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Comparisons of levels of polychlorinated dibenzo-*p*-dioxins/dibenzofurans in the surrounding environment and workplace of two municipal solid waste incinerators

Shun-I Shih [a,*], Ya-Fen Wang [b], Juu-En Chang [c,d], Jiun-Shiang Jang [c,d], Fan-Leng Kuo [c,d], Lin-Chi Wang [e], Guo-Ping Chang-Chien [e]

[a] *Department of Environmental Engineering, Kun Shan University of Technology, Yung-Kang, Tainan County 710, Taiwan*
[b] *Department of Bioenvironmental Engineering, Chung Yuan Christian University, Chung-Li 320, Taiwan*
[c] *Department of Environmental Engineering, National Cheng Kung University, Tainan 70101, Taiwan*
[d] *Sustainable Environment Research Centre, National Cheng Kung University, Tainan 70101, Taiwan*
[e] *Department of Chemical and Materials Engineering, Cheng Shiu University, 833 Kaohsiung, Taiwan*

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Abstract

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the surrounding environment (outdoor) and workplace air of two municipal solid waste incinerators (MSWIs, T and M) were characterized and compared. T and M represented two typical municipal solid waste incinerators in the north of Taiwan, which have different processes for controlling the PCDD/F emissions. The results of this study are summarized as follows. (1) The total PCDD/F and the total PCDD/F WHO-TEQ concentrations in the workplace air were 5–13 and 5–15 times higher than those in the outdoor air, respectively. Obviously, it is worthwhile to explore more on health risk assessment for exposure of PCDD/Fs emitted from MSWIs, particularly in the workplace air. (2) Mean total PCDD/F I-TEQ concentrations in the outdoor air ranged between 0.0216 and 0.155 pg I-TEQ/Nm^3 and averaged 0.0783 pg I-TEQ/Nm^3 (0.0828 pg WHO-TEQ/Nm^3) during two seasons for two MSWIs, which were 6.5-fold higher than that of a remote site (0.0119 pg I-TEQ/Nm^3 or 0.0132 pg WHO-TEQ/Nm^3) in Taiwan. However, the above outdoor air concentration levels in the MSWIs were still much lower than the air quality limitation of PCDD/Fs (0.6 pg I-TEQ/Nm^3) in Japan [http://www.env.go.jp/en/topic/pops/Appendix/00report/report.pdf]. (3) PCDFs were the primary toxicity distributors for PCDD/Fs in the outdoor air, since the ratios of PCDDs/PCDFs (I-TEQ) at all sampling sites ranged from 0.180 to 0.492 and were less than unity. (4) The OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF were the four dominant species in both workplace and outdoor air near MSWIs. (5) By spraying water on and wetting both the fly and bottom ashes, the mean total PCDD/F I-TEQ concentration in the workplace air was reduced 86.9% in the T MSWI. The above results indicate an appropriate improving action did inhibit the fugitive emission of PCDD/Fs and reduce the health risk of workers during work handling ashes in MSWIs.

Keywords: Municipal solid waste incinerator; PCDD/Fs; Surrounding environment; Workplace

1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) have been first found in the flue gases and in the fly ash of municipal solid waste incinerators (MSWIs) [2]. Thereafter, due to their acute toxicity and associated adverse health effects, they have been extensively studied in many countries [3–5]. In Northeast Spain, Abad et al. [3] reported that the levels of PCDD/Fs in urban and suburban sites vary from 10 to 357 fg I-TEQ/Nm^3, the lowest concentrations were found in rural areas, ranging from 5 to 125 fg I-TEQ/Nm^3. In Korea, Oh et al. [4] evaluated the PCDD/Fs from various incinerators and concluded that the PCDD/Fs emission of MSWIs exhibited a large variation (0.07–36.5 ng I-TEQ/Nm^3). The PCDD/Fs homologue patterns from these various incinerators were similar and the fraction of PCDFs was higher than that of PCDDs. In the USA, PCDD/Fs in the ambient air had been statistically investigated by Raun et al. [5]. Their results showed

* Corresponding author. Tel.: +886 6 205 0524; fax: +886 6 205 0540.
E-mail address: ssi10@mail.ksu.edu.tw (S.-I. Shih).

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a total annual mean dioxin and furan congener concentration of 1047 fg/m^3 and an annual mean total I-TEQ concentration of 15 fg I-TEQ/m^3 (16 fg WHO-TEQ/m^3).

In the past decades, based on the research completed in several industrialized countries, MSWIs had been identified as the largest contributors to the environmental levels of PCDD/Fs [6]. But, recently, the emission inventory of PCDD/PCDFs in Taiwan indicated that municipal waste incineration was not the highest contributor of PCDD/Fs released into the atmosphere [7]. Also, a long-term study by Schuhmacher and Domingo [8] indicated that the environmental level of PCDD/Fs showed that the MSWI was not the main cause of the atmospheric pollution by these compounds. In turn, human health risks for the population living in the vicinity of the facility after the introduction of a modern technology were negligible in comparison with the dietary PCDD/F exposure.

Regardless of whether municipal waste incineration was the main contributor for PCDD/F emissions or not, there is a great deal of research related to PCDD/F emissions of MSWIs, particularly in the ambient air. Nevertheless, among them still little research compared PCDD/Fs between the surrounding environment and workplace of the MSWI. Hu et al. [9] monitored PCDD/F levels in indoor environments and the blood of workers of three municipal waste incinerators in Taiwan. They concluded that blood concentrations of several PCDD/F congeners were significantly different among three incineration plants. The differences were not explained by the discrepancy in job contents, duration of employment, and time activity in these plants. Chen et al. [10] characterized PCDD/Fs in the surrounding environment and workplace of a secondary aluminum smelter. They concluded that the PCDD/F concentration of air in the workplace that workers breathe could be very high and the PCDD/Fs exposure of furnace maintenance workers may be serious. But, they did not point out how high the PCDD/F exposure of workers in the workplace was after all, especially the difference of PCDD/F concentrations in the ambient air between the workplace and the surrounding environment.

A case study of PCDD/Fs monitoring in and around an industrial waste incinerator in Korea concluded that the difference between the levels of PCDD/Fs in the blood of office and plant workers demonstrates that human exposure to PCDD/Fs occurs as a result of the operation of the incinerator [11]. The health risk associated with the incineration of municipal wastes needs to be evaluated in order to protect public health, especially in the workplace. This is due to the fact that the PCDD/F concentration in the workplace air is generally more than five times higher in magnitude than that in the ambient air of the MSWI surrounding environment. This study primarily attempts to set up information on the health risks of PCDD/Fs in the workplace and outdoor air of municipal solid waste incineration plants. Moreover, this investigation also discusses the influence of MSWI emissions on outdoor air and compares the levels and congener profiles of PCDD/Fs in the workplace and outdoor air.

2. Experimental

Table 1 lists the basic information of T and M MSWIs, and Figs. 1 and 2 illustrate the sampling sites of the surrounding environment (outdoor) and workplace in T and M MSWIs, respectively. In T MSWI, TA, TB, TC and TD are the sampling sites of the surrounding environment. They are located in the east, north, west and south sites of T MSWI with distances around 500 m away from the stack, respectively; TE, TF, TG and TH are the sampling sites of the workplace, TE is bottom ash bunker, TF is ash wetting unit, TG is ash conveyer and TH is common fly ash conveyer. In M MSWI, MA, MB, MC and MD are the sampling sites of the surrounding environment. They are located in the east, north, west and south sites of M MSWI with distances around 500 m away from the stack, respectively; ME, MF, MG and MH are the sampling sites of the workplace, ME is fly ash dumping platform, MF is fly ash conveyer, MG is bottom ash conveyer and MH is bottom ash bunker. The total numbers of the samples being analyzed was 30. It included four in summer and four in winter for each MSWI. So, the sub-total numbers of samples in the ambient air of MSWIs were 16. Then, 14 (6 + 8) samples belonging to Kenting and Pingtung were also analyzed.

