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Characteristics of PCDD/Fs in Flue Gas from MSWIs and HWIs: Emission Levels, Profiles and Environmental Influence

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ABSTRACT

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the flue gas from two types of waste incinerators (municipal solid waste incinerators and hazardous waste incinerators) the most commonly used in China were investigated in this study. The PCDD/F emissions from the stack gas of the waste incinerators showed a large variation from 0.016 to 0.29 ng I-TEQ m^{-3} with an average value of 0.103 ng I-TEQ m^{-3}. The emission factors of PCDD/Fs were in the range of 0.056–0.607 µg I-TEQ t^{-1} (average 0.235 µg I-TEQ t^{-1}) for municipal solid waste incinerators (MSWIs) and 0.07–3.27 µg I-TEQ t^{-1} (average 1.03 µg I-TEQ t^{-1}) for hazardous waste incinerators (HWIs), respectively. It was estimated that 10.2–88.0 mg I-TEQ (average 35.0 mg I-TEQ) and 1.13–53.0 mg I-TEQ (average 17.1 mg I-TEQ) of PCDD/Fs were annually released to the atmosphere, respectively from MSWIs and HWIs. The level and congener patterns of 17 species of PCDD/Fs in the ambient air and in soil samples around the MSWIs and HWIs were investigated to evaluate the influence of PCDD/F emission on the environment. The PCDD/F concentrations in the ambient air from the 5 MSWIs and 4 HWIs ranged from 0.088 to 13.9 pg I-TEQ m^{-3} with an average value of 0.185 pg I-TEQ m^{-3}. The soil samples in the vicinity of MSWIs and HWIs presented concentrations between 0.68 and 16.0 ng I-TEQ kg^{-1} with an average value of 3.98 ng I-TEQ kg^{-1}. This study showed that PCDD/F emission from the incineration plants had an obvious impact on the surrounding ambient air.

***Keywords*:** PCDD/Fs; Flue gas; Emission factor; Air; Soil.

INTRODUCTION

Municipal solid waste (MSW) incineration in China has been boosted more than twelve fold in the past decade, in response to the rapid increase of MSW (Lu *et al.*, 2017; Zhu *et al.*, 2018). In addition, the amount of hazardous waste (HW) in China is increasing greatly from 10.79 million tons in 2007 to 69.37 million tons in 2017 as the development of economy (National Bureau of Statistics of China, 2008, 2018). Hazardous waste, including industrial hazardous waste, medical waste, and household hazardous waste (Duan *et al.*, 2008), presents some particular characteristics of ignitability, corrosivity, reactivity, and toxicity. Trinh and Chang (2018) reported that municipal solid waste incinerators (MSWIs) were important emission sources of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs, also referred to as dioxin) to ambient air (Ni *et al.*, 2009; Hu *et al.*, 2014; Wang *et al.*, 2015; Bai *et al.*, 2017; Ngo *et al.*, 2018; Trinh and Chang, 2018). Dopico and Gomez (2015) and Zhan *et al.* (2019) also reported that the emission factor of PCDD/Fs from hazardous waste incinerators (HWIs) is significantly higher than those from MSWIs. Therefore, it is important to control the PCDD/F emissions from HWIs.

The emission of PCDD/Fs has recently been investigated extensively in the waste incineration process (Bie *et al.*, 2007; Choi *et al.*, 2008; Gao *et al.*, 2009; Ni *et al.*, 2009; Zhu *et al.*, 2018). Choi *et al.* (2008) investigated dioxins in gases emitted from MSWIs and HWIs, finding the concentration of PCDD/Fs ranged from 0.05 to 609.27 ng TEQ Nm^{-3}. Bie *et al.* (2007) discovered the average dioxin emission in the flue gases from MSWIs by applying a twin internal fluidized bed incinerator in Harbin was 0.02 ng I-TEQ Nm^{-3}. Ni *et al.* (2009) analyzed the congener profiles from various MSWIs, estimating the emission factors of PCDD/Fs in the stack gas to be 0.169–10.7 µg TEQ t^{-1} MSW with an average value of 1.73 µg TEQ t^{-1}MSW. Gao *et al.* (2009) revealed the stack gas emissions of PCDD/Fs from hospital waste incinerators (HWIs) exhibited a large variation (0.08–31.6 ng I-TEQ Nm^{-3}). Zhu *et al.* (2018) revealed that the concentrations of PCDD/F emissions in stack gas from 6 MSWIs were in the range of 0.007–0.059 ng TEQ Nm^{-3}. The emission factors of PCDD/Fs from 6 MSWIs varied from 0.027 to 0.225 mg I-TEQ t^{-1},

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with a mean value of 0.17 mg I-TEQ t^{-1} waste.

The PCDD/F concentrations and congeners distribution in the surrounding environment of different incinerators were investigated and studied in the past years (Oh *et al.*, 2006; Yan *et al.*, 2008; Li *et al.*, 2018; Zhou *et al.*, 2019). Oh *et al.* (2006) studied the influence of MSWI on the PCDD/Fs levels in ambient air and soil, and the concentration of PCDD/Fs in air samples and soil samples ranged from 0.22 to 1.16 ng I-TEQ Nm^{-3} and 1.25 to 75.0 ng I-TEQ kg^{-1}, respectively. Zhou *et al.* (2019) studied the characteristics of PCDD/Fs in surface soil around an iron and steel plant and the PCDD/Fs content in the soil ranged from 0.16 to 4.5 ng I-TEQ kg^{-1}. However, few studies focused on the emission characteristics of PCDD/Fs and PCDD/Fs levels in the ambient air and soil samples from HWIs, and the similarities and differences between MSWIs and HWIs were still obscure in terms of PCDD/Fs.

In order to investigate the emission characteristics of PCDD/Fs the in stack gas and PCDD/Fs levels in the ambient air, soil samples from MSWIs and HWIs in China, stack gas samples from 5 MSWIs and 4 HWIs and ambient air and soil samples around the plants were collected and analyzed. The objectives of this study were: (1) to compare the PCDD/F concentration of stack gas and congener profiles of PCDD/Fs from 5 MSWIs and 4 HWIs; (2) to investigate the emission factors of PCDD/Fs and the annual emission of PCDD/Fs; (3) to analyze the possible grouping of similar emissions and demonstrate the PCDD/F congeners profiles using principal component analysis (PCA); (4) to study the PCDD/F concentration and patterns in the ambient air and soil samples from MSWIs and HWIs.

MATERIALS AND METHODS

Sample Collection

Stack gas samples were collected from municipal solid waste incinerators (MW01–MW05) and hazardous waste incinerators (HW01–HW04) in China, and the basic information for these incinerators and their APCDs were given in Table 1. The MW01–MW05 treated municipal solid waste incinerators. The HW01 treated dye waste, coke, organic sludge, waste oil, paint waste water with rotary kiln. The HW02 dealt with waste gas and liquid produced during VCM production. The HW03 treated wastes composed of oily refuse, rock debris, mud, etc. The HW04 treated medical waste.

