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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/Nm^3, 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.

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/Nm^3. 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 $^{13}C_{12}$-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 Nm^3. 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 input side, 50 kg of MSW was taken from the grab bucket during feedind 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.

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.

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 ^{13}C-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, ^{13}C-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

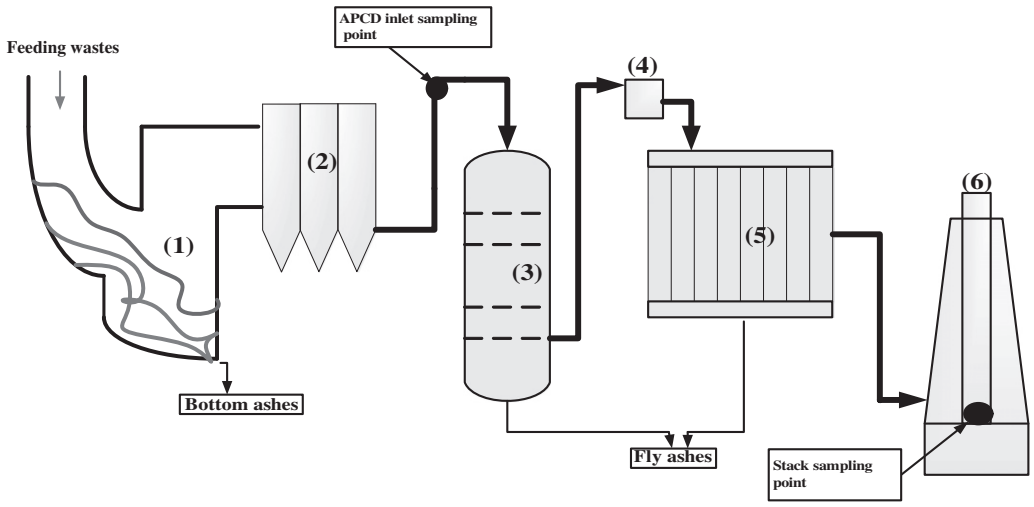

Fig. 1. Schematic overview process and sampling points of MSWI: (1) Furnace, (2) Boiler, (3) Semi-dry scrubber, (4) Injection of activated carbon, (5) Bag filter, (6) Stack.

Ministry of the Environment, similar to the values of MSWI in China (Bie et al., 2007; Jin et al., 2003) and some other countries (Hutzinger and Fiedler, 1993). However, the mean dioxin content from the bottom ash was 12.94 ng I-TEQ/kg, lower than that reported by Chen et al. (2006) (Taiwan: 16.2–52.3 ng I-TEQ/kg), Abad et al. (2003) (Spain: 20–30 ng I-TEQ/kg), and Kim et al. (2005) (Korea: 91 ng I-TEQ/kg). By the way, it is much lower than that in the fly ash as well. These results indicate that a high amount of dioxins are transferred into fly ash in the process of semi-dry lime scrubbing and bag filtration coupled with ACI adsorption for dioxins.

The PCDD/F levels in the MSW samplings are also listed in Table 2. The average concentration is around 15.56 ng I-TEQ/kg, slightly higher than that reported by Abad et al. (2000) (Spain: 4.4–13.3 ng I-TEQ/kg) and Makoto et al. (1998) (Japan: 1.3–16 ng I-TEQ/kg). It may be attributed to the fact that many countries have made great efforts to reduce the global PCDD/Fs emission levels.

3.2. Evaluation of dioxin removal efficiencies through APCDs

The dioxin removal efficiencies achieved by the MSWI at different sampling points are listed in Table 3. The dioxin concentration in flue gas before APCDs is 87.5 ng/Nm^3 or 3.87 ng I-TEQ/Nm^3. After passing the APCDs, the dioxin concentration decreases to 0.695 ng/Nm^3 or 0.078 ng I-TEQ/Nm^3 attributed to the function of the modern APCDs. The dioxin removal efficiency reached 99.2% with SDS + ACI + BF (in terms of total PCDD/F emissions) almost typically higher than those of the previous study, as shown in Table 3. It can be also seen that on the total PCDD/F I-TEQ emission, the dioxin removal efficiency (98%) is lower than that (99.2%) based on 17 congener concentrations owing to the distinct toxic equivalent factor (TEF) and removal efficiency of individual congener (Chang et al., 2002). In addition, the dioxin emission obtained in these samplings is far below the international emission standard limit established by the European Union Directive of 0.1 ng I-TEQ/Nm^3. But the dioxin emission of the previous study doesn't meet the standard limit (0.43 ng I-TEQ/Nm^3), as shown in Table 3. Based on these, it is concluded that the removal efficiency of the APCDs on PCDD/Fs emissions is adequate and different flue gas treatment equipment influences the emission of PCDD/Fs at the stack.