Each ambient air sample was collected using a PS-1 sampler (Graseby Andersen, GA) according to the revised EPA Reference Method T09A. The sampling flow rate was specified at ~0.225 m^3/min. Each sample was collected continuously

Table 1
Basic information of T and M MSWIs

	T MSWI	M MSWI
Level of PCDD/Fs in flue gas (pg/Nm^3)	Summer: 1383 Winter: 513	Summer: 8136 Winter: 558
Type of incinerator	Municipal solid waste incinerator	Municipal solid waste incinerator
Components of combustion gas	CO, CO_2, O_2, HCl, NO, NO_2, SO_2, SO_3, H_2O, NH_3	CO, CO_2, O_2, HCl, NO, NO_2, SO_2, SO_3, H_2O, NH_3
Combustion temperature	850–1050 °C	850–1050 °C
Main component of burning material	Paper, kitchen wastes, plastics, wood, leather and rubber, fiber and cloth	Paper, kitchen wastes, plastics, Wood, leather and rubber, fiber and cloth
Location	North of Taiwan	North of Taiwan
Capacity	1800 t/day	1500 t/day
High of the stack	150 m	150 m
Furnace	Von-Roll Stoker Type (four sets)	Horizontal HN type (four sets)
Capacity of furnace	18.75 t/h/set	15.625 t/h/set

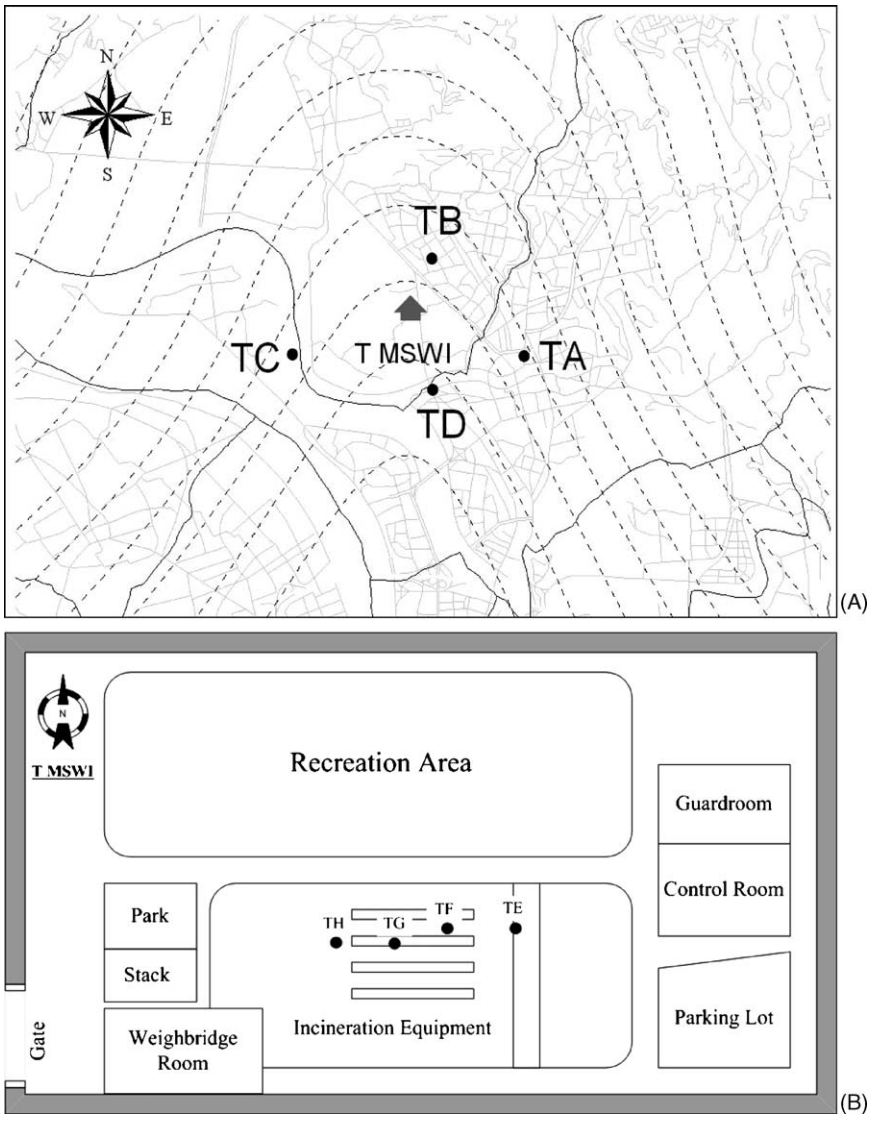

Fig. 1. Air sampling sites of the surrounding environment (outdoor) (A) and workplace (B) in T MSWI. *Note*: TA, TB, TC and TD are the sampling sites of the surrounding environment; TE is bottom ash bunker, TF is ash wetting unit, TG is ash conveyer and TH is common fly ash conveyer.

on 3 consecutive days. The PS-1 sampler was equipped with a quartz-fibre filter for sampling particle-phase PCDD/Fs and followed by a glass cartridge for sampling gas-phase PCDD/Fs, respectively. Prior to sampling, XAD-2 resin was spiked with PCDD/Fs surrogate standards. To ensure the collected samples were contamination-free, one trip blank and one field blank were also taken when the field sampling was conducted [12].

Analyses of ambient air samples followed the US EPA Reference Method T09A. All chemical analyses were performed in the Super Micro Mass Research and Technology Centre of Cheng Shiu Institute of Technology. This centre is the first lab certified by the Taiwan EPA to analyze PCDD/Fs in Taiwan and passes the international inter-calibration on PCDD/Fs in fly ash, sediment, mother's milk, human blood and cod liver. The sample analysis was performed according to the standard procedures. Each collected sample was spiked with a known amount of the internal standard. After being extracted for 24 h, the extract was concentrated, treated with concentrated sulfuric acid, and then followed by a series of sample cleanup and fractionation procedures. The eluate was concentrated to ~1 ml, then transferred to a vial, and then further concentrated to near dryness by using a nitrogen stream. Prior to PCDD/Fs analysis, the standard solution was added to the sample to ensure recovery during the analysis process.

A high-resolution gas chromatograph (HRGC), coupled with a high-resolution mass spectrometer (HRMS), were used for PCDD/Fs measurements. The HRGC is a Hewlett Packard 6970 Series gas chromatograph, equipped with a DB-5 (J&W Scientific, CA, USA) fused silica capillary column (60 m, 0.25 mm i.d., 0.25 μm film thickness), and splitless injection. An initial oven temperature was 150 °C. The temperature was programmed as follows: 150 °C, held for 1 min, increased by 30 °C/min to 220 °C, held for 12 min, increased at 1.5 °C/min to 240 °C, held for 20 min. Helium was used as the carrier gas. The HRMS is a Micromass Autospec Ultima (UK) mass spectrometer with a positive electron impact (EI+) source. The analyzer

