



## Effects of characteristics of waste incinerator on emission rate of halogenated polycyclic aromatic hydrocarbon into environments

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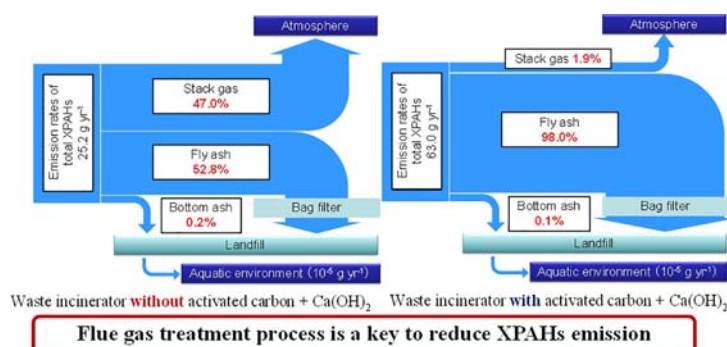
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### HIGHLIGHTS

- XPAH concentrations were affected by characteristics of waste incinerator.
- XPAH congener profile was similar in stack gas and urban air.
- Dominant emission pathway of XPAHs into atmosphere was stack gas.
- Emission rates of XPAHs were affected by flue gas treatment process.
- Incinerator-produced XPAHs have greater emission rates than PCDDs/Fs and PCBs.

### GRAPHICAL ABSTRACT



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### ABSTRACT

We determined the concentrations of halogenated polycyclic aromatic hydrocarbons (XPAHs), some of which are carcinogenic and/or mutagenic compounds, in fly and bottom ashes and stack gas collected from waste incinerators in Japan. The dominant XPAHs in stack gas were consistent with those in the urban atmosphere. The dioxin-like toxic equivalent (TEQ) concentration ranges of the XPAHs in stack gas, fly ash, and bottom ash were 0.00497–20.5 ng-TEQ m<sup>-3</sup>, 0.0541–101 ng-TEQ g<sup>-1</sup>, and 0.000914–2.00 ng-TEQ g<sup>-1</sup>, respectively. The TEQ concentrations of the XPAHs targeted in this study were higher than those of polychlorinated dibenzo-*p*-dioxins/dibenzofurans and polychlorinated biphenyls reported in the literature. The annual amounts of XPAHs produced in the waste incinerators ranged from 25.1 to 881 g. The mass balance of XPAHs in each waste incinerator was calculated to evaluate the emission rate of XPAHs from waste incinerators. Less than 6.7% of the XPAHs produced in the waste incinerators were emitted into the atmosphere from the facilities in which the flue gas was treated by using a combination of bag filter and activated carbon. In contrast, from the facility using a bag filter only, approximately 50% of the XPAHs produced were emitted into the atmosphere. Thus, the flue gas treatment process appears to be a key determinant of the emission rate of XPAHs produced during waste incineration.

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**Abbreviations:** BrPAHs, brominated polycyclic aromatic hydrocarbons; ClPAHs, chlorinated polycyclic aromatic hydrocarbons; IWI, industrial waste incinerators; MSWI, municipal solid waste incinerators; TEQ, toxic equivalent; XPAHs, halogenated polycyclic aromatic hydrocarbons; PCDD/Fs, polychlorinated dibenzo-*p*-dioxins/dibenzofurans; PCBs, polychlorinated biphenyls.

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## 1. Introduction

Halogenated polycyclic aromatic hydrocarbons (XPAHs), such as chlorinated and brominated polycyclic aromatic hydrocarbons (CIPAHS and BrPAHs), are characterized by the substitution of one to several hydrogen atoms on the parent PAHs by halogens (e.g., chlorine or bromine atoms). Like PAHs, XPAHs are unintentionally produced by combustion processes and have mutagenic and carcinogenic properties mediated through activation of the aryl hydrocarbon receptor (Boström et al., 2002; Colmsjö et al., 1984; Löfroth et al., 1985; Menzie et al., 1992; Ohura et al., 2007).

