



Characterization and mass balance of dioxin from a large-scale municipal solid waste incinerator in China

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ABSTRACT

The input and output samples from existing large-scale municipal solid waste incinerator (MSWI) were collected and analyzed for polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in this study, aiming to evaluate PCDD/F characteristic and the corresponding mass balance through the whole system. The grate-type MSWI is equipped with semi-dry scrubber, activated carbon injection, and bag filter as air pollutant control devices (APCDs). Results showed that on the output side, the stack gas, bottom ash and fly ash presented their mean dioxin levels of 0.078 ng I-TEQ/Nm³, 12.94 ng I-TEQ/kg and 858 ng I-TEQ/kg, respectively, and showed large similarities in congener profiles. Instead, on the input side, the municipal solid waste (MSW) presented a mean dioxin level of 15.56 ng I-TEQ/kg and a remarkable difference in congener profiles compared with those of the output. The dioxin mass balance demonstrated that the annual dioxin input value was around 5.38 g I-TEQ/yr, lower than the total output value (7.62 g I-TEQ/yr), signifying a positive dioxin balance of about 2.25 g I-TEQ/yr.

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1. Introduction

With the rapid development of the municipal population, economy and living standard in China, the production of municipal solid waste (MSW) has increased dramatically with a rate of about 9% by year. Municipal waste treatment is defined as a series of operations to give the waste produced in cities the most advisable treatment from an economic and environmental perspective, according to volume of treatment, recovery, cost, and legal regulations (Conesa et al., 2011). It covers certain procedures including landfill, compost, and incineration. Landfill is the dominant MSW disposal method, accounting for more than 80% of the MSW disposal in China (Chen et al., 2008). However, enhancement of people's environmental awareness, continuous increments of landfill costs and limited landfill area have forced some metropolis to take the incineration into account as an alternative technology for treating MSW due to its advantages including significant volume reduction (about 90%), mass reduction (about 70%), detoxification and resource recovery (Li et al., 2004). Therefore, the government has planned to construct about 100 commercial-scale MSW incinerators, including the approximate seventy already existing facilities (Ni et al., 2009).

PCDDs and PCDFs emitted from MSWI, commonly known as dioxins and furans, have caused much public concern owing to their

high toxicity and potential carcinogenic and mutagenic effects (Huang and Buekens, 1995). Consequently, stringent regulations governing the dioxin emissions have been enforced in recent years in many countries. The formation mechanisms and emissions of dioxins have also been investigated by a lot of researchers extensively. Many studies showed that PCDD/Fs were mainly formed by de novo mechanism that is in the low-temperature post-combustion zone of incinerators through some heterogeneous catalytic reactions that occur in the flue gas-fly ash environment (Chang and Huang, 1999; Chang and Lee, 1998; Huang and Buekens, 1995; Johnke and Stelzner, 1992). Everaert and Baeyens (2002) supported the dominant role of the de novo synthesis through the analysis of PCDD/Fs profiles from large scale thermal processes in general and MSWI in particular. They observed that the PCDF/PCDD ratio exceeded 1 and the degree of chlorination points towards the dominant presence of HpCDD and OCDD within the dioxin group, and of PeCDF, HxCDF and HpCDF within the furan group.

Incineration of MSW not only gives rise to flue gases containing dioxins, but also produces a certain amount of solid residues, mainly including bottom and fly ashes, which may also contain significant levels of dioxins. Dioxins not only existed in output samples, but also input samples from the MSWI. Nevertheless, limited studies have been completed so far to evaluate a dioxin input/output balance from a full-scale MSWI. In Italy, Giugliano et al. (2001) performed an extensive research on a full scale plant to evaluate the presence of dioxins and establishing a mass balance over the whole system. They observed that the most significant contribution to the total emissions of dioxins derived from filter fly ash (71.5%)

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followed by slag (22%) and boiler fly ash (3.3%), while sludge and stack gas were almost ignorable. In Spain, Abad et al. (2000) carried out two sampling collection episodes to examine dioxin mass balance by the analysis of PCDD/Fs in MSW, stack gas, fly ash and slag. In one collection episode the dioxin input value was 1.33 g I-TEQ/yr, lower than the total output one (4.64 g I-TEQ/yr), revealing a non-thermal destruction. In the other collection the input value is 9.62 g I-TEQ/yr, higher than the output one (1.92 g I-TEQ/yr), revealing a thermal destruction.

At present, many negative reports on adverse health effects associated with dioxin exposure and MSWIs make people apprehend the dioxins from MSWIs. As a result, it is greatly essential to assess the real situation of dioxin emissions from all the input and output derived from a modern MSWI. To the best of our knowledge, there is no previous analysis of dioxin characteristic profiles from different components constituting the dioxin inputs and outputs, and no data reported on a comprehensive study on dioxin balance in a modern MSWI in China. Therefore, the main objective of this study was to present characterization and mass balance of dioxin from large-scale MSWI.

