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# Levels and patterns of polycyclic aromatic hydrocarbons and polychlorinated biphenyls in municipal waste incinerator bottom ash in Zhejiang province, China

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# ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) were analyzed in bottom ash from municipal solid waste (MSW) incineration in six cities in Zhejiang province, where one-fourth of the MSW incinerators of China are located. Total PAH contents varied from 2222.53 to 6883.91 µg/kg. The patterns of PAHs were found to be very similar in all the samples, dominated by three-ring and fourring PAHs. Total PCB concentrations in bottom ash ranged from 1.00 to  $1.31 \,\mu g/kg$ , while the coplanar PCBs in the bottom ash were in the range of  $0.08-0.52 \,\mu$ g/kg. Among PCB congeners, low chlorinated PCBs contributed to the majority of total PCBs. Generally, PAH concentrations in cities with fluidized bed incinerator were less than those in cities with grate furnace incinerator. PAH and PCB levels were affected by both plastic content in MSW incinerator feed and combustion efficiency. However, further study is required to investigate the effect of these two variables deeply, as well as other influencing factors.

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# 1. Introduction

With the ongoing economic boost and the improvement of living standards in China, about 150 million tons of municipal solid waste (MSW) are being produced every year, followed by an annual increase of 8-10% [1]. While landfill sites for municipal solid waste treatment have become scarce, incineration thus gains popularity for its more volume and mass reduction advantage when compared to landfill treatment [2]. In China, there had been totally 69 municipal solid waste incinerator (MSWI) with a treatment capacity of 39,966 t/d by the end of 2006 [3].

However, MSW incineration is sometimes considered as a secondary pollution source. During incineration waste material is burned at high temperature and converted into ash, unburned solids, carbon dioxide, oxides of nitrogen, sulfur dioxide, ammonia and water [4]. A major concern regarding the operation of incinerators is the problem of solid residues. Large quantities of solid residues such as bottom ash and fly ash are produced during MSW incineration, of which approximately 80% are bottom ash [5]. Bottom ash may contain high concentration of toxic micropollutants. Although heavy metals are subjected to regulatory control in China under the Environmental Protection Act, relatively little attention

has been focused on emissions of organic micropollutants by routes other than air.

Polycyclic aromatic hydrocarbons (PAHs), a group of ubiquitous persistent organic pollutants (POPs) possessing carcinogenic, mutagenic and immunotoxic properties, have become an issue of increasing concern in recent decades [6]. PAH levels in bottom ash from municipal waste incinerators have been reported in London. UK since 1970s [7]. Johansson and van Bavel [8,9] observed that the sum of the 16 USEPA PAHs varied from 140 µg/kg up to more than 77,000 µg/kg in bottom ash from MSWI in Orebro, Sweden. In another study on MSWI bottom ash from Roma, Italy [10], the compositions of PAHs were investigated during optimization of combustion conditions in a rotary kiln incinerator. In China, the levels and distributions of PAHs in bottom ash were rarely studied. Shi et al. [11] investigated PAH levels (1961.0-2420.2 µg/kg) in two bottom ash samples in MSWI of Hangzhou.

Well-known sources of PCBs include those released by the use or disposal of industrial PCB products or formed as byproducts during municipal solid waste incineration [12-14]. MSW incineration processes have the potential to both produce and destroy PCBs. Previously, Sakai et al. [15-17] reported the results of substance flow analyses in a MSW incineration facility in Kyoto city. In those studies, they examined the inflow amounts of dioxin-like PCBs in solid municipal waste and the amounts released via emission gas and incineration residues. Vehlow et al. [18] have reviewed the PCBs in bottom ash from MSW. In comparison with the data of other pollutants such as heavy metals and polychlorinated dibenzo-p-dioxins

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and dibenzofurans (PCDD/Fs), those for PCBs in solid residues are less. Furthermore, few PCB data are available for bottom ash from municipal waste incineration in China.

