



Emissions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from an Electric Arc Furnace, Secondary Aluminum Smelter, Crematory and Joss Paper Incinerators

Jui-Chi Chiu¹, Yun-Hwei Shen^{1†}, Hsing-Wang Li^{2,3*}, Long-Full Lin⁴, Lin-Chi Wang⁵, Guo-Ping Chang-Chien⁵

¹ Department of Resources Engineering, National Cheng Kung University, Tainan, Taiwan

² Department of Environmental Engineering, National Cheng Kung University, Tainan, Taiwan

³ Sustainable Environment Research Center, National Cheng Kung University, Tainan, Taiwan

⁴ Department of Environmental Engineering, Kun Shan University, Tainan, Taiwan

⁵ Department of Chemical and Materials Engineering, Cheng Shiu University, Kaohsiung, Taiwan

ABSTRACT

Emissions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) from the stack flue gases, fly ashes and bottom ashes of various stationary sources were investigated. The mean total PCDD/F I-TEQ concentration of flue gas ranged from 0.00681 to 0.703 ng I-TEQ/Nm³. However, the emission factor of PCDD/F from various incinerators was 0.00827 to 3.50 µg I-TEQ/ton, whereas it was 5.36 µg I-TEQ/body for a crematory (CM). In addition, the mean total PCDD/F I-TEQ content in fly ash from an electric arc furnace (EAF) and a secondary aluminium smelter (secondary ALS) were 74.0, and 49.9 ng I-TEQ/kg, respectively, whereas they are 21.3 and 0.494 ng I-TEQ/kg for bottom ash. Meanwhile, the removal efficiency of PCDD/F by bag filters from EAF was –44.4% which is attributed to the “memory effect”. The indicatory PCDD/Fs of EAF, and secondary ALS have the same congeners (1,2,3,7,8,9-HxCDF, 2,3,7,8-TeCDF, and 1,2,3,7,8-PeCDF). In addition, CM, joss paper-A (JP-A) and joss paper-B (JP-B) incinerators have similar indicatory PCDD/F (2,3,4,6,7,8-HxCDF, OCDF, 1,2,3,4,6,7,8-HpCDD, and OCDD). The high contribution of total PCDD/F is from fly ash (61.1–95.3%) for metallurgical facilities (EAF, secondary ALS), whereas 99.9% contribution of stack flue gas is from JP-A and JP-B. In conclusion, continually monitoring various PCDD/F emission sources is necessary to understand current PCDD/F emission (flue gas, fly/bottom ash) and the related removal efficiency of existing air pollution control devices. Information about both emission factors of PCDD/Fs and indicatory PCDD/F congeners are useful for the establishment of control strategies and for use as fingerprints with regard to the dominant congeners from different emission sources.

Keywords: PCDD/Fs; Flue gas; Emission factor; Fly ash; Indicatory congeners.

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) were first detected in the flue gas and fly ash from municipal solid waste incinerators in 1977 (Olie, 1977). Since then, PCDD/Fs have received considerable attention due to concerns about the related adverse health effects and emission problems (US EPA,

2000). Various forms combustions, such as incinerations, are believed to be the main sources of PCDD/F to the environment (Rappe, 1992; US EPA, 2000). Furthermore, these pollutants can be transported over long distances in the atmosphere, and be deposited on the surrounding environment (Tysklind *et al.*, 1993; VanJaarsveld *et al.*, 1993; Lee *et al.*, 2009).

In Taiwan, the PCDD/F emission inventory has been reported that iron ore sintering, heat/power generation, electric arc furnaces, secondary aluminum smelter plants, crematories and ritual paper burning make contributions of 32%, 30%, 23%, 1.5%, 0.7% and 0.1% to the total emissions, respectively (Wang *et al.*, 2003a; Lin *et al.*, 2007). Regulations, control technology (devices), operating conditions and feeding materials can all affect PCDD/F emission and the related emission factors must be investigated yearly to develop appropriate control strategies

* Corresponding author. Tel: +886-6-2757575 ext. 65831;
Fax: +886-6-2752790

E-mail address: p5893102@nckualumni.org.tw

† Corresponding author. Tel: +886-6-275757 ext. 62820;
Fax: +886-6-2380421

E-mail address: yhshen@mail.ncku.edu.tw

(Wang *et al.*, 2003b; Li *et al.*, 2007a). Therefore, monitoring the PCDD/F emissions from various stationary sources is of great importance for environmental protection issue. For example, the removal efficiency of total PCDD/Fs and the international toxicity equivalence (I-TEQ) value were found to be 57% and 58%, respectively, after injecting powder activated carbon (PAC) in a hazardous waste incinerator (Karademir, *et al.*, 2004). However, the PAC injection was followed by various types of air pollution control devices to enhance the removal efficiency of PCDD/Fs, approaching the range of 92%–99% in a municipal waste incinerator (Tejima, *et al.*, 1996; Abad *et al.*, 2003). A combination of air pollution control devices, including a dry scrubber, activated carbon injection and bag filters have been frequently used, achieving overall removal efficiencies of up to 99% (Li *et al.*, 2008; Wang *et al.*, 2009).

This study investigates the characterization of PCDD/F emission from an electric arc furnace (EAF), a secondary aluminum smelter (secondary ALS), a crematory (CM) and two joss paper incinerators (JP-A, JP-B) in southern Taiwan. Meanwhile, the fly ash and bottom ash were collected to measure the PCDD/F content. In addition, the removal efficiencies of PCDD/F by bag filters were evaluated. Finally, the emission factors of PCDD/Fs from five emission sources were reported and the indicatory PCDD/F congeners were established.