In this study, the congener profiles of PCDD/Fs were presented and compared among the stack flue gases, fly ashes, bottom ashes and MSW of existing full scale MSWI. An accurate dioxin mass balance was also performed by analyzing the levels of PCDD/Fs in the input/output samples from a Chinese MSWI. Furthermore, we assessed the dioxin removal efficiency of the modern APCDs.

2. Materials and methods

2.1. Basic information on MSWI

The continuously operating modern MSWI in the south of China was investigated in 2010. The capacity of the MSWI is 1040 tons per day. The Martin grate-type MSWI consists of two identical incinerating units, each with its own heat recovery system, semi-dry scrubber (SDS), activated carbon injection (ACI), bag filter (BG) and stack, which is recognized as one of effective dioxin minimization technologies. Ca(OH)₂ emulsion are added to trap the acid gases in the SDS. So the treatment processes are employed for reducing PCDD/F emissions to comply with the limit of 0.1 ng I-TEQ/Nm³. Operating conditions and parameters of the MSWI are depicted in Table 1.

2.2. Sampling procedures

To evaluate PCDD/PCDF characteristics and mass balance of the different components constituting the dioxin inputs and outputs. The sampling and quantification process for PCDD/Fs emission in the stack gas, bottom ash, fly ash and MSW samples were carried out according to the American Standard Method EPA 23A, EPA1613B and relative Chinese standard measurement procedure. Samples was collected three times in each sampling site under normal operating conditions of the MSWI and the average value was determined from three values. In addition, the APCDs inlet flue gas samples were collected concurrently in order to access the dioxin removal efficiencies of the modern APCDs. The sampling sites and flow sheet are shown in Fig. 1.

The flue gases were sampled isokinetically and sampling time is about 120–180 min. Before sampling of flues gas, the ¹³C₁₂-labelled EDF-4504 with 100 ng/mL in nonane as sampling standard was spiked to XAD-2 resin. The sampled flue gas volumes were normalized to the dry condition of 760 mm Hg and 273 °C, and denoted as Nm³. For obtaining representative ash samples, the bottom and fly ash samples were collected simultaneously every one hour during flue gases sampling to reach a total of 2 kg. Meanwhile, on the in-

Table 1
Operating conditions and parameters of the MSWI.

Operating conditions	Parameters
Annual capacity (t/yr)	345600
Incineration units	2
Operation ^a (h/day)	24
Production of fly ashes (t/yr)	7460
Production of bottom ashes (t/yr)	84585
Flue gas per unit (Nm ³ /h)	80000
Waste flow rate (t/h)	20
Middle temp. of furnace (°C)	1050
Exit temp. of boiler (°C)	181
Exit temp. of scrubber (°C)	172
Exit temp. of bag filter (°C)	161
Fuel	MSW

^a 8640 h/yr.

put side, 50 kg of MSW was taken from the grab bucket during feeding MSW. Because of general inhomogeneity of the MSW, MSW sample was collected by fully mixed multipoint sampling method. The duration of the sampling campaign was three days.

2.3. Analytical procedures

All samplings, as well as complex chemical analyses were carried out by South China Institute of Environmental Sciences, Ministry of Environmental Protection. Each sample was spiked with a mixture of ¹³C-labeled PCDD/Fs internal standards. Then the spiked samples were extracted for 24 h with 250 ml toluene. Prior to clean-up process, they were treated with sulphuric acid and base repeatedly until transparent. The clean-up procedure was performed with two columns: multi-layer silica gel and basic alumina column. Prior to HRGC/HRMS analysis, ¹³C-labeled PCDD/Fs recovery standard mixture was spiked.

2.4. HRGC/HRMS analysis

Instrumental analysis were performed by HRGC/HRMS on a 6890 series gas chromatograph (Agilent, USA) coupled to a high resolution mass spectrometer (Waters, AutoSpec Ultima). One µl of sample was injected by an auto-sampler in splitless mode. The mass spectrometer was operated in the selected ion monitoring (SIM) mode using a positive electron impact (EI+) source at a resolving power of 10000 (10% valley definition). The source temperature was 270 °C. Helium at a constant flow rate of 1.2 mL/min was the carrier gas. Chromatographic separation was achieved with a DB-5MS fused-silica capillary column (60 m × 0.25 mm i.d., 0.25 µm film thickness). The GC temperature program was performed as follows: initially oven temperature began at 150 °C (held for 3 min), secondly increased at 20 °C/min to 230 °C (held for 18 min), thirdly at 5 °C/min to 235 °C (held for 10 min), finally at 4 °C/min to 320 °C (held for 3 min). The detailed quantitative determination of PCDD/Fs was referred to US EPA method 1613B. The international toxic equivalency quantity (I-TEQ) was calculated using the international toxicity equivalency factor (I-TEF) (NATO/CCMS, 1988).

3. Results and discussion

3.1. Dioxin contents in the output and input samples from the MSWI

Table 2 lists the PCDD/F concentrations measured in the stack flue gases, fly ashes and bottom ashes of the MSWI, in terms of total values and I-TEQ, respectively. Fly ash samples presented mean levels of around 858 ng I-TEQ/kg, which meets the environmental quality standards for soil (less than 1 ng I-TEQ/g) in Japan

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