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Monitoring and Identification of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in the Ambient Central Taiwan

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ABSTRACT

Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans (PCDD/Fs) in the ambient air of five sampling sites around central Taiwan were investigated. Principal components analysis (PCA), cluster analysis (CA) and the chemical mass balance (CMB) model were adopted to assess possible PCDD/F sources and their effects on the air quality levels. Field experimental results showed that the mean PCDD/F concentrations in the ambient air were 0.0526, 0.0591, 0.0339 and 0.0727 pg I-TEQ/Nm^3 in winter, spring, summer and autumn, respectively. Analysis of atmospheric isopleths for five sampling sites displayed that high PCDD/F concentrations were mostly close to electric arc furnace plants (EAF). Through PCA and CA, the congener profiles of ambient air sampling sites were close to medical waste incinerators (MWI), secondary aluminum smelters (ALS), EAF, waste open burning (OB) and crematories (CM). By using the CMB model, the dominant sources of PCDD/Fs in ambient air were EAF, medical waste incinerator/municipal solid waste incinerators (MWI/MSWI) and unleaded gasoline fuel vehicle/diesel fuel vehicle (UGFV/DFV), which contributed 22–23%, 17–31% and 9–22%, respectively. The above results revealed that ambient air was affected by surrounding PCDD/F sources (MWI, ALS, EAF, CM and OB). However, due to high stack height and low emission concentration, the impact of coal-fired power plant on the PCDD/F levels of ambient air was insignificant.

Keywords: PCDD/Fs; Ambient air; Principal components analysis; Cluster analysis; Chemical mass balance.

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) were discovered in both flue gas and fly ash of municipal solid waste incinerators in 1977 (Olie *et al*., 1977). PCDD/Fs have received much public concern over the last decade due to adverse health effects (U.S. EPA, 2000). Recently, many countries have become seriously concerned about the PCDD/F emissions from thermal processes (Evert and Baeyens, 2002; Wang *et al*., 2003a; Kao *et al*., 2006). Indeed, combustion was believed to be a major contributor of PCDD/F emissions to the atmosphere (Rappe, 1992; US EPA, 2000). However, the above toxic pollutants could be transported long distances from the sources to the receptors via the atmospheric dispersion and deposition, ultimately affecting the environment and human health (Tysklind *et al*., 1993; VanJaarsveld and Schutter, 1993; Lin *et al*., 2010).

Previous studies have shown that the PCDD/F concentration from the stacks of power plants are between 0.41 and 50 pg I-TEQ/Nm^3 (Fernández-Martínez *et al*., 2004; Lin *et al*., 2007; Hsieh *et al*., 2009; Wang *et al*., 2009a). Although the PCDD/F concentrations from power plants are low, the total amount of PCDD/F emission can not be neglected. This is due to the huge amount of volumetric flow rate in stack flue gas emitted from power plants. Therefore, from the previous studies, the coal-fired power plants contributed approximately 28% of PCDD/F emissions in Taiwan (Lin *et al*., 2007). Consequently, information on PCDD/F characteristics in ambient air near a coal-fired power plant is of great importance for the establishment of control strategies for the above air toxins.

The atmosphere is a very complex system; therefore, the management of air quality is a challenging issue. Mathematical dispersion models have been used widely in air pollution control to predict the effect of point, line or area source emissions (Hopke, 1985, 1991). But they have displayed certain limitations, especially for predicting short-term impacts. Therefore, the receptor model has been

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developed to assess the contribution of various emission sources on the receptors based on observations at sampling sites (Gordon, 1980). The most widely used receptor model is the chemical mass balance (CMB) which is composed of patterns of emissions from various classes of sources that are disparate enough that one can identify their contributions by measuring concentrations of many species in samples collected at a receptor site (Gordon, 1988). Previous study showed that gasoline vehicle exhaust, liquid gasoline, and gasoline evaporation contributed up to 50% of ambient volatile organic compounds by CMB analysis (Watson *et al*., 2001). In addition, CMB estimated a 6.3–24.6% contribution from open burning to total atmospheric polycyclic aromatic hydrocarbons (PAHs) at two sampling sites (Lai *et al*., 2009). Furthermore, over 70% of the PCDD/F TEQ value was attributable to residential emissions by the CMB model (Paradiž *et al*., 2008).

The main objective of the present study was to quantity the contribution to the ambient PCDD/F from various sources. Five sampling sites were chosen nearby the coal-fired power plant to collect air samples in four seasons. Principal components analysis (PCA), cluster analysis (CA) and the chemical mass balance (CMB) model were adopted to assess the contributions of possible PCDD/F sources and, particularly, the coal- fired power plant on the ambient air.

