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Polychlorinated dibenzo-*p*-dioxins and dibenzofurans in flue gas emissions from municipal solid waste incinerators in China*

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Abstract: Polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs) emissions in flue gas from two types of municipal solid waste incinerators (MSWIs) most commonly used in China were investigated in this study. The selected incinerators include two grate-type MSWIs: MSWI-A (350 t/d) and MSWI-B (150 t/d), and two fluidized bed MSWIs: MSWI-C (400 t/d) and MSWI-D (400 t/d), which are all equipped with semi-dry lime scrubber and bag filter except MSWI-D equipped with cyclone and wet scrubber (WS) as air pollutant control device (APCD). Results indicated that the emission concentration and the international toxic equivalents (I-TEQs) of the PCDD/Fs from the stacks were in the range of 1.210~10.273 ng/Nm^3 and 0.019~0.201 ng I-TEQ/Nm^3, respectively. They were greatly lower than the emission regulation standard of PCDD/Fs in China (1.0 ng I-TEQ/Nm^3). However, only the PCDD/Fs emission level from MSWI-C was below 0.1 ng I-TEQ/Nm^3. Although the homologue profiles were distinct, the contributions of the 2,3,7,8-subsituted congeners to the total I-TEQ were similar among all the investigated MSWIs. Two major 2,3,7,8-substituted congeners, 2,3,4,7,8-PeCDF and 1,2,3,7,8-PeCDD, account for 47% and 9% (average values) of the total I-TEQ values, respectively. The correlation between PCDD/Fs levels and composition of flue gas was also discussed.

Key words: Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), Municipal solid waste incinerator (MSWI), Grate, Fluidized bed

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INTRODUCTION

With the rapid development of economy and great improvement of living standards in China, a large amount of municipal solid waste (MSW) is generated, which reached to about 136.38 million tons in 2002 and keeps going up at a rate of 8%~10% per year (Li *et al.*, 2004; Zhu *et al.*, 2005). For disposing these MSW, three approaches, i.e., landfill, compost and incineration are applied in China. Among them, though landfill is the dominant one, which accounts for more than 80% of the MSW disposal (Li *et al.*, 2004), the amount of large-scale MSW incinerator (MSWI) plants is growing, especially in developed cities and its total capacity had been 13155 t waste per day by July 2002 (Zhu *et al.*, 2005) because of its advantages including significant volume reduction (~90%), mass reduction (~70%), toxicity reduction, and energy recovery (Yan *et al.*, 2006).

In 1988, the first MSWI, a Martin grate imported from Mitsubishi Co., Japan, was built in Shenzhen, China. Since then, more MSWI plants have been constructed in Beijing, Shanghai, Guangzhou and other big cities. Up to now, more than 140 MSWI plants are in operation or under construction (Bie *et al*., 2007). Three incineration technologies, i.e., grate, fluidized bed and rotary kiln, are now used in China, and the grate and fluidized bed type boilers contributed more than 80% of the total incinerators.

Although MSWI has many advantages, many pollutants, such as HCl, heavy metals, especially polychlorinated dibenzo-*p*-dioxins and dibenzofurans

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(PCDD/Fs) are emitted during the incineration process. A preliminary investigation on PCDD/Fs emission from flue gas of over 15 typical incineration facilities has shown that half of them exceeded 1.0 international toxic equivalents (I-TEQs) ng/Nm^3 at 11% (v/v) O_2, the standard of PCDD/Fs emission regulation in China (Tian and Ouyang, 2003). In this study, two grate-type incinerators and two fluidized bed incinerators (FBIs) were investigated for PCDD/Fs emission. The study aimed to determine the characteristic of PCDD/Fs distribution profiles in different types of MSWIs in China. The relationship between PCDD/Fs levels and compositions of flue gas was discussed. Furthermore, the suggestions on the control technology of PCDD/Fs in commercial-scale MSWI plants were presented.