METHODS

Sampling of PCDD/Fs

The stack flue gas samples were collected from an electric arc furnace (EAF), a secondary aluminium smelter (secondary ALS), a crematory (CM), and two joss paper incinerators (JP-A, JP-B). The sampling procedures followed those of the US EPA Modified Method 23 (US EPA, 1996). Prior to sampling, XAD-2 resin was spiked with PCDD/F surrogate standards prelabeled with isotopes. The fly ash samples were collected by the bag filters and bottom ash samples were collected directly from each furnace. The ash samples were well-mixed and diagonally sectioned for PCDD/F analyses.

Analysis of PCDD/Fs

The analyses of the stack flue gas samples were performed according to the US EPA Modified Method 23, while the fly/bottom ash samples were analysed with the US EPA Method 1613B (US EPA, 1994; 1996). In brief, samples were extracted with toluene for 24 hr and this was then followed by a series of sample cleanup procedures, including a multilayer silica gel column, alumina column and activated carbon column. The extract was transferred to a vial, and finally further concentrated by a N₂ gas stream.

The high-resolution gas chromatographs/high-resolution mass spectrometers (HRGC/HRMS) were used for PCDD/F analysis. The HRGC (Hewlett-Packard 6970 Series gas, CA) was equipped with a DB-5 fused silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25 μm) (J&W Scientific, CA) with a splitless injection, while the HRMS (Micromass Autospec Ultima, Manchester, UK) had a

positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring was used with the resolving power at 100,000. The electron energy and source temperature were specified at 35 eV and 250°C, respectively. The oven temperature program was set as followings: initially at 150°C (held for 1 min), then increased by 30 °C/min to 220°C (held for 12 min), and finally increased by 1.5 °C/min to 310°C (held for 20 min). Helium was used as the carrier gas. The recoveries of PCDD/F internal standards ranged from 65–102% which met the criteria within 25–130%.

Indicatory PCDD/Fs

The indicatory PCDD/F is used to figure out the major PCDD/F congener for different PCDD/F emission sources. The indicatory PCDD/Fs analysis is undertaken using the following equation (Yang *et al.*, 1998; Lee *et al.*, 2004; Lin *et al.*, 2006; Lin *et al.*, 2007):

$$\text{Ratio}_{ji} = (X_i / \Sigma X)_j / (X_i / \Sigma X)_{\min} \quad (1)$$

where the numerator is the mass fraction of the *i*th congener of emission source *j*, and the denominator is the minimum value of the mass fraction of the *i*th congener among all emission sources. A higher value of Ratio_{ji} means that the *i*th congener of emission source *j* is the one that has a greater contribution than the other emission sources. In this way, the highest three ratio values of PCDD/Fs for each source were recognized as the indicatory PCDD/Fs.

RESULTS AND DISCUSSION

PCDD/F Concentrations in the Stack Flue Gas

The mean PCDD/F concentrations in the stack flue gases of various sources are listed in Table 1. The mean total PCDD/F concentration ranged from 0.315 to 18.7 ng/Nm³, while the corresponding total I-TEQ value ranged from 0.00681 to 0.703 ng I-TEQ/ Nm³. These results are close to those of previous studies, such as, 0.28 ng I-TEQ/Nm³ for EAF, 3.3–22.7 ng I-TEQ/Nm³ for ALS and 0.166–4.13 ng I-TEQ/Nm³ for JP (Chen *et al.*, 2004; Lee *et al.*, 2005; Li *et al.*, 2007a; Hu *et al.*, 2009).

The results of the experiment showed that the total PCDD/F I-TEQ concentrations from secondary ALS, CM and JP-A were higher than the PCDD/F emissions standard for a stationary source (0.1 ng I-TEQ/Nm³) (EPA Taiwan, 2009). Therefore, using some air pollution control methods, such as activated carbon injection and bag filter, are necessary to reduce PCDD/F emissions from the above emission sources.

In addition, the PCDFs are the major contributors of PCDDs/PCDFs, because the PCDDs/PCDFs I-TEQ ratio is < 1. For CM, the mean total PCDD/F I-TEQ concentration was 0.139 ng I-TEQ/ Nm³, which is close to that found in a previous study (0.322 ng I-TEQ/Nm³) (Wang *et al.*, 2003c). In addition, the mean total PCDD/F I-TEQ concentration from JP-A and JP-B were 0.00681–0.703 ng I-TEQ/Nm³, which is slightly lower than in another earlier report (0.166–4.13 ng I-TEQ/Nm³) (Hu *et al.*, 2009).

Table 1. PCDD/F concentration in the stack flue gas from various sources.