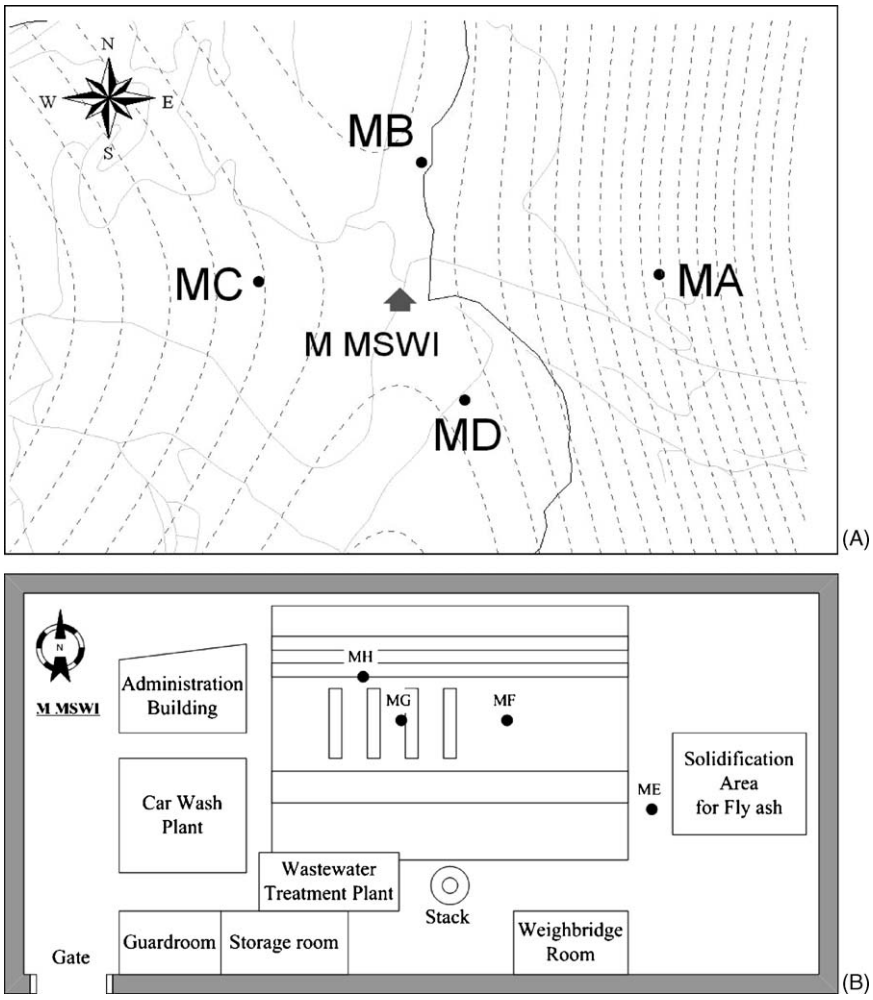

Fig. 2. Air sampling sites of the surrounding environment (outdoor) (A) and workplace (B) in M MSWI. *Note*: MA, MB, MC and MD are the sampling sites of the surrounding environment; ME is fly ash dumping platform, MF is fly ash conveyer, MG is bottom ash conveyer and MH is bottom ash bunker.

mode was selected ion monitoring (SIM) with a resolving power of 10,000. The electron energy was set at 35 eV, and the source temperature was set at 250 °C. An CTC A200S autosampler (CTC Analytics AG, GCPAL, Switzerland) was equipped with pull-up speed of 55 μl/s and injection speed of 55 μl/s. Syringes for analyses were washed with two kinds of solvents: *n*-hexane and dichloromethane. The injection volume was 2 μl. The temperature of the injector and the interface was 300 °C.

3. Results and discussion

3.1. PCDD/Fs in the air of surrounding environments of two MSWIs

Tables 2 and 3 summarize the PCDD/F concentrations in the outdoor air during summer and winter for T and M MSWIs, respectively. The results show that total PCDD/F concentrations of outdoor air during winter are lower than those during summer at all sampling sites in these two MSWIs. For T MSWI, the possible reason was it was rainy during the latest 2 sampling days, thus parts of PCDD/Fs were washed out by the rain and the levels of them were lowered accordingly. The results for M MSWI disagreed with that of the study of Fiedler [13], which reported that ambient air concentrations monitored over several years have shown a clear seasonal trend with higher PCDD/Fs levels in winter and lower concentrations during summer.

Indeed, studies on PCDD/F seasonal trend are clearly interesting and important, because the results provide clues to link with pollution sources. The PCDD/Fs emitted from some combustion sources (such as domestic heating) are greater in winter and certain atmospheric loss processes (e.g., photolysis) may also vary seasonally. Seasonal changes, with winter levels higher than summer levels (both total PCDD/Fs and total PCDD/F I-TEQ), have been reported by several other researchers [14–16]. However, this seasonal trend in air concentrations is suggested by many studies but not by all. Making a link to source(s) as controlling any seasonal trend is complex [17]. In our study, we categorize the raining into atmospheric loss processes for T MSWI and lack of domestic heating and photolysis (full of sunlight) in winter in Taiwan for M MSWI, which possibly led to lower levels of PCDD/Fs during winter.

From results shown in Table 2 (for T MSWI), there appears an interesting phenomenon during summer. When total PCDD/F concentration increased from 2.19 (TA) to 4.27 (TD) pg/Nm^3

Table 2
PCDD/F concentrations in the ambient air of surrounding environment (outdoor) of T MSWI

PCDD/PCDFs (pg/Nm^3)	Summer					Winter				
	TA	TB	TC	TD	Mean	TA	TB	TC	TD	Mean
2,3,7,8-TeCDD	0.00904	0.00375	0.00383	0.00349	0.00503	0.00436	0.00753	0.00597	0.00583	0.00592
1,2,3,7,8-PeCDD	0.0274	0.0149	0.0164	0.0137	0.0181	0.0106	0.00792	0.0134	0.0114	0.0108
1,2,3,4,7,8-HxCDD	0.0179	0.0160	0.0188	0.0162	0.0172	0.00921	0.00864	0.0120	0.0107	0.0101
1,2,3,6,7,8-HxCDD	0.0275	0.0353	0.0385	0.0363	0.0344	0.0160	0.0153	0.0233	0.0195	0.0185
1,2,3,7,8,9-HxCDD	0.0455	0.0274	0.0325	0.0498	0.0388	0.0117	0.0125	0.0197	0.0147	0.0147
1,2,3,4,6,7,8-HpCDD	0.151	0.239	0.271	0.472	0.283	0.0748	0.0722	0.138	0.0996	0.0962
OCDD	0.354	0.506	0.554	1.40	0.704	0.144	0.165	0.349	0.311	0.242
2,3,7,8-TeCDF	0.435	0.263	0.249	0.201	0.287	0.0186	0.0242	0.0253	0.0330	0.0253
1,2,3,7,8-PeCDF	0.0893	0.0518	0.060	0.0398	0.0602	0.0261	0.0224	0.0293	0.0347	0.0281
2,3,4,7,8-PeCDF	0.150	0.0869	0.109	0.0627	0.102	0.0343	0.0375	0.0392	0.0532	0.0411
1,2,3,4,7,8-HxCDF	0.259	0.224	0.243	0.147	0.218	0.0364	0.0345	0.0432	0.0441	0.0396
1,2,3,6,7,8-HxCDF	0.0988	0.0894	0.0975	0.0596	0.0863	0.0356	0.0359	0.0427	0.0447	0.0397
1,2,3,7,8,9-HxCDF	0.00600	0.00790	0.00965	0.00529	0.00721	0.00265	0.00300	0.00305	0.00301	0.00293
2,3,4,6,7,8-HxCDF	0.104	0.114	0.124	0.0696	0.103	0.0504	0.0436	0.0622	0.0566	0.0532
1,2,3,4,6,7,8-HpCDF	0.261	0.391	0.390	0.258	0.325	0.117	0.102	0.150	0.136	0.126
1,2,3,4,7,8,9-HpCDF	0.0295	0.0508	0.0524	0.0369	0.0424	0.0121	0.0154	0.0168	0.0201	0.0161
OCDF	0.128	0.506	0.241	1.40	0.569	0.0644	0.0695	0.0860	0.148	0.0920
PCDDs	0.632	0.842	0.935	1.99	1.10	0.271	0.289	0.561	0.473	0.399
PCDFs	1.56	1.78	1.58	2.28	1.80	0.398	0.388	0.498	0.573	0.464
PCDDs/PCDFs ratio	0.405	0.472	0.593	0.874	0.586	0.681	0.745	1.13	0.824	0.845
Total PCDD/Fs	2.19	2.63	2.51	4.27	2.90	0.668	0.677	1.06	1.05	0.864
PCDDs pg I-TEQ/Nm^3	0.0337	0.0220	0.0243	0.0267	0.0267	0.0142	0.0160	0.0199	0.0173	0.0169
PCDFs pg I-TEQ/Nm^3	0.173	0.121	0.134	0.0859	0.128	0.0342	0.0352	0.0405	0.0482	0.0395
PCDDs/PCDFs (TEQ) ratio	0.195	0.182	0.181	0.311	0.217	0.417	0.455	0.492	0.360	0.431
Total pg I-TEQ/Nm^3	0.206	0.143	0.159	0.113	0.155	0.0484	0.0513	0.0604	0.0655	0.0564
PCDDs pg WHO-TEQ/Nm^3	0.0471	0.0290	0.0320	0.0323	0.0351	0.0194	0.0198	0.0263	0.0227	0.0221
PCDFs pg WHO-TEQ/Nm^3	0.173	0.120	0.134	0.0847	0.128	0.0341	0.0352	0.0404	0.0481	0.0395
PCDDs/PCDFs (TEQ) ratio	0.273	0.241	0.238	0.381	0.283	0.569	0.564	0.651	0.473	0.564
Total pg WHO-TEQ/Nm^3	0.220	0.149	0.166	0.117	0.163	0.0535	0.0550	0.0667	0.0708	0.0615

(around two-fold), PCDD/F I-TEQ concentration decreased from 0.206 to 0.113 pg I-TEQ/Nm^3 (or from 0.220 to 0.117 pg WHO-TEQ/Nm^3) (around half). This phenomenon can be elucidated by Fig. 3, which indicates that the TD location had a different congener profile from the others. Meanwhile, it is apparent that there were two peaks in location TD. These two peaks represent concentrations of OCDD and OCDF, which both had the lowest toxic potency (toxic equivalency factor proposed by World Health Organization, WHO-TEF = 0.0001) [18], thus significantly lowering the total PCDD/F WHO-TEQ (or I-TEQ) concentration.