EXPERIMENTS

Sampling

Table 1 briefly lists the spec of the investigated MSWI. MSWI-A, MSWI-B and MSWI-C are all equipped with semi-dry lime scrubber and bag filter while MSWI-D is equipped with cyclone and wet scrubber (WS) as air pollutant control device (APCD). All sampling points are located downstream to APCD. Schematic of PCDD/Fs sampling system is shown in Fig.1. This sampling system was based on US EPA Method 0023A (1996). Sampling volume is approximately 2~4 Nm^3. All samplings were repeated once. The PCDD/Fs surrogate standards were added to the XAD-2 resin for checking the PCDD/Fs sampling efficiency. After each sampling, the system was rinsed with acetone and toluene, respectively, and the rinse was saved in brown glass bottle. Filter and XAD-2 resin also stored and maintained in dark below 4 °C on field until transferred to the laboratory.

PCDD/Fs analysis

Pretreatment of PCDD/Fs in the collected flue gas was conducted according to the US EPA Method 1613B (1994). The sample was spiked with known amounts of 13C-labelled internal standards, then Soxhlet extracted with toluene for 24 h. After extraction the sample was macro-concentrated by rotary evaporator to 1~2 ml, and then the sample extracts were exchanged into hexane. Sample clean up

Table 1 Spec of the investigated MSWI

Incinerator	Furnace type	Capacity (t/d)	Fuel type	Air pollutant control device
MSWI-A	Hydraulic ladder mechanical grate	350	MSW	Semi-dry lime scrubber+bag filter
MSWI-B	Martin grate	150	MSW	Semi-dry lime scrubber+bag filter
MSWI-C	Fluidized bed	400	Coal/MSW=20/80	Semi-dry lime scrubber+bag filter
MSWI-D	Fluidized bed	400	Coal/MSW=28/72	Cyclone+wet scrubber

Note: activated carbon was used to remove PCDD/Fs in the flue gas produced from MSWI-A and MSWI-B

