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Levels of polycyclic aromatic hydrocarbons in different types of hospital waste incinerator ashes

Lijuan Zhao, Fu-Shen Zhang⁎, Zhengping Hao, Hailin Wang

Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, 18 Shuangqing Road, Beijing 100085, China

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ABSTRACT

Waste ashes from three types of hospital waste (HW) incinerators, built in SARS (Severe Acute Respiratory Syndrome) period and currently running in China, were collected and polycyclic aromatic hydrocarbons (PAH) properties in the ashes were investigated. The mean ∑PAH levels in the waste ashes varied widely from 4.16 mg kg⁻¹ to 198.92 mg kg⁻¹, and the mean amounts of carcinogenic PAHs ranged from 0.74 to 96.77 mg kg⁻¹, exceeding the limits regulated by several countries. Among the three types of incinerators, two medium-scale incinerators generated relatively high levels of PAHs (mean ∑PAH 22.50 and 198.92 mg kg⁻¹) compared to small-scale and large-scale incinerators (mean ∑PAH 4.16 and 16.43 mg kg⁻¹). Bottom ashes were dominated by low molecular weight PAHs (LM-PAH; containing two- to three-ringed PAHs) and medium molecular weight PAHs (MM-PAH; containing four-ringed PAHs), while fly ashes were abundant in MM-PAH and high molecular weight PAHs (HM-PAH, containing five- to six-ringed PAHs). Statistical analysis indicated that there was a positive relationship (R² = 0.88) between organic matter and total PAHs thus it has the potential to be used as an indicator for PAHs in HW ashes. Moreover, it was found that PAHs in the ashes correlated highly with some metallic elements either positively (e.g. Fe, Ti, Mg) or negatively (Ca), indicating that these elements might promote or prevent PAH formation during HW combustion. Although bottom ash resulted from HW incinerators has not been classified as hazardous material, the results of this study indicated that this type of waste ash contained high levels of PAHs thus need special treatment before landfill.

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1. Introduction

The annual generation of hospital wastes (HW) in China is estimated to be 1.65 million tons (Liu et al., 2006), and the main disposal way for this type of waste is incineration. However, most of the HW incinerators were built in the serious situation for dealing HW generated during Severe Acute Respiratory Syndrome (SARS), thus many of the facilities were old and improperly designed and usually lacks emission control system. Currently, many hospital-scale incinerators in some small cities are still operating although China EPA has closed some on-site small incinerators which pose a potential threat to the environment. In recent years, several large-scale and properly designed HW incinerators, equipped with well-maintained air pollution control devices (APCDs), have been built in large cities.

Polycyclic aromatic hydrocarbons (PAHs) are known as an important species due to the carcinogenicity and mutagenicity of some of these compounds (IARC, 1983). In developed countries, efforts have been made to reduce PAH emission. It is estimated that total benzo[a]pyrene emissions of 33 European countries were reduced from 1300 tons in 1970 to 590 tons in 1995 (Pacyna et al., 2003). While in China, emissions of 16 PAHs from major sources have increased substantially from around

* Corresponding author. Tel.: +86 10 6284 9515; fax: +86 10 6289515. E-mail address: fszhang@rcees.ac.cn (F.-S. Zhang).

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Combustion process is one of the major sources of PAHs. Studies concerning the emissions of PAHs in the process of fossil fuels combustion, organic matters burning (wood, straw), solid waste incineration under incomplete combustion have been conducted (Davies et al., 1976; Low and Batley, 1988; Liu et al., 2000; Johansson and van Bavel, 2003a,b). Meanwhile, hospital waste was found to generate higher amount of PAHs compared to coal and municipal solid waste (MSW) because it contains high amount of plastic and rubber (gloves, containers, bags, packaging) (Levendis et al., 2001). Mastral (1999) concluded that such wastes promote more PAHs formation as compared to coal combustion at the same combustion conditions. Thus HW incineration is an important emission source of PAHs, especially small-scale incinerators which lack emission control system.