PCDD/Fs	EAF	RSD %	Secondary ALS	RSD %	CM	RSD %	JP-A	RSD %	JP-B	RSD %
2,3,7,8-TeCDD	0.00162	72.9	0.00895	58.3	0.00508	41.8	0.0164	49.9	0.000715	48.2
1,2,3,7,8-PeCDD	0.00638	91.5	0.0137	57.4	0.0173	61.2	0.0942	49.8	0.00151	53.7
1,2,3,4,7,8-HxCDD	0.00469	100	0.00574	37.2	0.0245	78.4	0.167	58.7	0.00207	46.8
1,2,3,6,7,8-HxCDD	0.014	106	0.00983	30.2	0.0663	83.0	0.349	80.8	0.00352	58.9
1,2,3,7,8,9-HxCDD	0.00797	108	0.00847	28.9	0.0521	82.8	0.283	76.4	0.00341	55.8
1,2,3,4,6,7,8-HpCDD	0.0552	109	0.0403	39.4	0.495	82.1	2.93	100	0.0392	61.3
OCDD	0.0343	74.8	0.0411	58.7	0.565	61.2	3.68	96.7	0.181	79.0
2,3,7,8-TeCDF	0.0221	67.0	0.648	42.6	0.0359	38.7	0.0853	52.5	0.00194	31.3
1,2,3,7,8-PeCDF	0.0229	85.7	0.163	36.3	0.0502	50.2	0.209	65.1	0.00221	27.2
2,3,4,7,8-PeCDF	0.0401	90.0	0.208	30.3	0.0834	56.5	0.351	67.9	0.00361	24.5
1,2,3,4,7,8-HxCDF	0.0327	98.4	0.0784	36.9	0.117	74.4	0.726	85.2	0.00369	25.7
1,2,3,6,7,8-HxCDF	0.0279	98.1	0.0719	38.6	0.132	75.1	0.669	87.6	0.00406	34.8
1,2,3,7,8,9-HxCDF	0.027	99.7	0.0777	32.4	0.0273	92.2	0.153	86.2	0.00116	50.2
2,3,4,6,7,8-HxCDF	0.00282	113	0.00378	46.6	0.216	81.0	1.26	98.1	0.00591	36.5
1,2,3,4,6,7,8-HpCDF	0.0677	102	0.229	25.2	0.686	82.7	4.14	109	0.0201	45.5
1,2,3,4,7,8,9-HpCDF	0.0102	105	0.0205	35.5	0.0978	81.5	0.681	104	0.00399	48.0
OCDF	0.0169	77.6	0.0642	48.7	0.389	74.1	2.90	117	0.0369	92.3
PCDDs	0.124	97.2	0.128	46.7	1.23	70.5	7.52	94.0	0.232	74.6
PCDFs	0.27	93.5	1.56	33.1	1.84	76.0	11.2	103	0.084	62.3
PCDDs/PCDFs ratio	0.443	–	0.0817	–	0.730	–	0.738	–	2.63	–
Total PCDD/Fs ng/Nm ³	0.394	94.6	1.69	33.0	3.06	73.5	18.7	99.6	0.315	70.9
PCDD ng I-TEQ/Nm ³	0.00808	92.8	0.0186	53.6	0.0335	69.5	0.176	63.1	0.00294	51.9
PCDFs ng I-TEQ/Nm ³	0.0332	91.0	0.203	32.8	0.105	67.8	0.526	83.4	0.00387	30.3
PCDDs/PCDFs (TEQ) ratio	0.227	–	0.0905	–	0.312	–	0.372	–	0.730	–
Total PCDD/F I-TEQ ng I-TEQ/Nm ³	0.0413	91.3	0.221	33.2	0.139	68.2	0.703	78.1	0.00681	39.6

Fig. 1 shows the congener profiles of the 17 2,3,7,8-substituted PCDD/Fs and each selected congener was normalized by the sum of 17 PCDD/Fs. For EAF, the dominant congeners were 1,2,3,4,6,7,8-HpCDF (17.1%) and 1,2,3,4,6,7,8-HpCDD (13.9%) (Fig. 1a). As for secondary ALS, the major congeners were 2,3,7,8-TeCDF (38.3%) and 1,2,3,4,6,7,8-HpCDF (13.5%) (Fig. 1b). For CM, the most abundant congeners were 1,2,3,4,6,7,8-HpCDF (22.4%), OCDD (18.5%), and 1,2,3,4,6,7,8-HpCDD (16.1%) (Fig. 1c). The OCDD (19.6–57.5%), 1,2,3,4,6,7,8-HpCDF (6.37–22.1%), 1,2,3,4,6,7,8-HpCDD (12.4–15.7%) and OCDF (11.7–15.5%) were the dominant congeners in JP-A and JP-B (Fig. 1d and 1e).

Emission Factors of Various PCDD/F Sources

The emission factors can be used to establish the PCDD/Fs inventory and to develop an integrated the PCDD/F control strategy. Table 2 lists the measured mean emission factors of PCDD/Fs from the five emission sources. The input rate of feeding materials from EAF, secondary ALS, JP-A, JP-B and CM are 72.0 ton/hr, 1.2 ton/hr, 4.0 kg/hr, 2.4 kg/hr, and 0.5 body/hr, respectively. The emission factor of PCDD/F from EAF, secondary ALS, JP-A and JP-B are 0.416, 3.50, 0.166 and 0.00827 $\mu\text{g I-TEQ/ton}$, respectively. These results are consistent

with those obtained in previous studies, which were 1.8–2.4 $\mu\text{g I-TEQ/ton}$ for EAF, 2.66–50.1 $\mu\text{g I-TEQ/ton}$ for ALS and 0.0203–0.176 $\mu\text{g I-TEQ/ton}$ for JP (Chen *et al.*, 2004; Lee *et al.*, 2005; Lin *et al.*, 2007; Li *et al.*, 2007a; Hu *et al.*, 2009). For CM, the emission factor is 5.36 $\mu\text{g I-TEQ/body}$, which is also close to the results in an earlier study (6.11–13.6 $\mu\text{g I-TEQ/body}$) (Wang *et al.*, 2003c).

