×10^{-4}	5.97×10^{-3}	2.32×10^{-4}	1.85×10^{-4}	2.59×10^{-4}	5.31×10^{-4}	1.37×10^{-4}	1.64×10^{-4}
	HQ_{inh}	2.26×10^{-6}	4.67×10^{-7}	1.85×10^{-6}	3.50×10^{-7}	5.49×10^{-7}	5.75×10^{-7}	1.86×10^{-5}	7.21×10^{-7}	5.74×10^{-7}	8.04×10^{-7}	1.65×10^{-6}	4.27×10^{-7}	5.08×10^{-7}
	HQ_{vap}	9.37×10^{-2}	1.94×10^{-2}	7.69×10^{-2}	1.45×10^{-2}	2.29×10^{-2}	2.39×10^{-2}	7.72×10^{-1}	3.00×10^{-2}	2.39×10^{-2}	3.35×10^{-2}	6.86×10^{-2}	1.78×10^{-2}	2.12×10^{-2}
	HI	1.17×10^{-1}	2.43×10^{-2}	9.64×10^{-2}	1.82×10^{-2}	2.87×10^{-2}	3.00×10^{-2}	**9.68×10^{-1}**	3.76×10^{-2}	2.99×10^{-2}	4.20×10^{-2}	8.60×10^{-2}	2.23×10^{-2}	2.65×10^{-2}
Adults	ADD_{ing}	7.40×10^{-7}	1.53×10^{-7}	6.07×10^{-7}	1.15×10^{-7}	1.80×10^{-7}	1.89×10^{-7}	6.10×10^{-6}	2.37×10^{-7}	1.89×10^{-7}	2.64×10^{-7}	5.42×10^{-7}	1.40×10^{-7}	1.67×10^{-7}
	ADD_{der}	3.01×10^{-8}	6.23×10^{-9}	2.47×10^{-8}	4.66×10^{-9}	7.33×10^{-9}	7.67×10^{-9}	2.48×10^{-7}	9.62×10^{-9}	7.66×10^{-9}	1.07×10^{-8}	2.20×10^{-8}	5.70×10^{-9}	6.78×10^{-9}
	ADD_{inh}	1.09×10^{-10}	2.26×10^{-11}	8.93×10^{-11}	1.69×10^{-11}	2.65×10^{-11}	2.78×10^{-11}	8.97×10^{-10}	3.48×10^{-11}	2.77×10^{-11}	3.89×10^{-11}	7.97×10^{-11}	2.06×10^{-11}	2.46×10^{-11}
	ADD_{vap}	4.53×10^{-6}	9.39×10^{-7}	3.72×10^{-6}	7.03×10^{-7}	1.10×10^{-6}	1.16×10^{-6}	3.73×10^{-5}	1.45×10^{-6}	1.15×10^{-6}	1.62×10^{-6}	3.32×10^{-6}	8.59×10^{-7}	1.02×10^{-6}
	HQ_{ing}	2.47×10^{-3}	5.11×10^{-4}	2.02×10^{-3}	3.83×10^{-4}	6.02×10^{-4}	6.30×10^{-4}	2.03×10^{-2}	7.89×10^{-4}	6.29×10^{-4}	8.81×10^{-4}	1.81×10^{-3}	4.68×10^{-4}	5.57×10^{-4}
	HQ_{der}	1.50×10^{-3}	3.11×10^{-4}	1.23×10^{-3}	2.33×10^{-4}	3.66×10^{-4}	3.83×10^{-4}	1.24×10^{-2}	4.81×10^{-4}	3.83×10^{-4}	5.37×10^{-4}	1.10×10^{-3}	2.85×10^{-4}	3.39×10^{-4}
	HQ_{inh}	1.27×10^{-6}	2.63×10^{-7}	1.04×10^{-6}	1.97×10^{-7}	3.10×10^{-7}	3.24×10^{-7}	1.05×10^{-5}	4.06×10^{-7}	3.24×10^{-7}	4.54×10^{-7}	9.30×10^{-7}	2.41×10^{-7}	2.87×10^{-7}
	HQ_{vap}	5.28×10^{-2}	1.10×10^{-2}	4.34×10^{-2}	8.20×10^{-3}	1.29×10^{-2}	1.35×10^{-2}	4.35×10^{-1}	1.69×10^{-2}	1.35×10^{-2}	1.89×10^{-2}	3.87×10^{-2}	1.00×10^{-2}	1.19×10^{-2}
	HI	5.68×10^{-2}	1.18×10^{-2}	4.66×10^{-2}	8.82×10^{-3}	1.39×10^{-2}	1.45×10^{-2}	**4.68×10^{-1}**	1.82×10^{-2}	1.45×10^{-2}	2.03×10^{-2}	4.16×10^{-2}	1.08×10^{-2}	1.28×10^{-2}

Ci (mg/kg), ADD (mg/kg/day),

$ADD_{ing} = \frac{C_i \times IngR \times EF \times ED}{BW \times AT} \times 10^{-6}$, $ADD_{der} = \frac{C_i \times SA \times SL \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6}$, $ADD_{inh} = \frac{C_i \times InhR \times EF \times ED}{BW \times AT \times PEF}$, $ADD_{Vap} = \frac{C_i \times InhR \times EF \times ED}{VF \times BW \times AT}$, $HQ = \frac{ADD_{ing/der/inh/vap}}{RfD_{ing/der/inh/vap}}$, $HI = HQ_{ing} + HQ_{der} + HQ_{inh} + HQ_{vap}$

The higher risk of Hg to children meant that the health of the children living in and around the vicinity of the CFL factory should be regularly monitored. The total health risk by CFL incident may be required to additionally monitor other sources such as emission gas, drinking water, beverage, farm products, and foods in Hanoi city. Street dust, which is the medium used in this health risk assessment of CFL incident, the combined Hg atmospheric pollutant can enter humans via inhalation of Hg vapor such as tobacco smoke, exhaust, and emission gas from several point-sources. Furthermore, the combined health risk by metals and chemicals might occur through the CFL incident.