OCDD and OCDF are two highly chlorinated congeners among the PCDD/Fs and preferred to occur in the particulate phase. The high levels of OCDD and OCDF at a sampling site generally mean that the total PCDD/Fs was quantitatively and qualitatively influenced by a certain major emission source (e.g., the flue gas emission). TD location was situated on the downwind side of T MSWI and was thus easily influenced by the flue gas emission during summer. This can be verified in Table 2, which shows that TD location exhibited the highest total PCDD/Fs concentration (4.27 pg/Nm^3) and high levels of OCDD and OCDF (both were 1.40 pg/Nm^3) during summer. Therefore, it shows a different congener profile at TD location, because the other three sampling sites were not so influenced by T MSWI.

As can be seen from Table 3 (for M MSWI), the higher the total PCDD/F concentration, the higher the total PCDD/F WHO-TEQ concentration is at most sampling sites during both summer and winter. In addition, the above results for associated toxicity were significantly lower than 0.6 pg I-TEQ/Nm^3, which was the dioxin emission standard in Japan [1]. Such information reveals that the Taiwan government has made a strong decision to control the PCDD/F emissions from MSWIs.

It is worthy to mention that, except at the TC location during winter for the T MSWI and MC location during winter for M MSWI (where PCDDs/PCDFs ratios equal to 1.13 and 1.17, respectively), concentrations of total PCDFs were all higher than those of total PCDDs. Meanwhile, the ratios of PCDDs/PCDFs (I-TEQ or WHO-TEQ) at all sampling sites were less than unity, meaning that PCDFs were the primary distributors of toxicity for PCDD/Fs in the surrounding environments.

PCDD/F emissions from most combustion processes are mixtures of 75 PCDD and 135 PCDF congeners. The mixture can be translated into profiles, which represent the distribution of individual PCDD/Fs. The comparison of a homologue pattern is a useful method to trace the source of contamination. Differ-

Table 3
PCDD/F concentrations in the ambient air of surrounding environment (outdoor) of M MSWI

PCDD/PCDFs (pg/Nm^3)	Summer					Winter				
	MA	MB	MC	MD	Mean	MA	MB	MC	MD	Mean
2,3,7,8-TeCDD	0.00108	0.00261	0.00229	0.00169	0.00192	0.00200	0.00339	0.00263	0.00270	0.00268
1,2,3,7,8-PeCDD	0.00379	0.00921	0.0109	0.00789	0.00795	0.00283	0.00381	0.00270	0.00325	0.00315
1,2,3,4,7,8-HxCDD	0.00672	0.0127	0.0119	0.0106	0.0105	0.00182	0.00299	0.00260	0.00244	0.00246
1,2,3,6,7,8-HxCDD	0.0124	0.0249	0.0266	0.0215	0.0214	0.00396	0.00562	0.00503	0.00620	0.00520
1,2,3,7,8,9-HxCDD	0.0158	0.0346	0.0196	0.0167	0.0217	0.00332	0.00443	0.00435	0.00490	0.00425
1,2,3,4,6,7,8-HpCDD	0.0990	0.194	0.181	0.179	0.163	0.0240	0.0360	0.0507	0.0315	0.0356
OCDD	0.308	0.491	0.458	0.457	0.429	0.0492	0.0672	0.140	0.0651	0.0804
2,3,7,8-TeCDF	0.0938	0.194	0.202	0.142	0.158	0.00877	0.0141	0.00777	0.0117	0.0106
1,2,3,7,8-PeCDF	0.0202	0.0367	0.0407	0.0306	0.0321	0.00982	0.0143	0.00872	0.0133	0.0115
2,3,4,7,8-PeCDF	0.0284	0.0570	0.0592	0.0477	0.0481	0.0135	0.0211	0.0120	0.0186	0.0163
1,2,3,4,7,8-HxCDF	0.0633	0.137	0.141	0.124	0.116	0.0128	0.0191	0.0116	0.0172	0.0152
1,2,3,6,7,8-HxCDF	0.0259	0.0563	0.0599	0.0522	0.0486	0.0138	0.0186	0.0124	0.0189	0.0159
1,2,3,7,8,9-HxCDF	0.00284	0.00548	0.00598	0.00483	0.00478	0.00148	0.00178	0.00244	0.00285	0.00214
2,3,4,6,7,8-HxCDF	0.0258	0.0721	0.0717	0.0657	0.0588	0.0160	0.0225	0.0140	0.0250	0.0194
1,2,3,4,6,7,8-HpCDF	0.102	0.243	0.252	0.230	0.207	0.0444	0.0527	0.0439	0.0638	0.0512
1,2,3,4,7,8,9-HpCDF	0.0155	0.0330	0.0392	0.0378	0.0314	0.00795	0.00879	0.00842	0.0107	0.00897
OCDF	0.0875	0.169	0.179	0.162	0.149	0.0345	0.0415	0.0567	0.0480	0.0452
PCDDs	0.447	0.769	0.710	0.694	0.655	0.0871	0.123	0.208	0.116	0.134
PCDFs	0.465	1.00	1.05	0.897	0.853	0.163	0.214	0.178	0.230	0.196
PCDDs/PCDFs ratio	0.960	0.766	0.676	0.774	0.794	0.534	0.576	1.17	0.505	0.696
Total PCDD/Fs	0.912	1.77	1.76	1.59	1.51	0.250	0.338	0.386	0.346	0.330
PCDDs pg I-TEQ/Nm^3	0.00777	0.0169	0.0158	0.0128	0.0133	0.00461	0.00703	0.00583	0.00606	0.00588
PCDFs pg I-TEQ/Nm^3	0.0376	0.0798	0.0828	0.0671	0.0668	0.0131	0.0195	0.0118	0.0183	0.0157
PCDDs/PCDFs(TEQ) ratio	0.206	0.211	0.191	0.190	0.200	0.353	0.360	0.492	0.331	0.384
Total pg I-TEQ/Nm^3	0.0454	0.0966	0.0986	0.0799	0.0801	0.0177	0.0266	0.0177	0.0244	0.0216
PCDDs pg WHO-TEQ/Nm^3	0.0938	0.0210	0.0209	0.0163	0.0169	0.00598	0.00887	0.00705	0.00763	0.00738
PCDFs pg WHO-TEQ/Nm^3	0.0376	0.0796	0.0826	0.0669	0.0667	0.0131	0.0195	0.0118	0.0183	0.0157
PCDDs/PCDFs(TEQ) ratio	0.250	0.264	0.252	0.243	0.252	0.459	0.455	0.598	0.417	0.482
Total pg WHO-TEQ/Nm^3	0.0469	0.101	0.103	0.0832	0.0835	0.0190	0.0284	0.0188	0.0259	0.0230

ent sources of PCDD/Fs can be usually characterized by their different congener patterns [17,19]. The 17 congener profiles of PCDD/Fs in the outdoor air during summer and winter for T and M MSWIs are illustrated in Figs. 3 and 4; the *y*-coordinate was normalized by dividing the concentration of each congener by that of total 2,3,7,8-PCDD/Fs. As shown in these two figures, except at the TD location during summer for T MSWI (Fig. 3), the congener profiles of these outdoor sampling sites did not

Table 4
Comparison of mean PCDD/F concentrations in the ambient air of four areas in Taiwan