Flue gas sampling was conducted using isokinetic samplers (Model KNJ23, KNJ corporation, Korea), complying with U.S. EPA Method 23a; and the specific sampling method was described in detail in previous study (Chen *et al.*, 2008). All sampling points were located downstream to APCDs. Ambient air sampling was performed in winter and summer at the boundary of the plants over 2 meters in height, and down the wind. Ambient air samples were collected according to U.S. EPA Method TO-9A (U.S. EPA, 1999), as described in detail by Chen *et al.* (2015). The sampling time and air volumes were about 24 h and 900–1100 m^3, respectively (Table 2). The soil samples (approximately 2 kg per sample) were taken from the upper 200 mm (MW01–MW05, HW02, HW04) and 200-400 mm (HW01, HW03) at the same sites.

Table 1. Basic information for the investigated waste incineration facilities

	Industrial facility	Number of Samples	Stack temperature (°C)	Technology and type of air pollution control device (APCD)	Capacity ($t\ h^{-1}$)	Average operating time ($h\ y^{-1}$)	Average emission flow rate ($m^3\ h^{-1}$)
MW01	Municipal waste	9 (M01–M09)	127	Fluidizedbed incinerator, SDS + AC+ FF	15	7200	70000
MW02	Municipal waste	9 (M10–M18)	169	Grate incinerator, SDS + AC+ FF	20	7200	82000
MW03	Municipal waste	9 (M19–M27)	169	Grate incinerator, SDS + AC+ FF	25	7200	94000
MW04	Municipal waste	6 (M28–M33)	165	Grate incinerator, SDS + FF	20	7200	92580
MW05	Municipal waste	6 (M34–M39)	120	Grate incinerator, SDS + AC+ FF	17.5	7200	60000
HW01	Hazardous industrial waste	3 (H01–H03)	70	Rotary kiln incinerator, SDS + AC+ FF	2.25	7200	25000
HW02	Hazardous industrial waste	3 (H04–H06)	115	Rotary kiln incinerator, SDS + FF	3.5	7200	24000
HW03	Hazardous industrial waste	3 (H07–H09)	76	Rotary kiln incinerator, SDS + AC+ FF	0.3	7200	3500
HW04	Medical waste	3 (H10–H12)	91	Rotary kiln incinerator, SDS + AC+ FF	1.5	7200	15000

Table 2. Information for the ambient air samples.

Sample	Sampling season	Sampling position	Sampling volume (m^3)	Wind and mean temperature (°C)	PCDD/Fs (pg I-TEQ m^{-3})
MW01-A1	Winter	Out of west gate	1007.8	Southwest, 9.3	0.433 (0.26,0.75,0.29)
MW01-A2	Summer	Out of west gate	1007.9	Southeast, 28.8	0.102 (0.088,0.077,0.14)
MW01-A3	Summer	By the chimney	1007.9	Southeast, 29.0	13.9 (3.89,11.1,26.7)
MW02-A1	Winter	Out of corner gate	911.1	Southwest, 10.5	0.207 (0.38,0.14,0.10)
MW02-A2	Winter	Out of west gate	908.3	Southwest, 9.2	0.421 (0.75,0.43,0.082)
MW03-A1	Winter	Out of east gate	1007.8	Southwest, 10.0	0.088 (0.11,0.11,0.043)
MW04-A1	Winter	Out of main gate	959.8	West, 5.6	0.114 (0.16,0.10,0.082)
MW05-A1	Winter	Out of main gate	984.3	Northwest, 5.0	0.34 (0.39,0.41,0.22)
HW01-A1	Winter	Out of main gate	1007.8	Southwest, 11.9	0.103 (0.11,0.11,0.090)
HW02-A1	Winter	Out of east gate	996.2	Southwest, 8.7	0.11 (0.12,0.14,0.070)
HW03-A1	Winter	Out of main gate	1007.8	Northwest,-2.4	0.229 (0.59,0.071,0.025)
HW03-A2	Summer	By the chimney	1007.9	South, 26.6	0.427 (0.25,0.27,0.76)
HW03-A3	Summer	Out of main gate	1007.9	East, 26.6	0.076 (0.13,0.048,0.049)
HW04-A1	Winter	By the maintenance shop	984.2	North, –2.0	0.175 (0.047,0.069,0.41)

Sample Extraction and Analysis

The PCDD/Fs analysis of stack gas and ambient air samples were performed according to the U.S. EPA Method 23 (U.S. EPA, 1995). Detailed steps of analysis method of the PCDD/Fs could be found from our previous studies (Chen *et al.*, 2014; Chen *et al.*, 2016). The soil samples were dried until constant weight and then they were ground and screened by a 2-mm sieve. About 500 g of each soil sample was finally homogenized through a 60-mesh sieve, and was refrigerated until analysis. And the detailed steps of pretreatment and analysis could be found from our previous study (Yan *et al.*, 2008).

Statistical Analysis

To better understand the congener profiles of dioxins in stack gases from various MSWIs and HWIs, principal component analysis (PCA) were used to evaluate the possible groupings of similar emissions and the dominant congeners in a defined grouping. Prior to analysis, the PCDD/F congeners levels were normalized with respect to the total PCDD/F concentration in the relevant sample. The statistical analyses were performed using the SPSS 20.0 software.

RESULTS AND DISCUSSION

TEQ Contribution of PCDD/Fs in Stack Gas

The gas emission levels of PCDD/Fs and the PCDF/PCDD weight ratio from 9 waste incineration facilities in China were investigated, and the results were presented in Fig. 1. The emissions of PCDD/Fs ranged from 0.016 to 0.29 ng I-TEQ m^{-3} with an average value of 0.103 ng I-TEQ m^{-3}. These values except HW02 and HW03 were all below the emission limit of national standard in China (0.1 ng I-TEQ m^{-3}) (China MEP, 2014). However, the PCDD/F emissions from HW02 and HW03 were 0.29 and 0.25 ng I-TEQ m^{-3} respectively, indicating more improvement space for PCDD/Fs control technologies in hazardous waste incinerators. The proportion of PCDF much higher than that of PCDD as in Fig. 1 indicating that PCDF was the main contributor to TEQ value in flue gas. Pham *et al.* (2019a) reported that concentrations of PCDD/Fs in flue gas were in the range of 13.4–17.6 pg TEQ m^{-3} (mean: 15.5 pg TEQ m^{-3}) for MSWIs and 12.7–438 pg TEQ m^{-3} (mean: 125 pg TEQ m^{-3}) for medical waste incinerators (MWIs), respectively. Zhu *et al.* (2018) reported that concentrations of PCDD/Fs in stack gas from MSWIs were in the range of 0.007-0.059 ng TEQ m^{-3}.