Most previous studies on PAHs have focused on the gaseous phase of emission sources (Levendis et al., 2001; Lee et al., 2002; Ferraz and Afonso, 2003; Sadhra and Wheatley, 2007). However, in the combustion processes, PAHs adsorbed on solid particles can also cause serious pollution when they evaporate into the atmosphere (Wild et al., 1992; Liu et al., 2000), as most of the PAHs are associated with particulate material and the PAHs either remain in the bottom ash or are removed from the flue gases by the precipitators to form fly ash. Lee et al. (2002) and Wheatley and Sadhra (2004) reported PAH levels and concentrations in clinical waste incineration bottom and fly ash in Taiwan and UK. To the best knowledge of the authors, however, only limited reports are available on PAHs content in various HW incinerator ashes in China, though thermal treatment of HW has dramatically increased after the outbreak of SARS.

In this study, the levels of PAHs in waste ashes from different types of HW incinerators were examined. The aim was to evaluate the environmental impacts of various HW incinerators currently operating in China. The data obtained in the study could assist HW incineration plants in selecting appropriate technologies in the future.

2. Materials and methods

2.1. Sampling

Three types of HW incinerators, including two medium-scale incinerators, one large-scale incinerator and one small-scale incinerator, were selected in this study. Table 1 shows the background information and operating conditions of the incinerators.

HWI-I is a special type of medium-scale incinerator, which combined more than ten small fixed grate furnaces. Part or all of the furnaces are operated according to the amount of hospital wastes collected. Although APCDs are equipped to each furnace, fly ash could hardly be collected in this type of incinerator, thus only bottom ash were obtained. Three mixed samples were collected every 10 days from the incinerator within a month. HWI-II is a typical small-scale incinerator run by a hospital, representing hundreds of incinerators running in China. This incinerator has no APCDs equipped, thus no fly ash could be collected. The sampling activities were conducted within 3 months, and 12 mixed samples were collected once a week. HWI-III is a medium-scale incinerator operated in a large city. Fly ashes were collected in the bag filters, but the residue mainly consists of unburned glasses, plastics and metallic matters, thus bottom ash could not be collected. The sampling activities were carried out within 1 month and 3 mixed samples were collected. HWI-IV, which was properly designed and has well-maintained APCDs, is a large-scale incinerator with the batch capacity of 40 tons per day. From this incinerator, six mixed samples were collected every half-month during 3 months.

All collected samples were dried at 30 °C for 24 h, crushed and sieved through a 0.9 mm mesh and stored at 4 °C for use. The amount of organic matter in the samples was estimated by weight loss on ignition at 550 °C for 6 h (Johansson and van Bavel, 2003a,b).

Heavy metals were analyzed by inductively coupled plasma optical emission spectroscopy (ICP-OES) after HNO3/HClO4/HF digestion (Yamasaki, 1997). The quality and precision of metallic element analysis were controlled using the reference material (NIST 1646), which was a sediment from the National Institute of Standards and Technology (USA). The recoveries of metallic element are between 87 and 117%.

2.2. Chemicals and materials

Silica gel (100 200 mesh) and alumina (Qingdao, China) were activated at 180 °C and 250 °C respectively for 12 h, then deactivated with 3% water. Sodium sulfate (Beijing, China) was baked at 450 °C before use. Acetone, n-hexane and dichloromethane (DCM) were all pesticide grade purchased from Tedia (USA). Standards of 16 US EPA priority PAHs, i.e. Naphthalene, Acenaphthylene, Acenaphthen, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benzo[a]anthracene,
the carrier gas at a constant flow rate of 1 ml/min. Splitless injection of 1 µl sample was conducted with an autosampler. The GC oven temperature was programmed from 50 °C held for 2 min followed by an increase to 300 °C at a rate of 6 °C/min, where it was held for 5 min. The injector and detector temperatures were 300 °C and 230 °C, respectively. Quantization was performed using the five-point calibration curve for individual components.

An internal standard containing Hexamethylbenzene was used for quantification. Calibration curves including five different concentrations were constructed using the internal standard method. The detection limits of these PAHs ranged from 0.02 to 0.03 mg kg\(^{-1}\).

### 2.5. Quality assurance and quality control

Laboratory quality control procedures include analyses of method blanks, spiked blanks, matrix spike duplicates, and sample duplicates. The recoveries for surrogate standards fell within a fairly narrow range of 72–100%.