3.4. Conclusion

This is the first study of Hg in street dust that was used to determine the pollution characteristic and identify the source of Hg in Hanoi. The street dust could be used to assess the pollution level, pollution sources identified, and human health risk assessment. In Hanoi, a significantly higher concentration was found in the center resulted in the pollution sources of Hg in these areas were identified. In central Hanoi (RR1), Hg discharged from honeycomb coal use for cooking leads to the health risk of adult women because common in cooking work. Since the case of the CFL factory fire outbreak, the wind blows Hg to downstream areas and deposited Hg in street dust with high Hg content resulted in the Hg pollution and human health risk effect, especially children near the CFL factory and the downstream of the wind. From this result, during the spring, Hg from the source was found higher in the central Hanoi (RR1) and seemed higher in the site located in the southwest direction, such as H22, H23, H24, H15, and H3 as typical sites. It may result from the northeast monsoon wind during the winter and spring, probably blow Hg from central Hanoi to these areas. Extend to larger areas of northern Vietnam, Hg emission from coal combustion areas mainly focus on Quang Ninh province (Chapter 2) could transfer to the inside land of Vietnam during the northeast monsoon wind and have risk effect to humans and living environment.

CHAPTER 4

Mercury in cigarettes collected from markets in Vietnam: The concentration, distribution, absorption ability of filter and human health risk assessment

4.1. Introduction

Tobacco has been used in the world for thousands of years by humans. Tobacco and its smoke contain more than 4000 different substances, including various carcinogens and toxic metals (Stration *et al.*, 2001). As a crop, tobacco is considered an indicator of environmental pollution. Air pollutants damage food crops' yield and nutritional quality and safety, imposing a major risk to food safety. In addition to this, the soil parent material is the most important influencing factor of heavy metal concentrations, and human activities are the main external sources of heavy metals (Wu *et al.*, 2020). Around 6.5 trillion cigarettes are sold worldwide each year, which translates to roughly 18 billion cigarettes per day (Martin, 2019). Furthermore, tobacco kills more than 8 million people each year. More than 7 million deaths result from direct tobacco use, while around 1.2 million results from non-smokers being exposed to secondhand smoke (World Health Organization, 2020). The research data on mercury (Hg) in the cigarette is still limited; meanwhile, the effect of Hg on human health has been known to mainly affect the nervous system, seriously affecting the fetus if exposed to mercury from the mother. Therefore, a study investigating Hg concentrations in commercial cigarettes is needed. These data will contribute to the management and prevention of tobacco-related harms to the community and human health.

Recently, several studies on heavy metals in tobacco have been implemented. (Yurdakok, 2015) recommended that research on Hg in breast milk should aim to reduce life-long exposure through precautionary measures such as prevention of exposure to cigarette smoke, use of unleaded gasoline, and prevention of air pollution with an effect at the community level. Artisanal and small-scale gold mining is known to be the major anthropogenic source of Hg pollution globally due to the direct use of Hg in the gold mining processes (Norvisa *et al.*, 2019; Addai-Arhin *et al.*, 2021). Besides, the improper management of factories or equipment containing mercury also leads to Hg leakage into the environment that is toxic to humans and the ecosystem (Bai, 2017) (United Nations Environment, 2019). Hg is volatile even at room temperature, making it a global pollutant. Kowalski and Wiercinski, 2009 reported that Hg was found in tobacco cigarettes and ranged from 2.95 – 10.2 ng Hg per single cigarette. Almost all mercury contained was released into the smoke (from 86.7 to 100%). However, they used a "mechanical lip" model to determine the mercury concentration in cigarette smoke. Airflow interruption by balancing the pump valve connected to the pump syringe leads Hg separately to mainstream and side-stream smoke. Therefore, the absorption of mercury by the filter had not been fully evaluated. Hg analysis in cigarette smoke is still hampered by the loss of smoke flow and smoking process descriptors' entanglement. Tobacco smoke analysis is also susceptible to cross-contamination of samples when conducting experiments. Therefore, this affects test results and is difficult to perform on many samples.

Thus, the aims of this study were to first investigate concentrations of Hg in marketed cigarettes collected from retail sale stores in Vietnam, and second, analyze the distribution concentrations of Hg in parts of the cigarette (cigarette filter, tobacco, and rolling paper), the absorption ability of Hg and from tobacco after smoking by filter materials was observed. And finally, the human health risk of Hg in cigarettes was estimation.

4.2. Materials and methods

4.2.1. Sampling and pretreatment

Marketed cigarettes were collected nine cigarette brands from convenience stores in Vietnam through 2019 (**Table 10**). In the other hand, we also collected cigarettes from other countries in the region with Vietnam for the comparison of Hg content in cigarettes include Asian countries (Korea: n=10, Japan: n=10, Indonesia: n=13, Thailand: n=4, and Taiwan: n=6) and European countries (Belgium: n=3, France: n=3, Italy: n=3, Finland: n=3, and the UK: n=3).

In other countries, the samples were collected from different brands of cigarettes. There were several cigarette brands of the same brand but that were collected from different countries. However, information from the cigarette labels indicated that the manufacturers were in the cigarette home country. Cigarette samples were dried at room temperature for two days, then homogenized using mortar and pestle. The homogenized cigarette samples were packaged in polypropylene tubes and kept at 4^{o}C in a refrigerator until analysis.