Sampling sits	Kenting (remote area) ($n=6$)	Pingtung (suburban area) ($n=8$)	T MSWI (surrounding environment of incineration plant) ($n=8$)	M MSWI (surrounding environment of incineration plant) ($n=8$)
PCDDs (pg/Nm^3)	0.0648	0.670	0.750	0.395
PCDFs (pg/Nm^3)	0.0792	0.522	1.13	0.526
PCDDs/PCDFs ratio	0.412	1.10	0.716	0.744
Total PCDD/Fs (pg/Nm^3)	0.144	1.19	1.88	0.921
PCDDs pg I-TEQ/Nm^3	0.00399	0.0174	0.0218	0.00974
PCDFs pg I-TEQ/Nm^3	0.00787	0.0521	0.0840	0.0413
PCDDs/PCDFs (TEQ) ratio	0.265	0.294	0.325	0.297
Total pg I-TEQ/Nm^3	0.0119	0.0695	0.106	0.0510
PCDDs pg WHO-TEQ/Nm^3	0.00535	0.0231	0.0285	0.0123
PCDFs pg WHO-TEQ/Nm^3	0.00785	0.0520	0.0838	0.0412
PCDDs/PCDFs (TEQ) ratio	0.350	0.391	0.424	0.375
Total pg WHO-TEQ/Nm^3	0.0132	0.0751	0.112	0.0535

n: Number of samples.

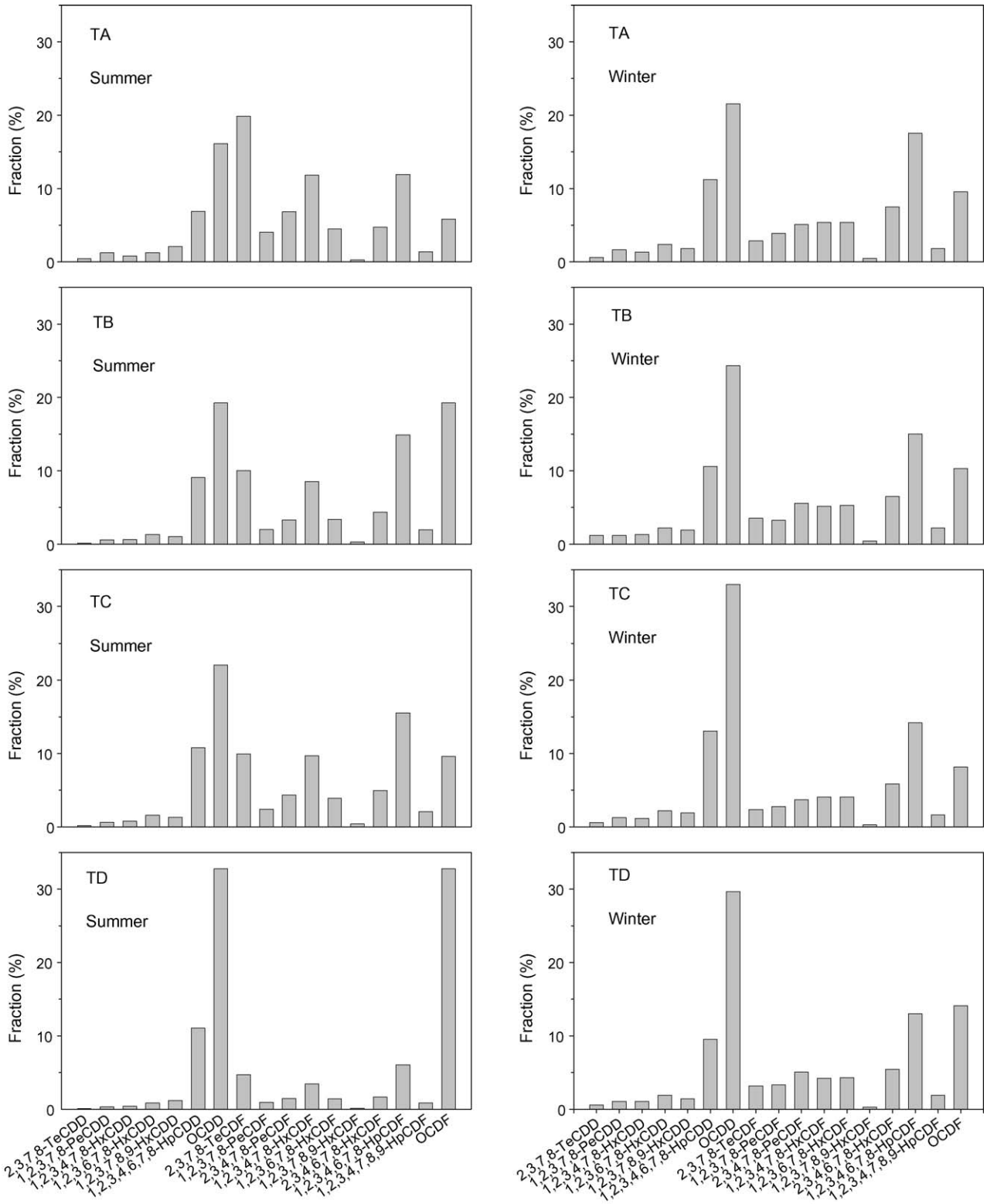

Fig. 3. Congener profiles of PCDD/Fs in the surrounding environment (outdoor air) during summer and winter of T MSWI.

exhibit significant differences. This result implies that the ingredients of municipal wastes and the performance of incineration plants were quite similar between T and M MSWIs.

Table 4 shows the comparisons of mean PCDD/F concentrations in the ambient air of four areas in Taiwan. Ken-ting is a remote area which was not quite influenced by anthropogenic activities all the way, and Pingtung is a representative of a suburban area in Taiwan. The total PCDD/F concentrations of T and M MSWIs were significantly higher than that of the Ken-ting area, around 13- and 6-fold higher (1.88/0.144 and 0.921/0.144).