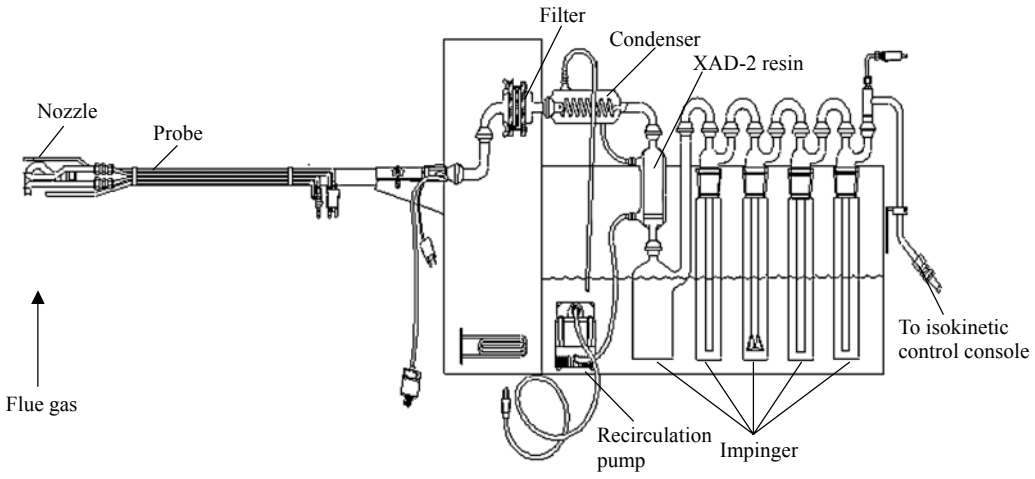

Fig.1 Schematic of system for PCDD/Fs sampling from flue gas

procedures involved a multi-silica gel column and a basic-alumina column. The elute was concentrated again and blown with nitrogen to approximately 20 μl. Before analysis the ^{13}C-labelled injection standards were added to the sample for calculating the recovery of ^{13}C-labelled internal standards. All solvents were purchased from Mallinckrodt Baker Inc., USA and were pesticide residue analysis grade. The analysis was performed by HRGC/HRMS on a 6890 Series gas chromatograph (Agilent, USA) and coupled to a JMS-800D mass spectrometer (JEOL, Japan). A DB-5 ms fused-silica capillary column (60 m×0.25 mm inside diameter, 0.25 μm film thickness) was used. The temperature program was as follows: 150 °C, held for 1 min; increased at a rate of 25 °C/min to 190 °C; then increased at a rate of 3 °C/min to 280 °C, held for 20 min. The carrier gas was helium at a constant flow rate of 1.2 ml/min. The mass spectrometer (MS) was operated at a resolution of 10000 under positive electron ionization (EI) condition (38 eV electron energy). One μl of sample was injected using an auto-sampler in splitless mode. The PCDD/Fs were quantified using a molecular ion (M), an M+2 ion or an M+4 ion. The I-TEQs was calculated using the international toxic equivalence factor (I-TEF) (NATO/ CCMS, 1988).

RESULTS AND DISCUSSION

PCDD/Fs concentration in the flue gas

PCDD/Fs concentration in the flue gas was shown in Table 2. The I-TEQ values were in the range of 0.019~0.201 ng I-TEQ/Nm^3, which were all lower than the standard limit of China (1.0 ng I-TEQ/Nm^3). In 2001, the State Environmental Protection Administration (SEPA) of China investigated the pollutants including PCDD/Fs emission from 15 MSWIs. The investigated results showed half data of PCDD/Fs concentration in the flue gas exceed 1.0 ng I-TEQ/Nm^3, indicating that the emission levels of PCDD/Fs from the MSWIs have been reduced over the past few years. However, half data of the emission results were slightly higher than the developed countries emission limit of 0.1 ng I-TEQ/Nm^3. So the control technologies for PCDD/Fs need to be improved if the emission limit was lowered to 0.1 ng I-TEQ/Nm^3.

Table 2 PCDD/Fs concentration in the flue gas (standard dry gas at 11% (v/v) O_2)

Incinerator	Concentration (ng/Nm^3)			TEQ value (ng I-TEQ/Nm^3)
	PCDDs	PCDFs	Total	
MSWI-A	2.4	2.0	4.4	0.20
	3.2	6.2	9.4	0.17
MSWI-B	2.2	4.2	6.4	0.11
	2.9	2.8	5.8	0.078
MSWI-C	0.75	0.87	1.6	0.026
	0.46	0.75	1.2	0.019
MSWI-D	2.1	2.7	4.8	0.072
	4.1	6.2	10.0	0.16

Note: two sampling were done for each MSWI

Among all MSWIs investigated, the emission levels of PCDD/Fs from MSWI-C were the lowest. MSWI-C is an FBI using coal as auxiliary fuel. The relatively high sulfur in the coal can inhibit the PCDD/Fs formation, which has been confirmed by many studies (Gullett *et al*., 1992; 1998; 2000). One of the emission levels of MSWI-D, which is also an FBI using MSW and coal as fuel, was 0.162 ng I-TEQ/Nm^3. The relatively high emission level may be due to the poor APCD, which is composed of cyclone and wet scrubber. Takaoka *et al*.(2003) investigated the behavior of PCDD/Fs in WS system of two MSWIs, and reported that PCDD/Fs concentration in the flue gas at outlet of WS was higher than that at inlet of WS in both MSWIs.

Profiles of homologue and congener of PCDD/Fs in the flue gas

Profiles of PCDD/Fs homologue in the flue gas from MSWI-A to MSWI-D were shown in Fig.2. For FBIs (MSWI-C and MSWI-D), the PCDFs levels were all greater than those of PCDDs and the lower chlorinated furans were dominant for PCDFs, which was similar to the emission characteristic of PCDD/Fs from Chinese and Korean MSWIs (Oh *et al*., 1999; Yan *et al*., 2006). However, for the grate fired incinerators (MSWI-A and MSWI-B), the PCDDs levels were slightly greater than those of PCDFs for one sampling, and the ratios of PCDDs and PCDFs are 1.15 and 1.03, respectively, but for another sampling, similar ratios of PCDDs and PCDFs were observed as those of FBIs. The difference of the homologue profiles in the flue gas from FBIs and grate fired incinerators may be explained by different prevailing

