

Correlation between heavy metals and polyaromatic hydrocarbons of particulate matter in the flue gas of municipal solid waste incinerators in north Viet Nam

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Abstract. This study aimed to determine the content of heavy metals (Cu, Ni, Pb, Zn, Cd, Cr, and As) and polyaromatic hydrocarbon (PAHs), and focus on their relationship in particulate matter contained in the flue gas of the incinerators. Samples were collected in the exhaust gas of 5 municipal solid waste incinerators located in Bac Giang, Bac Ninh, Hai Duong, Ha Noi, and Hai Phong. Heavy metals and PAHs were analyzed by ICP-MS and GC-MS, respectively. The heavy metal content shows a significant difference between incinerators, varying from 368 to $6.13 \times 10^3 \mu\text{g}/\text{Nm}^3$. Here, the predominant element in the PM samples was zinc, accounting for up to 80% of the total content of studied elements, while Pb and Cu contribution were approximately 10 to 20 %, respectively. The contribution of Cd was stable at roughly 5% of the total heavy metal content. Meanwhile, total PAHs exhibited a fairly low concentration, varying from 0.774 to $27.4 \mu\text{g}/\text{Nm}^3$, of which PAHs have a carcinogenic potential ranging from 21.3 to 73.2% of total PAHs. However, the low molecular PAH content decreased when heavy metal concentration increased.

Keywords: PAHs, heavy metal, PM, solid waste incinerator, North Viet Nam

Classification numbers: 3.4.5, 3.6.1, 3.7.1

1. INTRODUCTION

Recently, Viet Nam has faced a great challenge in solid waste management due to the explosion in population. Its population has raised to more than 100 million ton in 2022, creating huge stress on the environment by solid waste, especially municipal solid waste (MSW). Solid waste is produced more than 18 million tons per year, and this volume is expected to grow quickly in this decade. Many solid waste treatment techniques have been applied, including

landfilling, incineration, waste reduction, and composting. However, the lack of pre-treatment methods (e.g. waste classification) leads to many difficulties in MSW treatment in Viet Nam. Solid waste incineration is one of the common methods of reducing waste in Viet Nam which brings many advantages compared to other methods, such as including short treatment time, and small occupied space. Despite many benefits, municipal waste incinerators have been identified as a source of environmental pollution due to their by-product (e.g. volatile organic substances) produced during the combustion [1 - 4].

Among various potential toxic substances released from municipal solid waste incinerators (MSWI), this study focuses on trace metals (Cu, Ni, Pb, Zn, Cd, Cr, and As) and PAHs. Heavy metals have been studied due to their persistence during combustion and their toxic behaviours in the ecosystem and their impact on human life [5 - 10]. Meanwhile, the insufficient air supply and incineration time lead to incomplete combustion, which could be an important factor in the forming of organic compounds such as dioxin, PAHs, etc. In the literature, many studies focused on the occurrence, toxicity, and distribution of PAHs in the environment after the first announcement of the United States Environmental Protection Agency (US EPA) in the late 1990s [11 - 13]. The formation and fate of PAHs, from anthropogenic sources (e.g. residues of incinerator), have not been truly studied yet. Few studies have investigated the effect of metals on PAH formation during combustion [14, 15]. However, those studies provided the influence of heavy metal on the formation and distribution of PAHs in the fly or bottom ash of the MSWI only.

In this study, the main investigated points including: i) the distribution of heavy metals and PAHs in particulate matter contained in flue gas for different working MSWI in North Vietnam; ii) highlight the correlation between heavy metals and PAHs.

2. MATERIALS AND METHODS

2.1. Sampling collection

Sample were collected at the municipal waste incinerators from different provinces in the north of Viet Nam, including Ha Noi (HN), Hai Phong (HP), Bac Ninh (BN), Bac Giang (BG), and Hai Duong (HD). These provinces were chosen due to their population, and socioeconomic situation which could influence dramatically to the waste incineration composition and volume. The sampling map was shown in Figure 1 and the information about incinerators was presented in Table 1.