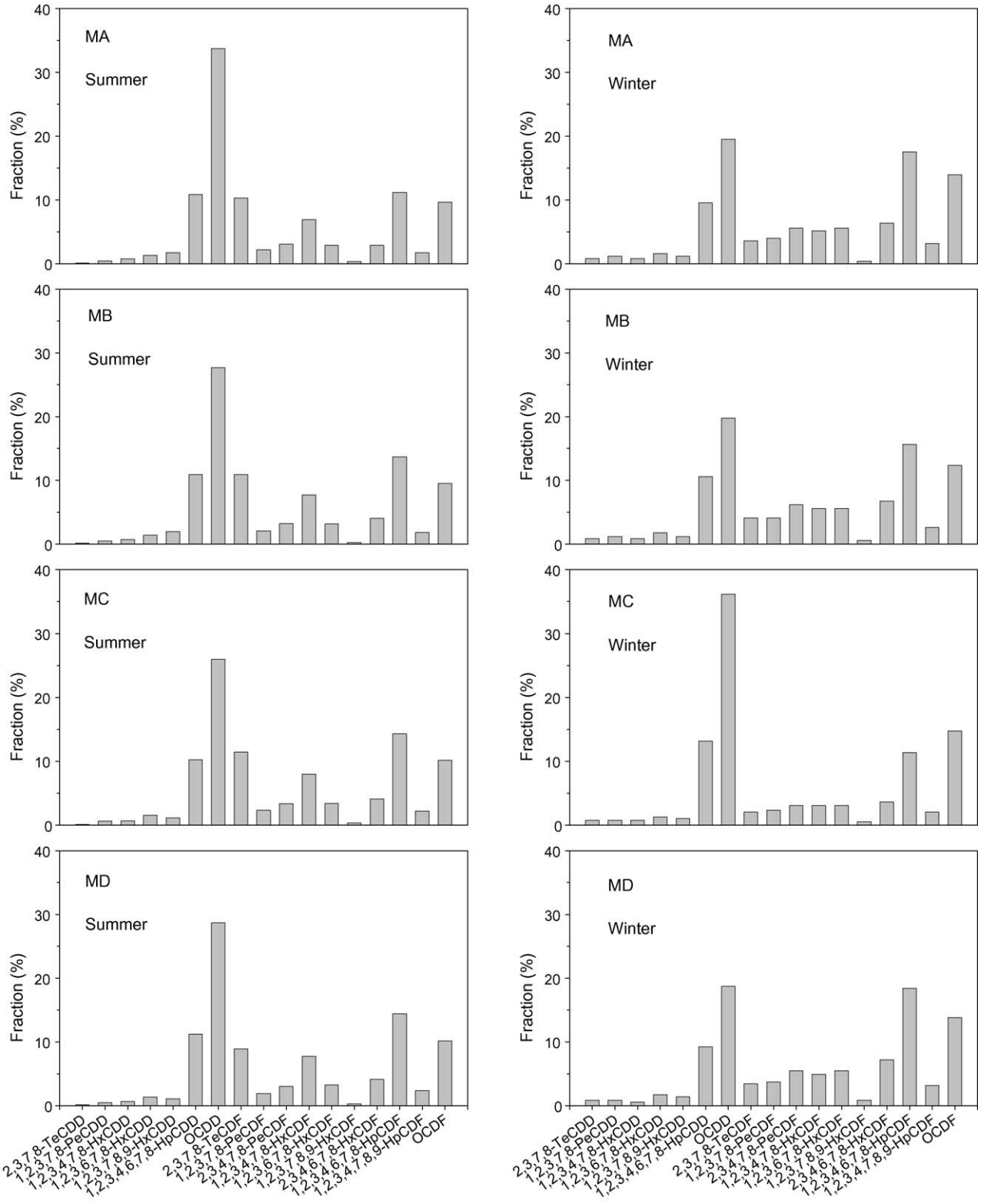

Fig. 4. Congener profiles of PCDD/Fs in the surrounding environment (outdoor air) during summer and winter of M MSWI.

However, the higher total PCDD/F concentration of Pingtung is probably due to the existence of open burning during sampling.

Comparing the PCDD/F I-TEQ concentrations in the ambient air of T and M MSWIs (Taiwan, this study) with various countries (Table 5, including Germany, Japan, and Spain), we found that the remote area of Taiwan (Ken-ting) has the lowest PCDD/F I-TEQ concentration (0.0119 pg I-TEQ/Nm^3). For the rural areas, the PCDD/F concentrations ranged between 0.018 and 0.070 pg I-TEQ/Nm^3, which were the lowest levels next to the remote area. For suburban, urban, or MSWI influenced

Table 5
Comparisons of PCDD/F I-TEQ concentrations in the ambient air of various countries

Area/city, country	pg I-TEQ/Nm^3	Reference
Remote area (Kenting, Taiwan)	0.0119	This study
Rural area (Germany)	0.021	[22]
Rural area with elevated regions (Black Forest, Germany)	0.018	[22]
Rural area, near MWI (Spain)	0.05	[23]
Rural area (Germany)	0.025–0.070	[13]
Suburban area (Tsukuba, Japan)	0.39	[21]
Suburban area (Tanzawa, Japan)	0.26	[21]
Suburban area (Germany)	0.056	[22]
Suburban area (Pingtung, Taiwan)	0.0695	This study
Urban area (Tokyo, Japan)	0.45	[21]
Urban area (Yokohama, Japan)	0.54	[21]
Urban area (Germany)	0.083	[22]
Urban area with high traffic influence (Spain)	0.26	[23]
Urban area (Germany)	0.070–0.350	[13]
Surrounding environment of T incinerator (Northern Taiwan)	0.106	This study
Surrounding environment of M incinerator (Northern Taiwan)	0.0510	This study
MWI influence zone (Spain)	0.55	[23]
MWI influence zone, high traffic and industrial influence (Spain)	0.28	[23]
High industrial activity (Spain)	0.52	[23]

Table 6
PCDD/F concentrations in the workplace air of T MSWI

PCDD/PCDFs (pg/Nm^3)	Summer					Winter				
	TE	TF	TG	TH	Mean	TE	TF	TG	TH	Mean
2,3,7,8-TeCDD	0.286	0.335	0.0158	0.0263	0.166	0.0465	0.0303	0.0129	0.0136	0.0258
1,2,3,7,8-PeCDD	0.618	0.719	0.0406	0.0633	0.360	0.0849	0.0621	0.0309	0.0356	0.0534
1,2,3,4,7,8-HxCDD	0.505	0.613	0.0361	0.0612	0.304	0.0504	0.0443	0.0266	0.0312	0.0381
1,2,3,6,7,8-HxCDD	1.16	1.48	0.0851	0.136	0.715	0.101	0.0859	0.0572	0.0688	0.0782
1,2,3,7,8,9-HxCDD	1.29	1.59	0.0641	0.160	0.776	0.0756	0.0616	0.0420	0.0499	0.0573
1,2,3,4,6,7,8-HpCDD	9.00	11.7	0.593	0.934	5.56	0.544	0.465	0.306	0.322	0.409
OCDD	22.5	28.8	1.27	1.96	13.6	1.18	1.02	0.742	0.611	0.888
2,3,7,8-TeCDF	4.95	5.61	0.513	0.731	2.95	0.302	0.206	0.0905	0.0993	0.174
1,2,3,7,8-PeCDF	2.15	2.33	0.142	0.218	1.21	0.330	0.232	0.116	0.121	0.200
2,3,4,7,8-PeCDF	2.59	2.96	0.200	0.329	1.52	0.370	0.267	0.153	0.169	0.240
1,2,3,4,7,8-HxCDF	3.93	4.31	0.381	0.629	2.31	0.284	0.219	0.139	0.154	0.199
1,2,3,6,7,8-HxCDF	2.22	2.47	0.176	0.280	1.29	0.304	0.231	0.138	0.154	0.207
1,2,3,7,8,9-HxCDF	0.211	0.244	0.0155	0.0251	0.124	0.0258	0.0217	0.0123	0.0126	0.0181
2,3,4,6,7,8-HxCDF	2.16	2.48	0.194	0.323	1.29	0.311	0.264	0.162	0.177	0.229
1,2,3,4,6,7,8-HpCDF	6.19	7.24	0.604	1.04	3.77	0.751	0.611	0.469	0.481	0.578
1,2,3,4,7,8,9-HpCDF	0.907	1.13	0.0784	0.164	0.570	0.114	0.0888	0.0661	0.0668	0.0839
OCDF	2.79	3.61	0.315	0.814	1.88	0.474	0.420	0.505	0.329	0.432
PCDDs	35.4	45.2	2.10	3.34	21.5	2.08	1.77	1.22	1.13	1.55
PCDFs	28.1	32.4	2.62	4.55	16.9	3.27	2.56	1.85	1.76	2.36
PCDDs/PCDFs ratio	1.26	1.40	0.804	0.734	1.05	0.638	0.691	0.658	0.642	0.657
Total PCDD/Fs	63.5	77.6	4.72	7.89	38.4	5.35	4.33	3.07	2.90	3.91
PCDDs pg I-TEQ/Nm^3	1.00	1.21	0.0618	0.105	0.594	0.118	0.0862	0.0447	0.0502	0.0748
PCDFs pg I-TEQ/Nm^3	2.82	3.20	0.242	0.387	1.66	0.333	0.247	0.142	0.156	0.220
PCDDs/PCDFs (TEQ) ratio	0.355	0.378	0.255	0.271	0.315	0.355	0.349	0.314	0.322	0.335
Total pg I-TEQ/Nm^3	3.83	4.40	0.304	0.492	2.26	0.452	0.333	0.187	0.206	0.295
PCDDs pg WHO-TEQ/Nm^3	1.29	1.54	0.0810	0.135	0.762	0.160	0.116	0.0595	0.0675	0.101
PCDFs pg WHO-TEQ/Nm^3	2.82	3.19	0.242	0.386	1.66	0.333	0.246	0.142	0.156	0.219
PCDDs/PCDFs (TEQ) ratio	0.458	0.483	0.335	0.349	0.406	0.480	0.472	0.419	0.433	0.451
Total pg WHO-TEQ/Nm^3	4.11	4.73	0.323	0.521	2.42	0.493	0.363	0.201	0.223	0.320

Note: TE is bottom ash bunker, TF is ash wetting unit, TG is ash conveyer and TH is common fly ash conveyer.