## 3. Results and discussion

### 3.1. PAH levels in the HW ashes

PAH levels in the HW bottom ashes from the two different types of incinerators (HWI-I and HWI-II) are shown in Table 2. The mean \(\sum PAH\) (16 US EPA priority PAHs) concentrations in the carrier gas at a constant flow rate of 1 ml/min. Splitless injection of 1 µl sample was conducted with an autosampler. The GC oven temperature was programmed from 50 °C held for 2 min followed by an increase to 300 °C at a rate of 6 °C/min, where it was held for 5 min. The injector and detector temperatures were 300 °C and 230 °C, respectively. Quantization was performed using the five-point calibration curve for individual components.

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### Table 2 – PAH concentrations in the HW bottom ashes (mg kg\(^{-1}\))

| PAH               | HWI-I (n=3) |   |   |   |   |   |   |   |   |   |   |
|-------------------|------------|---|---|---|---|---|---|---|---|---|---|
|                   | Mean       | Median | SD | Mean | Median | SD | Mean | Median | SD | Mean | Median | SD |
| Naphthalene       | 1.30       | 1.02 | 0.65 | 1.28 | 1.27 | 0.45 |       |       |    |       |       |    |
| Acenaphthylene    | 0.23       | 0.20 | 0.16 | 0.12 | 0.09 | 0.08 |       |       |    |       |       |    |
| Acenaphthene      | 0.10       | 0.11 | 0.04 | 0.44 | 0.48 | 0.11 |       |       |    |       |       |    |
| Fluorene          | 0.48       | 0.38 | 0.20 | 0.95 | 0.93 | 0.21 |       |       |    |       |       |    |
| Phenanthrene      | 4.93       | 4.26 | 3.30 | 3.32 | 2.53 | 1.82 |       |       |    |       |       |    |
| Anthracene        | 0.79       | 0.62 | 0.39 | 1.25 | 1.18 | 0.29 |       |       |    |       |       |    |
| Fluoranthene      | 2.64       | 2.24 | 1.67 | 1.30 | 1.03 | 0.57 |       |       |    |       |       |    |
| Pyrene            | 2.21       | 1.86 | 1.36 | 1.36 | 0.99 | 0.69 |       |       |    |       |       |    |
| Benzo[a]anthracene* | 1.76 | 1.52 | 1.18 | 1.51 | 1.47 | 0.37 |       |       |    |       |       |    |
| Benzo[k]fluoranthene* | 0.11 | 1.11 | 0.52 | 0.76 | 0.81 | 0.26 |       |       |    |       |       |    |
| Benzo[a]pyrene    | 0.95       | 0.95 | 0.45 | 1.12 | 1.20 | 0.50 |       |       |    |       |       |    |
| Indeno[1,2,3-cd]pyrene | 0.64 | 0.50 | 0.31 | 1.06 | 1.03 | 0.24 |       |       |    |       |       |    |
| Dibenzo[a,h]anthracene* | 1.11 | 0.94 | 0.70 | 1.03 | 0.70 | 0.88 |       |       |    |       |       |    |
| Benzo[g,h]perylene | 1.02       | 0.85 | 0.61 | 0.82 | 0.81 | 0.11 |       |       |    |       |       |    |
| \(\sum_{\text{carcinogenic}}\) PAH | 8.82       | 7.63 | 5.93 | 6.13 | 5.95 | 2.45 |       |       |    |       |       |    |
| \(\sum_{16\ \text{US EPA PAH}}\) | 22.50 | 19.07 | 14.23 | 16.43 | 15.15 | 6.43 |       |       |    |       |       |    |

* Carcinogenic PAHs.