Bottom ash problem is especially serious in Zhejiang province, which is one of the most developed areas in East China. Till 2006, there were 17 MSW incinerators in six cities with the treatment capacity of 10,150 t/d, accounting for 24.6% in amount and 25.4% in treatment capacity of China [3,19]. Nowadays, about 30% of MSW in Zhejiang are treated by incineration resulting in a large volume of bottom ash, which accounted for about 30% of those generated in the whole country [3,19]. The environmental impact of MSW incineration has received a lot of attention. However, the issues have mainly been focused on the gas emissions or fly ash, many of which deal with heavy metals. There are few studies on the POPs in the bottom ash of MSWI [20,21].

The objective of this study was to conduct a comprehensive investigation on PCB and PAH levels and patterns in bottom ash from municipal waste incinerators in six cities (Hangzhou, Ningbo, Wenzhou, Jinhua, Shaoxing, and Jiaxing) of Zhejiang province, China. PAHs and PCBs were selected due to their strong carcinogenicity and mutagenicity. All the six cities with incinerators in Zhejiang province were sampled and analyzed to give all-round view on PAH and PCB distributions in bottom ash. Factors affecting the difference of PAHs and PCBs among the MSW incinerations in six cities were also discussed.

### 2. Materials and methods

#### 2.1. Sample collection and preparation

Bottom ash was sampled from six typical incinerators located in six different cities of Zhejiang province between June and August of 2008. The six cities included Hangzhou (HZ), Ningbo (NB), Wenzhou (WZ), Jinhua (JH), Shaoxing (SX) and Jiaxing (JX), as shown in Fig. 1. Detailed information about the six incinerators was listed in Table 1 [19]. The sampling time at each incinerator location lasted for 5 days. Samples were collected once every half hour during 7 h when MSW incineration was under steady state operating conditions each day. Approximately 5 kg bottom ash sample was taken from each incinerator every day. Meanwhile, about 10kg MSW sample was withdrawn from MSW pits of incinerator plants during the same sampling time. All about 25 kg bottom ash sample of each incinerator was mingled, dried at 50 °C, crushed and sieved through a 4 mm mesh, mixed thoroughly and stored in polyethylene bags at 4 °C ready for PAH, PCB and property analysis. The composition of MSWI feed [19] and the property of bottom ash were listed in Table 2.

# 2.2. Sample extraction and clean-up

Detailed information of the sampled incinerators<sup>a</sup>.

Ten grams of bottom ash were taken randomly from the prepared sample which had been crushed, sieved and mingled thoroughly. The samples were extracted for PAHs and PCBs in a



Fig. 1. Map of sampling locations.

Soxhlet apparatus for 24 h with toluene [8,9,22]. Before extraction, a measured aliquot of surrogate standard mixture ( $d_8$ -naphthalene,  $d_{10}$ -acenaphthene,  $d_{10}$ -phenanthrene,  $d_{12}$ -chrysene,  $d_{12}$ -perylene, 2,4,5,6-tetrachloro-m-xylene, PCB198 and nine <sup>13</sup>C-labeled PCB congeners PCB15, PCB28, PCB52, PCB118, PCB153, PCB180, PCB194, PCB208, PCB209) was added to each sample. Following extraction, the apparatus was air cooled for 30 min. The extracts were concentrated to approximately 2 mL by rotary evaporator and were solvent-exchanged to hexane, and then concentrated to 1–2 mL for the clean-up procedure.

For PAH analysis, clean-up and fractionation were achieved using columns packed with deactivated silica gel as described by Johansson and van Bavel [8,9] and Lundstedt et al. [23]. For PCB analysis, the column for clean-up was filled with sequential layers of KOH silica gel, 44% H<sub>2</sub>SO<sub>4</sub> silica gel, 22% H<sub>2</sub>SO<sub>4</sub> silica gel, AgNO<sub>3</sub> silica gel, and dried sodium sulfate. Hexane (50 mL) was used to prewash the multilayer column, and the extracts that had been concentrated by rotary evaporation flowed through the column. The target compounds were eluted from the column with 120 mL hexane at 2.5 mL/min [21,22]. The elutions were subsequently concentrated to 1 mL for analysis.