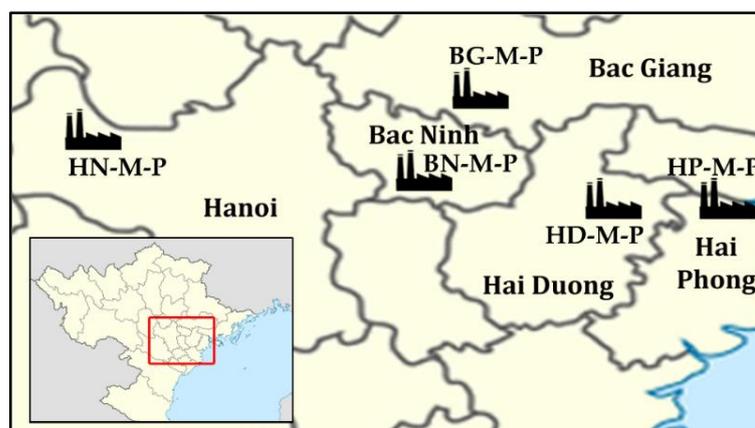


Figure 1. Sampling map.

Table 1 General information on the municipal waste incinerators in this study.

Sample	Plant	Province	Location	Year of Operation	Operation condition	Air Pollution Control
BG-M-P (n = 1)	Hung Phat Urban Environment Co., Ltd	Bac Giang	21°09'23.0"N 106°14'18.0"E	2015	IC: 3.0 AOT: 8030 AEF: 13000	1. Filter bag 2. Scrubber with tap water
BN-M-P (n = 1)	Thuan Thanh Environmental JSC - Plant 2	Bac Ninh	21°01'07.6"N 105°25'28.3"E	2019	IC: 5.0 AOT: 8030 AEF: 22000	1. Scrubber with lime water
HD-M-P (n = 1)	Hai Duong APT – Seraphin environmental JSC	Hai Duong	20°58'14.0"N 106°26'49.0"E	2016	IC: 2.5 AOT: 8030 AEF: 18000	1. Scrubber with lime water
HN-M-P (n = 1)	Thanh Cong Environmental Co-operative	Ha Noi	21°07'33.1"N 105°25'28.3"E	2019	IC: 4.5 AOT: 8030 AEF: 20000	1. Cyclone
HP-M-P (n = 1)	Green consulting science, technology, and environmental treatment Co., Ltd	Hai Phong	20°54'04.0"N 106°46'11.0"E	2019	IC: 1.0 AOT: 7300 AEF: 14500	1. Scrubber with tap water 2. Electrostatic Precipitator
<i>IC = Incineration Capacity (ton/h); AOT = Average operating time (h/year); AEF = average emission flow rate (Nm³/h).</i>						

A total of 5 particulate matter samples were collected from the sampling sites from March 20 to July 20, 2020. Particulate matter (PM, with an aerodynamic diameter equal or less than 10 µm) was collected from exhausted gas on a quartz fibre filter (Advance, QR-100, size 110 mm, Japan) with an isokinetic equipment system (C5000 ESC, USA) with suitable sampling train and in compliance with US EPA Method 201 [16]. The isokinetic system included all the supporting devices (e.g., sampling train, sampling probe, filter heating system, etc.). The sampling location was defined upon the stack specifications with diameter higher than 300 mm as described in US sampling procedure [16]. The sampling location was defined as described in US procedure Method 5 and 5A. Prior to being used, the quartz fibre filters were placed in a clean volumetric flask and shaken for 1 hour with dichloromethane (DCM). The filter was then dried by Nitrogen flow in a laminar flow hood, put in the Petri dish, and placed in the desiccator for 48 hours.