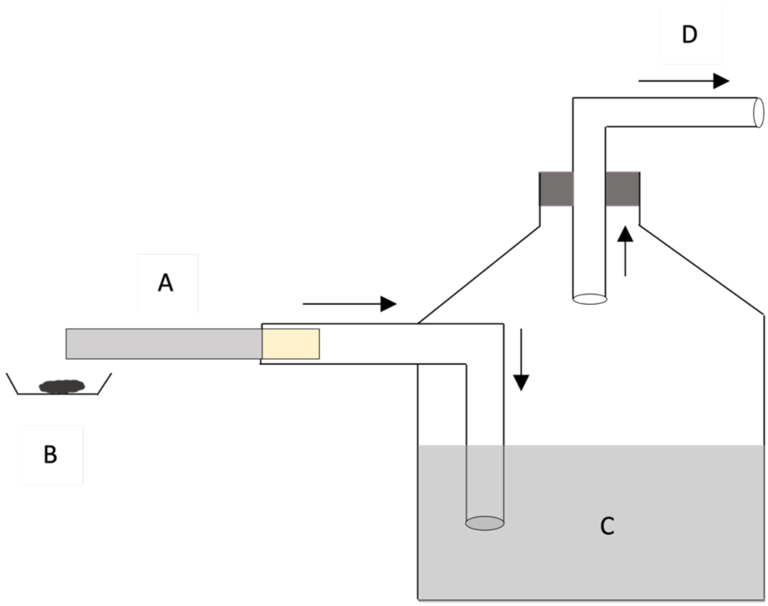

Figure 15. Model of smoking. A - cigarette, B - sample boat for ash accumulation, C – water scrubber, D – pump.

To comprehend Hg in parts of a cigarette after smoking, we conducted an analysis of these heavy metals in filters and the ash after smoking, using a physical model of smoking (**Figure 15**). In this case, a rubber tube was connected to the filter of the cigarette. The pump was run non-stop, the tobacco was burned until finished to keep all the smoke through the filter. Therefore, this experiment conducted a survey only in the mainstream smoke to estimate tobacco's transfer abilities to smoke.

4.2.2. Mercury analysis methods

Approximately 50 mg sample was weighed using an analytical/electronic balance (Practum124-1SJP, Sartorius, Göttingen, Germany). Hg was determined using pyrolysis atomic absorption spectrometry with gold amalgamation by a model of mercury analyzer 3000 (Nippon Instrument Corporation, Tokyo, Japan). Hg was determined at a wavelength of 253.7 nm by thermal combustion. The oxygen gas was purchased from Kumamoto Sanso Corporation (Kumamoto, Japan), and the flowing gas was set at 0.2 L/min. Two calibration curves were prepared using a Hg standard solution (1000 mg/L, Wako Pure Chemical Company, Osaka, Japan), to calculate Hg concentration in samples. The low calibration curves were prepared at six points of 0.2, 0.5, 1, 5, 10, and 20 ng Hg, and the high calibration curves were prepared at six points of 10, 20, 50, 100, 200, and 500 ng of Hg. The calibration curves were described based on the real Hg concentration of analysis with the area of absorbance (ABS) from the atomic absorption spectroscopy of a mercury analyzer (MA 3000, NIC, Tokyo, Japan). The correlation coefficients were 0.9999 for both calibration curves at high and low Hg concentrations. The method limit of detection (MDL) was calculated using the formula: 3SD of a blank sample (n = 20), while the method limit of quantification (MQL) was calculated using 10SD. The MDL and MQL values for the Hg analysis method in cigarettes were informed at 0.03 ng/g and 0.09 ng/g, respectively.

4.2.3. Human health risk assessment

To understand the health risk assessment of Hg, a health risk assessment model was used to calculate their health risk to humans. Freitas *et al.*, (2011), Benson *et al.*, (2017), and Ismail *et al.*, (2017) and applied the US EPA model for inhalation route to estimate the human health risk of heavy metals in cigarettes. These reports which estimated the human health risk of heavy metals in cigarettes used their concentrations in cigarettes based on the unit of tobacco leaf (mg/g cigarette). In this study, we modified the human health risk of Hg using the concentrations of Hg that were estimated in the entire cigarette (mg/cigarette: tobacco leaf, rolling paper, and filter).

Based on a study by Rodgman and Green, (2003), the average daily concentration (ADC: μg/m^3), non-cancer health effects (HI) were evaluated using formulas (1) to (2).

$$\text{ADC } (\mu g/m^3) = \frac{C_m(\mu\text{g/cigarette})\times \text{SF (cigarette/day)}}{VB\ (m^3\ of\ air\ breathed/day)} \tag{1}$$

Where C_m (μg/cigarette) is the Hg contents; C_m was derived from the total content of Hg per cigarette (tobacco leaf, roll paper, and filter). SF (cigarettes/day) is the daily smoking frequency (packed-a-day smoker = 20 cigarettes/day), and VB (m^3/day) is the daily volume of air breathed by adults (20 m^3/day).

The potential non-cancer health effect (HI) was evaluated by comparing the ADC to non-cancer risks for the inhalation route of exposure Hg using the reference concentration (RfC = 3.0×10^{-4} mg/m^3).

$$\text{HI} = \text{ADC } (\mu g/m^3 \times mg/1000) \times \text{RfC } (mg/m^3) \tag{2}$$

An HI value ≥ 1 suggests a potential for adverse health effects, while an HI value < 1 suggests no health effects (Vorhees *et al.*, 1997).

4.3. Results and discussion

4.3.1. Cigarette properties

The filtered cigarette is made from three main parts: cigarette filter, roll paper, and tobacco. According to our investigation, in the markets, some brands did not have a filter. However, all collected samples contained a filter. There was an average 0.829 ±0.129 g/cigarette (tobacco leaf: 0.633 ±0.098 g/cigarette; filter: 0.158 ±0.050 g/cigarette; and roll paper: 0.038 ±0.006 g/cigarette). Tobacco accounted for 76.36% of the total weight of the whole cigarette on average. Filters and rolling paper accounted for 19.06% and 4.58% of one filtered cigarette's total weight, respectively.