#### 2.3. PAH analysis

PAHs were analyzed by Varian GC 3800/Saturn 2200 ion trap mass spectrometer in the modes of EI/MS and selected ion storage (SIS). Chromatographic separation was conducted on VF-5 ms capillary column ( $30 \text{ m} \times 250 \,\mu\text{m}$  i.d. with a film thickness of 0.25  $\mu$ m). The heating procedure for the oven temperature was as follows: start temperature  $60 \,^\circ\text{C}$  held for 1 min;  $60\text{-}210 \,^\circ\text{C}$  at

	-				
Sampling location	Sample name	Type of incinerator	Treatment capacity (t/d)	Bottom ash pretreatment	
				Water quenched	Magnetic separation
Hangzhou	HZ	Grate furnace	650	Yes	Yes
Ningbo	NB	Grate furnace	1000	Yes	Yes
Wenzhou	WZ	Grate furnace	350	Yes	Yes
Jinhua	JH	Fluidized bed	400	No	No
Shaoxing	SX	Fluidized bed	400	No	No
Jiaxing	JX	Fluidized bed	1000	No	No

Table 1

#### Table 2

Composition of MSWI feed<sup>a</sup> and property of bottom ash from six incinerators (%).

	MSWI feed	1		Bottom as	sh					
	Plastic	Paper	Wood	Textile	Leaf	Slag/earth	Kitchen waste	Metal	рН	Organic matter
HZ	9.52	0.03	1.65	16.51	0.28	44.20	27.79	0.02	11.14	4.07
NB	29.50	3.93	5.18	12.43	1.04	26.93	20.71	0.28	9.76	9.21
WZ	7.90	5.30	4.60	14.35	1.33	36.56	29.72	0.24	10.23	4.02
JH	12.65	3.29	7.59	14.47	0.76	38.90	21.76	0.58	9.08	2.48
SX	8.60	2.90	5.30	12.70	1.10	40.00	29.10	0.30	11.81	1.03
JX	1.73	0.61	3.29	12.13	1.54	41.00	39.38	0.31	10.60	1.21

<sup>a</sup> From Ref. [19].

#### Table 3

Amount of PAHs in bottom ash from the six cities ( $\mu$ g/kg).

Compound	PEF <sup>a</sup>	HZ	NB	WZ	JH	SX	JX
Naphthalene		235.77	955.80	296.42	222.26	134.50	203.45
Acenaphthylene		52.48	32.57	119.96	26.27	23.56	25.84
Acenaphthene		32.51	117.47	61.06	38.38	21.53	28.03
Fluorene		156.08	530.06	308.80	145.02	101.68	122.44
Phenanthrene		1075.70	2554.76	1293.33	550.98	508.91	683.49
Anthracene		1076.29	440.57	181.67	553.52	512.04	78.10
Fluoranthene		546.71	982.16	496.57	217.59	297.35	369.71
Pyrene		435.74	1124.80	399.38	183.32	253.68	304.71
Benz[a]anthracene <sup>b</sup>	0.1	118.66	52.63	109.13	73.30	77.30	96.66
Chrysene <sup>b</sup>	0.01	156.39	46.39	155.14	79.38	110.28	122.42
Benzo[b]fluoranthene <sup>b</sup>	0.1	136.50	ND <sup>c</sup>	72.16	58.76	95.96	70.52
Benzo[k]fluoranthene <sup>b</sup>	0.1	74.71	ND	13.04	0.34	35.60	59.72
Benzo[a]pyrene <sup>b</sup>	1	66.34	46.70	58.99	51.56	50.13	58.36
Indeno[1,2,3 <i>-cd</i> ]pyrene <sup>b</sup>	0.1	62.68	ND	55.78	ND	ND	ND
Dibenzo[a,h]anthracene <sup>b</sup>	1 <sup>d</sup>	ND	ND	ND	ND	ND	ND
Benzo[g,h,i]perylene		58.10	ND	ND	46.90	ND	50.83
Total 16 PAHs		4284.65	6883.91	3621.44	2247.56	2222.53	2274.28
Total carcinogenic PAHs		615.28	145.72	464.24	263.34	369.27	407.69
Total B[a]Peq		121.03	63.13	90.34	72.98	78.58	85.31

<sup>a</sup> Potency equivalence factor (PEF) values from Ref. [37].

<sup>b</sup> Carcinogenic PAHs.