3.3. Characteristics of PCDD/F congener distribution from input to output

The congener profile of PCDD/Fs is often referred to as 'fingerprint' or 'signature'. Fingerprinting of dioxins has been extensively employed in source identification, atmospheric transport and

Table 2
Dioxin concentrations in the output and input from the MSWI (*n:* sampling times).

Output/input	PCDD	PCDF	Total PCDD/F	I-TEQ total PCDD/F	PCDFs:PCDDs
Output:					
Stack gas (ng/Nm^3) (n = 3)	0.247	0.448	0.695	0.078	1.82
Fly ash (ng/kg) (n = 3)	2700.5	5220.7	7921.2	858.2	1.94
Bottom ash (ng/kg) (n = 3)	148.8	103.5	252.3	12.94	0.70
Input:					
Municipal solid waste (ng/kg) (n = 3)	2084.5	246.8	2331.3	15.56	

Table 3
Dioxin removal efficiencies before and after APCDs.

	SDS + ACI + BF	DSI + ACI + BF (Chang et al., 2002)
Dioxin concentration at the APCD inlet (ng/Nm^3)	87.5	47.5
Dioxin concentration at the stack (ng/Nm^3)	0.695	1.63
Dioxin removal efficiency (%)	99.2	96.6
Dioxin concentration at the APCD inlet (ng I-TEQ/Nm^3)	3.87	4.93
Dioxin concentration at the stack (ng I-TEQ/Nm^3)	0.078	0.43
Dioxin I-TEQ removal efficiency (%)	98	91.3

SDS: semi-dry scrubber. ACI: activated carbon injection. DSI: dry sorbent injection. FF: bag filter.

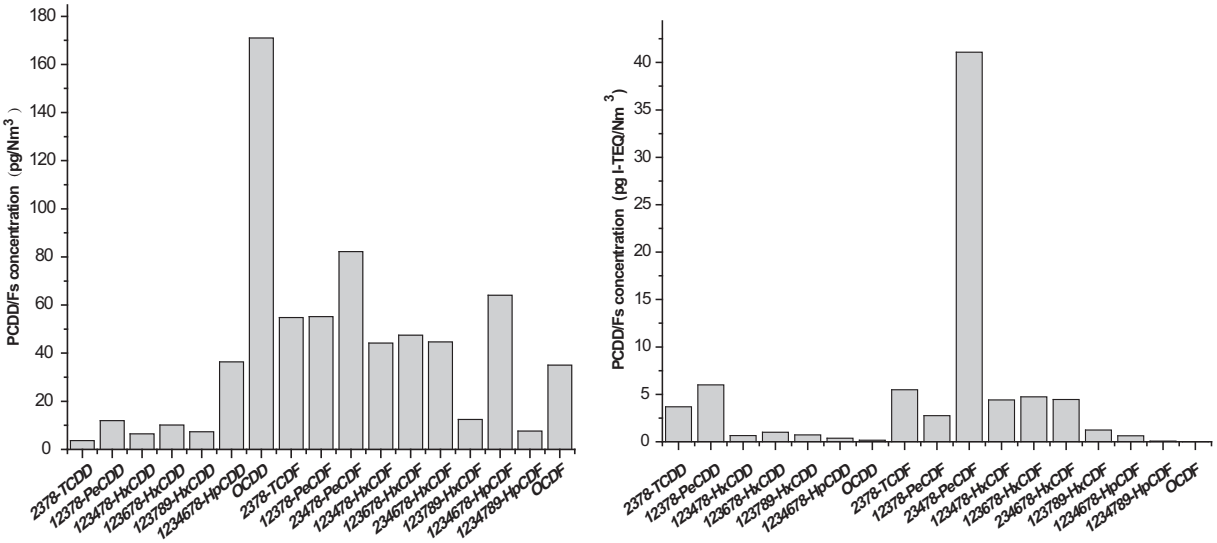

Fig. 2. Congener profiles of seventeen 2,3,7,8-chlorinated substituted PCDD/Fs containing in the stack flue gas.