4.3.2. The distribution of mercury in cigarette

Based on the experiment layout, after smoking, the cigarette left ash, smoke, and filter. Thus, we supposed that Hg from tobacco leaves and rolling paper go to three pathways: to be trapped by filter, to remain in the ash, and to transfer to smoke. Therefore, we estimated the ratio of Hg from one cigarette to smoke after smoking.

The distribution of Hg concentrations in parts of cigarettes is shown that the mean concentrations of Hg in tobacco of cigarette samples collected from Vietnam were 27.23 ±7.67 ng/g. The filter contained mean concentrations of 0.13 ±0.13 ng/g of Hg. For rolling paper, Hg was not detected (< MDL). The results showed that Hg contents in cigarettes are mostly contained in tobacco leaf (99.84 %). As discussed above, tobacco accounted for an average of 76.36% of the total weight of the whole cigarette, indicating that these heavy metals in cigarettes mainly come from tobacco leaf. In addition, tobacco is made from the leaf of the tobacco plant (*Nicotiana tabacum*). Hg may absorb from the soil by the plant's roots and transport to the leaf. However, the percentage concentrations of Hg in cigarette filters were relatively lower. Toxic heavy metals are the major source of environmental pollution in this new millennium. Hg is the most common toxic heavy metal in the environment (Yurdakok, 2015). This explains the presence of Hg in cigarette rolling papers and filters at low concentrations.

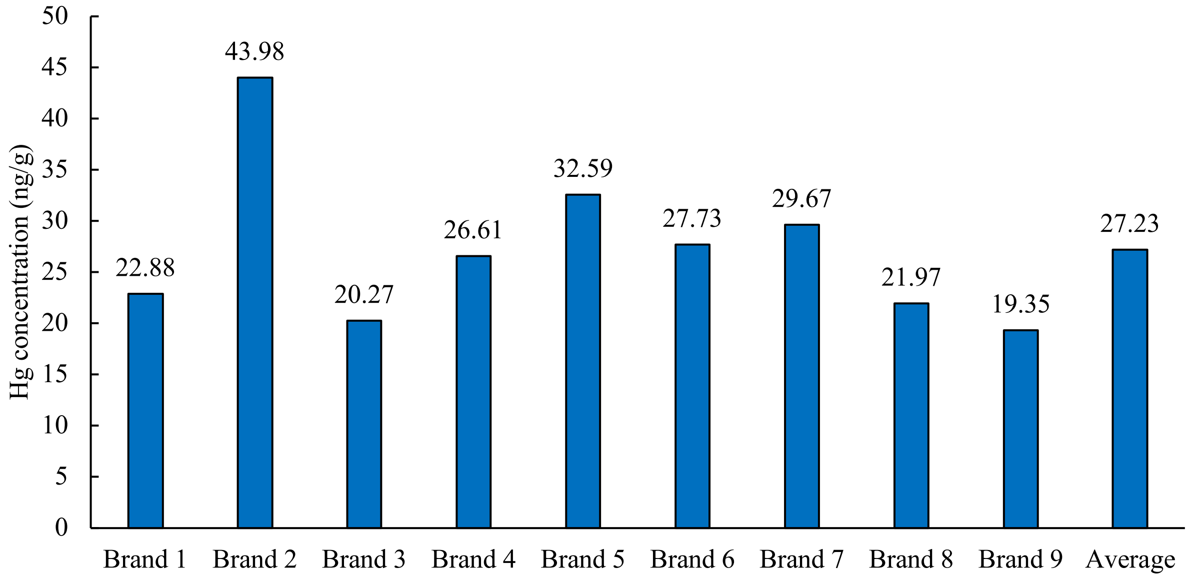

Figure 16. The concentration of Hg in cigarette collected from Vietnam.

As discussed above, Hg was mostly contained in tobacco at different concentrations. The concentrations of Hg in tobacco are shown in **Figure 16**. The results showed that Hg concentrations in tobacco cigarettes from these countries are non-uniformly distributed. The highest total Hg concentration in tobacco was found in cigarettes from Vietnam (average: 27.23 ±6.69 ng/g) that almost similar to cigarettes from Thailand (average: 27.80 ±1.06 ng/g), and Belgium (average: 23.56 ±6.88 ng/g). Hg concentrations in tobacco of cigarettes from these countries were higher than those of Korea (average: 20.89 ±3.48 ng/g), Finland (average: 19.66 ±1.58 ng/g), Japan (average: 18.38 ±2.6 ng/g), France (average: 18.01 ±1.68 ng/g), and the UK (average: 17.72 ±1.92 ng/g) who also had similar Hg concentrations in tobacco of cigarettes. Meanwhile, the Hg in tobacco of cigarettes from Indonesia (average: 14.19 ±2.77 ng/g) and Italy (12.37 ±0.81 ng/g) recorded the lowest concentrations (**Figure 17**). In this study, the source of tobacco raw material was not investigated. However, it is supposed that the tobacco material from different countries has different Hg pollution levels in the environment, such as in the soil, in drainage water, in fertilizer used for tobacco cultivation. Regardless of the different concentrations of these metals, their cigarette content is remarkable because these heavy metals in the cigarettes have negative health effects on both primary and secondary smokers following continuous exposure.