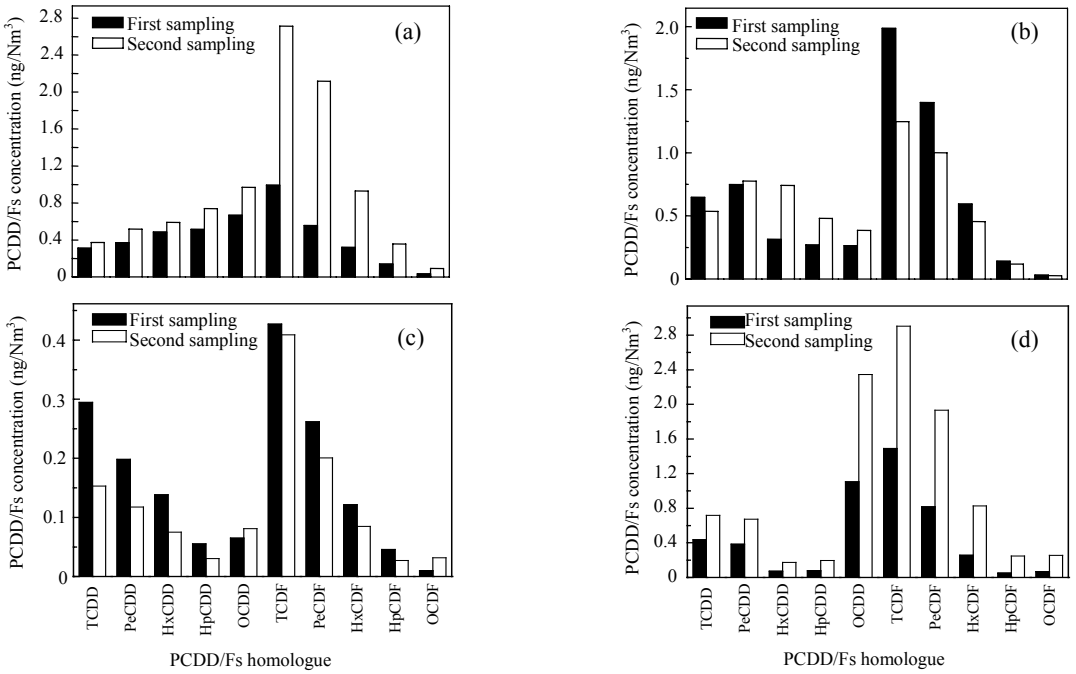

Fig.2 Profiles of PCDD/Fs homologue in the flue gas from (a) MSWI-A, (b) MSWI-B, (c) MSWI-C and (d) MSWI-D

mechanisms of PCDD/Fs formation.

Although the profiles of PCDD/Fs homologue in the flue gas from FBIs and grate fired incinerators were different, the 2,3,7,8-substituted congeners distribution patterns were similar for all MSWIs (Fig.3). The comparison between this data and others obtained from Harbin of China, Korea and Japan was shown in Fig.4. The very similar distribution of 2,3,7,8-substituted congeners was found in Chinese MSWI. For example, 1,2,3,4,6,7,8-hepta-chlorinated dibenzo-*p*-dioxin (HpCDD) and octa-chlorinated dibenzo-*p*-dioxin (OCDD) were the major congeners for PCDDs. 2,3,4,7,8-penta-chlorinated dibenzofuran (PeCDF) and 1,2,3,4,6,7,8-hepta-chlorinated dibenzofuran (HpCDF) were the predominant congeners for PCDFs. However, there was different distribution for PCDD/Fs emission from MSWI in Korea and Japan. 2,3,4,6,7,8-hexa-chlorinated dibenzofuran (HxCDF) and 1,2,3,4,6,7,8-HpCDF amounted to more than 54% of total PCDFs for MSWI in Korea. 1,2,3,4,6,7,8-HpCDF and octa-chlorinated dibenzofuran (OCDF) were the major congeners for PCDFs of MSWI in Japan.