As in Fig. 2, the 50% for HWIs was much higher than that for MSWIs, indicating that HWIs were generally characterized by higher emission levels than MSWIs in China.Three potential factors were identified to affect the concentration and the congener distributions of PCDD/Fs in incinerators: the types of furnace, the composition of APCDs, and waste composition (chlorine content in the waste) (Wang *et al.*, 2003; Gao *et al.*, 2009). Wastes incinerated at a low temperature yield a higher TEQ value than a high temperature. Besides, the absence or removal of chlorine in wastes effectively decreased the emission TEQ values (Wikstrom *et al.*, 1996; Nzihou *et al.*, 2012). Many studies focused on the relationship between chlorine and PCDD/Fs, and the effect of chlorine content in the feeding waste on PCDD/Fs formation was obviously. Wang *et al.* (2003) and Li *et al.* (2017) reported that MSW contains 0.2–0.8% chlorine, while the content of chlorine in hazardous waste is generally higher than municipal solid waste, which accelerates the formation of PCDD/F. The samples that did not contain chlorine or were not combusted with chlorides exhibited low emission TEQ values (Wikstrom *et al.*, 1996; Nzihou *et al.*, 2012).

Estimated Emission Factor and Emission of PCDD/Fs from MSWIs and HWIs

Much work so far focused on the gas emission of PCDD/Fs from MSWIs and HWIs in China by using the emission factor (EF) to calculate the annual emission (EA) of PCDD/Fs (Li *et al.*, 2017; Zhu *et al.*, 2018; Pham *et al.*, 2019a, b).

$$\text{EF } (\mu\text{g I-TEQ } t^{-1}) = [\text{concentration in flue gas (ng I-TEQ } m^{-3}) \times \text{flue gas flow rate } (m^3 \cdot h^{-1})]/[\text{capacity level } (t\ h^{-1}) \times 10^3] \quad (1)$$

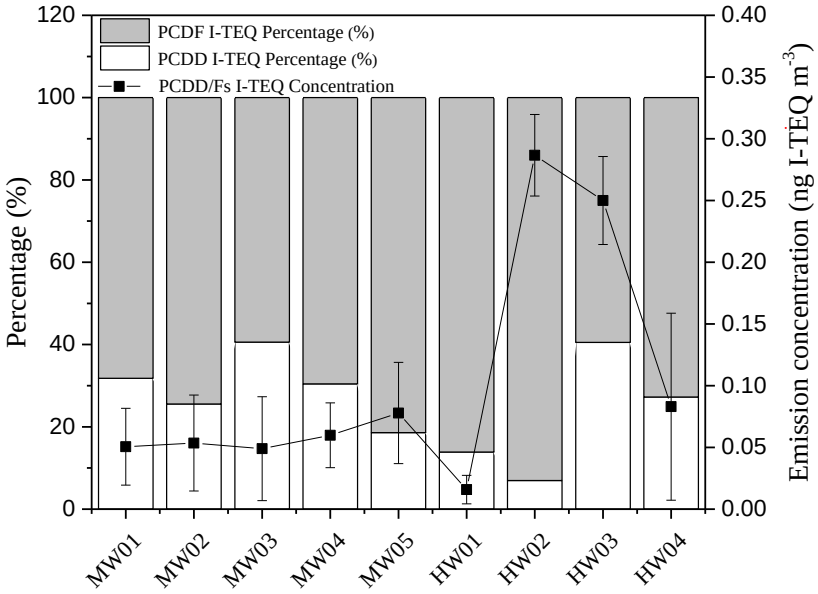

Fig. 1. Emission levels of PCDD/Fs from the 9 waste incinerators.

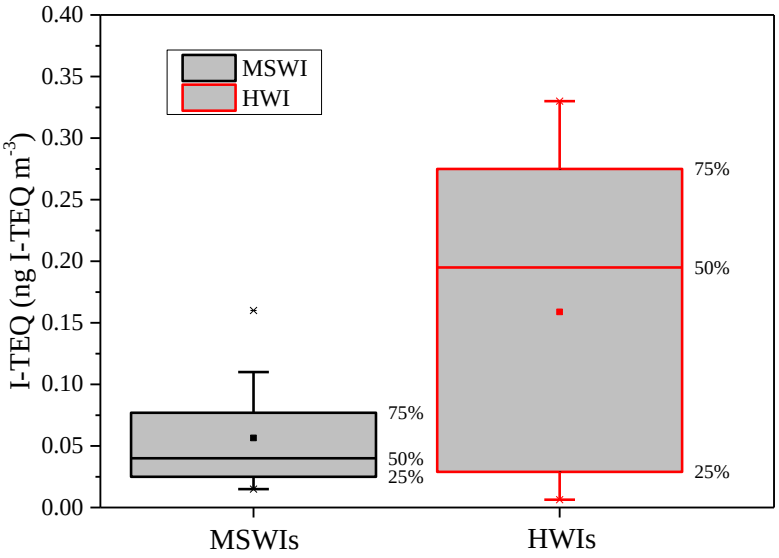

Fig. 2. PCDD/Fs statistical parameters for stack gas samples from MSWIs and HWIs: mean (▪), 25%, 50% and 75%.

EA (mg I-TEQ y^{-1}) =
[emission factor (μg I-TEQ t^{-1}) × capacity level (t h^{-1}) × average operating (h y^{-1})]/10^3 (2)

Estimated emission factors and annual emission amounts of PCDD/Fs (mean concentrations and the ranges) from flue gas of the 5 MSWIs, 4 HWIs and the cases from literatures were presented in Table 3.

In this study, the emission factors of PCDD/Fs to the atmosphere from 5 MSWIs varied from 0.056 to 0.607 μg I-TEQ t^{-1} MSW (mean: 0.235 μg I-TEQ t^{-1}). It was estimated that 10.2-88.0 mg I-TEQ of PCDD/Fs (mean: 35.0 mg I-TEQ of PCDD/Fs) were annually released to the atmosphere from MSWIs. In particular, the mean EF values and EA values of the MSW were similar, respectively. These values were in good consistent with the study of Zhu *et al.* (2018), in which the EF and EA values ranged in 0.027–0.225 μg I-TEQ t^{-1} (mean value of 0.17 μg I-TEQ t^{-1}) and in 5.36–101 mg I-TEQ (mean: 35.0 mg I-TEQ), respectively.

The EF values ranged from 0.07 to 3.27 μg I-TEQ t^{-1} for HWIs (mean: 1.03 μg I-TEQ t^{-1}), varying significantly for different HWIs. PCDD/Fs annual released to the atmosphere were estimated as 1.13–53.0 mg I-TEQ (mean: 17.1 mg I-TEQ) in HWI. The noticeable EF value of HW03 among HWIs could be attributed to the particularly treated wastes, which were composed of oily refuse, rock debris, mud, etc.

Until 2018, 352 MSWIs which had a yearly capacity of 89.4 million tons were running in China, then the estimated annual emission amounts of PCDD/Fs from flue gas was 22.3 g TEQ. And the scale of HWI treatment reached 6.06 million tons per year, the estimated annual emission amounts of PCDD/Fs from flue gas was 6.51 g TEQ.