The sample volume was recorded at the end of the sampling procedure. PM filters were wrapped in an aluminium sheet, and stored in a sealable polypropylene bag for transportation. The sample was then placed in a desiccator for 48 h at the ambient temperature (25 ± 2 °C). The particle mass was identified by the weight of the filters before and after using a high precision balance (Adam AEA-210 LC). The 1/4 filter was used to determine the heavy metal concentration and the 1/2 filter was analysed PAH.

2.2. Analysis method and quality control

Heavy metals analysis: Back in the laboratory, the 1/4 filter was placed into a Teflon digestion vessel. Filter was then digested with a 10 mL mixture of HNO₃/HCl (1:3, v/v) using a microwave system (D-728 Enningen, Herretstrasse 1) for heavy metal treatment (Cu, Ni, Pb, Zn, Cd, Cr, and As). The solution was heated to 120°C to almost dry and then redissolved with HNO₃ 1 %. Sample concentration was measured using Inductively Coupled Plasma Mass

Spectrometry (ICP-MS, ELAN 9000, Perkin Elmer, USA). The quantification and recoveries were calculated using ELAN software. The method accuracy was identified by the analysis of reference material (National Metrology Institute of Japan-NMIJ, CRM 7302-a, Japan), giving the recovery of 86 - 94 %.

PAHs analysis: 1/2 filter was placed in thimbles of the Soxhlet extractor, spiked with 6 deuterated surrogate standards including: naphthalene-d₈, acenaphthylene-d₈, phenanthrene-d₁₀, fluoranthene-d₁₀, pyrene-d₁₀, benzo[a]pyrene-d₁₀, and benzo[ghi]perylene-d₁₂ (10 ng each compound, AccuStandard®). The analytical components in samples were extracted using a Soxhlet extractor by the mixture of acetone/hexane (1:1, v/v) as extraction solvent. The aliquot was rotary evaporated and solvent-exchanged into hexane with a small portion of n-decane (300 µL). The exact constituted in 5 mL hexane and was loaded onto a glass chromatographic column packed with 5.00 g silica gel (Silica Gel 60 N, 63 - 210 µm; Kanto Chemical Co., Inc.), modified with 10 % activated carbon (AC), this portion of AC was sandwich between two small portions of sodium sulphate for eliminating any water contain. The column was prewashed with 30 mL hexane before being used in the cleaning process. After loading, the column was rinsed with 20 mL of hexane to avoid any aliphatic interferences. Analytes were eluted with 30 mL of dichloromethane/hexane (1:3, v: v). The eluate was collected into a pear-shaped flask. An internal standard (10 ng of chrysene-d₁₂, AccuStandard®) was spiked and the eluate was constituted in 1.00 mL hexane before GC-MS analysis. A total of 18 PAHs were identified using gas chromatography coupled with the mass system (GC 8890; 5977B GC/MSD, Agilent®) with a HP-5 ms Ultra Inert GC column (30 m × 0.25 mm × 0.25 µm; Agilent®). The list of target compounds and its abbreviation were presented in Table 2. Concentrations of the target compound were reported to four significant figures and were treated as not detected (nd) if below the MDLs. The sum of all the 18 target compounds was expressed as total PAHs (Σ18PAHs). The recovery of surrogate standards ranged from 70 to 120 %. The method was confirmed by using the Standard Reference Material 2585 samples (NIST, Gaithersburg, MD, USA; n = 3, RSD < 25 %). The method detection limits (MDLs) were calculated as three-time of the standard deviations of blank levels, ranging from 0.02 to 0.05 µg/Nm³.

The method blank for both heavy metals and PAHs were conducted with the clean filter (following the procedure mentioned above) giving less than 2 % of the sample signal.

3. RESULTS AND DISCUSSION

3.1. Distribution of heavy metal and PAHs in PM

Table 2 showed the concentration of each of the 7 heavy metals (Cu, Ni, Pb, Zn, Cd, Cr, and As) and 18 PAHs in the particulate matter contained in the flue gas of MSWIs in this study.