PCDD/F Contents in Fly Ash and Bottom Ash

The mean PCDD/F contents in fly ash and bottom ash are listed in Table 3. The mean total PCDD/F content in fly ash from EAF and secondary ALS were 1039, and 639 ng/kg, respectively, while the corresponding I-TEQ values were 74.0, 49.9 ng I-TEQ/kg. As for bottom ash, the total PCDD/F content from EAF, secondary ALS, JP-A and JP-B were 528, 8.87, 8.36 and 5.98 ng/kg, respectively, and the corresponding I-TEQ values were 21.3, 0.494, 0.457 and 0.148 ng I-TEQ/kg. The highest total PCDD/F I-TEQ value (74.0 ng I-TEQ/kg) in fly ash from EAF was lower than that (194 ng I-TEQ/kg) from a municipal solid waste incinerator (Giugliano *et al.*, 2002). Since the total flue gas PCDD/F concentration is low, the fly ash contents are also expected to be low.

Fig. 2 shows the congener profiles of the 17 2,3,7,8-substituted PCDD/Fs and each selected congener was

normalized by the sum of 17 PCDD/Fs. For EAF, the major congeners in fly ash were OCDF, 1,2,3,4,6,7,8-HpCDF and 1,2,3,7,8,9-HxCDF (Fig. 2a); in bottom ash they were OCDD, 1,2,3,4,6,7,8-HpCDD, and OCDF (Fig. 2b). As for secondary ALS, the dominant congeners in fly ash were OCDD, 2,3,7,8-TeCDF and OCDF (Fig. 2c); in bottom ash they were OCDF, OCDD and 1,2,3,4,6,7,8-HpCDF (Fig. 2d). For JP-A and JP-B, the most abundant

congeners were OCDD, OCDF, 1,2,3,4,6,7,8-HpCDF, and 1,2,3,4,6,7,8-HpCDD (Fig. 2e and 2f).

PCDD/F Removal Efficiency by Bag Filter

The gas samples were taken at the same time before and after the bag filters to evaluate the efficiencies of the filters in EAF. The mean PCDD/F emission rate before and after the bag filter were 277 and 400 µg/hr, respectively, while

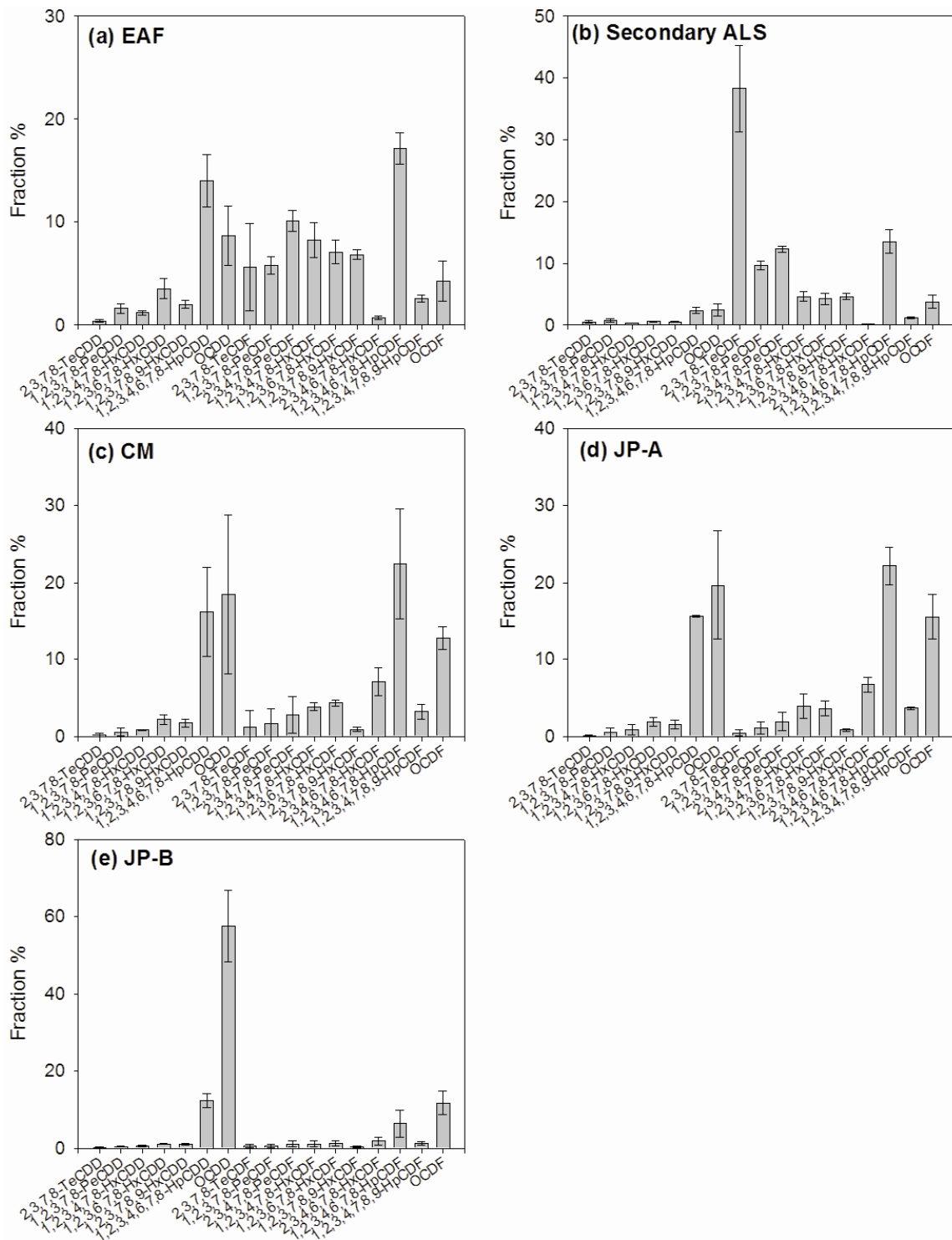


Fig. 1. Congener profiles of seventeen 2,3,7,8-substituted PCDD/Fs in flue gases from various sources.