Table 3. Mean emission factor and emission of PCDD/Fs from 5 MSWIs, 4 HWIs and cases from the literatures.

Classification		Emission factor (μg I-TEQ t^{-1})	Annual emission amount (mg I-TEQ·y^{-1})	References
MW01		0.238 (0.121–0.607)	25.7 (13.1–65.5)	This study
MW02		0.221 (0.090–0.533)	31.9 (13.0–76.8)	This study
MW03		0.244 (0.056–0.489)	44.0 (10.2–88.0)	This study
MW04		0.278 (0.171–0.509)	40.0 (24.7–73.3)	This study
MW05		0.266 (0.106–0.549)	33.5 (13.4–69.1)	This study
HW01		0.176 (0.07–0.356)	2.85 (1.13–5.76)	This study
HW02		0.199 (1.72–2.26)	50.1 (43.2–53.0)	This study
HW03		2.92 (2.33–3.27)	6.30 (5.04–7.06)	This study
HW04		0.83 (0.26–1.9)	8.96 (2.81–20.5)	This study
China	Municipal waste	0.17 (0.027–0.225)	35.0 (5.36–101)	Zhu *et al.*, 2018
	Hospital Waste	63.3 (0.78–474)	4.87	Gao *et al.*, 2009
	Medical waste	1.92 (0.033–4.9)	3.15 (0.217–18.8)	Li *et al.*, 2017
VietNam	Municipal waste	0.111 (0.096–0.127)	0.3 (0.2–0.4)	Pham *et al.*, 2019a
	Industrial waste	0.991 (0.663–1.43)	6.4 (4.3–9.2)	
	Medical waste	16.4 (1.68–57.5)	23.6 (2.4–82.7)	

Congener Profiles of PCDD/Fs in Stack Gas

Fig. 3 shows the scope plot of principal component analysis, for relative contributions of the 17 toxic dioxin congeners. The first principal component (PC1) took into account 48.0% of the variability of the data set, and the second principal component (PC2) accounted for 43.7% of the total variance. PC1 and PC2 accounted for 91.7% of the total variance. The score plot of PCA indicated that all investigated MSWIs and HWIs could be assigned to three groups. Group1 on the left were samples from HW01 and HW02, group 2 in the middle were samples from MSWIs and HW04, and group 3 on the right included samples from HW03. As in Fig. 3, the distribution of PCDD/F congeners was similar between the samples from MSWIs, but different between the samples from MSWIs and HWIs. Moreover, samples from HWIs were also distinct.

Fig. 4 presented the congener profiles of PCDD/Fs in each group. Congener profiles of 17 PCDD/F congeners in the samples were analyzed to understand the formation pathways and release behavior of dioxins in the investigated facilities. The mass ratios of PCDFs to PCDDs ($R_{F/D}$) was always used to estimate the formation of dioxins by the de novo synthesis ($R_{F/D} > 1$) or by precursor formation ($R_{F/D} < 1$) (Luijk *et al.*, 1994; Huang and Buekens, 1995; Tuppurainen *et al.*, 2000; Everaert and Baeyens, 2002; Pham *et al.*, 2019b). The congener distributions were obtained by normalizing each congener to the total weight of all 2,3,7,8-substituted PCDD/F congeners. In Group 1, the proportion of PCDFs to the total amount of PCDD/Fs was 88.8% with a high proportion of 2,3,4,7,8-PeCDF (28.4%), 1,2,3,4,7,8-HxCDF (20.6%), 1,2,3,6,7,8-HxCDF (14.1%) and 2,3,4,6,7,8-HxCDF (10.3%). In Group 2, 2,3,4,7,8-PeCDF was the most abundant congener (35.3%) with higher fraction of 2,3,4,6,7,8-HxCDF (10.2%), and the total contribution of 2,3,7,8-substitued PCDFs was 70.8%. In Group 3, the congener 2,3,4,7,8-PeCDF contributed dominantly to the total TEQ with 31.4%. Other significant contributor were as follows: 2,3,7,8-TCDD (24.6%), 2,3,7,8-TCDF (17.5%) and 1,2,3,7,8-PeCDD (14.4%). PCDF isomer distributions indicated that low-chlorinated dibenzofurans were more than the high-chlorinated dibenzofurans. The ratios of PCDFs to PCDDs for all gas samples were over 1, implying the dominant role of de novo synthesis (Everaert and Baeyens, 2002; Ooi and Lu, 2011).

Concentration and TEQ Distribution of PCDD/F Congeners in Stack Gas from MSWIs and HWIs

Emission concentrations of 17 PCDD/Fs congeners and TEQ values in stack gas from 5 MSWIs and 4 HWIs were presented in Figs. 5 and 6. As shown in Fig. 5, the contribution of OCDD (37.6%) and 1,2,3,4,6,7,8-HpCDD (14.2%) in terms of total concentration were higher than other congeners. Notably, OCDD is the dominant congener in stack gas. However, for TEQ concentration, the 2,3,4,7,8-PeCDF, 2,3,4,6,7,8-HxCDF and 1,2,3,7,8-PeCDD were the dominant congeners of PCDD/Fs in stack gas, and accounted for 38.5%, 9.4% and 8.86%, respectively. They contribute over 50% of the total TEQ. Li *et al.* (2017) and Zhu *et al.* (2018) also found that 2,3,4,7,8-PeCDF was the largest contributor in MWIs, this conclusion was supported by the previous works (Bie *et al.*, 2007; Choi *et al.*, 2008; Gao *et al.*, 2009; Yoon *et al.*, 2017; Trinh and Chang, 2018; Zhu *et al.*, 2018).

The congener profiles in the stack gas from HWIs were shown in Fig. 6. According to these TEQ results, the congener profiles of HWIs could be classified into three main groups: (1) PCDFs were more abundant than PCDDs with 2,3,4,7,8- PeCDF (28.4%), 1,2,3,4,7,8-HxCDF(20.6%) and 1,2,3,6,7,8-HxCDF(14.1%) as major congeners (e.g., HW01 and HW02); (2) the percentages of PCDFs and PCDDs were 59.17% and 40.83%, respectively, with 2,3,4,7,8-PeCDF (31.4%) and 2,3,7,8-TCDD (24.61%) as major congeners (e.g., HW03); (3) PCDFs were more abundant than PCDDs with high proportions of 2,3,4,7,8-PeCDF (37.34%) congener (e.g., HW04). We concluded that the de novo synthesis was always dominant.