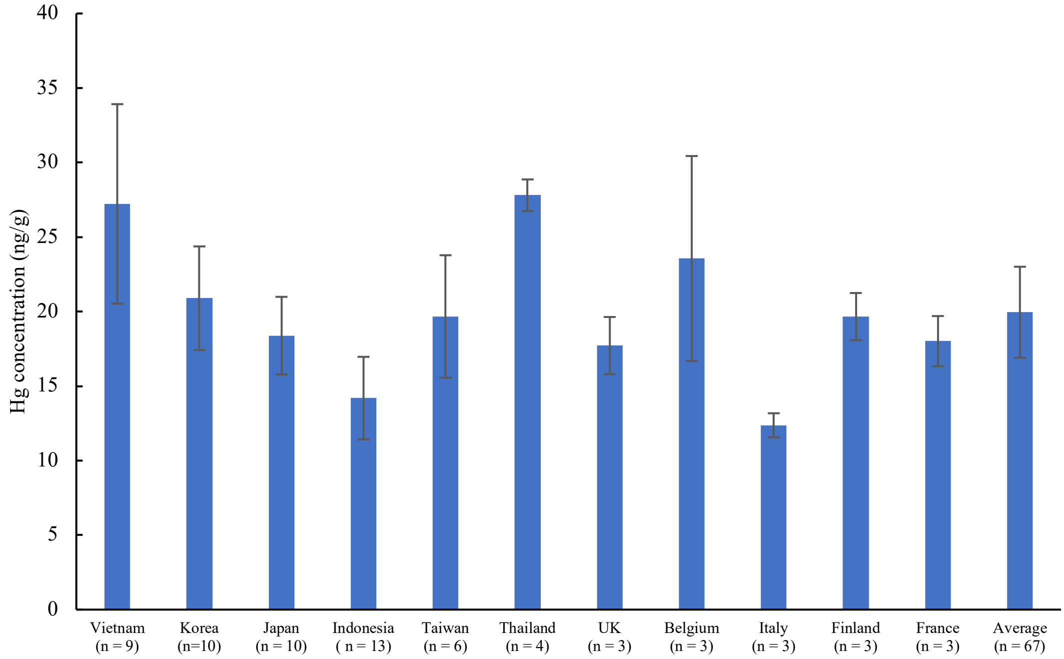

Figure 17. The average Hg concentration in tobacco of cigarette from Vietnam and several other countries.

Based on Hg concentration in parts of cigarettes, the amount of Hg per one cigarette was also evaluated (**Figure 18**). The results showed that the average content of Hg per one cigarette was 15.28 ±2.77 ng/cigarette. It was realized that the higher the concentration of Hg in tobacco of cigarettes and the higher the weight of the cigarette, the greater the concentration of the Hg content per one cigarette. All the cigarettes contained an amount of tobacco below one gram, the reason that Hg levels in a cigarette are lower than their respective evaluated concentrations. The weight of the cigarette was indicated by the manufacturer on the package label. Generally, the cigarettes from Indonesia had the highest weight of tobacco (897.40 mg/cigarette). Also, the Hg concentrations in tobacco and per one cigarette were the same trends that reflected the distribution of these heavy metals mostly in tobacco of cigarettes.

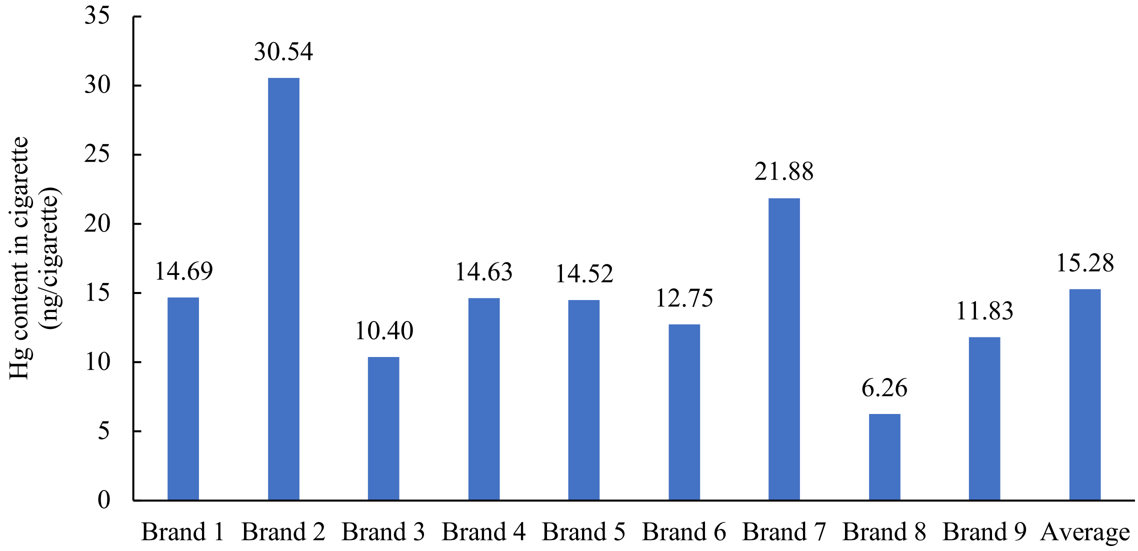

Figure 18. The content of Hg in cigarette (ng/cigarette) collected from Vietnam.

Comparison of Hg concentration in the cigarettes of the present study with other studies is shown in **Table 11**. The data of Hg in cigarettes is still limited, there were few studies of Hg in marketed cigarettes from some previous studies in the United States of America (Panta *et al.*, 2008) (Panta *et al.,* 2008; Swani *et al.,* 2009; Fresquez *et al.,* 2015), Canada (Hammond and O'Connor, 2008), and Poland (Kowalski and Wiercinski, 2019) showed that the Hg concentrations in cigarettes from these studies are almost equal to those of the present study.

Table 10. Hg content in tobacco of marketed cigarettes from the different countries.