Table 2. PCDD/F emission factor from various sources.

PCDD/Fs	EAF	Secondary ALS	CM	JP-A	JP-B
2,3,7,8-TeCDD	0.0163	0.142	0.281	0.00386	0.000864
1,2,3,7,8-PeCDD	0.0643	0.216	0.964	0.0222	0.00183
1,2,3,4,7,8-HxCDD	0.0472	0.0909	0.897	0.0393	0.00251
1,2,3,6,7,8-HxCDD	0.141	0.155	2.43	0.082	0.00427
1,2,3,7,8,9-HxCDD	0.0803	0.134	1.91	0.0666	0.00415
1,2,3,4,6,7,8-HpCDD	0.556	0.637	18.0	0.692	0.0477
OCDD	0.346	0.650	20.4	0.87	0.220
2,3,7,8-TeCDF	0.224	10.2	1.33	0.0201	0.00235
1,2,3,7,8-PeCDF	0.231	2.58	1.85	0.0493	0.00269
2,3,4,7,8-PeCDF	0.404	3.29	3.07	0.083	0.00439
1,2,3,4,7,8-HxCDF	0.329	1.24	4.32	0.171	0.00449
1,2,3,6,7,8-HxCDF	0.281	1.14	4.85	0.158	0.00494
1,2,3,7,8,9-HxCDF	0.272	1.23	1.01	0.0362	0.00141
2,3,4,6,7,8-HxCDF	0.0284	0.0598	7.92	0.297	0.00718
1,2,3,4,6,7,8-HpCDF	0.682	3.62	25.1	0.979	0.0244
1,2,3,4,7,8,9-HpCDF	0.102	0.325	3.57	0.1608	0.00485
OCDF	0.171	1.02	14.1	0.686	0.0446
PCDDs	1.25	2.03	44.9	1.77	0.282
PCDFs	2.72	24.7	67.1	2.64	0.101
PCDDs/PCDFs ratio	0.459	1.29	0.669	0.344	1.77
Total PCDD/Fs $\mu\text{g}/\text{ton}$	3.97	26.8	112	4.41	0.383
PCDDs $\mu\text{g I-TEQ}/\text{ton}$	0.0812	0.295	1.49	0.0416	0.00357
PCDFs $\mu\text{g I-TEQ}/\text{ton}$	0.335	3.21	3.87	0.124	0.00470
PCDDs/PCDFs (TEQ) ratio	0.242	1.43	0.384	0.1653	0.436
Total PCDD/F I-TEQ $\mu\text{g I-TEQ}/\text{ton}$	0.416	3.50	5.36	0.166	0.00827

Table 3. PCDD/F content in fly ash and bottom ash from various sources.

PCDD/Fs	EAF		Secondary ALS		JP-A	JP-B
	Fly ash	Bottom ash	Fly ash	Bottom ash	Bottom ash	Bottom ash
2,3,7,8-TeCDD	1.11	0.255	1.67	0.0300	0.0257	0.00867
1,2,3,7,8-PeCDD	6.22	2.01	3.45	0.0373	0.107	0.0273
1,2,3,4,7,8-HxCDD	6.06	2.62	2.50	0.0230	0.0767	0.0243
1,2,3,6,7,8-HxCDD	14.3	13.6	4.45	0.0350	0.126	0.0577
1,2,3,7,8,9-HxCDD	12.0	9.77	4.19	0.0313	0.0827	0.0233
1,2,3,4,6,7,8-HpCDD	73.0	110	42.0	0.293	0.450	0.361
OCDD	79.8	124	107	1.63	3.18	3.63
2,3,7,8-TeCDF	23.2	4.12	105	0.741	0.429	0.163
1,2,3,7,8-PeCDF	34.5	7.17	33.5	0.498	0.318	0.0997
2,3,4,7,8-PeCDF	69.4	17.0	48.0	0.424	0.386	0.105
1,2,3,4,7,8-HxCDF	59.9	13.1	23.4	0.290	0.239	0.0837
1,2,3,6,7,8-HxCDF	69.7	15.1	20.5	0.329	0.230	0.0847
1,2,3,7,8,9-HxCDF	102	29.5	27.3	0.341	0.0867	0.0160
2,3,4,6,7,8-HxCDF	8.91	2.93	4.39	0.0657	0.255	0.104
1,2,3,4,6,7,8-HpCDF	200	58.9	90.5	1.30	0.574	0.345
1,2,3,4,7,8,9-HpCDF	67.2	21.8	16.9	0.250	0.0770	0.0807
OCDF	211	96.1	105	2.56	1.71	0.763
PCDDs	192	262	166	2.08	4.05	4.14
PCDFs	846	266	474	6.79	4.31	1.84
PCDDs/PCDFs ratio	0.227	0.987	0.349	0.306	0.940	2.24
Total PCDD/Fs ng/kg	1039	528	639	8.87	8.36	5.98
PCDD $\text{ng I-TEQ}/\text{kg}$	8.27	5.08	5.04	0.0622	0.115	0.0401
PCDFs $\text{ng I-TEQ}/\text{kg}$	65.7	16.2	44.8	0.432	0.341	0.108
PCDDs/PCDFs (TEQ) ratio	0.126	0.313	0.112	0.144	0.338	0.372
Total PCDD/F I-TEQ $\text{ng I-TEQ}/\text{kg}$	74.0	21.3	49.9	0.494	0.457	0.148