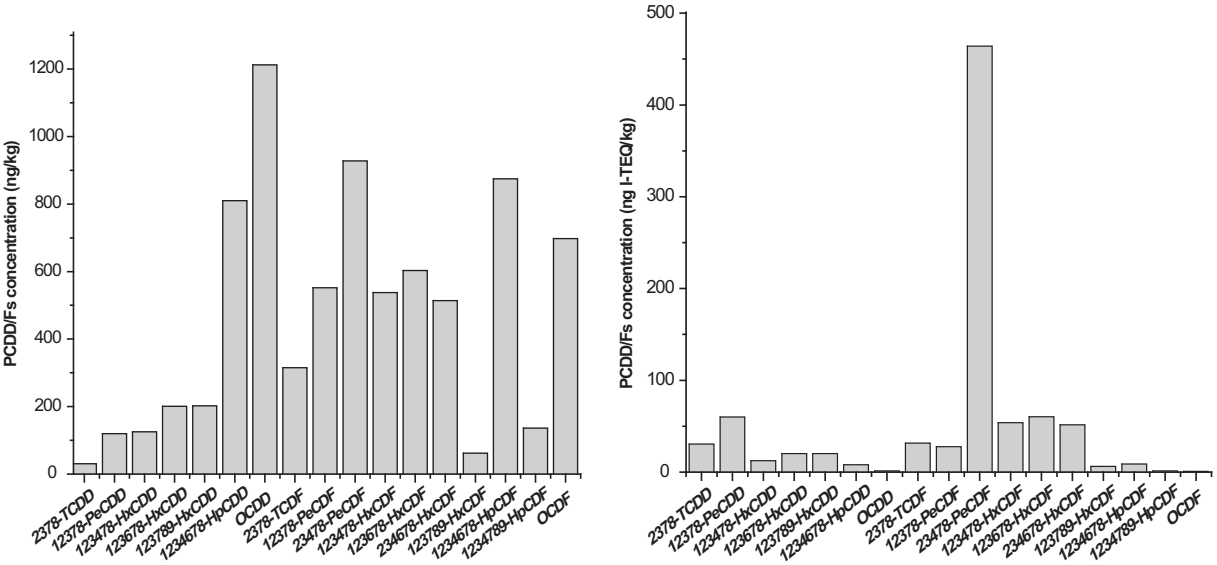

Fig. 3. Congener profiles of seventeen 2,3,7,8-chlorinated substituted PCDD/Fs containing in the fly ash.

transformation studies as well as formation mechanism elucidation. Figs. 2–5 show average congener-specific 2,3,7,8-PCDD/PCDF distributions in stack gas, fly ash, bottom ash and MSW samples in both concentration (left column) and TEQ (right column) units. Table 4 provides the complete data of the analyses done. As shown in Figs. 2–4 and Table 4, large similarities could be presented in both concentration and TEQ profiles. In concentration units, it can be observed that as the chlorinated-level increases, the concentration of the 2,3,7,8,-PCDD congener increases, but the concentration of the 2,3,7,8,-PCDF congener presents irregularities. For PCDDs 1,2,3,4,6,7,8-HpCDD and OCDD are the predominant congeners and for PCDFs 2,3,4,7,8-PeCDF, 1,2,3,4,6,7,8-HpCDF and OCDF are the major congeners. These profiles are in accordance with emission patterns reported for MSWI (Abad et al., 2002; Ni et al., 2009; Wang et al., 2003). In TEQ profiles, these profiles are characterized by a major content 2,3,4,7,8-PeCDF amounting to around 50% of the total I-TEQ PCDD/Fs. Another feature was observed for PCDD/F distributions. As listed in Table 2, the ratios of PCDFs to PCDDs for the stack gas and fly ash are 1.82 and 1.94, respectively. These results indicate that the de novo synthesis plays the dominant role in the low-temperature post-combustion zone.

As shown in Fig. 5 and Table 4, remarkable differences could be found compared with those output profiles in Figs. 2–4. The PCDD levels are significantly higher than those of PCDF and the MSW concentration profile is clearly dominated by a high content of OCDD followed by 1,2,3,4,6,7,8-HpCDD. The fraction of OCDD and HpCDD reaches around 90% of the total PCDD, which is consistent with the profile reported by Abad et al. (2000).