2,3,4,7,8-PeCDF and 1,2,3,7,8-penta-chlorinated dibenzo-*p*-dioxin (PeCDD) were two major 2,3,7,8-substituted congeners, and their contributions were up to 47% and 9% (average values) of the total TEQ values, respectively, due to their high toxicity (I-TEF value is all 0.5). Kato and Urano (2001) also found that 2,3,4,7,8-PeCDF and 1,2,3,7,8-PeCDD contributed to about 49% of the I-TEQ values. The contribution of 2,3,4,7,8-PeCDF was stable, though the contribution of 1,2,3,7,8-PeCDD fluctuated. Several researchers have reported a linear correlation between the 2,3,4,7,8-PeCDF concentration and TEQ values (Fiedler *et al*., 2000; Kato and Urano, 2001; Iino *et al*., 2003). The positive correlation was obtained from this paper (Fig.5), which was different from the results of Kato and Urano (2001). In their study, the correlation between the 2,3,4,7,8-PeCDF concentration and I-TEQ values could be described by the following Eq.(1). The difference coefficient maybe attributed to different MSW composition, furnace type, etc. In this study, the correlation could be described by Eq.(2).

$$[\text{I-TEQ of PCDD/Fs}]=1.4[2,3,4,7,8\text{-PeCDF}], \quad (1)$$

$$[\text{I-TEQ of PCDD/Fs}]=2.1[2,3,4,7,8\text{-PeCDF}]. \quad (2)$$

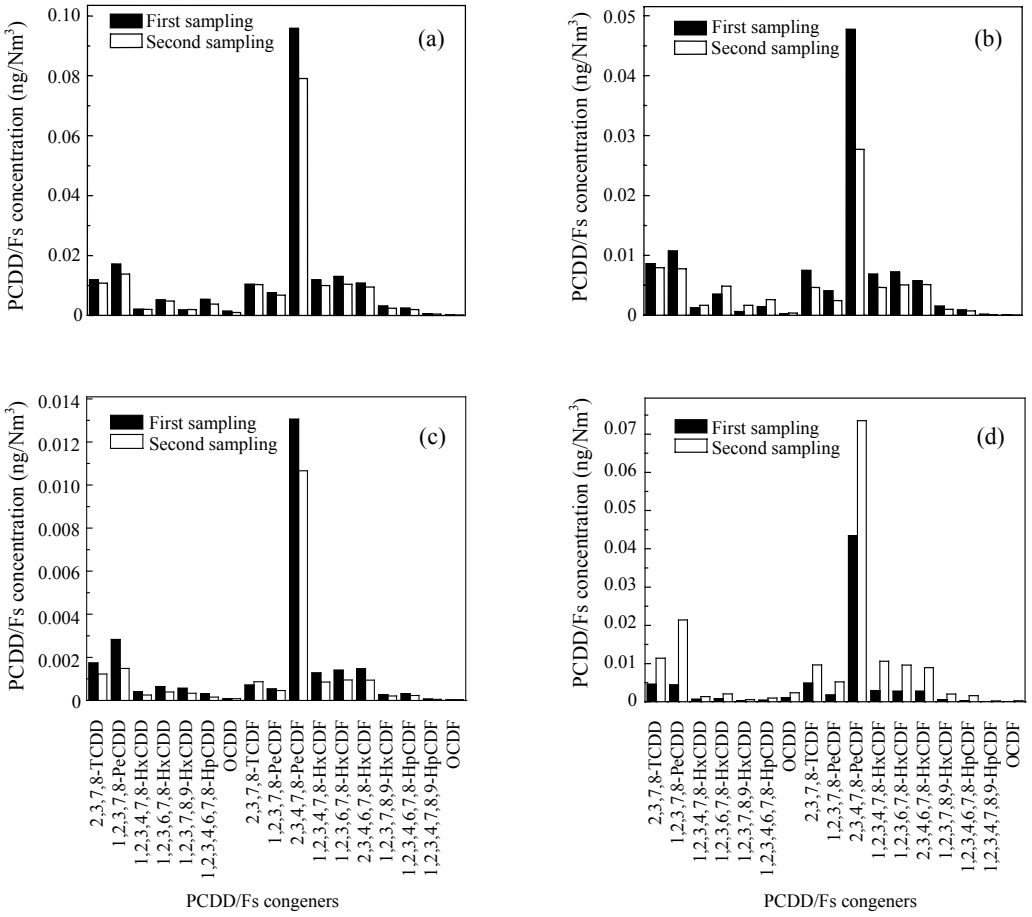

Fig.3 Profiles of PCDD/Fs congeners in the flue gas from (a) MSWI-A, (b) MSWI-B, (c) MSWI-C and (d) MSWI-D