METHODS

PCDD/F Sampling

PCDD/F concentrations in ambient air were collected by a standard semi-volatile sampling train (General Metal works PS-1) according to the revised US EPA Reference Method TO9A (U.S. EPA, 1999). The PS-1 sampler was equipped with a quartz fiber filter (particle-phase PCDD/Fs), followed by a glass cartridge with polyurethane foam (PUF, gas-phase PCDD/Fs). Prior to sampling, the PUF was spiked with PCDD/F surrogate standards pre-labeled with isotopes. A total of 20 samples were collected from sampling sites A, B, C, D, and E over four seasons (Fig. 1). Sampling meteorological information is shown in Table 1. Each sample was set at a flow rate of 0.225 m^3/min and was sampled for three consecutive days.

The sampling sites were designed according to the understanding of local stationary PCDD/F emission sources and the results of the Industrial Source Short Term modeling (ISCST) to assess the possible sites with maximum PCDD/F concentrations in this region. Similar methodology has been widely used to simulate ambient air concentrations at specified receptor points for various sources (Yang *et al*., 1998; Mi *et al*., 2001; Lee *et al*., 2003a).

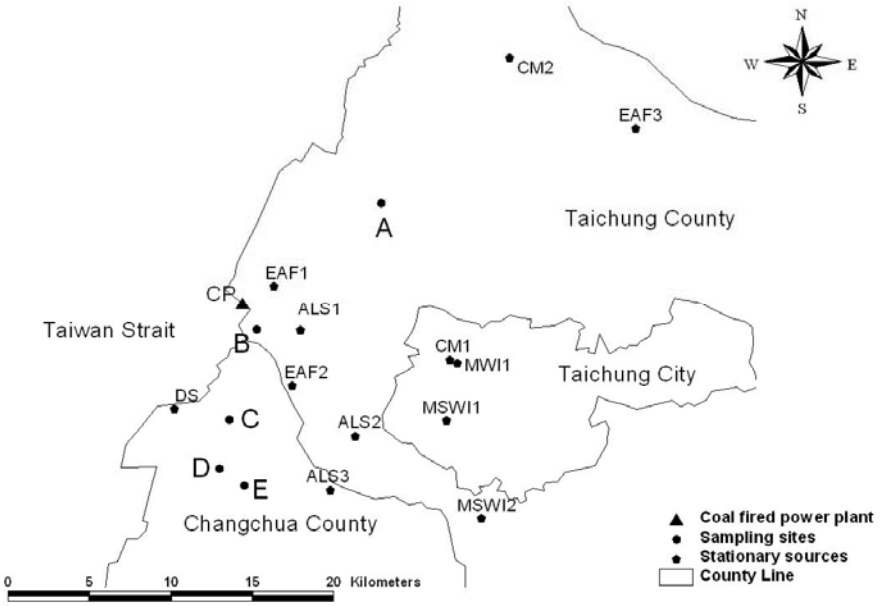

Fig. 1. Sampling sites located in central Taiwan.

Table 1. Meteorological information during PCDD/Fs sampling periods.

Season	Winter	Spring	Summer	Autumn
Sampling period	11/25/2008–11/27/2008	02/24/2009–02/26/2009	07/07/2009–07/09/2009	08/25/2009–08/27/2009
Sampling number	5	5	5	5
Average temperature	21.5°C	20.0°C	29.3°C	29.0°C
Direction of prevailing wind	North-northeast	Northwest and West-northwest	West-northwest and South- southwest	South-southwest and South

Analysis of PCDD/Fs

The analyses for PCDD/F ambient air samples were performed according to the US EPA Reference Method TO9A (U.S. EPA, 1999). Essentially, samples were extracted with toluene for 24 hr and this was then followed by a series of sample cleanup procedures. The extract was transferred to a vial, and finally further concentrated by a N_2 gas stream.

A high-resolution gas chromatographs/high-resolution mass spectrometers (HRGC/HRMS) was 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 10,000. The electron energy and source temperature were specified at 35 eV and 250°C, respectively. The oven temperature program was set according to the following: 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.

Principal Components Analysis

Principal components analysis (PCA) has been widely used as an assessment method to identify possible pollution sources. The data is divided into cases and variables. In this study, the cases that were used for the PCDD/F emission sources were classified according to the position of their corresponding coordinates with respect to the factor axis. In addition, the variables that were used the fraction of seventeen PCDD/F congeners are used to characterize the case. In the score plot, the cases with similar patterns will be located close to each other, while those which have divergent patterns will be distant (Jambu, 1991; Wang *et al*., 2008).