3.4. Dioxin mass balance from input to output

It is necessary to finish a dioxin mass balance from input to output aimed at assessing the real situation of the modern MSWI. A dioxin mass balance is quite a difficult project. Initially, three times sampling campaigns, covering the input and output, were designed to acquire representative scientific data. Then the total PCDD/Fs

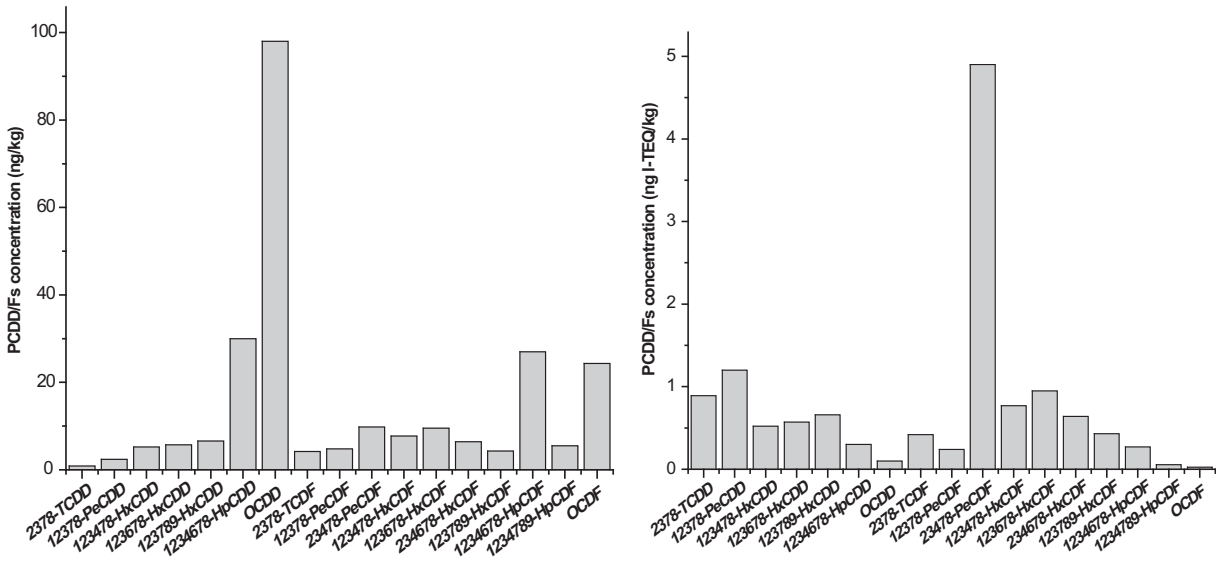

Fig. 4. Congener profiles of seventeen 2,3,7,8-chlorinated substituted PCDD/Fs containing in the bottom ash.

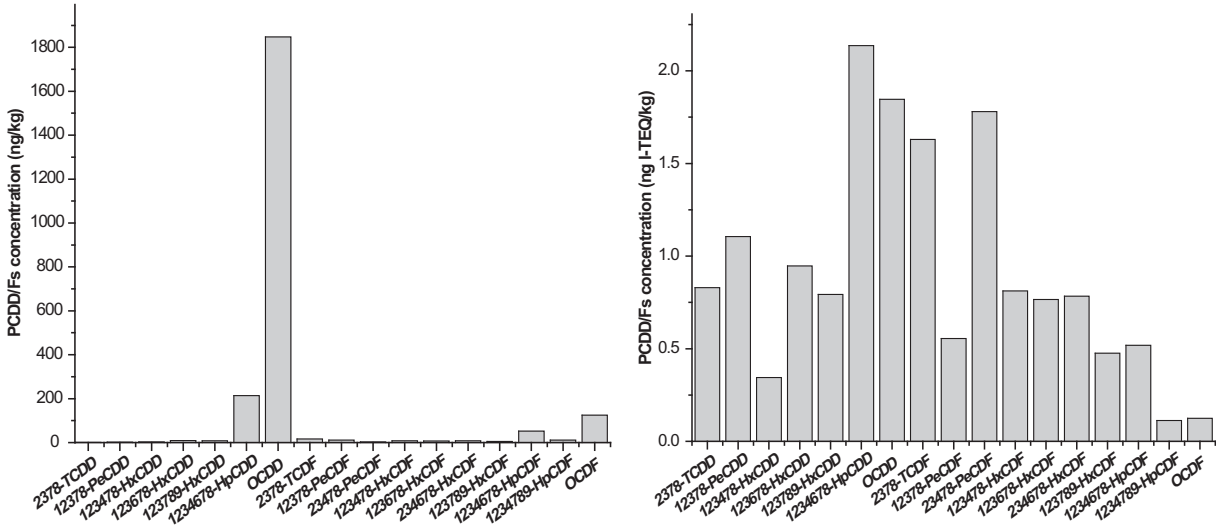

Fig. 5. Congener profiles of seventeen 2,3,7,8-chlorinated substituted PCDD/Fs containing in the MSW from the output of the MSWI.