 $^{\circ}$  ND, not detected. The detection limits for 16 PAHs were in the range of 0.45–0.62  $\mu$ g/kg.

<sup>d</sup> Value from Ref. [38].

10 °C/min; 210–260 °C at 1.5 °C/min; 260–290 °C at 3 °C/min. All injections were made in the splitless mode. The carrier gas was helium (99.999%) at a flow rate of 1.0 mL/min. Two microliters of the final extract were injected on the GC column with an injector temperature of 300 °C. Two to three specific ions of each PAH were used to identify every compound, and the base peak corresponding to the molecular mass was used for quantification. In the described protocol, the surrogate standard was used to quantify 16 target compounds. The operating parameters of the ion trap mass spectrometry (ITMS) system were set as follows: electron impact at 70 eV, scan rate at 0.5 s, RF storage level at 45 u, axial modulation at 4.5 V, ion source temperature at 200 °C, transfer line temperature at 280 °C, manifold temperature at 50 °C, 50  $\mu$ A for the filament emission current, electron multiplier voltage at 1650 kV and resonant mode for collision-induced dissociation (CID) [11,24].

The 16 PAHs considered as primary environmental concern to USEPA were naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenz[a,h]anthracene, and benzo[g,h,i]perylene.

# 2.4. PCB analysis

PCBs were analyzed by GC 3800/Saturn 2200 ion trap mass spectrometry in mode of MS/MS and multiple reaction monitoring (MRM). GC conditions were the same as those of PAH analysis except for injector temperature (300 °C) and heating procedure for the oven temperature: start temperature 80 °C held for 1 min;  $80-160 \circ C$  at  $15 \circ C/min$  and held for 1 min;  $160-310 \circ C$  at  $3 \circ C/min$ . The operating parameters of the ITMS system were the same as those of PAH analysis [24].

Twenty-eight PCB congeners with 2–10 chlorine atoms were analyzed. Their IUPAC numbers were 8, 18, 28, 44, 52, 66, 77, 81, 101, 105, 114, 118, 123, 126, 128, 138, 153, 156, 157, 167, 169, 170, 180, 187, 189, 195, 206 and 209. Among them, PCB8 was DiCB, PCB18 and PCB28 were TrCBs, PCB44, PCB52, PCB66, PCB77, PCB81 were TeCBs, PCB101, PCB105, PCB114, PCB118, PCB123, PCB126 were PeCBs, PCB128, PCB138, PCB153, PCB156, PCB157, PCB167, PCB169 were HxCBs, respectively.

# 2.5. Statistical analysis

All the statistical analysis was performed using SPSS software version 13.0. One-way analysis of variance (ANOVA) was used for statistical comparisons and Pearson coefficient was used for correlation analysis. A value of p < 0.05 was considered to be statistically significant.

### 3. Results and discussion

### 3.1. Total 16 PAHs in MSWI bottom ash

Total PAHs in MSWI bottom ash were detected in the range from 2222.53  $\mu$ g/kg in SX city to 6883.91  $\mu$ g/kg in NB city (Table 3). The PAH levels were much higher than those in soil from Taizhou (330–790  $\mu$ g/kg) [25], Dalian (796  $\mu$ g/kg) [6] in China, and Switzerland (50–619  $\mu$ g/kg) [26]. Thus a particular attention should be

Com	oarison o	of PAH	contents in	bottom a	ash from	different	areas be	etween	this study	v and	literatures	(ug/)	kg).
										,		(10)	

	Total 16 PAHs	Total carcinogenic PAHs	Benzo[a]pyrene	Reference
Zhejiang, China	2222.53-6883.91	145.72-615.28	46.70-66.34	This study
Orebro, Sweden	480-3590	89-438	11–59	[8,9]
Roma, Italy	44.3-79.1	7.0-14.4	0.4-1	[10]
Vernaison, France	126.3-209.4	24.3-40.4	6-10	[27]
London, UK	NA <sup>a</sup>	47–292	147	[7]

<sup>a</sup> NA, not available.

paid to PAHs when bottom ash is reused or landfilled. When compared with other incineration plants, the PAH levels were consistent with those from one MSW plant in Hangzhou, China, with the range of 1961.0–2420.2  $\mu$ g/kg [11]. Compared with the data from other countries (Table 4), total PAH concentrations in this study were in the similar range as those found in Orebro, Sweden (480–3590  $\mu$ g/kg) [8,9], while were higher than those in Roma, Vernaison and London [7,10,27].