Country	Hg concentration (ng/g)			References
	Mean ±SD	Min	Max	
Vietnam	27.23 ±6.69	19.35	43.98	This study
Korea	20.89 ±3.48	16.74	28.31	This study
Japan	18.38 ±2.60	15.47	22.87	This study
Indonesia	14.19 ±2.77	10.32	18.97	This study
Taiwan	19.67 ±4.11	16.57	27.93	This study
Thailand	27.80 ±1.06	26.38	28.92	This study
UK	17.72 ±1.92	15.60	19.35	This study
Belgium	23.56 ±6.88	15.81	28.94	This study
Italy	12.37 ±0.81	11.57	13.18	This study
Finland	19.66 ±1.58	17.84	20.66	This study
France	18.01 ±1.68	16.66	19.90	This study
USA	-	17.9	24.9	(Fresquez *et al.*, 2015)
Canada	26.8	-	-	(Hammond and O'Connor, 2008)
USA	13.0 ±1.3	-	-	(Panta *et al.*, 2008)
USA	-	20	21	(Swani, Juud and Orsini, 2009)
Poland	-	6.74	10.56	(Kowalski and Wiercinski, 2009)

(-) data not available.

4.3.3. The absorption ability of Hg by cigarette filter

In this study, average Hg concentrations in the filter were found at 0.26 ±0.24 ng/g cigarette. The results showed that the concentrations of Hg in filters after smoking were higher than before smoking, indicating that the filter of cigarettes had abilities to absorb Hg from the smoking process.

Basically, the filter of the cigarettes is made of cellulose acetate plastic (Ce). In recently, there are several brands that add activated carbon to the cigarette filter (Ce-C). In this study, the cigarette samples collected from Vietnam, there was only one cigarette sample filter made from Ce-C, while eight remain cigarette samples filter was made from Ce. Therefore, we evaluated the difference of trapping ability between Ce filters (15) and Ce-C filters (14) using 29 cigarettes (Ce: Japan 5, Korea 2, and Vietnam 8; Ce-C: Japan 5, Korea 8, and Vietnam 1). The results of the comparison of adsorption (trapping) ability of Hg by the cigarette filter are indicated in **Table 12**. Before smoking, the Hg content in Ce-C (Hg 0.044 ±0.017 ng/cigarette), was significantly higher than that of Ce (Hg 0.024 ±0.013 ng/cigarette), indicate Hg probably potential in active carbon that made from charcoal in Ce-C filter materials.

After smoking, the Hg content of both Ce (Hg 0.044 ±0.029 ng/cigarette), and Ce-C (Hg 0.053 ±0.023 ng/cigarette, was found to be significantly higher compared with before smoking. The Hg content in the Ce filter after smoking was significantly higher than before smoking. However, it was no significant difference with Ce-C filters. The difference of Hg content between Ce and Ce-C after smoking was identified as Hg 0.026 and 0.003 ng/cigarette. However, the levels of Hg in whole cigarettes were evaluated as Hg was 15.28 ng/cigarette, the ratio of trapped metals by the filter was estimated as Hg 0.07%. On the other hand, Hg had lower absorption in both types of filters (Ce: 0.15%, Ce-C: 0.02%). Even though Ce has a relatively higher trapping ability compared with Ce-C, both filters did not have enough trapping ability for Hg.

Table 11. Hg content in filter and the differences between the means of two groups.

Filter	Smoking process	Hg (ng/cigarette)
Ce	A: Before	0.024 ±0.013
	C: After	0.050 ±0.017#
Ce-C	B: Before	0.050 ±0.017#
	D: After	0.053 ±0.023

Ce: Filter that was made by cellulose acetate plastic only. Ce-C: Filter that was made by cellulose acetate plastic added activated carbon. Data were represented mean ±SD.
*: $p < 0.01$ (A vs C, B vs D), #: $p < 0.01$(A vs B, C vs D).

However, the results showed the Ce filter was higher trapping than Ce-C filter just in the case of Hg in this study. It was reported that Ce-C filter was more effected trapping of other toxic substant such as Pb, Cd compared to Ce filter (Dinh *et al.*, 2021), although the trapping abilities of these metals was far lower than the total metals content in whole cigarette by both Ce and Ce-C filters.

As far as the authors know, this experiment was the first of its kind to be performed regarding Hg. The experiment showed that Hg was not found in ash. Therefore, almost Hg from tobacco and rolling paper also flowed with smoke when smoking. When a cigarette is smoked, the temperature in the cigarette's tip reaches a high temperature (World Health Organization, 2019); at such a temperature, Hg easily vaporizes and becomes swept into a stream by smoke. Hg in particular easily vaporizes at high temperatures and this may explain why Hg absorption ability was the lowest. In the designed experiment, the pump was kept running non-stop to keep all smoke flowing through the filter. Therefore, cigarette smoke contains harmful substances such as heavy metals that go to mainstream smoke. In fact, when humans smoke, the smoke of cigarettes separately becomes two flows that are mainstream and side-stream; meanwhile, both affect the active smoker and passive smoker in different levels that belong to smoking frequency and types of tobacco.

4.3.4. Human health risk assessment

The ADCs (μg/m^3) were calculated using formula (1), and the estimated non-carcinogenic health risk (HI) values of Hg were calculated using formula (2). Both data are shown in **Table 12**. The ADCs of Hg were evaluated by C_m, SF (cigarette/day), and VB (m^3 of air breathed/day).

Therefore, ADC and C_m had the same numerical value because of SF (20 cigarette/day) and VB (20 m^3 of air breathed/day); however, its unit was different (mg/m^3).

Table 12. Human health risk of Hg in cigarette from Vietnam.