### Table 3 – PAH concentrations in the HW fly ashes (mg kg\(^{-1}\))

| PAH               | HWI-III (n=3) |   |   |   |   |   |   |   |   |   |   |   |
|-------------------|---------------|---|---|---|---|---|---|---|---|---|---|---|
|                   | Mean     | Median | SD | Mean | Median | SD | Mean | Median | SD | Mean | Median | SD |
| Naphthalene       | 2.39     | 2.02 | 0.65 | 2.17 | 1.15 | 2.30 |       |       |    |       |       |    |
| Acenaphthylene    | 2.27     | 2.22 | 0.30 | 0.02 | 0.02 | 0.01 |       |       |    |       |       |    |
| Acenaphthene      | 0.03     | 0.03 | 0.01 | nd   | nd   | –    |       |       |    |       |       |    |
| Fluorene          | 0.28     | 0.32 | 0.08 | 0.23 | 0.19 | 0.08 |       |       |    |       |       |    |
| Phenanthrene      | 18.65    | 22.38 | 6.75 | 0.58 | 0.38 | 0.41 |       |       |    |       |       |    |
| Anthracene        | 2.22     | 2.53 | 0.69 | 0.29 | 0.29 | 0.12 |       |       |    |       |       |    |
| Fluoranthene      | 25.62    | 29.70 | 8.49 | 0.27 | 0.27 | 0.09 |       |       |    |       |       |    |
| Pyrene            | 20.58    | 23.73 | 6.87 | 0.17 | 0.14 | 0.05 |       |       |    |       |       |    |
| Benzo[a]anthracene* | 7.25 | 7.82 | 1.66 | nd   | nd   | –    |       |       |    |       |       |    |
| Benzo[k]fluoranthene* | 13.47 | 11.21 | 4.73 | 0.32 | 0.33 | 0.01 |       |       |    |       |       |    |
| Benzo[a]pyrene    | 12.19    | 13.72 | 4.38 | 0.23 | 0.23 | 0.03 |       |       |    |       |       |    |
| Indeno[1,2,3-cd]pyrene | 19.41 | 20.05 | 13.30 | 0.27 | 0.27 | 0.02 |       |       |    |       |       |    |
| Dibenzo[a,h]anthracene* | 6.38 | 7.96 | 3.76 | nd   | nd   | –    |       |       |    |       |       |    |
| Benzo[g,h]perylene | 30.32 | 34.63 | 14.36 | 0.19 | 0.19 | 0.01 |       |       |    |       |       |    |
| \(\sum_{\text{carcinogenic}}\) PAH | 96.77 | 103.04 | 21.29 | 0.74 | 0.33 | 0.64 |       |       |    |       |       |    |
| \(\sum_{16\ \text{US EPA PAH}}\) | 198.92 | 220.59 | 58.97 | 4.16 | 2.51 | 3.69 |       |       |    |       |       |    |

nd: not detected, below limit of detection;–: no data.

* Carcinogenic PAHs.
two ashes are 22.50 mg kg\(^{-1}\) and 16.43 mg kg\(^{-1}\) respectively, much higher than concentrations observed in other literatures about medical waste incinerator bottom ashes, such as 3.17 mg kg\(^{-1}\) and 0.45 mg kg\(^{-1}\) (Lee et al., 2002; Wheatley and Sadhra, 2004). Among the 16 PAHs, phenanthrene is the dominating PAH species in both HW bottom ashes, in the ranges of 13.9–29.9% and 19.5–22.3%, respectively. In addition, fluoranthene, pyrene and chrysene are also prevalent in most samples.

An interesting result from Table 2 is that medium-scale incinerator (HWI-I) generated higher amount of PAHs than small-scale incinerator (HWI-II), i.e., 22.50 mg kg\(^{-1}\) and 16.43 mg kg\(^{-1}\) respectively. This could attributed to the operating conditions of this type incinerator, which are suitable for PAHs formation. For example, the operating temperature in HWI-I is generally below 800 °C, while that in HWI-II is below 400 °C. Therefore, medium-scale HW incinerators currently running in China may cause more serious environmental problems than small-scale ones.

The levels of PAHs in the two types of HW fly ashes are significantly different (Table 3). The mean \(\sum\)PAH concentration contained in the fly ashes from HWI-III was 198.92 mg kg\(^{-1}\), about 50 times higher than that from HWI-IV (4.16 mg kg\(^{-1}\)). It was also much higher than those reported in literatures, i.e. 0 and 47.0 mg kg\(^{-1}\) (Lee et al., 2002; Wheatley and Sadhra, 2004). The prominent species in the two types of fly ashes are quite different. Nine PAH species tend to predominate in ash samples from HWI-III, i.e., benzo[g,h,i]perylene, fluoranthene, benzo[b]fluoranthene, pyrene, indeno[1,2,3-cd]pyrene, phenanthrene, benzo[k]fluoranthene, chrysene and benzo[a]pyrene in descending order. While naphthalene is the most abundant species in samples from HWI-IV, the percentage of this species to total PAHs in the range of 23.33% to 57.95%. These results indicate that fly ashes from medium-scale incinerators (HWI-III) contain much higher amounts of PAHs than from large-scale incinerators (HWI-IV). This could be partly attributed to the different exact operating temperatures of the two incinerators. Although the designed incineration temperatures were similar for the two types of incinerators (Table 1), the real running temperatures in HWI-III were much lower than in HWI-IV because of the structural difference between the two incinerators. First, the color of the ash from HWI-IV was grey while that from HWI-III was black, indicating uncompleted burning in HWI-III. Second, the loss of on ignition (weight percent at 600 °C for 3 h) of the two types of ashes were quite different, i.e. 20% for HWI-III ash and 3% respectively for HWI-III ash, indicating that HWI-IV ash contained much more unburned organic matter.