The PAH levels in MSWI bottom ash from different cities followed the sequence of NB>HZ>WZ>JX>JH>SX. Interestingly, it was found that the PAH concentrations in cities (JH, SX and JX) with fluidized bed incinerator were less than those in cities (HZ, NB and WZ) with grate furnace incinerator. It seemed that fluidized bed incinerator had better combustion efficiency.

Organic matter content is an indicator for combustion efficiency. As shown in Table 2, organic matter contents in bottom ashes varied from 1.03% to 9.21%, indicating differential conditions of incinerator operation among the six cities in Zhejiang province. In general, optimal combustion control did not achieve in most incinerators especially in grate furnaces. The highest level of organic matter (9.21%) in bottom ash was found in NB, which could be partly due to the incomplete combustion of MSW in the incinerator. This might be a possible explanation for the finding that the PAH concentration in the bottom ash of NB city was the highest among the six samples, since PAHs are typical products of incomplete combustion [18]. It has been reported that PAHs are associated with organic carbon in MSWI ash [28]. A positive correlation ( $R^2 = 0.95$ , p < 0.05) was observed between organic matter content and total PAH amount. Such correlation would be useful as the results from analysis of organic matter could serve as an indicator for PAH content.

In addition, pH of the bottom ash is also an indicator for combustion efficiency. At high temperature, carbonates and other compounds in MSW could be thermal decomposed to oxides such as CaO, resulting in high pH above 11.5 in bottom ash. From the relatively low pH values in Table 2, a conclusion could be drawn that the combustion efficiencies were not high at all the six incinerators. However, there was no significant difference between pH values in bottom ashes from fluidized bed incinerators and those from grate furnace incinerators.

Furthermore, MSWI feed composition is also a key factor for combustion efficiency. Co-combustion studies found a positive impact of plastics on the burnout by increasing the heating value of the fuel. However, large quantities of plastic could not burn easily, and thus might increase the final PAH concentration in bottom ash. Previous studies have demonstrated that PAHs could be generated from incineration of combustible materials in MSW such as plastics including high-density polyethylene (HDPE), polypropylene (PP), polyvinyl chloride (PVC) [29,30], PS (polystyrene) [31] and rubber [32,33]. As shown in Table 2, the highest plastic percentage in the MSWI feed was found in NB, which could lead to the highest PAH content in the incinerator. Furthermore, PVC was used most frequently in all the six cities based on our survey. A positive correlation ( $R^2 = 0.72$ , p < 0.05) was observed between PAH and plastic contents, indicating the plastic content of input MSW had effect on PAH formation during incineration.

#### 3.2. Carcinogenic PAHs in MSWI bottom ash

Since several PAH homologues are considered mutagenic or genotoxic and some may be carcinogenic, the USEPA issued a guidance document for PAHs in which some were classified as carcinogens [34]. The levels of carcinogenic PAHs were between 145.72 and 615.28 µg/kg as depicted in Table 3. There was no significant correlation between the total amount of PAHs and carcinogenic PAHs. The sample with the highest amount of total PAHs in NB city contained the lowest percentage of carcinogenic PAHs. The highest value of carcinogenic PAHs was found in HZ bottom ash. The percentages of carcinogenic PAHs found in ash extracts were between 2.1% (NB) and 17.9% (JX). With respect to the individual carcinogenic PAH, benz[a]anthracene and chrysene were dominant in all the samples. Table 4 shows the reported values for carcinogenic PAHs in MSWI ash ranging from 7 to 438 µg/kg [7–10,27]. The levels of carcinogenic PAHs in this study seemed to be in the similar range (145.72–615.28 µg/kg).