Brand code	C_m (ng/cigarette)	ADC (ng/m^3)	HI
Brand 1	14.74	14.74	0.049
Brand 2	30.65	30.65	0.102
Brand 3	10.44	10.44	0.035
Brand 4	14.68	14.68	0.049
Brand 5	14.57	14.57	0.049
Brand 6	12.79	12.79	0.043
Brand 7	21.96	21.96	0.073
Brand 8	06.29	06.29	0.021
Brand 9	11.87	11.87	0.040
Average	15.33 ±7.11	15.33 ±7.11	0.051 ±0.024

The average HI value of Hg was 0.051 ±0.024 (ranged from 0.021 to 0.102). The results showed that the HI values were less than 1, indicate no significant effected by Hg.

Although the HI values of Hg in cigarettes investigated in this study were relatively no significant human health effect, the health of smokers should still be attended to because of their direct and long-term inhalation exposure.

Although, the HI showed there no significant human health effected. Smokers also have more human health risk effect by inhalation of Hg from other sources such as Hg in street dust that was described in Chapter 2 and Chapter 3. The hazard index of Hg in street dust through inhalation of street dust that contaminated Hg and inhalation of Hg from street dust with Hg in cigarettes were shown in Figure 19 and Figure 20. The results showed that the hazard index of Hg inhalation from cigarettes was much higher than street dust in the normal condition sampling (March 2019). This indicated Hg from cigarettes was more serious effect than inhalation of Hg in street dust via inhalation and vaporization also.

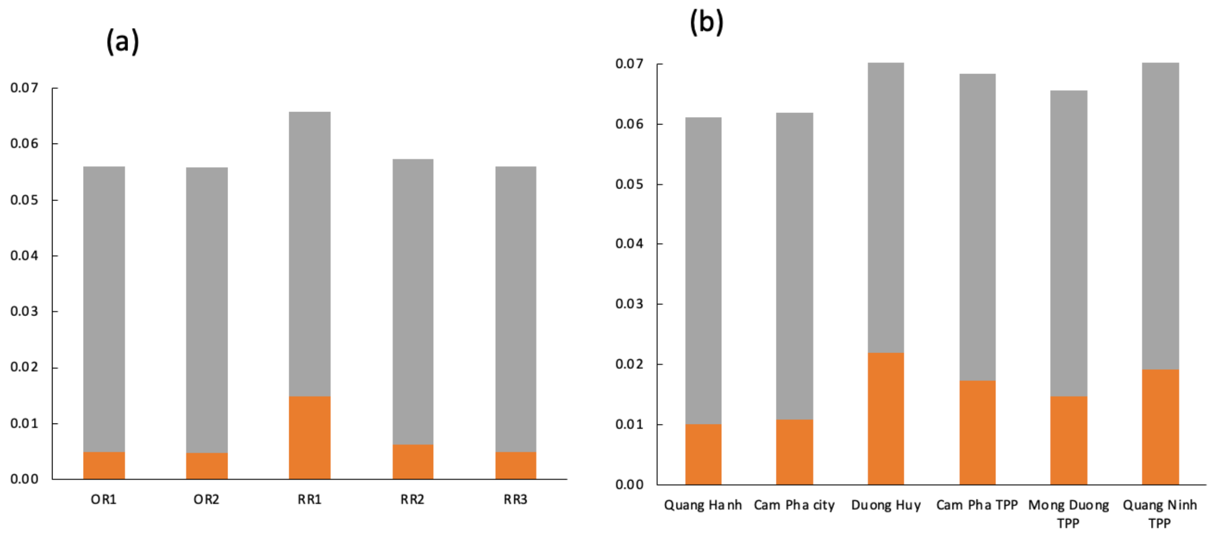

Figure 19. The hazard index to adults of Hg in street dust and cigarette in Quang Ninh (a) and Hanoi (b) during March 2019.

HI for Inhalation of Hg from dust (HQ_{inh} + HQ_{vap});

HI for Inhalation of Hg from cigarette.

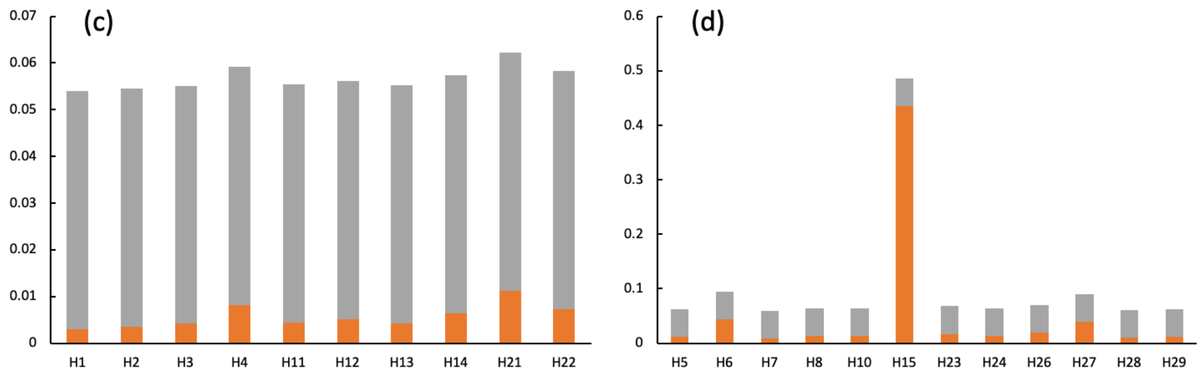

Figure 20. The hazard index to adults of Hg in street dust and cigarette in Hanoi after CFL factory fire outbreak ((c) RF sites (d) PO sites).

HI for Inhalation of Hg from dust (HQ_{inh} + HQ_{vap});

HI for Inhalation of Hg from cigarette.

The comparison of HQ_{inh} and HQ_{vap} with HI of cigarettes through inhalation of Hg showed that cigarette was more human health risk effects than street dust. In notices, HQ_{vap} in dust was highest and become most serious way to effect to human compared with another pathway. Therefore, it is necessary to have more study in the case of exposure Hg from many potential sources of Hg in the living environment.