Generally, the total concentration of heavy metals in PM ranges from 368 to 6.13×10^3 µg/Nm³. The MSWI of Hanoi revealed the highest content of total heavy metal, which was about 20 times higher than this of Bac Giang. Meanwhile, Bac Ninh and Hai Phong indicate similar content of heavy metal escape from chimneys on particulate matter. The average concentration following this order Zn>Pb>Cu>Cd>Cr>As>Ni. However, the concentration of individual heavy metals illustrated high variation among incinerators. These variations could be due to different factors, which were feeding materials, incineration technique, post-combustion treatment technique, etc.

Regarding the PAHs content, Σ18PAHs in PM varied from 0.774 µg/Nm³ (HD-M-P) to 27.4 µg/Nm³ (BG-M-P). The distribution of individual PAH or PAH rings varied significantly

between different incinerators (Table 2). The 5 to 6-ring PAH compounds accounted for roughly 40 % of total PAHs emission in PM. This observation has not been recorded in the literature. However, Li *et al.* [22] observed the dominance of 2 to 3-ring PAH compounds in the exhausted gas of laboratory scale MSWI. This high proportion of low-ring PAHs comparing with others in gaseous phase could lead to its low proportion in the particulate phase.

Table 2. Concentrations of heavy metals and PAHs in particulate matter of MSWI.

Sample	Bac Giang	Bac Ninh	Hai Duong	Ha Noi	Hai Phong
	($\mu\text{g}/\text{Nm}^3$)				
Copper (Cu)	20.0	489	241	575	52.0
Nikkel (Ni)	4.50	0.562	0.199	1.62	3.41
Lead (Pb)	0.108	512	34.8	1.36×10^3	148
Zinc (Zn)	278	1.83×10^3	499	3.82×10^3	1.67×10^3
Cadmium (Cd)	8.61	150	44.2	357	50.4
Chromium (Cr)	51.5	5.38	2.27	14.8	17.5
Arsenic (As)	5.16	0.766	1.22	8.67	7.02
$\Sigma 7$ Heavy metals	368	2.99×10^3	822	6.13×10^3	1.95×10^3
Naphthalene (Nap)	2.06	1.04	0.627	nd	0.738
1-Methylnaphthalene (1-Me-Nap)	nd	nd	nd	nd	nd
2-Methylnaphthalene (2-Me-Nap)	0.172	nd	nd	nd	0.085
Acenaphthylene (Acy)	0.170	nd	nd	nd	nd
Acenaphthene (Ace)	0.171	nd	nd	nd	nd
Fluorene (Flu)	1.940	nd	nd	nd	nd
Phenanthrene (Phe)	0.355	nd	nd	0.691	0.597
Anthracene (Ant)	nd	nd	nd	0.609	nd
Fluoranthene (Flt)	0.610	0.062	0.025	1.68	0.624
Pyrene (Pyr)	0.580	0.098	nd	2.42	0.568
Benzo(a)anthracene* (BaA)	5.00	nd	nd	0.430	0.523
Chrysene* (Chr)	6.52	0.094	nd	0.698	1.04
Benzo(b)fluoranthene* (BbF)	2.18	0.162	0.058	1.37	0.694
Benzo(k)fluoranthene* (BkF)	0.830	nd	nd	1.37	0.134
Benzo(a)pyrene* (BaP)	2.33	0.076	nd	4.01	0.554
Dibenzo(a,h)anthracene* (DA)	1.34	nd	nd	nd	nd
Indeno(1,2,3-cd)pyrene* (IP)	1.01	nd	0.064	8.12	1.73
Benzo(g,h,i)perylene (BP)	2.17	0.091	nd	0.389	1.74
$\Sigma 7$ carcPAHs	19.2	0.333	0.121	16.0	4.67
$\Sigma 18$ PAHs	27.4	1.62	0.774	21.8	9.06
% 2-3 ring	18 %	64 %	75 %	6 %	16 %
% 4 ring	46 %	15 %	7 %	24 %	30 %
% 5 ring	36 %	21 %	18 %	70 %	54 %

BG: Bac Giang; BN: Bac Ninh; HD: Hai Duong; HN Ha Noi; HP: Hai Phong, P: particulate matter 10; M: municipal waste incinerator; nd: not detected, below limit of detection, *: carcinogenic PAHs.