Benzo[*a*]pyrene, the most potent carcinogenic PAH, is usually measured as an indicator PAH. A comparison between this study and other investigations from literatures was shown in Table 4. Levels of PAHs found in the MSWI bottom ash in this study were similar to those reported in the literatures. In order to evaluate the toxicity of PAHs, carcinogenic toxic equivalent quantity (TEQ) labeled as B[*a*]Peq was calculated by multiplying the concentration of carcinogenic PAH by the corresponding potency equivalence factor (PEF), which were then summed [35,36]. As shown in Table 3, the total B[*a*]Peq of PAHs ranged from 63.13 µg TEQ/kg in NB to 121.03 µg TEQ/kg in HZ. The toxicity profiles were similar among these six cities, mainly due to the dominant levels of benzo[*a*]pyrene with the highest PEF value (1.0) in all the samples.

#### 3.3. PAH patterns in MSWI bottom ash

When relative concentration ratios of the 16 PAHs were compared, the distributions of PAH homologues in different samples were found to be very similar. This could be observed in Table 3 and Fig. 2, where the PAH distributions of different rings were shown. Phenanthrene, anthracene and fluoranthene were the predominant homologues in bottom ash, consistent with previous studies by di Palo et al. [10] and Dugenest et al. [27], who also found that phenanthrene, fluoranthene were the most abundant PAHs in bottom ash.

With regard to the grouped PAHs, the levels of three-ring and four-ring PAHs were dominant, while those of five-ring and sixring PAHs were relatively lower in all bottom ash samples (shown in Fig. 2). A small difference was observed in the NB MSWI bottom ash, as shown in Table 3 and Fig. 2. This sample showed relatively larger amount of pyrene and two-ring PAHs when compared with the other samples.

# 3.4. PCBs in MSWI bottom ash

PCB concentrations in MSWI bottom ash from the selected six cities were listed in Table 5. Total PCB concentrations ranged from 1.00 to  $1.31 \mu g/kg$ , while the coplanar PCBs were in the range of



**Fig. 2.** PAH distributions of different rings in bottom ash from MSWI in six cities of Zhejiang province.

0.08–0.52  $\mu$ g/kg. The PCB levels were lower than those in natural soil in Taizhou (11–100  $\mu$ g/kg) [25], Shenyang (6.4–15.2  $\mu$ g/kg) [37] in China, and Switzerland (1.1–12  $\mu$ g/kg) [38]. When compared with those in incineration plant reported in the literatures (mean concentration: 0.3–95  $\mu$ g/kg) [18], the PCB levels in MSWI bottom ash from Zhejiang province were also relatively low. However, the total PCB and coplanar PCB values were higher than those from Kyoto in Japan [17], and the coplanar PCBs were in the same level as compared with those from Ibaraki in Japan [39] as shown in Table 5. TEQ values were also given and compared with those from literatures. NB ash had the highest TEQ value (1.52 pg TEQ/g), which was similar with those observed in Kyoto and Ibaraki in Japan [17,39]. However, TEQ values from other five cities in Zhejiang, ranging from 0.06 to 0.12 pg TEQ/g, were one order magnitude lower than those from Kyoto and Ibaraki in Japan [17,39].

The total PCB contents in MSWI bottom ash from the six cities followed the sequence of NB>HZ>JX>SX>WZ>JH. Compared with the average PCB concentration  $(1.06 \ \mu g/kg)$  released from fluidized bed incinerators in JH, SX and JX cities, the mean value of PCB concentrations  $(1.17 \ \mu g/kg)$  obtained from MSW grate furnace incinerators in HZ, NB and WZ cities was a little higher, though no statistically significant difference was observed (p > 0.05). In addition, taking the sampling and analytical errors into consideration, PCBs in bottom ash from either fluidized bed incinerator or grate furnace incinerator were in the same level. Yang et al. [40] also compared the levels of PCBs and PCDD/Fs formed from different types of MSW incinerators. They found that PCBs and PCDD/Fs from fluidized bed incinerator were lower when compared with those from stocker incinerator and gasified and melting incinerator.

