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Size-dependent emission characteristics of airborne parent and halogenated PAHs from municipal solid waste incinerators in Shenzhen, China

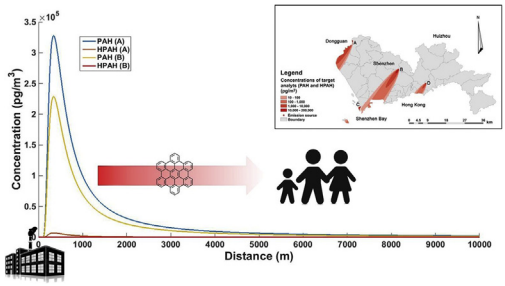
Wen-Bo Shu, Yi-Bo Zhao, Hong-Gang Ni*, Hui Zeng

Shenzhen Key Laboratory of Circular Economy, Shenzhen Graduate School, Peking University, Shenzhen 518055, China

HIGHLIGHTS

- High-ring and low-ring PAHs dominated in $PM_{2.5}$ and in PM_{10}, respectively.
- The size distributions of ΣPAH and ΣHPAH were characterized by bimodal peaks.
- PAH and HPAH from MSWI cannot be transported to far from incinerators.
- Overall, the intensity and extent of impacts from MSWIs might be opposite.
- The damage from incinerator to be far less than originally feared.

GRAPHICAL ABSTRACT



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ABSTRACT

Two waste incinerators were selected for investigation of size-dependent emission characteristics of airborne parent and halogenated PAHs (PAHs and HPAHs) and incidence of these pollutants from trash incineration. The concentrations of total PAHs (gas and particles with aerodynamic diameter 0.43 −10 μm) in ambient air of Shenzhen incinerators were at the lower end of the global range while those of HPAHs were higher than those of urban air in other studies. High-ring PAHs dominated in $PM_{2.5}$ (66% −86%), while low-ring PAHs dominated in PM_{10} (83%−86%). As for PAHs in gaseous phase, low-ring PAHs were collectively account for 86%−97%. ΣHPAH mainly enriched in coarse particles (>83%). The size distributions of ΣPAH and ΣHPAH were both characterized by bimodal peaks dominate in 9.0−10 μm and subordinate in 4.7−5.8 μm. PAHs and HPAHs enrichment in the coarse particles indicates that particle-bound PAHs and HPAHs from incinerators cannot travel great distances. Model simulation results showed the peak of airborne PAHs and HPAHs occurred in approximate 300 m from incinerator, then their concentrations reduced sharply. The extent of affected areas by municipal solid waste incinerators (MSWIs) seem very large, intensity of impacts can be neglected for the very low level of pollutants. Although waste incineration is perceived as most polluting way to manage waste, our study found the damage from incinerator to be far less than originally feared.

1. Introduction

With the population growth and rapid urbanization, the amount of municipal solid waste (MSW) in China is increasing sharply at an

* Corresponding author.
E-mail address: nihg@pkusz.edu.cn (H.-G. Ni).

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annual rate of 8–10% (He et al., 2010) and had grown to over million tons (Cheng and Hu, 2010). Accordingly, the disposal of MSW is urgently to be solved due to potential environmental issues. In general, the methods for MSW disposal primarily consist of landfill, composting and incineration (Peng et al., 2016a; Tian et al., 2012). Among them, incineration has been considered as a promising method for effective energy recovery, significant volume reduction (by up to 70% in weight and 90% in volume) and high degree detoxicity (Jin et al., 2013; Tian et al., 2012). Moreover, those toxic substances generated during incineration can be reduced with cleaning treatments. For example, incineration of MSW was addressed for reduction of the release of produced persistent organic pollutants (Verma et al., 2015). However, experience of the negative contrast indicated incineration is the most polluting way (far more polluting than landfills and coal burning) to make energy or to manage waste. For example, to make the same amount of energy, trash incinerators release 28, 2.5, 2, 3, 6–14 and 6 times as much dioxin, carbon dioxide, carbon monoxide, nitrogen oxides, mercury, and lead than coal power plant, respectively, according to the figures compiled by the Energy Justice Network mostly based on the published information by the U.S. EPA. (http://www.energyjustice.net/incineration/worsethancoal). There have long been controversies over incidence of pollutants emissions from municipal solid waste incinerators (MSWI).

A large number of reports on characteristics of various pollutants from MSWI have been published so far, but these publications mainly focused on determination of target compounds in MSWI samples including flue gas, fly ash, and residual ash, seldom on the space-time influence of the toxic pollutants from incinerators (Sakai et al., 2001; Tian et al., 2012; Wheatley et al., 1993; Wikstrom and Marklund, 2000). Also, nothing to date exists on particle size distribution of pollutants from incinerators and implications for dispersion range on the effect of the wind. A possible explanation for this knowledge gap may result from detection limits becoming an issue, when particulate-bound target compounds are split into multiple fractions (Zhang et al., 2012a). Again, wind velocity and wind direction are also key impact factors to the dispersion range of pollutants from incinerators. Unfortunately, these meteorological factors were not considered in the majority of previous field surveys. To deal with this problem, appropriate environmental model should be a good choice.