Among the 18 analysed PAHs in this study, 7 species (Benzo(a)anthracene, Chrysene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)pyrene, Dibenzo(a,h)anthracene, Indeno(1,2,3-cd)pyrene) were regarded as potential carcinogenic compounds (Table 2) [17]. In the PM sample, the total 7 carcinogenic PAH content ($\Sigma 7\text{carcPAH}$) varied widely, highest at $19.2 \mu\text{g}/\text{Nm}^3$ (BG-M-P), lowest at $0.211 \mu\text{g}/\text{Nm}^3$ (HD-M-P). These $\Sigma 7\text{carcPAH}$ s could contribute up to 70 % of total PAHs measured in the PM sample. In 2015, Li *et al.* [21] found that $\Sigma 7\text{carcPAH}$ in Fly Ash of MSWI (rotary kiln type) of China accounted for just less than 3 % of total PAH emission. Regarding the two most potential carcinogenic PAHs which are Benzo(a)pyrene-BaP and Dibenzo(a,h)anthracene-DA, the concentration of BaP in PM was high in comparison with other PAHs compounds and it could be found in 4 over 5 studied MSWI. Further treatment for PAHs should be seriously considered to eliminate the impact of these compounds on the environment, especially soil at surrounding area of the treatment facilities.

3.2. Correlation between total heavy metals and PAHs in the PM

The relationship between the heavy metal and PAHs in the flue gas was presented in Figure 2 and Table 3. Wey *et al.* [19] concluded that the influence of heavy metals on the formation of PAHs decreased by the order of $\text{Pb} > \text{Zn} > \text{Cd} > \text{Cr}$. Although Wey *et al.* [19] focused on fly ash, a similar trend could be indicated in this study.

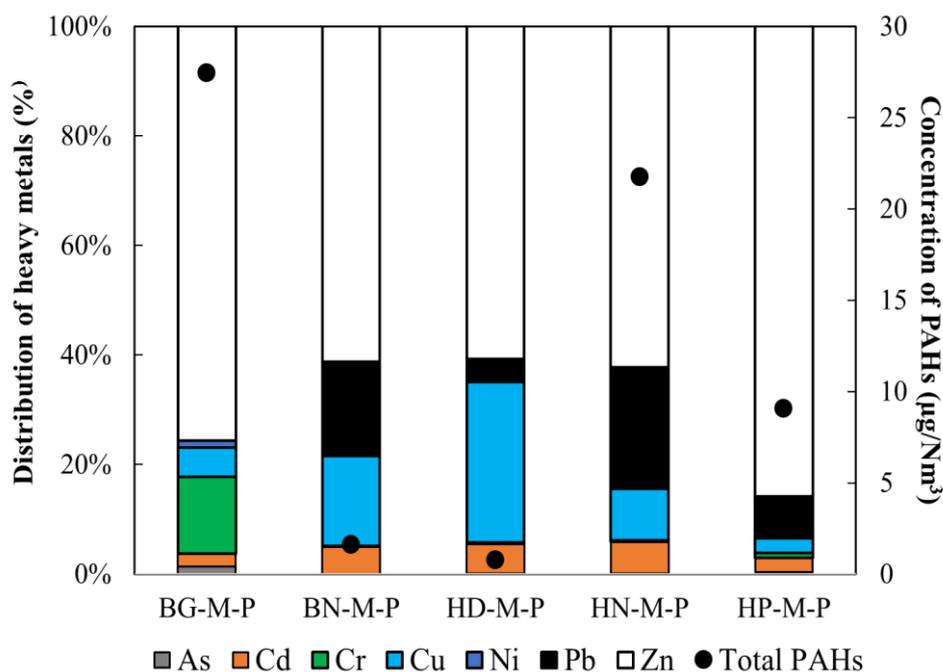


Figure 2. Total PAHs and Distribution of investigated elements.