Influence of PCDD/F Emissions on the Surrounding Atmosphere

The concentrations and the PCDD/Fs patterns in the ambient

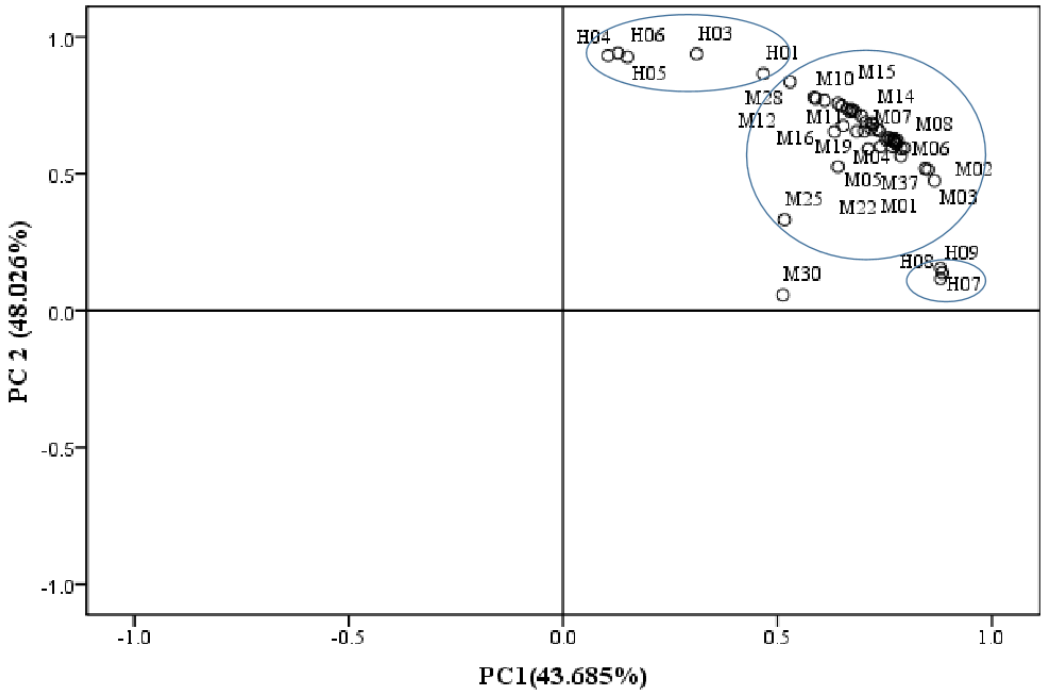

Fig. 3. Score plot of principal component analysis of 17 toxic PCDD/F emission.

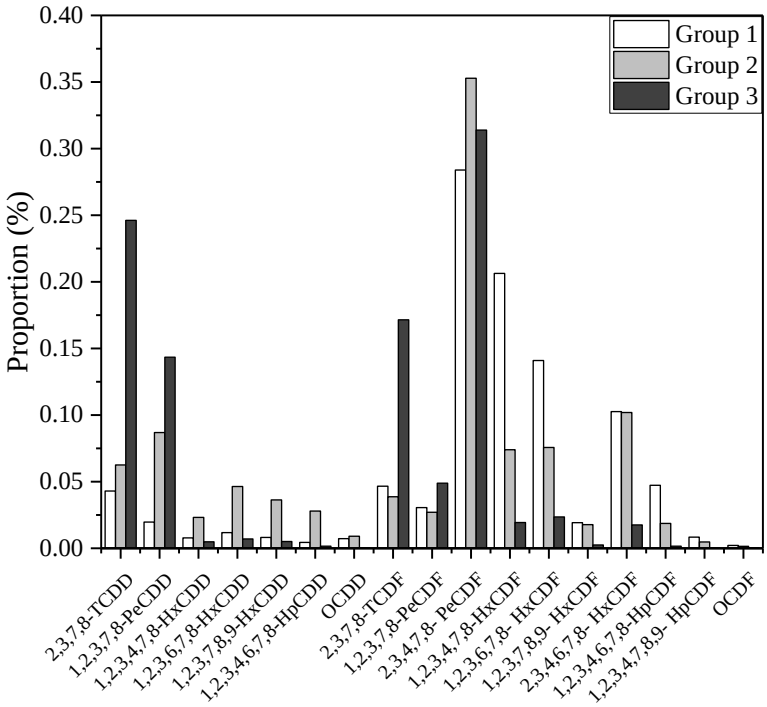

Fig. 4. Representative congener patterns of PCDD/Fs in each group (Group 1: H01–H06, Group 2: M01–M39 and H10–H12, Group 3: H07–H09).

air were studied to determine if the PCDD/F emissions from each plant affecting their surrounding environment.

As could be seen from Table 2, the PCDD/F concentrations in the ambient air varied between 0.088 and 13.9 pg I-TEQ m^{-3} with an average value of 0.185 pg I-TEQ m^{-3}. Among the monitoring sites, MW01-A3 showed the highest PCDD/F concentration, which was roughly two order of magnitude higher than those in other sites. The possible factor was due to the location that was by the chimney. These values except MW01-A3 which was by the chimney were all below the emission limit of national standard in Japan (0.6 pg I-TEQ m^{-3}). Xu *et al.* (2009) founded that the PCDD/F concentrations in the ambient air varied from 0.059 to 3.03 pg I-TEQ m^{-3} near the MSWIs in eastern China. Oh *et al.* (2006) reported that

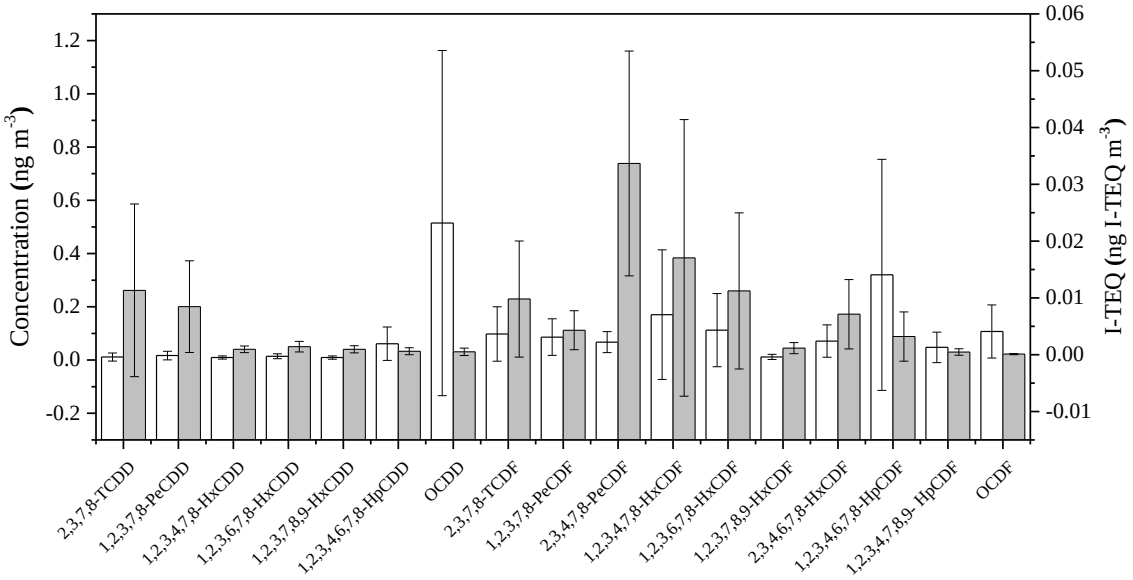

Fig. 5. PCDD/F congeners distribution and TEQ in stack gas from MSWIs.

the levels of PCDD/Fs in ambient air near a MSWI in Korea were 0.22–1.16 pg I-TEQ m^{-3} with an average of 0.66 pg I-TEQ m^{-3}. Li *et al.* (2018) reported that the concentration of PCDD/Fs in surrounding atmosphere of MSWIs in China was 0.05–0.12 pg I-TEQ m^{-3}.