Waste incineration and vehicle exhaust are two major sources of halogenated polycyclic aromatic hydrocarbons (HPAHs) (Freire et al., 2009; Haglund et al., 1987; Horii et al., 2008; Ma et al., 2009; Nilsson and Ostman, 1993). These two sources have a very intimate relationship with urbanization. It is therefore, Shenzhen, a typical rapid urbanization region even in the world, provided very favorable conditions for study incidence of HPAHs derived from the two major sources. This study is one part of our ongoing research program about source apportionment and incidence of HPAH in urban atmosphere.

HPAHs are a category of compounds with one or more halogen substituent attached to the aromatic rings of corresponding parent PAHs (Fu et al., 1999), such as chlorinated PAHs (Cl-PAHs) and brominated PAHs (Br-PAHs). Like PAHs, HPAHs are a class of incineration byproducts and ubiquitous in the environment (Horii et al., 2008; Lewtas, 2007). PAHs and HPAHs have mutagenic and carcinogenic properties (Freire et al., 2009; Horii et al., 2008; Lewtas, 2007; Peng et al., 2016b). In reality, HPAH have higher toxicity in comparison of their parent PAHs, especially for those HPAH with more halogen atoms. Recent studies indicated that the environmental persistence of HPAHs also increases with the number of halogen substituents.

The present study investigated incidence of PAHs and HPAHs from incinerators, mindful of these incineration byproducts cannot be removed completely from flue gas even with advanced cleaning equipment. The tasks of the present study were to (1) analyze the size distributions of atmospheric particle-bound PAHs and HPAHs from incinerators; (2) estimate the PAHs and HPAH emission ratios via gaseous and particle phase from incinerator; (3) examine the temporal trends and spatial variation characteristics of airborne parent and HPAHs emission load from MSWI. The present study provides the preliminary data on the incidence of airborne parent and HPAHs emitted from MSWI in urban Shenzhen, China. We attempt to simulate and visualize emission patterns of targeted pollutants change in future with improving incineration techniques, growing population and wastes. These results necessarily facilitate the control policies of pollution associated with MSWI in urbanized areas.

2. Materials and methods

2.1. Sample collection and preparation

All samples were collected outside selected residential apartments around the two incinerators with the largest capacity in urban Shenzhen, South China. Sampling was conducted at two heights: two rooftop levels of 5 and 25 m above the ground. Instead of collection samples directly from MSWI, sampling points were set at the downwind of MSWI (2000 m to A and 500 m to B, respectively) as an alternative method for assessment of impacts on the ambient air environment (Fig. 1 and Fig. S1 in supplementary data, SD thereafter). Size-segregated particle samples were collected using an Ambient Eight Stage Cascade Impactor Sampler (Thermo Fisher Scientific Inc, Waltham, MA USA) loaded with 81 mm diameter glass fiber filters (Whatman International, Maidstone, England) in the fractions of <0.43, 0.43–0.65, 0.65–1.10, 1.10–2.10, 2.10–3.30, 3.30–4.70, 4.70–5.80, 5.80–9.00, and 9.00–10.0, and >10 μm at a flow rate of 28 L min^{-1} with a conventional vacuum pump. Each sample was collected over 24 h, from March 7 to 12 for MSWI A, April 2 to 7 in 2016 for MSWI B. Sampling was made at heights 25 m for MSWI A and 5 m for MSWI B above ground, respectively. $PM_{2.5}$ and gaseous samples were collected using a high volume air sampler with $PM_{2.5}$ cutter housing a glass fiber filter (GFF; 180 mm × 230 mm; Whatman International, Maidstone, England) and a polyurethane foam plug (PUF; 6.5 cm diameter and 8.0 cm thick). In total, 80 size segregated particle samples were collected by the Eight Stage Cascade Impactor Sampler and 20 pairs ($PM_{2.5}$ + gas) were collected with TH-1000H Automatic Large Volume TSP Sampler (Tianhong Instruments Co., Ltd, China) at a constant flow rate of 1200 L min^{-1} with a conventional vacuum pump.

Each sample was spiked with surrogate standards (naphthalene-d_8, acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, and perylene-d_{12}, Table S1) and Soxhlet extracted for 24 h with 150 mL of a 3:1 by volume mixture of dichloromethane and hexane. The extracts were concentrated to 2 mL with a rotary evaporator, and then purified and fractionated on a chromatography column (300 mm × 10 mm i.d.) packed with alumina: silica gel (6:12, v:v). The column was eluted with a mixture of dichloromethane and hexane (60 mL; 3:7, v:v) after passage of 6 mL of hexane (this 6 mL fraction was discarded). The 60 mL fraction which contains the most of the target compounds was concentrated and further reduced to a final volume of 0.5 mL with a gentle N_2 stream. A known amount of the internal standards (e.g., 2-fluorobiphenyl, and *p*-terphenyl-d_{14}) was added to the extracts prior to instrumental analysis.