Chemical Mass Balance

The Chemical Mass Balance (CMB) air quality model has been applied to air resources management. The CMB model consists of a least-squares solution to a set of mass-balance equations that express each receptor chemical concentration as a linear sum of products of sources profile abundances and receptor concentration (Watson *et al*., 2001). In addition, CMB is the receptor model used for meteorological condition, chemical transformation mechanisms of pollutants and emissions rate of pollutants to assess the contribution of each source to receptor concentrations (Gordon, 1980; Hopke, 1985; Gordon, 1988; Hopke, 1991; Watson *et al*., 2001; U.S. EPA, 2004).

RESULTS AND DISCUSSIONS

PCDD/F Concentration in the Ambient Air

The mean total-PCDD/F concentrations in the ambient air of a coal-fired power plant were 0.873, 0.843, 0.464 and 0.903 ng/Nm^3 in winter, spring, summer and autumn, respectively, while the corresponding total PCDD/F-I-TEQ concentrations were 0.0526, 0.0591, 0.0339 and 0.0727 pg I-TEQ/Nm^3 (Table 2). The above PCDD/F concentration levels were similar to previous studies in southern Taiwan near two municipal solid waste incinerators and one coal-fired power plant (0.0319–0.0847 pg I-TEQ/Nm^3), but slightly higher than those in northern Taiwan near a heavy oil-fueled power plant (0.004–0.022 pg I-TEQ/Nm^3) (Hsieh *et al*., 2009; Wang *et al*., 2009a). Previous studies have reported that the PCDD/F concentration in winter was four to eight times higher than in summer in rural Germany due to the demand for domestic heating, while atmospheric PCDD/F concentrations also varied with the seasons, because of photolysis and its effects on chemical reactions (Hippelein *et al*., 1996; Lohmann and Jones, 1998). In addition, the present results were between 5.5% and 12.1% in magnitude compared with Japan regulated PCDD/F concentration in ambient air (0.6 pg I-TEQ/Nm^3) (MOE Japan, 2008).

By utilizing Arcview GIS software (Environmental Systems Research Institute), the atmospheric isopleths of five sampling sites are displayed in Fig. 2. For example, the prevailing winds at the sampling time in autumn were from south southwest and south, therefore, the northeast side of the EAF2 plant has higher PCDD/F isopleths concentrations (Fig.2).

Fig. 3. shows the congener profiles of the seventeen 2,3,7,8-substituted PCDD/Fs and each selected congener was normalized by the sum of seventeen PCDD/Fs. The dominant congeners are OCDD, 1,2,3,4,6,7,8-HpCDF, OCDF, and 1,2,3,4,6,7,8-HpCDD, which is similar to the congener profiles of the stack flue gas of the fly ash treatment plant (Waelz process) in previous study (Li *et al*., 2007). Several studies reported that the Waelz process which is thermal treatment of residues containing diverse organic and inorganic pollutants is a high potential for PCDD/F emission (Kim *et al*., 2003; Mager *et al*., 2003; Chi *et al*., 2008; Yu *et al*., 2009, 2010). The above results revealed that the PCDD/Fs in the ambient air of five sampling sites were affected by the EAF plants.

Principal Components Analysis and Cluster Analysis

To clarify the influence of PCDD/F in the ambient air by the coal-fired power plant, the congener profiles of PCDD/Fs in ambient air together with those of other possible PCDD/F sources (stationary sources, mobile sources and open burning) were analyzed by PCA (Lee *et al.*, 2004a). The score plots from PCA for twenty sampling sites and ten possible PCDD/F sources are shown in Fig. 4. The PCA of factor 1 explains 36.6% of total variance, while factor 2 accounts for 25.1%; together, they account for 61.7% of total variance. The results revealed that the congener profiles of ambient air sampling sites were close to medical waste incinerators (MWI), secondary aluminum smelters (ALS), and electric arc furnaces (EAF). Previous studies reported that ALS and EAF have become the major PCDD/F emission source in Taiwan (Chen *et al*., 2004; Lee *et al*., 2005).

Table 2. Mean PCDD/F concentration in the ambient air (N = 5).