4.4. Conclusion

This study provides a survey of Hg in marketed cigarettes collected from Vietnam. According to the authors, this is the first research on the distribution concentration of Hg in three parts of cigarettes separately: filter, tobacco, and rolling paper. This study found even the lower Hg trapping efficacy of active carbon additive filters was similar as compared to those without active carbon, indicating the addition of activated carbon to the filter was no contributes to a minimal extent to the health of smokers for Hg. Notably, the ability to absorb Hg was not significantly different between these types of filters. The novelty of this study's key point was we found in the fact that the cigarettes filter could not absorb Hg well. The human health risk of Hg in cigarette showed that no effect by Hg in cigarettes from Vietnam and other countries.

This study provides valuable data and information on Hg in marketed cigarettes for tobacco companies, public health protection agencies, public health environment organizations, and the general smoking population concerning the effect of Hg in marketed cigarettes.

CHAPTER 5

Conclusions

5.1. Conclusions

In this study, the application of street dust was used to identify the Hg pollution and human health risk assessment in a case study of Quang Ninh and Hanoi, Vietnam. This is the first study on Hg in street dust in those areas. Street dust was confirmed as a good tool for environmental pollution identification and human health risk assessment. The high Hg concentration in street dust was found in the areas that have sources of Hg pollution. In Quang Ninh, the high concentration of Hg was found in street dusts near the non-ferrous factory, coal mines areas, and thermal power plants areas. That indicated that Hg from those activities probably were main sources that make Hg pollution in Quang Ninh province and could be found in street dust. The high Hg concentrations in street dust near the non-ferrous factory and coal mines, and thermal power plants may have more effect on adult men, who are main employment working in the mines and the factories. In Hanoi, the high concentration was found in central Hanoi (RR1: 151.42 ±61.86 ng/g) may result from honeycomb coal that common use in restaurants and food streets. The Igeo and enrichment factor values showed the degree of anthropogenic sources of Hg in street dust in those areas that were identified as contaminated and resulted from significant anthropogenic sources.

The human health risk assessment showed that during spring, the HQ of Hg in street dust in central Hanoi (RR1) was higher than other areas, probably due to honeycomb coal using for cooking; this may effect adult women who commonly work with daily cooking works.

After the CFL factory incident, Hg in street dusts were evaluated. The Hg concentration in street dust was found extremely high in site H15 (4450 ng/g). The higher Hg concentration in street dust was found followed the north-east direction followed the monsoon wind direction was demonstrated in this study. It could be conducted that movement of Hg in this case probably affect the downstream areas of the wind.

Street dust samples collected from March 2019 in both Hanoi and Quang Ninh may not pose any significant non-carcinogenic human health risk although contamination of Hg because the HQ and HI values were below one. However, from CFL factory incident, Hg in street dust probably significant effect to the human, especially children. This means the environment agencies need to control and management of the TPPs, non-ferrous manufacturing, and CFL factories to reduce Hg emits into the environment.

Although the HQ values and HI values of Hg in street dusts were no significant health risk effect by Hg in street dust. Humans in those areas can exposure Hg from other sources such as food, beverage, and tobacco cigarette. In the addition study on Hg in marketed cigarettes in Vietnam, the combined human health risk of street dust and cigarette smoke showed the higher risk of Hg in street dust and cigarette smoke might more effect on adult men. Especially, the inhalation of Hg from dust through inhalation and vaporization, combined with inhalation of Hg in cigarettes may effect to adult men who are smoker.

From the results of this study, in future works, the effect of Hg on the adult men who work in the coal mine and TPPs from the Hg in dust exposure will continue to conduct. Besides, the effect of Hg in household dust on adult women who commonly use honeycomb coal for cooking and on children also. Besides, the Hg in indoor dust such as sources, distributions, and accumulations will be implemented. And from the CFL factory incident, the follow up of Hg in Hanoi area will continue to research.

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SUPPLEMENT DATA

Table S1. The concentration of reference metal (Fe) in street dust in Quang Ninh.

Area	Sample site	Fe concentration (µg/g)
Quang Hanh	Q1	5821.63
	Q2	5585.35
Cam Pha city	Q3	6953.69
	Q4	6897.39
	Q5	6991.46
	Q6	6578.95
Duong Huy	Q17	6523.55
	Q18	5681.39
	Q19	6852.52
	Q20	7065.50
	Q21	6753.64
	Q22	6622.64
Cam Pha TPP	Q7	7479.46
	Q8	6937.34
	Q9	6758.04
	Q10	7274.76
Mong Duong TPP	Q11	7086.19
	Q12	6763.96
	Q13	7198.94
	Q14	6993.13
	Q15	6726.50
	Q16	6910.20
Quang Ninh TPP	Q23	7064.01
	Q24	7515.72
	Q25	6917.55

Table S2. The concentration of reference metal (Fe) in street dust in Hanoi city.

Areas	Sample site	Fe concentration (μg/g)
OR1	H1	8903.86
	H2	7872.17
	H3	6759.96
	H4	8820.28
OR2	H5	7605.22
	H6	7830.50
	H7	8883.13
	H8	8154.27
	H9	8496.82
	H10	8779.32
RR1	H26	9076.95
	H27	8908.36
	H28	8781.76
	H29	8935.90
RR2	H19	8760.09
	H20	8913.76
	H21	8042.05
	H22	8894.02
	H23	9225.86
	H24	8687.83
	H25	9066.08
RR3	H11	9147.69
	H12	10009.89
	H13	9201.39
	H14	8919.32
	H15	8845.14
	H16	9115.52
	H17	9440.61
	H18	9209.75