Copper and cadmium suggested no significant effect on the formation of PAHs. The high contribution of Cu and Cd in Hai Duong and Bac Ninh incinerators over other provinces did not go with the raising of PAH content. Other elements contribute small proportions of total heavy metals. The results in Table 3 indicated that Ni, Cr, and As have a strong correlation ($r > 0.75$) with total PAHs in PM. The high concentration of these elements in Bac Giang's incinerator also

promoted the formation of PAHs, leading to the highest total concentration of PAHs recorded in 5 study MSWIs. Wey *et al.* [20] observed that when the organic compound-CI was added to the feedstock, Cr would be the main factor influencing the formation of PAHs. This observation may support our hypothesis about the relationship of Cr with PAHs because municipal solid waste in Viet Nam often contains high proportions of plastic of different types. This high amount of resin could be a factor for Cr function as a key factor for the formation of PAHs.

Table 3. Correlation coefficient (r) between trace elements and PAHs in PM.

	Cu	Ni	Pb	Zn	Cd	Cr	As	PAHs
Cu	1							
Ni	-	1						
Pb	0.847	- 0.293	1					
Zn	0.759	- 0.231	0.958*	1				
Cd	0.873	-0.357	0.997**	0.951*	1			
Cr	-	0.900*	- 0.271	- 0.336	- 0.329	1		
As	-	0.575	0.488	0.574	0.444	0.387	1	
PAHs	-	0.741	0.251	0.162	0.197	0.844	0.710	1

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

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

This study also highlighted the importance of the Air Pollution Control (APC) system to the removal of PAHs after combustion by comparing the content of particle-bound PAHs. In fact, the APC systems have been applied to remove fly ash and fine particles, the better particle removal efficiency of APC technique could decrease the PM-bound PAHs in the flue gas. Here, various APC techniques have been applied in different studied incinerators (Table 1). Likely, the use of the cyclone was not a sufficient method to remove the PAHs reflecting the huge number of PAHs escaping via PM in the flue gas of Hanoi's incinerators. The lower PAHs removal efficiency of cyclone comparing with scrubber has been highlighted in the study of Wey *et al.* [19]. In another way, among all case studies in this research, the scrubber with lime water was the most suitable technique to remove particulates, leading to decreasing the PM - bound PAHs after the combustion, satisfying both the installation method and manipulation, especially for the low-cost incinerator in Viet Nam. Further studies should be conducted on the sample before and after APC system to fully assess the efficiency of APC system on removal of PAHs.

4. CONCLUSION

This study presented the concentration of 7 heavy metals (Cu, Ni, Pb, Zn, Cd, Cr, and As) and 18 PAHs in PM of flue gas of MSWI in North Viet Nam. The total heavy metal and PAHs concentration ranged from 368 to $6.13 \times 10^3 \mu\text{g}/\text{Nm}^3$ and 0.774 to $27.4 \mu\text{g}/\text{Nm}^3$, respectively. The correlation between different heavy metals elements (Ni, Cr and As) and PAHs was highlighted, reflecting the influence of individual trace on the fate of PAHs after escaping from the APC system. Ni, Cr, and As were found to have a great impact on the formation of PAHs. The pollution treatment technique was also investigated, suggesting the importance of scrubber in removing PM – bound PAHs in the flue gas. To get the full picture of the formation, interaction, and fate of heavy metal and PAHs during the incineration, the combustion technique (temperature, feeding rate, fuel supply, etc.), chemical components of solid waste or feedstock, and some other factors must be assessed. It suggested the necessity for further study including all incineration residues (fly ash, bottom ash, flue gas) at a higher sample set.

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CRedit authorship contribution statement. All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by Viet Quoc Pham, Thi Phuong Mai Nguyen, and Phuong Thu Le. The first draft of the manuscript was written by Hue Thi Nguyen and Viet Quoc Pham and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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

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