On the other hand, HWI-III used activated carbon for acidic gases removal. Previous study (Zhou et al., 2005) showed that with increasing the concentration of activated carbon, the concentrations of PAHs in flue gas decreased apparently, while the concentration of PAHs in fly ash increased. Thus, injected activated carbon absorbing on ash particles contributed to the high level of PAHs in ash from HWI-III.

Moreover, HWI-IV is a modern large-scale incinerator and operated at high temperature in the second combustion chamber, promoting the complete decomposition of PAHs. In addition, enough retention time (above 2 s) make the organic matters burn completely in this incinerator.

### 3.2. Carcinogenic PAHs in the HW ashes

Among the 16 individual PAHs, 7 species, including benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene and dibenzo[a,h]anthracene, are regarded as potential carcinogenic compounds. The mean \(\sum\)carcinogenic PAHs content in the ashes from the four types of incinerators varied widely, highest for 96.77 mg kg\(^{-1}\) (HWI-III), lowest for 0.74 mg kg\(^{-1}\) (HWI-IV) (Table 4), implying that these PAH species are greatly related to incinerator type. Thus far, the National Environmental Protection Agency of China has not issued generic guidelines for PAHs in soil, therefore regulated values reported in literatures (Environment Canada, 2003; Johansson and van Bavel, 2003a,b; Vane et al., 2007) were cited so as to evaluate the potential risk of these ashes in case they are applied to soil. It can be seen that the mean \(\sum\)carcinogenic PAHs of the ashes are all above the limits for soil use. In particular, fly ash from HWI-III exceeded the limits of the three countries for about 100 times. Generally, bottom ashes are

### Table 4 – Comparison of carcinogenic PAH levels in this study with generic guidelines regulated by different countries

| Sensitive land use limit (mg kg\(^{-1}\)) | Carcinogenic PAHs in this study (mg kg\(^{-1}\)) |
|------------------------------------------|-----------------------------------------------|
| Netherlands\(^a\) | Sweden\(^b\) | Canada\(^c\) | HWI-I | HWI-II | HWI-III | HWI-IV |
| 0.12 | 0.30 | 0.50 | 8.82 | 6.13 | 96.77 | 0.74 |

a: Vane et al., 2007.

b: Johansson and van Bavel, 2003a,b.

c: Environment Canada, 2003.

![Fig. 1 – Triangular diagram of the 16 PAHs in the HW bottom ashes.](image-url)
regarded as less toxic waste materials since they contain less toxic heavy metals and the leachability of which are much lower compared with fly ashes. However, the results of this study indicated that HW bottom ash is a special type of waste ash, which contains high concentrations of PAHs.

3.3. Molecular weight distribution of the PAHs in the HW ashes

In order to assess PAH homologue distribution in each collected sample, the 16 PAH species were divided into three groups according to the molecular weight, i.e. low molecular weight (LM-PAHs; containing two- to three-ringed PAHs), medium molecular weight (MM-PAHs; containing four-ringed PAHs) and high molecular weight (HM-PAHs; containing five- to six-ringed PAHs). Relatively high proportions of LM-PAHs (34.1–50.6%) and MM-PAHs (30.8–43.3%) were detected in bottom ashes from HWI-I and HWI-II (Fig. 1), which is in consistent with some previous studies on medical waste incineration bottom ash and municipal solid waste incineration bottom ash (Johansson and van Bavel, 2003a,b, Wheatley and Sadhra, 2004). However, the distribution patterns of PAHs in HW fly ashes are quite different (Fig. 2). For fly ashes from HWI-III, high proportion of HM-PAHs (52.3–55.5%), followed by MM-PAHs (32.0–34.3%), were found. These results are similar with previous report on medical waste incinerator fly ash (Lee et al., 2002). In contrast, LM-PAHs were the dominating species in fly ashes from HWI-IV, which was quite different from that of HWI-III. As mentioned, HWI-IV is a large-scale incinerator. The operation conditions of this type of grate prohibit the formation of MM-PAH and HM-PAH. These results indicated that incinerator type affects not only the amount of PAHs, but also the molecular weight distribution patterns of the PAHs.