It could be seen that PCDD/F concentrations was high in winter and low in summer in the same sites. The possible factors were the difference of waste composition, temperature conditions. It could be seen that PCDD/F concentrations of the samples collected by the chimney were higher than the concentrations of the samples collected in the boundary of the plants. we could observe a trend of declining PCDD/Fs levels as the distance increased. If PCDD/Fs levels tended to decrease as the distance from a certain site increase, then the site could be considered a source of PCDD/Fs (Oh *et al.*, 2006). Comparing the sample MW01-A3 and HW03-A2, we can see that when the PCDD/F concentrations in the stack gas emitted from the plant were relatively low, the corresponding levels in the ambient air were also lower (Hu *et al.*, 2014).

As could be seen in Fig. 7, the concentration of 2,3,4,7,8-PeCDF in the ambient air was higher than other congener of PCDD/Fs, and the PCDD/Fs congener profiles of them were similar with each other, the result was similar with the previous study by Chen *et al.* (2015) and Oh *et al.* (2006).

Fig. 8 shows comparison of PCDD/Fs patterns in ambient air and stack gas, the relative importance of the individual congener was normalized to the total I-TEQ value. Among all PCDD/F congeners, 2,3,4,7,8-PeCDF was the most predominant (21.1%–40.5% to the ∑PCDD/Fs), and the content of PCDFs was higher than PCDDs, Ratios of PCDF/PCDD varied from 0.688 to 0.831, and the less chlorinated PCDD/Fs were the main congeners in the ambient air, these were also true with peer investigation (Oh *et al.*, 2006; Xu *et al.*, 2009; Hu *et al.*, 2014; Chen *et al.*, 2015). Besides, PCDD/Fs patterns in ambient air were similar to the patterns in the stack gas samples. These results indicated that PCDD/F emission from the plants had an obvious impact on the environments surrounding the plants.

Congener Profiles and Levels of PCDD/Fs in Surrounding Soil

Table 4 summarized the content of PCDD/Fs in soils. The I-TEQ values of PCDD/F ranged from 0.68 to 3.76 ng I-TEQ kg^{-1} in soils in the vicinity of MSWIs, with an average and a median value of 1.73 and 1.51 ng I-TEQ kg^{-1}. The I-TEQ values of PCDD/F ranged from 0.91 to 16.0 ng I-TEQ kg^{-1} in soils in the vicinity of HWIs, with an average and a median value of 7.93 and 7.41 ng I-TEQ kg^{-1}. The values of TEQ in the soil samples in the vicinity of MSWIs and HWIs were comparable with those in surrounding soil of MSWI in China (0.39–5.04 pg I-TEQ g^{-1}) (Yan *et al.*, 2008) and (7.62–15.5 ng I-TEQ kg^{-1}) (Li *et al.* 2018), the iron and steel plant (0.16–4.5 ng I-TEQ kg^{-1}) (Zhou *et al.*, 2019), hospital waste incinerator (0.46–2.63 ng I-TEQ kg^{-1}) (Li *et al.*, 2012) and lower than those in the vicinity of electronic wastes disposal sites in Indian (8–99 ng I-TEQ kg^{-1}) (Chakraborty *et al.*, 2018). For the ratio of the PCDF/PCDD (F/D) ranged from 0.05–1.72 with a mean value of 0.52. Among 11 surrounding soil samples, there were 9 samples whose F/D ratio below 1, suggesting combustion was not the mainly source of PCDD/Fs in surrounding soil (Li *et al.*, 2018). The 2 surrounding soil samples (F/D = 1.51, 1.72) from HW03 and HW04, indicating abundant enrichment of PCDFs in these soil sample, and the high TEQ of PCDD/Fs was formed because of the long-term deposition of pollutants in the vicinity of HWIs (Nganai *et al.*, 2014).

As shown in Fig. 9. For mass concentration of PCDD/Fs, among all soil samples, OCDD was the predominant congener, accounting for 28.3%–77.7% of the total concentration of 17 congeners for soil samples, followed by the high-chlorinated congeners 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF, and OCDF, which was consistent with those previously reported by other investigators (Yan *et al.*, 2008; Meng *et al.*, 2016; Zhou *et al.*, 2019; Li *et al.*, 2018).

