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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 four-ring PAHs. Total PCB concentrations in bottom ash ranged from 1.00 to 1.31 μg/kg, while the coplanar PCBs in the bottom ash were in the range of 0.08–0.52 μg/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.

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 10 kg 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

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

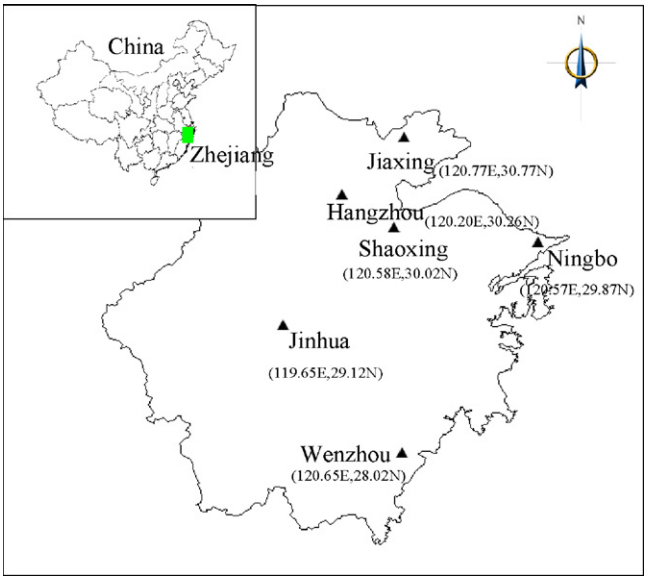

Fig. 1. Map of sampling locations.

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 ^{13}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_2SO_4 silica gel, 22% H_2SO_4 silica gel, $AgNO_3$ 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 m × 250 μm i.d. with a film thickness of 0.25 μm). The heating procedure for the oven temperature was as follows: start temperature 60 °C held for 1 min; 60–210 °C at

Table 1
Detailed information of the sampled incinerators[a].

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

[a] From Ref. [19].