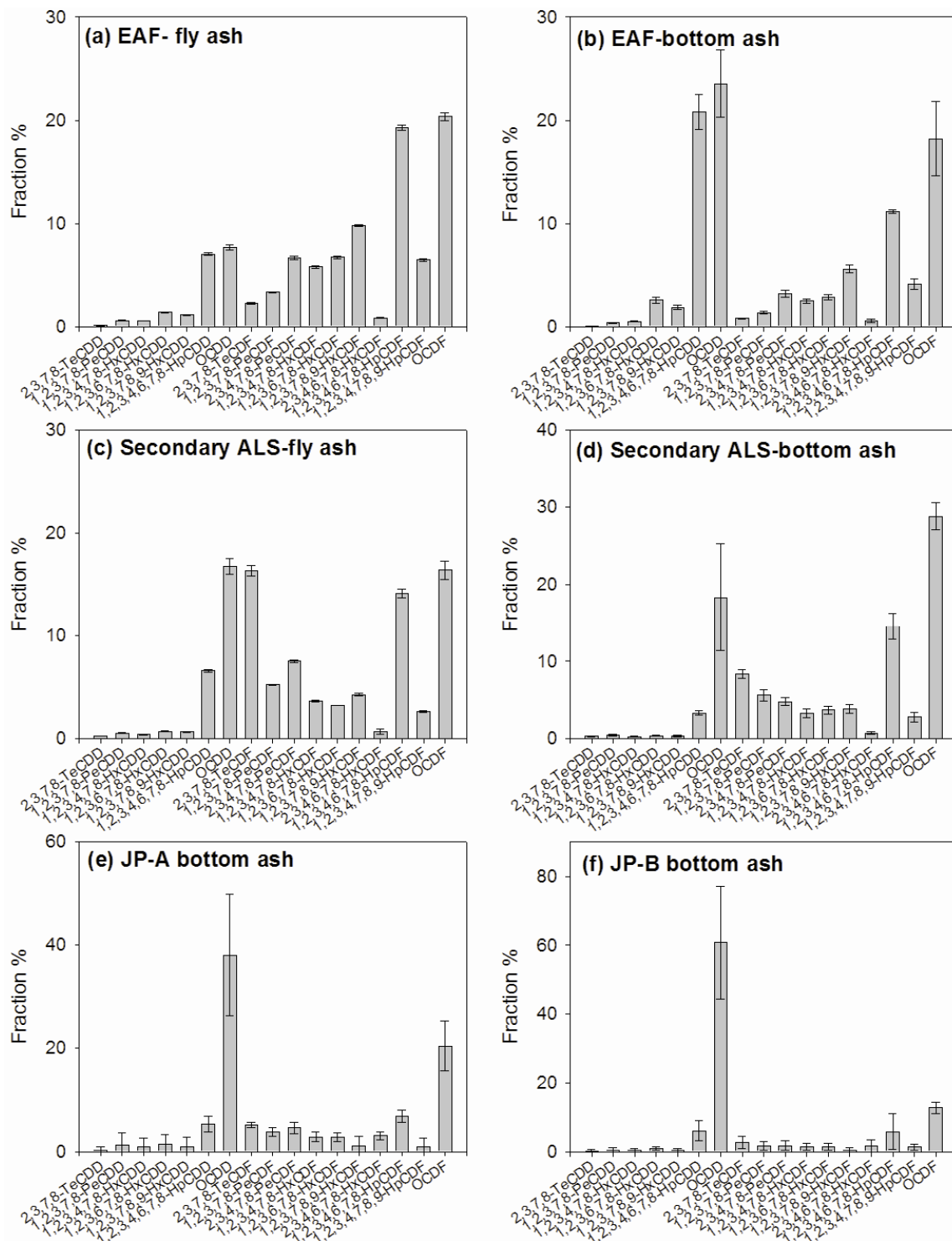


Fig. 2. Congener profiles of seventeen 2,3,7,8-substituted PCDD/Fs in fly ash and bottom ash from various sources.

the corresponding I-TEQ values were 21.7 and 42.1 μg I-TEQ/hr. The results show that, there was a reduction of -44.4 and -94.0% for total PCDD/Fs and total PCDD/Fs TEQ, respectively. This phenomenon was recognized as being due a “memory effect”, in which the wet scrubber made up of plastic material enhanced the PCDD/F adsorption and subsequent desorption (Kim *et al.*, 2001; Löthgren and Bavel, 2005). Therefore, the bag filters must

be replaced regularly in order to reduce PCDD/F emissions.

Indicatory PCDD/Fs

The indicatory PCDD/F that had the three highest ratio values of PCDD/Fs from various sources are listed in Table 4. A comparison with other PCDD/F emission sources was made, and these were selected from previous studies to determine the indicatory PCDD/Fs from the

Table 4. Indicatory PCDD/F of various PCDD/F emission sources.