3.4. Distribution of PAHs in size-fractioned HW ashes

Fig. 3 illustrates the PAH distribution patterns in different size particles of the ashes. For bottom ashes, PAH content in the fine particles (<0.15 mm) was much lower than that in the coarse particles (0.15 mm < Dp < 0.9 mm), suggesting that PAHs had a preference to absorb onto coarse particles. In case of fly ashes, PAH content in the fine particles was higher than that in the coarse particles, which was consistent with the results found in other literature (Liu et al., 2000). Fine fly ash particles have higher surface area than that of the coarse particles, thus more condensed PAHs could be absorbed in fine particle. It was found that the content of HM-PAH contained in the fine particle was much higher than that in coarse particle, suggesting that high molecular PAHs tend to adsorb onto fine fly ash particles.
3.5. Correlation between organic matter and total PAHs

The correlation between PAHs and organic matter content is of concern as organic matter content is an indicator of combustion efficiency. Fig. 4 illustrates that a positive correlation ($R^2=0.88$) between organic matter content and total PAH amount was observed. Such correlation would be useful as the results from analyzing of organic matter could serve as an indicator of PAH content. In addition, organic matter content in HW bottom ashes from HWI-II was observed varied widely from 1.0% to 53.2%, indicating inconsistent conditions in the incinerator operating.

3.6. Effects of metallic elements on PAH formation

Previous study showed that some heavy metals, such as Ba, Mg, Cr, V, U, Pb, Zn, Mn either catalyzed or prevented the formation of PAHs in fly ash during coal incineration (Wey et al., 2000; Arditsoglou et al., 2004). In this study, the correlation coefficients between various metals and PAHs were calculated so as to investigate the possible catalytic effects of some metallic elements on the formation of PAHs during HW incineration. The results show that no relationship between total PAH amount and metal content was found in the fly ash samples. However, high positive correlations were observed between PAHs and metallic elements (Fe, Ti and Mg) for bottom ashes, with correlation coefficients of 0.93, 0.73 and 0.88, respectively, suggesting that these metals might promote the formation of PAHs during HW combustion (Fig. 5). In contrast, Ca showed negative correlation ($R^2=0.80$) with PAHs indicating that calcium may have the effect of preventing PAH formation in the incineration process.

4. Conclusions

The results of this study demonstrated that PAHs generated from HW incineration was greatly affected by incinerator types. Among the three types of incinerators, two medium-scale incinerators generated high levels of PAHs, with mean total PAH values of 22.50 and 198.9 mg kg$^{-1}$, much higher than PAHs in ashes from large-scale (4.16 mg kg$^{-1}$), and even small-scale (16.43 mg kg$^{-1}$) incinerators. Although bottom ash has not been classified as a hazardous material, according to this study, bottom ashes from HW incinerator, especially from medium-scale and small-scale incinerators, contained high levels of PAHs thus need special treatment before landfill. Of the PAH species, the mean $\sum$carcinogenic PAHs in the examined ashes are all above the limits for soil application, especially for ashes from two medium-scale incinerators, which exceed the limit values for 100 times, suggesting that these ashes must be pre-treated before they are disposed by landfill. Organic matter content was found to have a positive relationship ($R^2=0.88$) with the total PAH thus has a potential to be used as an indicator for PAH levels in HW ashes. Furthermore, it was found that PAH concentrations in the ashes

![Fig. 4 – Relationship between PAHs and organic matter content.](image)

![Fig. 5 – Relationships between metallic elements and total PAHs in HW bottom ashes.](image)
correlate highly with some metallic elements either positively (e.g., Fe, Ti, Mg) or negatively (Ca), indicating that these elements may promote or prevent PAHs formation during HW combustion.

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