	Winter	RSD%	Spring	RSD%	Summer	RSD%	Autumn	RSD%
2,3,7,8-TeCDD	0.00231	19.0	0.00378	34.8	0.00209	47.3	0.00500	64.7
1,2,3,7,8-PeCDD	0.00631	26.4	0.00986	31.1	0.00542	64.4	0.0134	57.4
1,2,3,4,7,8-HxCDD	0.00519	21.1	0.00743	47.9	0.00383	60.3	0.00909	55.6
1,2,3,6,7,8-HxCDD	0.0103	19.5	0.0148	52.7	0.00648	42.7	0.0160	46.0
1,2,3,7,8,9-HxCDD	0.00750	15.4	0.0114	45.7	0.00539	48.7	0.0125	50.0
1,2,3,4,6,7,8-HpCDD	0.0618	9.87	0.0848	45.1	0.0355	41.5	0.0846	38.4
OCDD	0.195	24.7	0.199	22.3	0.108	44.7	0.168	24.8
2,3,7,8-TeCDF	0.0235	25.7	0.0290	27.1	0.0185	59.0	0.0428	39.0
1,2,3,7,8-PeCDF	0.0306	21.7	0.0322	25.5	0.0216	73.1	0.0490	39.8
2,3,4,7,8-PeCDF	0.0446	14.9	0.0515	32.6	0.0300	64.2	0.0602	43.1
1,2,3,4,7,8-HxCDF	0.0576	13.1	0.0442	32.7	0.0287	69.6	0.0599	36.5
1,2,3,6,7,8-HxCDF	0.0477	15.6	0.0418	38.4	0.0255	69.5	0.0528	42.5
1,2,3,7,8,9-HxCDF	0.00362	24.6	0.00272	33.3	0.00194	32.5	0.00498	28.9
2,3,4,6,7,8-HxCDF	0.0490	16.6	0.0513	59.6	0.0259	52.8	0.0583	43.2
1,2,3,4,6,7,8-HpCDF	0.163	16.4	0.136	43.7	0.0762	48.7	0.147	33.6
1,2,3,4,7,8,9-HpCDF	0.0228	13.7	0.0227	57.6	0.00975	49.1	0.0235	34.2
OCDF	0.142	30.1	0.101	54.0	0.0592	50.2	0.0953	24.0
PCDDs	0.289	15.6	0.331	28.9	0.167	39.2	0.309	31.3
PCDFs	0.585	17.9	0.512	41.0	0.297	49.9	0.594	35.1
PCDDs/PCDFs ratio	0.493	–	0.646	–	0.562	–	0.520	–
Total PCDD/Fs	0.873	15.5	0.843	35.3	0.464	43.7	0.903	33.0
PCDDs pg I-TEQ/Nm^3	0.00858	19.9	0.0131	31.0	0.00684	52.9	0.0165	56.2
PCDFs pg I-TEQ/Nm^3	0.04399	15.1	0.0460	34.6	0.0270	62.4	0.0562	40.9
PCDDs/PCDFs(TEQ) ratio	0.195	–	0.286	–	0.253	–	0.294	–
Total PCDD/F I-TEQ pg I-TEQ/Nm^3	0.0526	15.5	0.0591	33.0	0.0339	60.4	0.0727	44.1