Among the six cities, NB had the largest amount of PCBs in MSW residues  $(1.31 \mu g/kg ash)$ . This could partly result from the incomplete combustion, which was indicated by the highest organic matter. Moreover, concentration of coplanar PCBs had a strongly positive correlation with PVC found by Katami et al. [41]. In the present study, the highest PCBs in bottom ash from NB MSWI

# Table 5 Concentration of PCBs in bottom ashes $(\mu g/kg)$ in six cities from Zhejiang province.

Study area	Total PCBs	Co-PCBs	TEQ (pg TEQ/g)	Reference
Zhejiang, China				This study
HZ	1.16	0.52	0.10	
NB	1.31	0.28	1.52	
WZ	1.04	0.24	0.10	
JH	1.00	0.10	0.06	
SX	1.06	0.08	0.06	
JX	1.14	0.21	0.12	
Kyoto, Japan	0.62	0.06	0.85	[17]
Ibaraki, Japan	NA <sup>a</sup>	0.25	2.40	[39]

<sup>a</sup> NA, not available.



Fig. 3. Compositions of PCB homologues in bottom ash from MSWI in six cities of Zhejiang province.

were also in correspondence with the highest amount of plastic contents in MSW input. However, JX sample with the lowest content of plastic (1.73%) had a relatively high PCB concentration. In addition, there was no significant correlation between PCB concentrations and organic matter contents when the highest organic matter (9.21%) was ignored. Thus the effects of plastic content and combustion efficiency cannot be distinguished from each other when considering the formation of PCBs, and further study needs to be done to determine the importance of these two variables as well as other factors.

### 3.5. Congener distributions of PCBs in MSWI bottom ash

To compare PCB congener patterns, all data were normalized to the total sum of PCBs. From Fig. 3, similar profiles of PCB congeners were observed in all the six cities. It was also found that less chlorinated congeners such as DiCBs, TrCBs and TeCBs were more prevalent accounting for 61.7–92.3% of the total PCB concentrations, indicating that low chlorinated PCBs contributed to the majority of the total PCBs in bottom ash. Compared with the other five samples, bottom ash from NB MSWI had much higher percentage of HxCBs. Large amounts of PCB congeners at lower chlorination level were found, suggesting the initial PCB formation that had not been followed by further chlorination [42].

Among 12 coplanar PCB congeners, the most potent toxic PCB126 with the highest TEF was not detected in all the samples, while the most frequent coplanar congener detected was PCB105, followed by PCB169. PCB105 contributed 40.2–97.9% to the total coplanar PCB concentration. However, due to the low TEF value, the contribution of PCB105 to the total TEQ reached the highest value of 12.2%. On the contrary, PCB169 had the highest TEQ contribution (49.3–98.2%) while the concentration was lower as compared with PCB105. The PCB congeners with high TEQ contribution are of special concerns, rather than the high concentration congeners.

# 4. Conclusions

The levels of PAHs and PCBs in bottom ash from the six cities in Zhejiang province varied widely. The total PAH levels ranged from 2222.53 to 6883.91  $\mu$ g/kg, followed by the sequence of NB > HZ > WZ > JX > JH > SX. The patterns of PAH homologues were found similar in all the ashes and were dominated by low molecular weight PAHs such as phenanthrene, anthracene and fluoranthene. The total PCB concentrations in bottom ash from the MSW incineration ranged from 1.00 to 1.31  $\mu$ g/kg, and the total PCB contents in the six cities followed the sequence of NB > HZ > JX > SX > WZ > JH. Among PCB congeners, low chlorinated congeners DiCBs, TrCBs and TeCBs were more predominant, accounting for 62.7–92.3% of the total PCBs. Statistical analysis indicated that there was a positive relationship ( $R^2$  = 0.95, p < 0.05) between organic matter content and total PAHs, while no significant correlation was observed

between organic matter and PCBs, indicating that organic matter content has the potential to be served as an indicator for PAHs rather than PCBs in bottom ash. Moreover, when compared with grate furnace incinerator, fluidized bed incinerator seemed to have higher combustion efficiency, resulting in lower PAH concentration in bottom ash. Both plastic content in MSWI feed and combustion efficiency had important effects on the final levels of PAHs and PCBs in bottom ash. However, further study is required to investigate the effect of these two variables deeply, as well as other influencing factors. In conclusion, although bottom ash from MSWI has not been classified as hazardous material, the results of this study indicate that this kind of waste contains high levels of PAHs as well as some PCB congeners with high TEQ, and thus needs special treatment before landfill or other land use.

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