In previous studies, the concentrations of XPAHs produced during the incineration of wastes were comparable to those of polychlorinated (PCDD/Fs) and polychlorinated biphenyls (PCBs). The dioxin-like toxic equivalent (TEQ) concentrations of CIPAHS in fly ash collected from waste incineration facilities ranged from <0.0000300 to 104 ng-TEQ g<sup>-1</sup>, which were approximately 1.5 to 30 times those of PCDD/Fs and PCBs (Abad et al., 2000; Everaert and Baeyens, 2002; Horii et al., 2008). Waste incineration facilities are an important emission source of XPAHs into the urban atmosphere (Horii et al., 2008; Kitazawa et al., 2006; Ma et al., 2009; Yoshino and Urano, 1997). We previously reported the occurrence and profiles of XPAHs in fly and bottom ashes (Horii et al., 2008), whereas information about these compounds in stack gas samples is limited (Horii et al., 2011). The XPAHs in fly and bottom ashes end up in landfills, and stack gas is emitted into the atmosphere. Therefore, among these sources, stack gas is considered to be the most important emission pathway of XPAHs from a waste incinerator into the environment. Furthermore, to control XPAH emissions and reduce their impacts on humans and other organisms, it is necessary to identify the factors that affect the behavior of XPAHs in waste incinerators. To understand their behavior, a mass balance analysis should be carried out, which requires a set of concentrations of XPAHs in fly and bottom ashes and stack gas in the waste incinerator facility.

To perform a mass balance analysis of the XPAHs in waste incinerators, we simultaneously collected stack gas, fly ash, and bottom ash from each of three municipal solid waste incinerators (MSWIs) and four industrial waste incinerators (IWI) and determined the concentrations of 33 XPAHs. To identify potential factors that affect the behavior of XPAHs in the waste incinerators, the relationship among the emission rates, compositions of the XPAHs, and characteristics of the waste incineration facilities (e.g., incinerator type, exhaust gas treatment process, and capacity to treat waste) were also investigated. This is the first report to simultaneously determine the concentrations of XPAHs in a set of samples (stack gas, fly ash, and bottom ash) collected from an individual incinerator, which should be more useful information compared with an experiment to determine those in incomplete samples to understand the behavior of XPAHs unintentionally generated by waste incinerators.

## 2. Materials and methods

### 2.1. Chemicals

Analytical standards for 24 CIPAHS and 9 BrPAHs (Table 1) either were purchased from Tokyo Chemical Industry Co. (Tokyo, Japan) or Sigma-Aldrich (St. Louis, MO, USA) or were synthesized in our laboratory. Isotope-labeled fluoranthene-<sup>13</sup>C<sub>6</sub>, chrysene-<sup>13</sup>C<sub>6</sub> and benzo[a]pyrene-<sup>13</sup>C<sub>4</sub> were purchased from Cambridge Isotope Laboratories (Andover, MA, USA) and used as recovery standards. Chrysene-*d*<sub>12</sub> and benzo[a]pyrene-*d*<sub>12</sub> were purchased from AccuStandard (New Haven, CT, USA) and used as internal standards. Reduced copper and the solvents toluene, nonane, dichloromethane, and hexane were obtained from Wako Pure Chemical Industries (Tokyo, Japan).

### 2.2. Sampling

We collected a set of samples (stack gas, fly ash, and bottom ash) from each of three MSWIs and four IWIs in Japan in 2012 by using a

modified version of the Japanese Industrial Standard (JIS) K0311 sampling method for PCDD/Fs and PCBs (JIS, 2005). MSWIs 1–3 had continuous-feed stoker-type incinerators with capacities of 1875–3750 kg h<sup>-1</sup>. IWIs 1–3 had fixed-grate batch-type incinerators with capacities of 200–2000 kg h<sup>-1</sup>. IWI 4 was a continuous-feed fluidized-bed incinerator with a capacity of 2000 kg h<sup>-1</sup>. Most of the incinerators used bag filters, activated carbon, and Ca(OH)<sub>2</sub> for exhaust gas treatment, whereas IWI 2 used only a bag filter. Details about the waste incinerators (e.g., stack gas flow rates, flow rates of fly and bottom ashes, and operating times) are summarized in Table 2. The sampling period was 4 h to capture the variability of the XPAH emission rate over time.