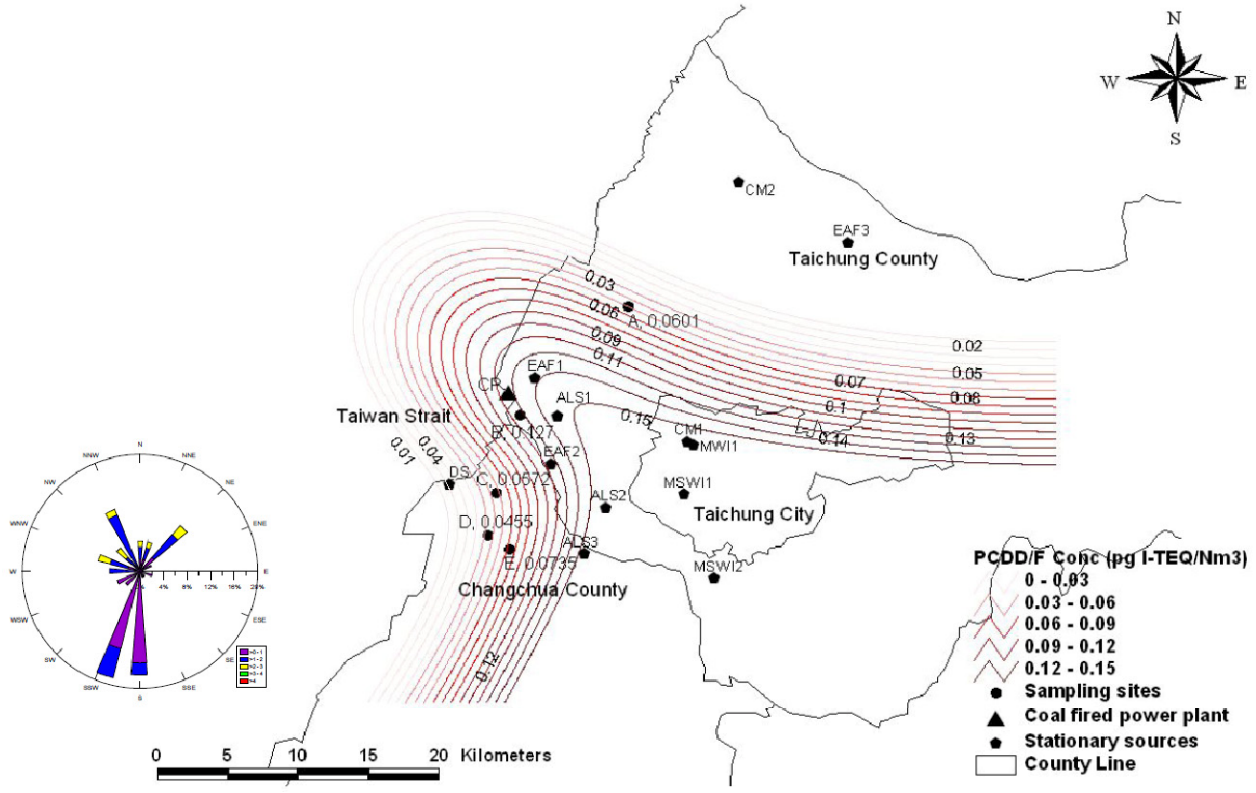

Fig. 2. The isopleths of PCDD/F concentration in the ambient air of the coal-fired power plant and regional wind rose.