PCDD/F emission sources	Indicatory PCDD/Fs	Ratio Values
EAF	1,2,3,7,8,9-HxCDF	46.9
	2,3,7,8-TeCDF	12.3
	1,2,3,7,8-PeCDF	11.5
Secondary ALS	2,3,7,8-TeCDF	83.9
	1,2,3,7,8,9-HxCDF	31.4
	1,2,3,7,8-PeCDF	19.1
CM	2,3,4,6,7,8-HxCDF	31.6
	OCDF	9.93
JP-A	1,2,3,4,6,7,8-HpCDD	8.85
	2,3,4,6,7,8-HxCDF	30.0
	OCDF	12.1
JP-B	1,2,3,4,6,7,8-HpCDD	8.57
	OCDD	23.7
	OCDF	9.14
	2,3,4,6,7,8-HxCDF	8.38

present study (US EPA 2001; Lee *et al.*, 2004; Lin *et al.*, 2006; Lin *et al.*, 2007). The indicatory PCDD/Fs of EAF, and secondary ALS have the same congeners: 1,2,3,7,8,9-HxCDF, 2,3,7,8-TeCDF, and 1,2,3,7,8-PeCDF. In addition, CM, JP-A and JP-B have similar indicatory PCDD/Fs: 2,3,4,6,7,8-HxCDF, OCDF, 1,2,3,4,6,7,8-HpCDD, and OCDD. However, the results of the indicatory PCDD/F of emission sources in this study are dependent on the database used.

The Fate of PCDD/Fs

The contribution fractions of total PCDD/F in fly ash, bottom ash and stack flue gas from EAF were approximately 61.1%, 38.9% and 0.00587%, respectively, whereas they were 95.3%, 3.31% and 1.37% from secondary ALS. These results show that fly ash from metallurgical facilities (EAF, secondary ALS) contributed a high total PCDD/F mass that is consistent with the results in a previous study (Li *et al.*, 2007b). However, JP-A and JP-B contribute over 99.9% of total PCDD/F from stack flue gas and only 0.00947% is from bottom ash. Therefore, the high contribution of total PCDD/F in fly ash from the metallurgical facilities (EAF, secondary ALS) should be carefully treated, while JP-A and JP-B need to be equipped with some air pollution control devices (bag filters) to reduce PCDD/F emission.

CONCLUSIONS

The mean total PCDD/F I-TEQ concentration of five different sources ranged from 0.00681 to 0.703 ng I-TEQ/Nm³, while the emission factor of PCDD/Fs ranged from 0.00827 to 3.50 µg I-TEQ/ton (5.36 µg I-TEQ/body for CM). Furthermore, in the indicatory PCDD/Fs analysis, the EAF, and secondary ALS have the same congeners, while CM, JP-A and JP-B have similar indicatory PCDD/Fs. In addition, the highest contribution of total PCDD/Fs is from fly ash (61.1–95.3%) for metallurgical

facilities (EAF, secondary ALS), while 99.9% of the stack flue gas is from JP. Hence, fly ash from the metallurgical facilities should be carefully treated and JP incinerators need to be equipped with some air pollution control devices (such as bag filters) to reduce PCDD/F emission. Moreover, the removal efficiency of PCDD/F by bag filters was –44.4%, and this result was due to the PCDD/F memory effect with these filters. Therefore, bag filters must be replaced regularly in order to reduce PCDD/F emissions.

ACKNOWLEDGMENTS

The authors are sincerely grateful to Mr. Hung-Chi Jiang and Ms. Xin Lu for their help in the sampling and analysis work.

REFERENCES

- Abad, E., Caixach, J. and Rivera, J. (2003). Improvements in Dioxin Abatement Strategies at a Municipal Waste Management Plant in Barcelona. *Chemosphere* 50: 1175–1182.
- Chen, S.J., Lee W.S., Chang-Chien, G.P., Wang, L.C., Lee, W.J., Kao, J.H. and Hu, M.T. (2004). Characterizing Polychlorinated Dibenzop-dioxins and Dibenzofurans in the Surrounding Environment and Workplace of a Secondary Aluminum Smelter. *Atmos. Environ.* 38: 3729–3732.
- EPA Taiwan. (2009). Available at <http://ivy5.epa.gov.tw/epalaw/index.aspx>.
- Giugliano, M., Cernuschi, S., Grosso, M., Miglio, R. and Aloigi, E. (2002). PCDD/F Mass Balance in the Flue Gas Cleaning Units of a MSW Incinerator Plant. *Chemosphere* 46: 1321–1328.
- Hu, M.T., Chen, S.J., Lai, Y.C., Huang, K.L. Chang-Chien, G.P. and Tsai, J.H. (2009). Characteristics of Polychlorinated Dibenzop-dioxins/Dibenzofuran from Joss Paper Burned in Taiwanese Temples. *Aerosol Air Qual. Res.* 9: 367–377.
- Karademir, A., Bakoglu, M., Taspinar, F. and Ayberk, S. (2004). Removal of PCDD/Fs from Flue Gas by a Fixed-bed Activated Carbon Filter in Hazardous Waste Incinerator. *Chemosphere* 38: 1201–1207.
- Kim, S.C., Jeon, S.H., Jung, I.R., Kim, K.H., Kwon, M.H., Kim, J.H., Yi, J.H., Kim, S.J., You, J.C. and Jung, D. H. (2001). Removal Efficiencies of PCDDs/PCDFs by Air Pollution Control Devices in Municipal Solid Waste Incinerators. *Chemosphere* 43: 773–776.
- Lee, W.J., Shih, S.I., Li, H.W., Lin, L.F., Yu, K.M., Lu, K., Wang, L.C., Chang-Chien, G.P., Fang, K. and Lin, M. (2009). Assessment of Polychlorinated Dibenzop-dioxins and Dibenzofurans Contribution from Different Media to Surrounding Duck Farms. *J. Hazard. Mater.* 163: 1185–1193.
- Lee, W.S., Chang-Chien, G.P., Wang, L.C., Lee, W. J., Wu, K.Y. and Tsai, P.J. (2005). Emission of Polychlorinated Dibenzop-dioxins and Dibenzofurans from Stack Gases of Electric Arc Furnaces and Secondary Aluminum