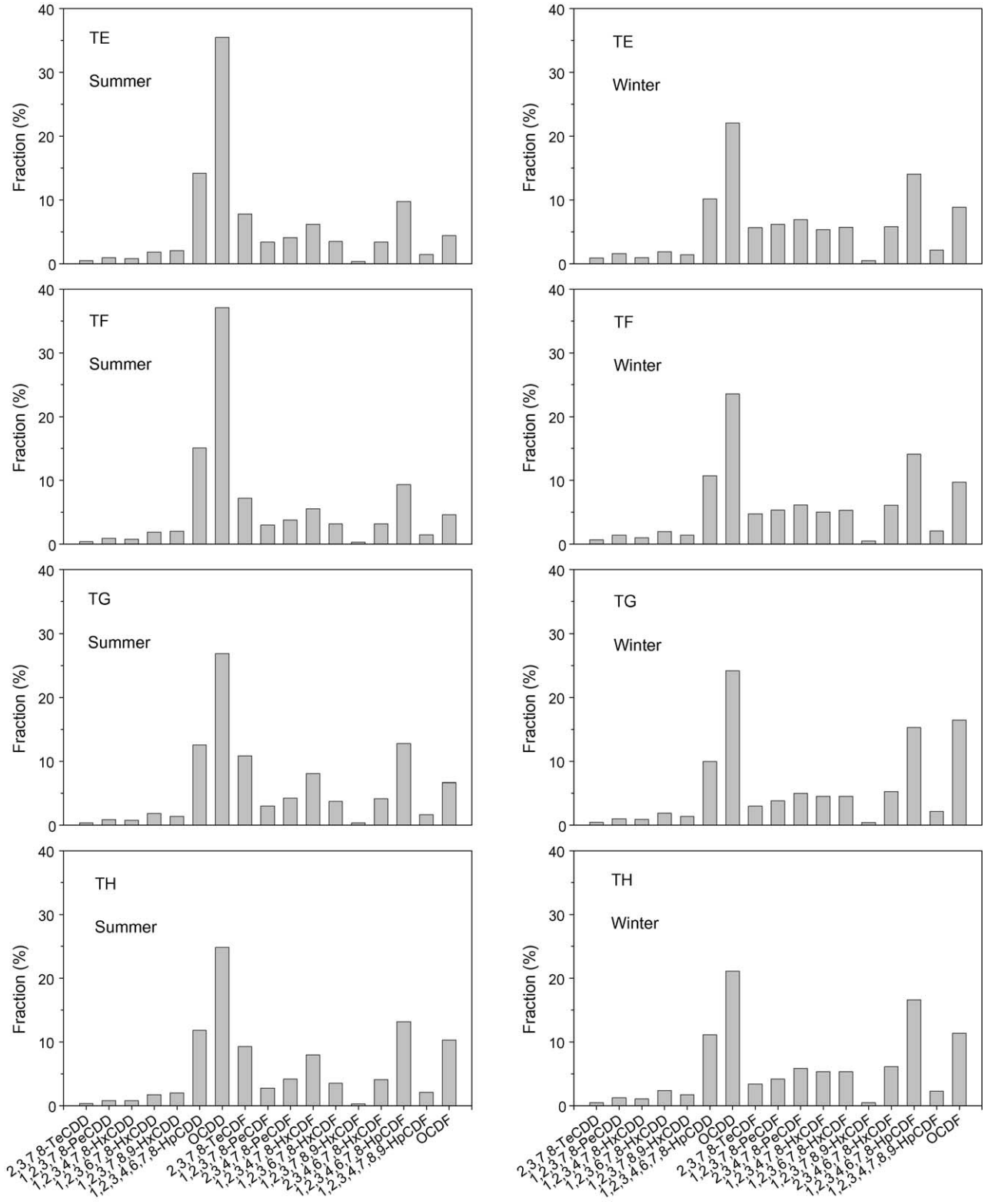

Fig. 5. Congener profiles of PCDD/Fs in the workplace air during summer and winter of T MSWI. *Note*: TE is bottom ash bunker, TF is ash wetting unit, TG is ash conveyer and TH is common fly ash conveyer.

zones, the PCDD/F I-TEQ concentrations are generally greater than those in the former two areas, but they did not show a significant difference among these three areas. Nevertheless, different countries show different levels. For instance, the PCDD/F I-TEQ concentrations in the ambient air of both suburban and urban areas in Japan are greater than those of Germany. Speaking of Asia, the PCDD/F I-TEQ concentrations of T and M MSWIs (Taiwan) were lower than those of the urban area in Tokyo (0.45 pg I-TEQ/Nm^3) and in Yokohama (0.54 pg I-TEQ/Nm^3) (Japan).

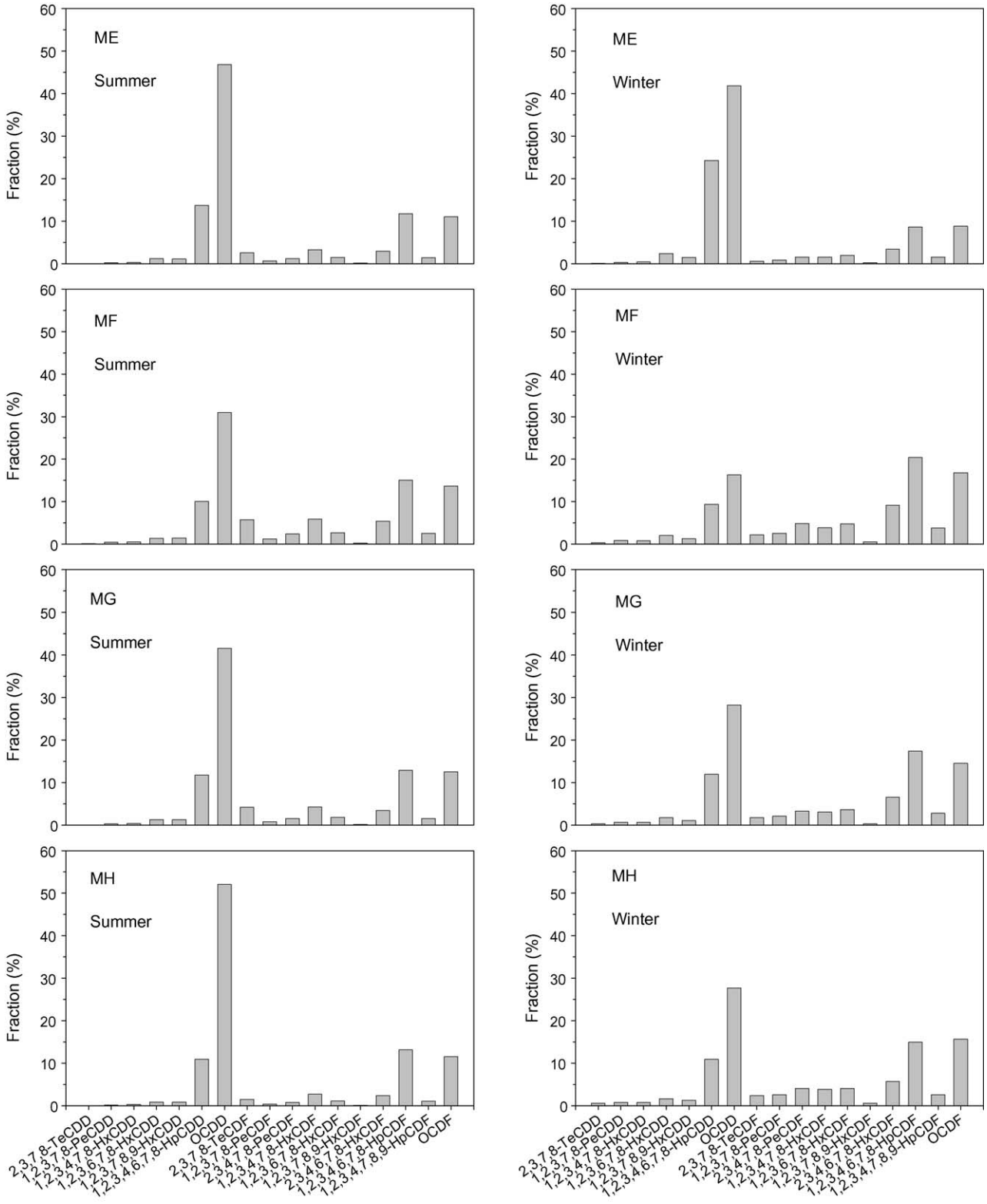

Fig. 6. Congener profiles of PCDD/Fs in the workplace air during summer and winter for M MSWI. *Note*: ME is fly ash dumping platform, MF is fly ash conveyer, MG is bottom ash conveyer and MH is bottom ash bunker.