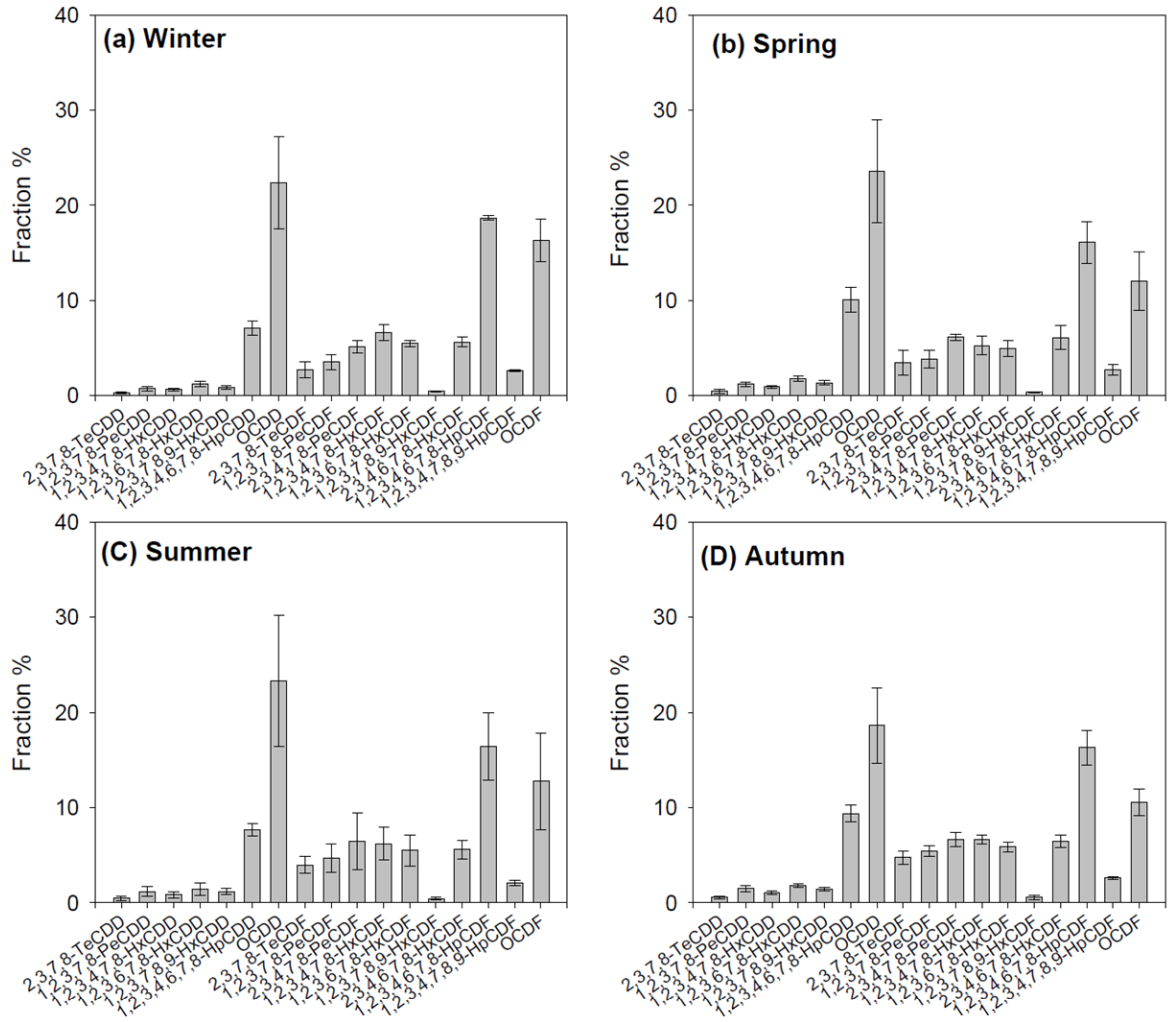

Fig. 3. Congener profiles of seventeen 2,3,7,8-subsituted PCDD/Fs in ambient air in four seasons.

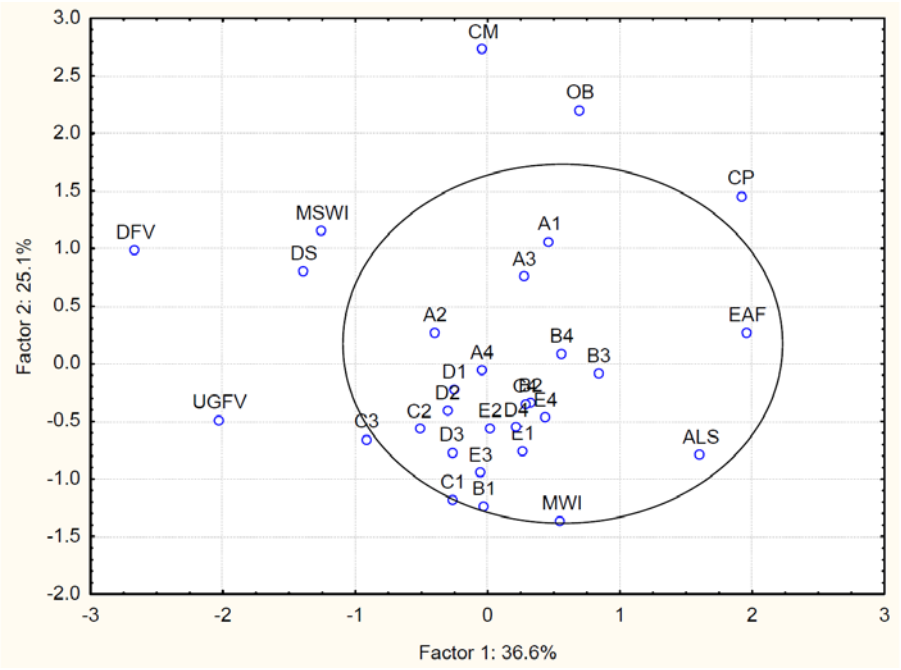

Fig. 4. Principal components plot of the ambient atmospheric samples and other emission sources.

The results of cluster analysis were similar to PCA, and are displayed in Fig. 5. Data points of ambient air sites cluster first with those of MWI, and then with open burning (OB) and crematories (CM). Previous studies have shown that PCDD/F emissions from crematories, medical waste incinerators and joss paper (semi-open systems) did significantly affect the surrounding environment (Lee *et al*., 2003b; Wang *et al*., 2003c; Hu *et al*., 2009; Wang *et al*., 2009b). The above results indicate that the ambient air samples were influenced significantly by stationary emission sources (MWI, ALS, EAF and CM) and open burning (OB). In addition, ambient air samples were mainly influenced by PCDD/F emissions from metallurgical facilities and MWIs (Lee *et al*., 2004b; Wang *et al*., 2008). However, the effect of the coal fired power plant (CP) on ambient air quality was insignificant.