- Smelters. *J. Air Waste Manage. Assoc.* 55: 219–226.
- Lee, W.S., Chang-Chien, G.P., Wang, L.C., Lee, W.J., Tsai, P.J., Wu, K.Y. and Lin, C. (2004). Source Identification of PCDD/Fs for Various Atmospheric Environments in a Highly Industrialized City. *Environ. Sci. Technol.* 38: 4937–4944.
- Li, H.W., Lee, W.J., Huang, K.L. and Chang-Chien, G.P. (2007a). Effect of Raw Materials on Emissions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from the Stack Flue Gases of Secondary Aluminum Smelters. *J. Hazard. Mater.* 147: 776–784.
- Li, H.W., Lee, W.J., Tsai, P.T., Mou, J.L., Chang-Chien, G.P. and Yang, K.T. (2008). A Novel Method to Enhance Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans Removal by Adding Bio-solution in EAF Dust Treatment Plant. *J. Hazard. Mater.* 150: 83–91.
- Li, H.W., Wu, Y.L., Lee, W.J. and Chang-Chien, G.P. (2007b). Fate of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in a Fly Ash Treatment Plant. *J. Air Waste Manage. Assoc.* 57: 1024–1031.
- Lin, L.F., Lee, W.J. and Chang-Chien, G.P. (2006). Emission of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from Various Industrial Sources. *J. Air Waste Manage. Assoc.* 56: 1707–1715.
- Lin, L.F., Lee, W.J., Li, H.W., Wang, M.S. and Chang-Chien, G.P. (2007). Characterization of Inventory of PCDD/F Emissions from Coal-fired Power Plants and other Sources in Taiwan. *Chemosphere* 68: 1642–1649.
- Löthgren, C.J. and Bavel, B.V. (2005). Dioxin Emissions after Installation of a Polishing Wet Scrubber in a Hazardous Waste Incineration Facility. *Chemosphere* 61: 405–412.
- Olie, K., Vermeulen, P.L. and Hutzinger, O. (1997). Chlorodibenzo-*p*-dioxins and Chlorodibenzofurans are Trace Components of Fly Ash and Flue Gas of some Municipal Incinerators in the Netherlands. *Chemosphere* 6: 455–459.
- Rappe, C. (1992). Sources of PCDDs and PCDFs. Introduction. Reactions, Levels, Patterns, Profiles and Trends. *Chemosphere* 25: 41–44.
- Tejima, H., Nakagawa, I., Shioda, T.A. and Maeda, I. (1996). PCDDs/PCDFs Reduction by Good Combustion Technology and Fabric Filter with/without Activated Carbon. *Chemosphere* 32: 169–175.
- Tysklind, M., Fängmark, I., Marklund, S., Lindskog, A., Thaning, L. and Rappe, C. (1993). Atmospheric Transport and Transformation of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans. *Environ. Sci. Technol.* 27: 2190–2197.
- U.S. EPA (1994). Tera-through Octa Chlorinated Dioxins and Furans by Isotope Dilution HRGC/HRMS, Revision B, Method-1613B.
- U.S. EPA (1996). Sampling Method for Polychlorinated Dibenzo-*p*-Dioxins and Polychlorinated Dibenzofurans Emissions from Stationary Sources, Method 23A.
- U.S. EPA (2000). Exposure and Human Health Reassessment of 2,3,7,8-Tetrachlorodibenzo-*p*-Dioxin (TCDD) and Related Compounds National Academy Sciences (NAS) Review Draft, EPA/600/P-00/001Bb.
- U.S. EPA (2001). Database of Sources of Environmental Releases of Dioxin like Compounds in the United States, EPA/600/C-01/012.
- VanJaarsveld, J.A. and Schutter, M.A.A. (1993). Modelling the Long-range Transport and Deposition of Dioxins; First Results for NW Europe. *Chemosphere* 27: 131–139.
- Wang, L.C., Lee, W.J., Lee, W.S., Chang-Chien, G.P. and Tsai, P.J. (2003b). Effect of Chlorine Content in Feeding Wastes of Incineration on the Emission of Polychlorinated Dibenzo-*p*-dioxins/Dibenzofurans. *Sci. Total Environ.* 302: 185–198.
- Wang, L.C., Lee, W.J., Lee, W.S., Chang-Chien, G.P. and Tsai, P.J. (2003c). Characterizing the Emission of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from Crematories and their Impacts to the Surrounding Environment. *Environ. Sci. Technol.* 37: 62–67.
- Wang, L.C., Lee, W.J., Tsai, P. J., Lee, W.S. and Chang-Chien, G.P. (2003a). Emission of Polychlorinated Dibenzo-*p*-dioxins/Dibenzofurans from Stack Flue Gases of Sintering Plants. *Chemosphere* 50: 1123–1129.
- Wang, Y.H., Lin, C., and Chang-Chien, G.P. (2009). Characteristics of PCDD/Fs in a Particles Filtration Device with Activated Carbon Injection. *Aerosol Air Qual. Res.* 9: 317–322.
- Yang, H.H., Lee, W.J., Chen, S.J. and Lai, S.O. (1998). PAH Emission from Various Industrial Stacks. *J. Hazard. Mater.* 60: 159–174.

Received for review, June 29, 2010

Accepted, October 21, 2010