Correlation PCDD/Fs levels with the composition of flue gas

It is well known that various factors can affect PCDD/Fs emission. Composition of flue gas is one of the factors. Table 3 shows the composition of flue gas and the PCDD/Fs levels in MSWI-A and MSWI-C. CO, O_2, water and other compounds were continuously measured for about 30 min by using a flue gas analyzer during the PCDD/Fs sampling process. Although there are only 4 datasets available in Table 3, the similar results were found.

CO value, one of the parameters to judge the combustion quality, has been used as a parameter to evaluate PCDD/Fs emissions in several studies. Sakurai *et al*.(2000) found that new FBIs with well controlled combustion emitted less PCDD/Fs in flue gas and fly ash, while old FBIs with high CO concentration had higher PCDD/Fs emission. However, other studies indicated no significant correlation between CO values and the PCDD/Fs levels in MSWIs or pilot incinerators (Weber and Hagenmaier, 1999; Weber *et al.*, 2002). In the present study, CO values showed no positive correlation with PCDD/Fs levels. In MSWI-A, the CO values were lower than the detect limit of the flue gas analyzer. The case was on the contrary for MSWI-C. But the PCDD/Fs levels in the MSWI-A were higher than those of MSWI-C.

O_2 content also affected the PCDD/Fs formation. Chang and Huang (2000) investigated the effect of O_2 content on the PCDD/Fs formation in MSW fly ash. They found that the PCDD/Fs formation increased with the increase of O_2 content in electrostatic precipitator (ESP) fly ash. We also found a positive correlation between PCDD/Fs levels (TEQ value) and O_2 content as shown in Table 3. In China, the PCDD/Fs emission standard for MSWIs was based on standard dry gas corrected to 11% (v/v) O_2. If the O_2 content was higher than 11% (v/v), the PCDD/Fs levels would

be higher than the detected value. It was reported that PCDD/Fs emission was minimal when the O_2 content was between 6%~9% (v/v) (Yan *et al*., 2006). So O_2 content might be a parameter for PCDD/Fs controlling.

As MSWI flue gas contains water, experiments were performed in several studies to compare the formation of PCDD/Fs with and without water in the gas flow (Addink and Olie, 1995). Stieglitz *et al*.(1990) reported that PCDD/Fs formation levels from carbon/fly ash at 300 °C with water increased compared to those without water. However, Jay and Stieglitz (1991) found a decrease of the total PCDD/Fs amounts formed in the presence of water with charcoal/$MgSiO_2$/$CuCl_2$ in air at 300 °C. Li *et al*.(2006) investigated the effect of water vapor on catalyzed de novo formation of PCDD/Fs. Their results indicated that water vapor could promote the PCDD/Fs formation. We also found a positive correlation between water and TEQ values of PCDD/Fs as shown in Table 3.