Chemical Mass Balance

The source profiles used for the CMB model are listed in Table 3. The mean PCDD/F contribution fractions from surrounding possible PCDD/F sources on the ambient air are listed in Table 4. In winter and spring, the major PCDD/F contribution of 67–70% was from secondary aluminium smelters (ALS); while the next at 11–17% was from the crematory sources (CM). However, during the summer and autumn period, the dominant PCDD/F contribution was from EAFs at 22–23%, with second most from MWI/MSWI at 17–31%, finally, 9–22% from UGFV/DFV. In addition, the fly ash treatment plant and open burning also provided about 6–16% and 3–4% of contributions during summer and autumn, respectively. The above results were similar with the PCA and cluster analysis (MWI, ALS, EAF, CM and OB). In previous PCDD/F inventory, the sources of PCDD/Fs are EAF, OB, ALS, CM and MWI which contributed for 23%, 8.2%, 1.5%, 0.7% and 0.4%, respectively (Lin *et al*., 2007). Consequently, the ambient air was affected by surrounding stationary PCDD/F sources. However, the coal-fired power plant (CP) again did not have significant PCDD/F contributions on the ambient atmosphere.

CONCLUSIONS

1. Mean total PCDD/F-I-TEQ concentrations in the ambient air of the coal-fired plant were 0.0526, 0.0591, 0.0339 and 0.0727 pg-I-TEQ/Nm^3 in winter, spring, summer and autumn, respectively. The present corresponding total PCDD/F-I-TEQ concentrations were between 5.5% and 12.1% in magnitude compared with Japan regulated PCDD/F concentrations in ambient air (0.6 pg I-TEQ/Nm^3).
2. Atmospheric isopleths of five sampling sites showed that the EAF2 plant at the northeast side had higher PCDD/F concentrations.
3. The dominant congeners in ambient air are OCDD, 1,2,3,4,6,7,8-HpCDF, OCDF, and 1,2,3,4,6,7,8-HpCDD.
4. The PCA results revealed that the congener profiles of ambient air sampling sites were close to MWI, ALS and EAF. For the cluster result, data points of ambient air sites cluster first with those of MWI and then with open burning (OB) and crematory (CM).
5. By using the CMB model, the contributions of PCDD/Fs on the ambient air were 22–23% from EAF, 17–31% from MWI/MSWI, and 9–22% from UGFV/DFV in summer and autumn. The above results revealed that the ambient air was affected by the surrounding PCDD/F sources (MWI, ALS, EAF, CM and OB); however, the coal-fired power plant (CP) did not have a significant impact on the atmospheric PCDD/Fs.

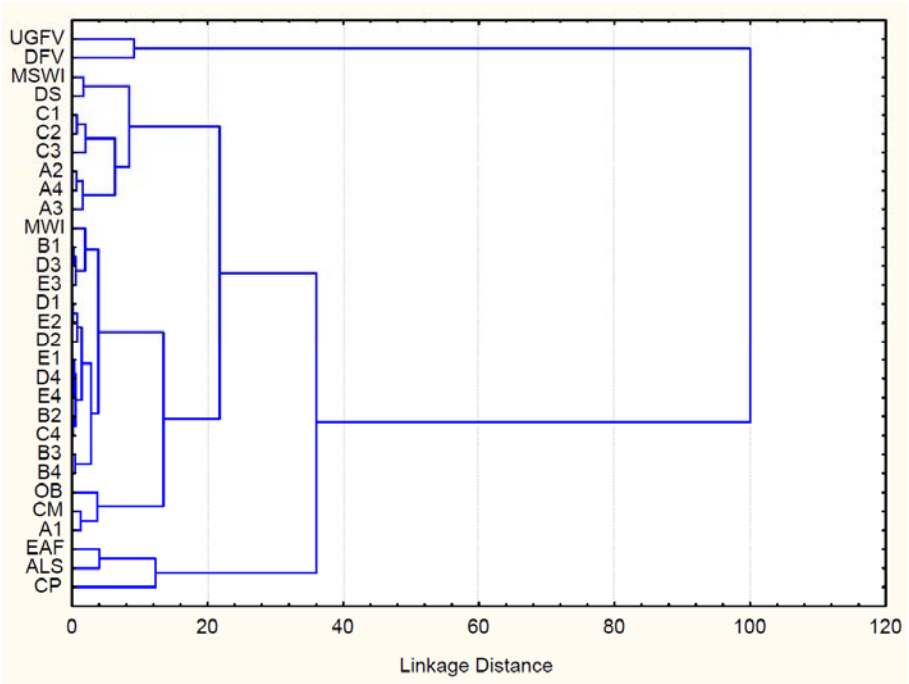

Fig. 5. Cluster analysis of the ambient atmospheric samples and other emission sources.