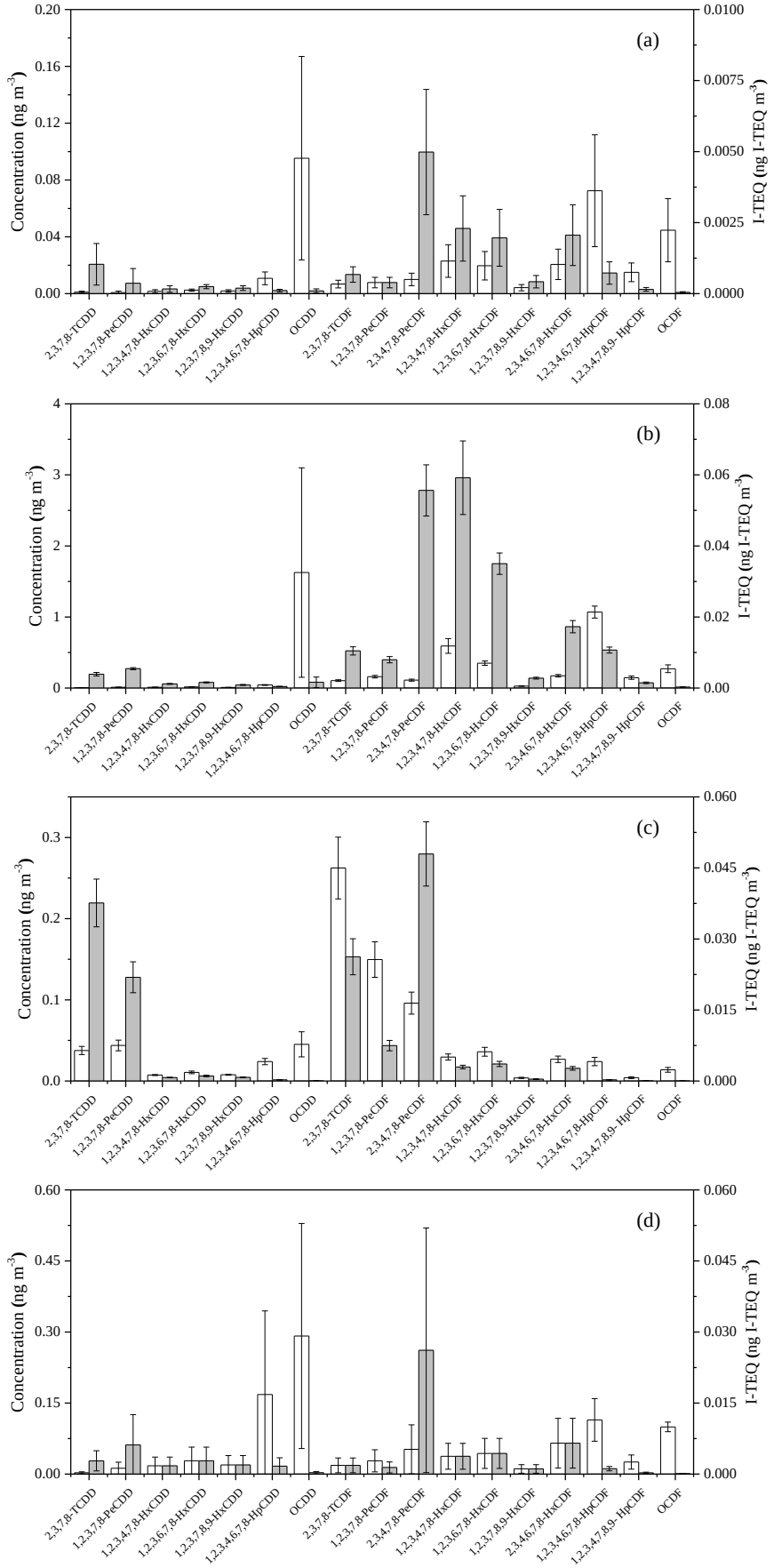

Fig. 6. PCDD/F congeners distribution and TEQ in stack gas from HWIs (a: HW01, b: HW02, c: HW03, d: HW04).

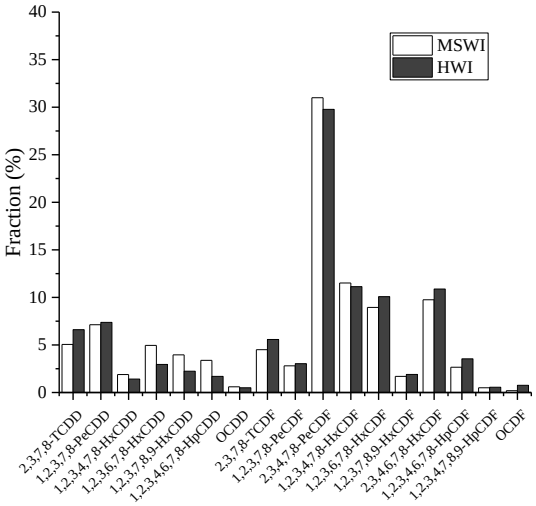

Fig. 7. PCDD/F congener profiles in the ambient air of MSWI and HWI.

CONCLUSIONS

In this study, all of the investigated MSWIs exhibited a small variation in PCDD/F emissions to the atmosphere (0.051–0.078 ng I-TEQ m^{-3} with an average value of 0.058 ng I-TEQ m^{-3}), and the emission levels of 5 MSWIs were below the current standard in China (0.1 ng I-TEQ m^{-3}). However, the emission of PCDD/Fs from the investigated HWIs ranged from 0.016–0.29 ng I-TEQ m^{-3} with an average value of 0.159 ng I-TEQ m^{-3}. Two HWIs had emission levels above the current standard in China, indicating that incineration technologies should be further improved. The emission factors of PCDD/Fs to the atmosphere were in the range of 0.056 to 0.607 μg I-TEQ t^{-1} MSW (mean: 0.235 μg I-TEQ t^{-1}) and 0.07–3.27 μg I-TEQ t^{-1} HW (mean: 1.03 μg I-TEQ t^{-1}), respectively. It was estimated that 10.2–88.0 mg (mean: 35.0 mg) and 1.13–53.0 mg (mean: 17.1 mg) I-TEQ of PCDD/Fs were annually released from MSWIs and HWIs to the atmosphere in China, respectively. Based on principal