### 2.3. Analytical procedures

The concentrations of XPAHs in stack gas and in fly and bottom ashes were determined according to an established method (JIS, 2005; Horii et al., 2008). Briefly, the samples were spiked with 2.5 ng of each recovery standard (fluoranthene-<sup>13</sup>C<sub>6</sub>, chrysene-<sup>13</sup>C<sub>6</sub> and benzo[a]pyrene-<sup>13</sup>C<sub>4</sub>) and were extracted by a liquid–liquid extraction method or a Soxhlet extraction method. The extracts were purified and fractionated by using an activated carbon cartridge column (Carboxene 1016, 200 mg, Supelco, St. Louis, MO, USA) connected to a silica gel cartridge column (Supelclean LC-Si, 2 g, Supelco). The cartridge columns were eluted with 20 mL of 10% dichloromethane/hexane. After removal of the silica gel cartridge, the activated carbon cartridge was reversed and then eluted with 120 mL of toluene. The toluene fraction containing XPAHs was concentrated after spiking with 2.5 ng of each internal standard (chrysene-*d*<sub>12</sub> and benzo[a]pyrene-*d*<sub>12</sub>).

The concentrations of XPAHs were determined by gas chromatography–high-resolution mass spectrometry (JMS-800D, JEOL, Tokyo, Japan) using electron-impact ionization and selected-ion monitoring. To separate compounds, we used a 60-m DB-5 ms fused silica capillary column (0.25 mm id × 0.25 μm film thickness; Agilent Technologies, Santa Clara, CA, USA). An aliquot (2 μL) of sample was injected in the splitless mode. The injector temperature was 280 °C. The column oven temperature was programmed as follows: hold at 80 °C for 1 min, ramp to 170 °C at 20 °C min<sup>-1</sup>, ramp to 260 °C at 4 °C min<sup>-1</sup>, ramp to 270 °C at 1 °C min<sup>-1</sup>, ramp to 320 °C at 8 °C min<sup>-1</sup>, and hold at 320 °C for 10 min. The TEQ concentrations of XPAHs were calculated based on toxic potencies relative to benzo[a]pyrene (Ohura et al., 2007, 2009).

### 2.4. Quality assurance/quality control

The calibration curves for XPAHs were linear over the range 0.4–100 ng mL<sup>-1</sup> (0.4, 1, 4, 20, 100 ng mL<sup>-1</sup>) of standard solutions (*R*<sup>2</sup> > 0.99). Recoveries of the recovery standards ranged from 69% to 107% in all samples. The limit of quantification of the instrument was calculated as 3 times the standard deviation from five injections of blank samples that had a signal-to-noise ratio of 3–10 (Table S1). Peaks were identified by comparison of the retention times of samples and standards if the signal-to-noise ratio was >3, and the peaks were quantified if the target/qualifier ion ratios were within 15% of theoretical values. None of the XPAHs analyzed in this study were detected in blank samples. During the stack gas sampling, no breakthrough of XPAHs was observed.

## 3. Results and discussion

### 3.1. Total concentrations of XPAHs in stack gas, fly ash, and bottom ash

The concentrations of 33 XPAHs were determined in stack gas (gas phase) and fly and bottom ashes (solid phases) collected from the seven waste incinerator facilities. Fig. 1 shows the total concentrations of XPAHs (sum of concentrations of each XPAH measured in this study), and Table S1 lists the concentrations of individual XPAHs. The

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