Table 3. Source profile used for CMB model.

	UGFV	DFV	MSWI	MWI	EAF	ALS	CP	OB	CM	DS
2,3,7,8-TeCDD	0.00909	0.00501	0.00156	0.000403	0.0142	0.00619	0.0776	0.0122	0.0303	0.00108
1,2,3,7,8-PeCDD	0.00406	0.00424	0.00610	0.00635	0.0243	0.00998	0.0439	0.0309	0.0537	0.00582
1,2,3,4,7,8-HxCDD	0.00425	0.00596	0.0123	0.00743	0.00787	0.00597	0.00390	0.0253	0.0213	0.00720
1,2,3,6,7,8-HxCDD	0.00870	0.0117	0.0412	0.0130	0.0155	0.00901	0.0303	0.0360	0.0236	0.0382
1,2,3,7,8,9-HxCDD	0.00541	0.0215	0.0301	0.0127	0.0106	0.00706	0.00714	0.0230	0.0293	0.0238
1,2,3,4,6,7,8-HpCDD	0.0653	0.140	0.286	0.0694	0.0239	0.0208	0.0374	0.138	0.0507	0.229
OCDD	0.516	0.648	0.410	0.120	0.0277	0.0341	0.0547	0.105	0.0284	0.281
2,3,7,8-TeCDF	0.0303	0.0126	0.0146	0.0168	0.247	0.222	0.0615	0.110	0.173	0.00432
1,2,3,7,8-PeCDF	0.0145	0.00504	0.00489	0.0418	0.148	0.0751	0.0160	0.0837	0.138	0.0113
2,3,4,7,8-PeCDF	0.0106	0.0112	0.0136	0.0557	0.187	0.129	0.253	0.0876	0.143	0.0186
1,2,3,4,7,8-HxCDF	0.0120	0.0189	0.0272	0.0867	0.0664	0.0738	0.103	0.102	0.0702	0.0450
1,2,3,6,7,8-HxCDF	0.0128	0.00859	0.0231	0.0770	0.0650	0.0677	0.0639	0.0574	0.0791	0.0331
1,2,3,7,8,9-HxCDF	0.00348	0.00500	0.00431	0.0304	0.0335	0.00594	0.0381	0.00691	0.00480	0.00333
2,3,4,6,7,8-HxCDF	0.0149	0.0125	0.0382	0.0854	0.0602	0.0698	0.0547	0.0424	0.0536	0.0361
1,2,3,4,6,7,8-HpCDF	0.133	0.0390	0.0490	0.229	0.0450	0.155	0.108	0.106	0.0811	0.161
1,2,3,4,7,8,9-HpCDF	0.00425	0.00590	0.0156	0.0428	0.0107	0.0291	0.0130	0.0107	0.0103	0.0255
OCDF	0.151	0.0446	0.0213	0.105	0.0128	0.0799	0.0333	0.0242	0.00935	0.0754

Notes. Unleaded gasoline fuel vehicle (UGFV); diesel fuel vehicle (DFV); municipal solid waste incinerator (MSWI); medical waste incinerator (MWI); electric arc furnace (EAF); secondary aluminum smelter (ALS); coal fire power plant (CP), Crematory (CM); open burning (OB) and fly ash treatment plant (DS); (Wang *et al*., 2003b; Lee *et al*., 2004a; Li *et al*., 2007)

Table 4. Mean PCDD/F contribution for ambient air sampling site from different PCDD/F sources by CMB.

Sources	Winter	Spring	Summer	Autumn
UGFV/DFV	9%	12%	22%	9%
MSWI/MWI	5%	–	17%	31%
ALS	70%	67%	17%	12%
EAF	5%	3%	22%	23%
DS	–	1%	6%	16%
CM	11%	17%	12%	6%
CP	–	–	–	–
OB	–	–	4%	3%
Total	100%	100%	100%	100%

Notes. Chemical mass balance (CMB); unleaded gasoline fuel vehicle (UGFV); diesel fuel vehicle (DFV); municipal solid waste incinerator (MSWI); medical waste incinerator (MWI); secondary aluminum smelter (ALS); electric arc furnace (EAF); fly ash treatment plant (DS); Crematory (CM); coal boil stream (CP), and open burning (OB).

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