input via MSW was compared with the sum of those measured in the output via all three types (stack gas, fly ash and bottom ash). Table 1 lists annual amount of waste treated, annual production of bottom ash and fly ash as well as flow rates of flue gas released into the environment, which are adopted to calculate the annual dioxin mass balance under steady running condition of the MSWI.

Fig. 6 shows the annual PCDD/F mass balance and emission factors across the whole MSWI. In the output balance, three forms of matrixes (stack gas emission, fly ash and bottom ash samples) over the three sampling campaigns yielded three average values from a total of 9 values. The results indicated that bottom ash, fly ash and stack gas presented an average of 1.10, 6.42 and 0.108 g I-TEQ/yr, respectively. The mean PCDD/F emission factors of the whole MSWI system were 3.17 μg I-TEQ/t-waste for bottom ash, 18.52 μg I-TEQ/t-waste for fly ash, and 0.312 μg I-TEQ/t-waste for stack gas. Total emission factors of 22 μg I-TEQ/t-waste were calculated, much lower than the results reported for similar facilities (Huang and Buekens, 1995; Wang et al., 2010). Expressed in percentage distribution, stack gas emissions constituted a minor fraction of the total dioxin output of around 1.41%, whereas the greatest contribution was attributed to fly ashes with 84.16% followed by bottom ashes with 14.42%. However, as shown in Fig. 6, the annual dioxin input value was around 5.38 g I-TEQ/yr, lower than the total output value (about 7.62 g I-TEQ/yr), signifying a positive dioxin balance of about 2.25 g I-TEQ/yr. The result was quite close to that reported by Abad et al. (2000). They also found a positive balance of about 3.31 g I-TEQ/yr. Accordingly, it can be inferred that the dioxins present in the waste are destroyed during the thermal process but regenerated again in the post-combustion zone by de novo synthesis mainly.

It is well known that complex factors can affect dioxin emission. The first to consider is the temperature in the furnace. When high temperature is maintained, destruction efficiency is high and the emission of PCDD/F at the exit of the furnace is expected to be very low relatively. It is generally considered that at temperature above 850 °C any dioxins/furans present in the waste will be destroyed (McKay, 2002). Then the raw gas at the exit of incineration chamber must go through the treatment of APCD before being released into the environment for the stringent limits to be achieved. The type of

Table 4
Mean concentrations of 2,3,7,8-substituted PCDD/Fs in the input and output samples.

PCDD/Fs	Input		Output					
	MSW		Stack gas		Fly ash		Bottom ash	
	ng/kg	ng I-TEQ/kg	ng/Nm^3	ng I-TEQ/Nm^3	ng/kg	ng I-TEQ/kg	ng/kg	ng I-TEQ/kg
2,3,7,8-TCDD	0.83	0.83	0.004	0.004	30.5	30.5	0.89	0.89
1,2,3,7,8-PeCDD	2.21	1.11	0.012	0.006	120	60.0	2.40	1.20
1,2,3,4,7,8-HxCDD	3.45	0.35	0.006	0.001	125	12.5	5.20	0.52
1,2,3,6,7,8-HxCDD	9.47	0.95	0.010	0.001	201	20.1	5.70	0.57
1,2,3,7,8,9-HxCDD	7.93	0.79	0.007	0.001	202	20.2	6.60	0.66
1,2,3,4,6,7,8-HpCDD	213.60	2.14	0.036	0.000	810	8.1	30.00	0.30
OCDD	1847.00	1.85	0.171	0.000	1212	1.2	98.00	0.10
2,3,7,8-TCDF	16.30	1.63	0.055	0.005	315	31.5	4.20	0.42
1,2,3,7,8-PeCDF	11.10	0.56	0.055	0.003	552	27.6	4.80	0.24
2,3,4,7,8-PeCDF	3.56	1.78	0.082	0.041	928	464.0	9.80	4.90
1,2,3,4,7,8-HxCDF	8.12	0.81	0.044	0.004	538	53.8	7.70	0.77
1,2,3,6,7,8-HxCDF	7.66	0.77	0.047	0.005	603	60.3	9.50	0.95
2,3,4,6,7,8-HxCDF	7.84	0.78	0.045	0.004	514	51.4	6.40	0.64
1,2,3,7,8,9-HxCDF	4.76	0.48	0.012	0.001	61.7	6.2	4.30	0.43
1,2,3,4,6,7,8-HpCDF	51.90	0.52	0.064	0.001	875	8.8	27.00	0.27
1,2,3,4,7,8,9-HpCDF	11.20	0.11	0.008	0.000	136	1.4	5.50	0.06
OCDF	124.40	0.12	0.035	0.000	698	0.7	24.30	0.02
Sum of 17 congeners	2331.33	15.56	0.695	0.078	7921.2	858.2	252.29	12.94