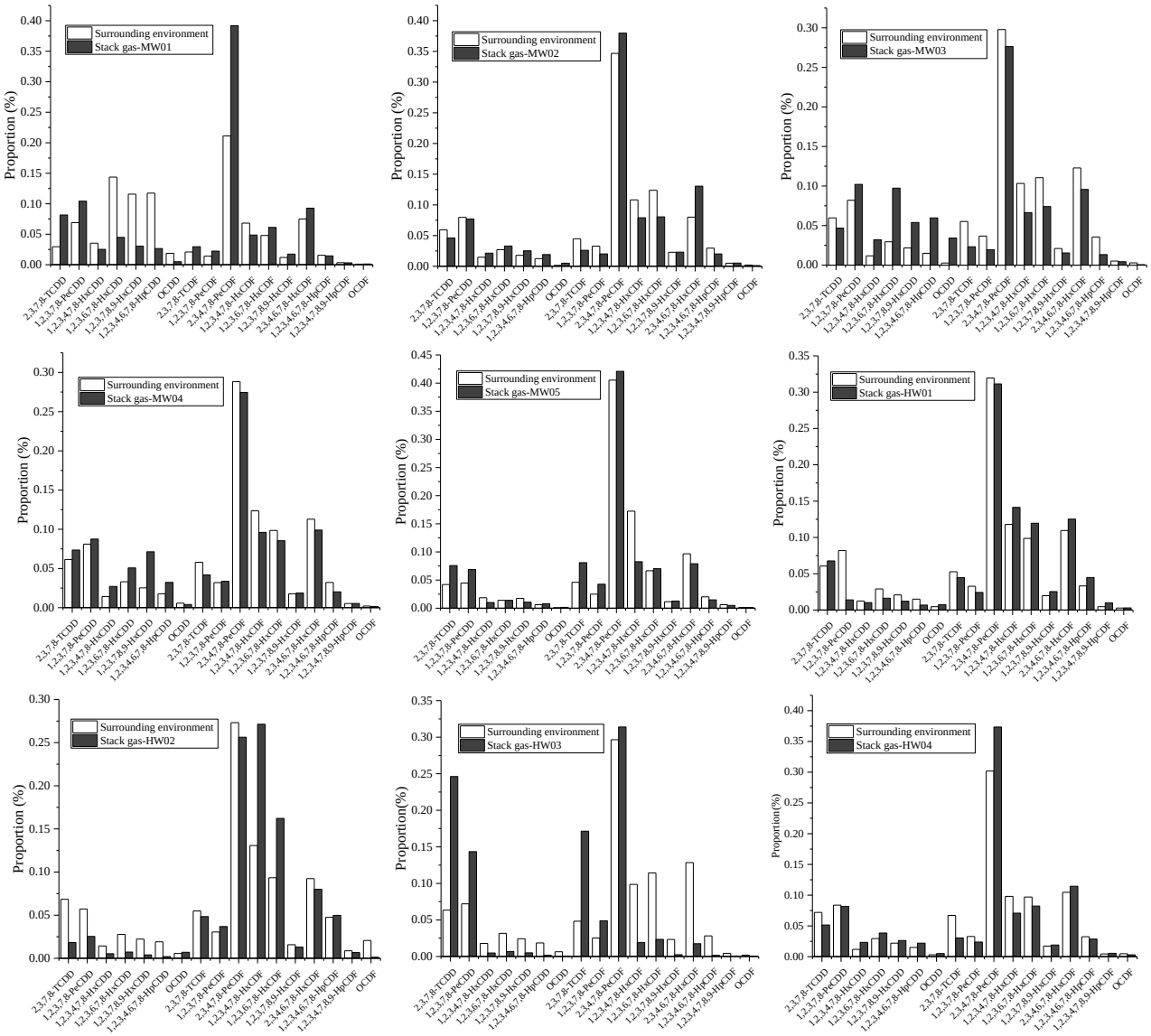

Fig. 8. Comparison of PCDD/Fs patterns in ambient air and stack gas.

Table 4. Content of PCDD/Fs (ng I-TEQ kg^{-1} dw) in soil samples in the vicinity of the MSWIs and HWIs.

PCDDs & PCDFs	MW01-S1	MW02-S1	MW02-S2	MW03-S1	MW04-S1	MW05-S1	MW05-S2	HW01-S1	HW02-S1	HW03-S1	HW04-S1
2,3,7,8-TCDD	0.14	0.13	0.05	0.10	0.15	0.09	0.03	0.12	0.15	0.74	0.24
1,2,3,7,8-PeCDD	0.33	0.23	0.17	0.31	0.21	0.14	0.25	0.14	1.68	1.12	0.93
1,2,3,4,7,8-HxCDD	1.06	0.37	0.25	0.42	0.38	0.15	1.16	0.16	6.70	1.16	1.70
1,2,3,6,7,8-HxCDD	0.59	0.99	0.93	1.03	0.56	0.27	7.20	0.35	53.09	1.91	2.53
1,2,3,7,8,9-HxCDD	0.59	0.63	0.62	1.09	1.01	0.28	5.21	0.28	29.63	1.37	2.56
1,2,3,4,6,7,8-HpCDD	3.72	20.82	11.71	11.27	11.90	2.06	118.63	6.79	164.72	11.27	34.18
OCDD	21.30	132.66	78.10	28.70	40.94	29.19	260.58	34.53	323.09	46.25	139.78
2,3,7,8-TCDF	1.00	1.13	0.58	0.87	0.61	0.54	0.30	0.50	0.25	2.72	1.85
1,2,3,7,8-PeCDF	1.73	0.80	0.57	0.78	0.56	0.45	0.40	0.40	0.90	3.72	3.21
2,3,4,7,8- PeCDF	0.80	0.58	0.50	0.44	0.54	0.29	0.31	0.38	1.34	1.67	2.65
1,2,3,4,7,8-HxCDF	1.62	2.41	1.38	1.60	1.81	0.53	3.59	1.13	22.63	9.29	12.28
1,2,3,6,7,8-HxCDF	0.99	1.13	0.85	0.91	1.66	0.46	1.05	0.73	3.82	4.60	11.81
1,2,3,7,8,9-HxCDF	0.27	0.23	0.24	0.14	0.41	0.12	0.36	0.17	2.21	0.96	2.25
2,3,4,6,7,8-HxCDF	1.33	0.84	0.93	1.54	0.97	0.39	0.27	0.56	1.34	3.97	17.84
1,2,3,4,6,7,8-HpCDF	2.85	5.26	8.06	3.21	4.65	1.40	4.77	2.36	24.38	25.62	113.29
1,2,3,4,7,8,9-HpCDF	0.51	0.79	0.58	0.48	0.76	0.14	0.77	0.15	7.17	4.04	17.23
OCDF	3.12	5.87	14.08	18.34	5.02	1.06	7.39	2.14	41.56	39.72	129.66
∑PCDD	27.74	155.83	91.83	42.91	55.15	32.18	393.06	42.37	579.07	63.83	181.92
∑PCDF	14.21	19.04	27.78	28.30	16.98	5.39	19.22	8.52	105.59	96.31	312.06
Ratio of F/D	0.51	0.12	0.30	0.66	0.31	0.17	0.05	0.20	0.18	1.51	1.72
∑PCDD/Fs	41.95	174.87	119.61	71.21	72.13	37.57	412.28	50.89	684.66	160.13	493.98
∑PCDD/Fs (ng I-TEQ kg^{-1})	1.63	1.75	1.29	1.46	1.51	0.68	3.76	0.91	16.0	5.42	9.39

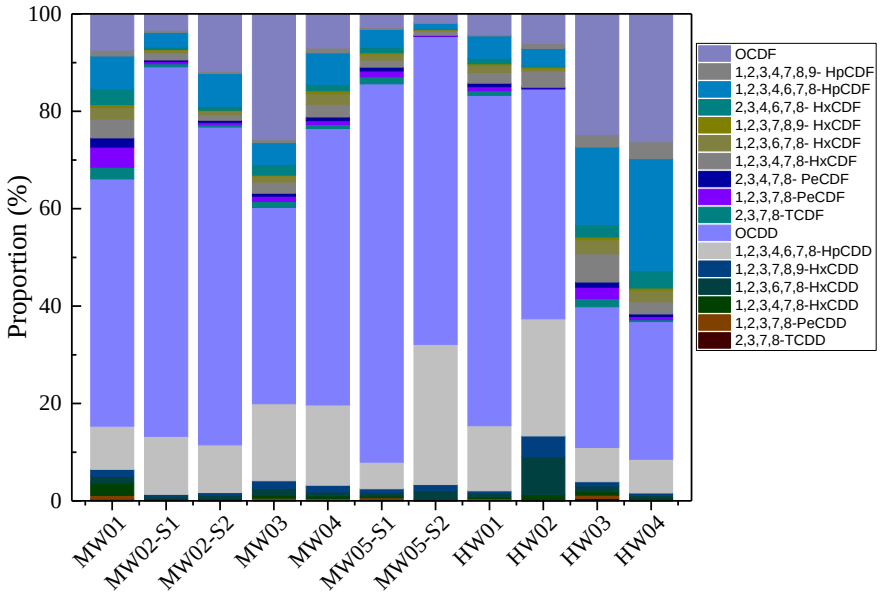

Fig. 9. Composition profiles of PCDD/Fs in the soil samples in the vicinity of MSWIs and HWIs.

component analysis, most MSWIs belonged to Group 2, and 4 investigated HWIs could be divided into three groups because of different feeding wastes. Our results indicated that PCDD/F emission from the plants had an obvious impact on the PCDD/F concentration and patterns in ambient air. The I-TEQ values of PCDD/F in soil samples in the vicinity of MSWIs were lower than those around the HWIs.

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