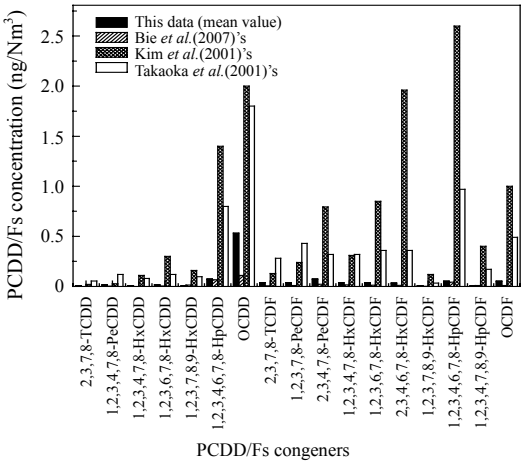

Fig.4 Comparison between this data and other data from the literature

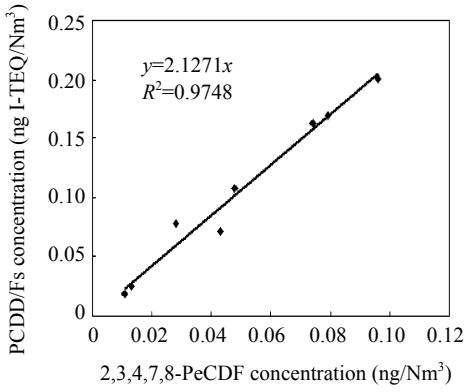

Fig.5 Relationship between 2,3,4,7,8-PeCDF concentration and TEQ values in MSWI-A to MSWI-D

It has been reported that co-firing coal with MSW could reduce PCDD/Fs emissions due to the increased flue gas SO_2 concentration (Ryan *et al*., 2006). Introducing SO_2 into the furnace section of a full-scale incinerator (SO_2 concentrations from 20 to 2000 mg/m^3) resulted in a decline of PCDD/Fs concentration in flue gas (Ruokojärvi *et al*., 2004). The conversion of metal chloride in the fly ash to sulfates may be the main mechanism of PCDD/Fs suppressing, which had been confirmed in the pilot plant TAMARA (Hunsinger *et al*., 2006). The chloride concentrations in the fly ash decreased significantly from ~200 mg/g down to below the detection limit of about <3 mg/g. Table 3 showed the S/Cl mole ratios in flue gas from MSWI-A were both about 0.2, however, those in flue gas from MSWI-C were 9.3 and 6.9, respectively. The increased S/Cl mole ratio in flue gas would lead the decrease of PCDD/Fs, which could be calculated from the data of Table 3.

CONCLUSION

The flue gas from two grate-type MSWIs and two fluidized bed MSWIs were analyzed for PCDD/Fs. The total concentration and TEQ values of PCDD/Fs emission were in the range of 1.210~

Table 3 Composition of flue gas and the PCDD/Fs levels (standard dry gas at 11% (v/v) O_2)

Incinerator	Composition						PCDD/Fs levels	
	CO (mg/Nm^3)	O_2 (%, v/v)	Water (%, v/v)	NO_x (mg/Nm^3)	SO_2 (mg/Nm^3)	HCl (mg/Nm^3)	PCDD/Fs (ng/Nm^3)	PCDD/Fs (ng I-TEQ/Nm^3)
MSWI-A	<1	12.8	18.2	119.5	53.1	185.0	4.4	0.200
	<1	11.5	16.8	129.5	70.0	179.7	9.4	0.170
MSWI-C	757.4	6.9	10.6	99.4	220.5	13.5	1.6	0.026
	1102.7	6.3	9.7	96.2	256.4	21.3	1.2	0.019

Note: The values except PCDD/Fs levels were the average values during 30 min sampling

10.273 ng/Nm^3, 0.019~0.201 I-TEQ ng/Nm^3, respectively, which were all greatly lower than the standard of the PCDD/Fs emission regulation (1.0 ng I-TEQ/Nm^3) in China, however, only the PCDD/Fs emission levels from MSWI-C below 0.1 ng I-TEQ/Nm^3.

Although the profiles of PCDD/Fs homologue in the flue gas from different type MSWIs were different, the 2,3,7,8-substituted congeners distribution patterns were similar for all MSWIs. Two major 2,3,7,8-substituted congeners were 2,3,4,7,8-PeCDF and 1,2,3,7,8-PeCDD. The positive relationship between the 2,3,4,7,8-PeCDF concentration and TEQ values could be described by the following equation: [I-TEQ of PCDD/Fs]=2.1[2,3,4,7,8-PeCDF].

It was observed that a correlation exists between PCDD/Fs emission and the contents of O_2, water, S/Cl mole ratio, but not CO in flue gas, so these parameters could be adjusted for PCDD/Fs emission controlling in MSWIs.

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