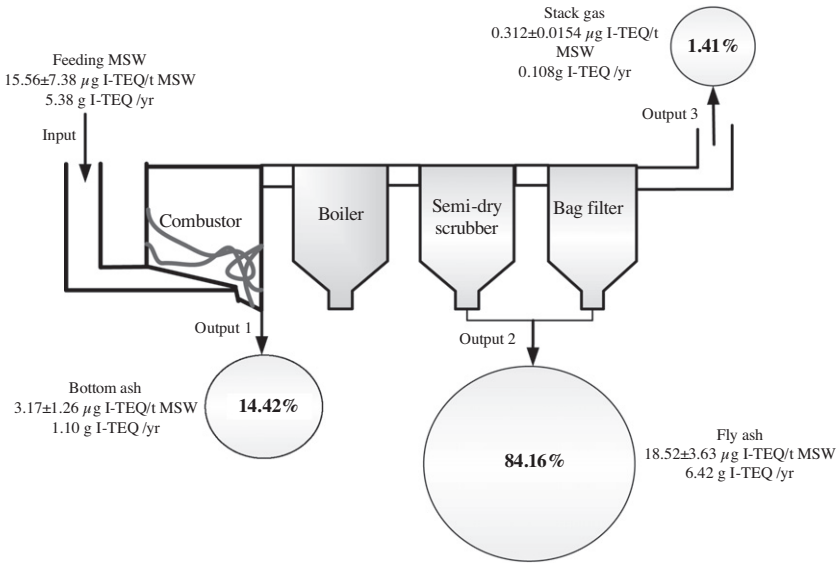

Fig. 6. PCDD/F mass balance and emission factors in the modern MSWI: percent (%) is the proportion of individual output to the total output.

APCD is one of the factors. The higher the removal efficiency of APCD, the lower are the dioxin emission concentrations. The most effective flue gas treatment is the combination of a semi-dry scrubber, a bag filter coupled with active carbon injection (Abad et al., 2002; McKay, 2002). Lastly the flue gas temperature in different components of APCD is also important influential parameter. Heterogeneous catalytic reactions including de novo and precursor synthesis occur in the temperature window (200–400 °C) (Shin et al., 1999). The scrubber and bag filter should be operated below 200 °C. From the above results, it can be seen that the MSWI we investigated emitted safely low dioxins, which may be attributed to high combustion temperature (>1000 °C), effective off-gas treatment (SDS + ACI + BF), and low flue gas temperature in the APCD (<180 °C).

4. Conclusions

The detail PCDD/PCDF Characteristics and mass balance of the different components constituting the dioxin inputs and outputs were investigated through three sampling campaigns. The results indicated that the dioxin removal efficiency reached 99.2% with SDS + ACI + BF. The dioxin emission was 0.078 ng I-TEQ/Nm3, which was far below the international emission standard limit established by the European Union Directive of 0.1 ng I-TEQ/Nm^3. Fly ash samples presented mean levels of around 0.858 ng I-TEQ/g, which meets the environmental quality standards for soil (less than 1 ng I-TEQ/g) in Japan Ministry of the Environment The output samples showed large similarities in congener profiles. Instead, on the input side, the municipal solid waste (MSW) showed a remarkable difference in congener profiles compared with those of the output The dioxin mass balance indicated that the annual dioxin input value was around 5.38 g I-TEQ/yr, lower than the total output value (about 7.62 g I-TEQ/yr), signifying a positive dioxin balance of about 2.25 g I-TEQ/yr. Technical measures that can maintain high combustion temperature (>850 °C), take SDS + ACI + BG as APCD, and keep low flue gas temperature in different components of APCD (<180 °C) are effective to reduce dioxin emission well below 0.1 ng I-TEQ/Nm^3.

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