3.2. PCDD/Fs in the workplace air of two MSWIs

Analytical results of PCDD/F concentrations in the workplace air of T and M MSWIs are listed in Tables 6 and 7, respectively. As shown in these two tables, concentrations of individual or total PCDD/Fs in the workplace air during winter were all lower than those during summer. Especially at locations with ash wetting unit (TF) and bottom ash bunker (TE) for

Table 7
PCDD/F concentrations in the workplace air of M MSWI

PCDD/PCDFs (pg/Nm^3)	Summer					Winter				
	ME	MF	MG	MH	Mean	ME	MF	MG	MH	Mean
2,3,7,8-TeCDD	0.00947	0.0169	0.0103	0.0114	0.0120	0.00434	0.0166	0.00447	0.00251	0.00698
1,2,3,7,8-PeCDD	0.0266	0.0621	0.0328	0.0398	0.0403	0.0133	0.0449	0.00828	0.00421	0.0177
1,2,3,4,7,8-HxCDD	0.0373	0.0671	0.0452	0.0636	0.0533	0.0184	0.0404	0.00775	0.00377	0.0176
1,2,3,6,7,8-HxCDD	0.135	0.181	0.141	0.207	0.166	0.0949	0.103	0.0225	0.00909	0.0574
1,2,3,7,8,9-HxCDD	0.125	0.192	0.140	0.199	0.164	0.0594	0.0635	0.0143	0.00660	0.0360
1,2,3,4,6,7,8-HpCDD	1.54	1.34	1.31	2.50	1.67	0.968	0.468	0.156	0.0587	0.413
OCDD	5.26	4.13	4.63	11.9	6.48	1.67	0.814	0.368	0.150	0.751
2,3,7,8-TeCDF	0.284	0.766	0.467	0.342	0.465	0.0231	0.110	0.0231	0.0127	0.0422
1,2,3,7,8-PeCDF	0.0686	0.164	0.0903	0.0874	0.103	0.0354	0.129	0.0267	0.0142	0.0513
2,3,4,7,8-PeCDF	0.131	0.328	0.170	0.172	0.200	0.0616	0.243	0.0432	0.0218	0.0924
1,2,3,4,7,8-HxCDF	0.372	0.784	0.476	0.626	0.565	0.0609	0.192	0.0398	0.0207	0.0784
1,2,3,6,7,8-HxCDF	0.171	0.362	0.202	0.257	0.248	0.0783	0.239	0.0483	0.0223	0.0970
1,2,3,7,8,9-HxCDF	0.0150	0.0364	0.0191	0.0206	0.0228	0.00803	0.0249	0.00353	0.00288	0.00984
2,3,4,6,7,8-HxCDF	0.331	0.723	0.386	0.539	0.495	0.137	0.457	0.0847	0.0305	0.177
1,2,3,4,6,7,8-HpCDF	1.32	2.01	1.43	3.01	1.94	0.344	1.02	0.227	0.0813	0.418
1,2,3,4,7,8,9-HpCDF	0.159	0.345	0.177	0.250	0.233	0.0623	0.190	0.0364	0.0139	0.0757
OCDF	1.24	1.82	1.40	2.65	1.78	0.352	0.842	0.189	0.0854	0.367
PCDDs	7.13	5.99	6.31	14.9	8.58	2.83	1.55	0.581	0.235	1.30
PCDFs	4.09	7.34	4.82	7.95	6.05	1.16	3.45	0.722	0.306	1.41
PCDDs/PCDFs ratio	1.74	0.816	1.31	1.88	1.44	2.43	0.450	0.805	0.768	1.11
Total PCDD/Fs	11.2	13.3	11.1	22.9	14.6	3.99	5.00	1.30	0.541	2.71
PCDDs pg I-TEQ/Nm^3	0.0732	0.109	0.0771	0.115	0.0936	0.0396	0.0652	0.0150	0.00730	0.0318
PCDFs pg I-TEQ/Nm^3	0.202	0.465	0.262	0.304	0.308	0.0677	0.243	0.0457	0.0216	0.0945
PCDDs/PCDFs (TEQ) ratio	0.362	0.236	0.294	0.379	0.318	0.585	0.268	0.328	0.339	0.380
Total pg I-TEQ/Nm^3	0.275	0.574	0.339	0.419	0.402	0.107	0.308	0.0607	0.0289	0.126
PCDDs pg WHO-TEQ/Nm^3	0.0817	0.137	0.0893	0.124	0.108	0.0448	0.0870	0.0188	0.00927	0.0400
PCDFs pg WHO-TEQ/Nm^3	0.201	0.463	0.261	0.302	0.307	0.0674	0.242	0.0455	0.0215	0.0941
PCDDs/PCDFs (TEQ) ratio	0.406	0.295	0.342	0.412	0.364	0.664	0.359	0.413	0.432	0.467
Total pg WHO-TEQ/Nm^3	0.283	0.600	0.350	0.426	0.415	0.112	0.329	0.0643	0.0307	0.134

Note: ME is fly ash dumping platform, MF is fly ash conveyer, MG is bottom ash conveyer and MH is bottom ash bunker.

T MSWI (Table 6), this trend is more significant than the other sampling sites. The means of total PCDD/Fs I-TEQ concentration during two seasons for two MSWIs, 2.26, 0.295, 0.402 and 0.126 pg I-TEQ/Nm^3, all fell into the range (0.08–3.01 pg I-TEQ/m^3) that Hu et al. [9] had reported in their study.

In T MSWI (Table 6), total PCDD/F concentrations at four sampling sites (TE-TH) ranged from 4.72 to 77.6 pg/Nm^3 during summer, and ranged from 2.90 to 5.35 pg/Nm^3 during winter. The means were 38.4 and 3.91 pg/Nm^3, respectively. Regarding toxicity, total PCDD/F I-TEQ concentrations ranged between 0.304 and 4.40 pg I-TEQ/Nm^3 (or 0.323 and 4.73 pg WHO-TEQ/Nm^3) during summer, and ranged between 0.187 and 0.452 pg I-TEQ/Nm^3 (or 0.201 and 0.493 pg WHO-TEQ/Nm^3) during winter. The means were 2.26 and 0.295 pg I-TEQ/Nm^3 (or 2.42 and 0.320 pg WHO-TEQ/Nm^3), respectively. Although total PCDD/F WHO-TEQ concentrations at locations with ash wetting unit (TF) and bottom ash bunker (TE) significantly decreased from summer to winter (from 4.73 to 0.363 and from 4.11 to 0.493 pg WHO-TEQ/Nm^3, respectively), TE and TF locations still had the highest total PCDD/F WHO-TEQ concentration among the four locations during winter and need to be improved. However, by spraying water on and wetting both the fly and bottom ashes during winter, the mean total PCDD/F I-TEQ concentration in the workplace air was reduced by 86.8%, from 2.42 to 0.320 pg WHO-TEQ/Nm^3 (or 86.9%, from 2.26 to 0.295 pg I-TEQ/Nm^3) in the T MSWI. The above results indicate that an appropriate improving action did inhibit the fugitive emission of PCDD/Fs and reduce the health risk of workers during handling work of ashes in MSWIs.

Fig. 5 shows the congener profiles of PCDD/Fs during summer and winter. As shown in this figure, there is no significant difference between summer and winter. Except at the TE, TF and TG sampling site in summer, where 2,3,7,8-TeCDF was the third dominant congener, OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF were generally the four dominant congeners.

Fig. 6 shows the congener profiles of PCDD/Fs in the workplace air of M MSWI during summer and winter. As shown in this figure, the congener profiles are very similar during summer and winter, and no significant difference was detected. Furthermore, comparing Fig. 6 with Fig. 5, there is also no significant difference between T and M MSWIs. OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF were generally the four dominant congeners in both MSWIs. This result agrees with the study of Lee et al. [20], in which they found the four congeners mentioned above were the predominant groups among

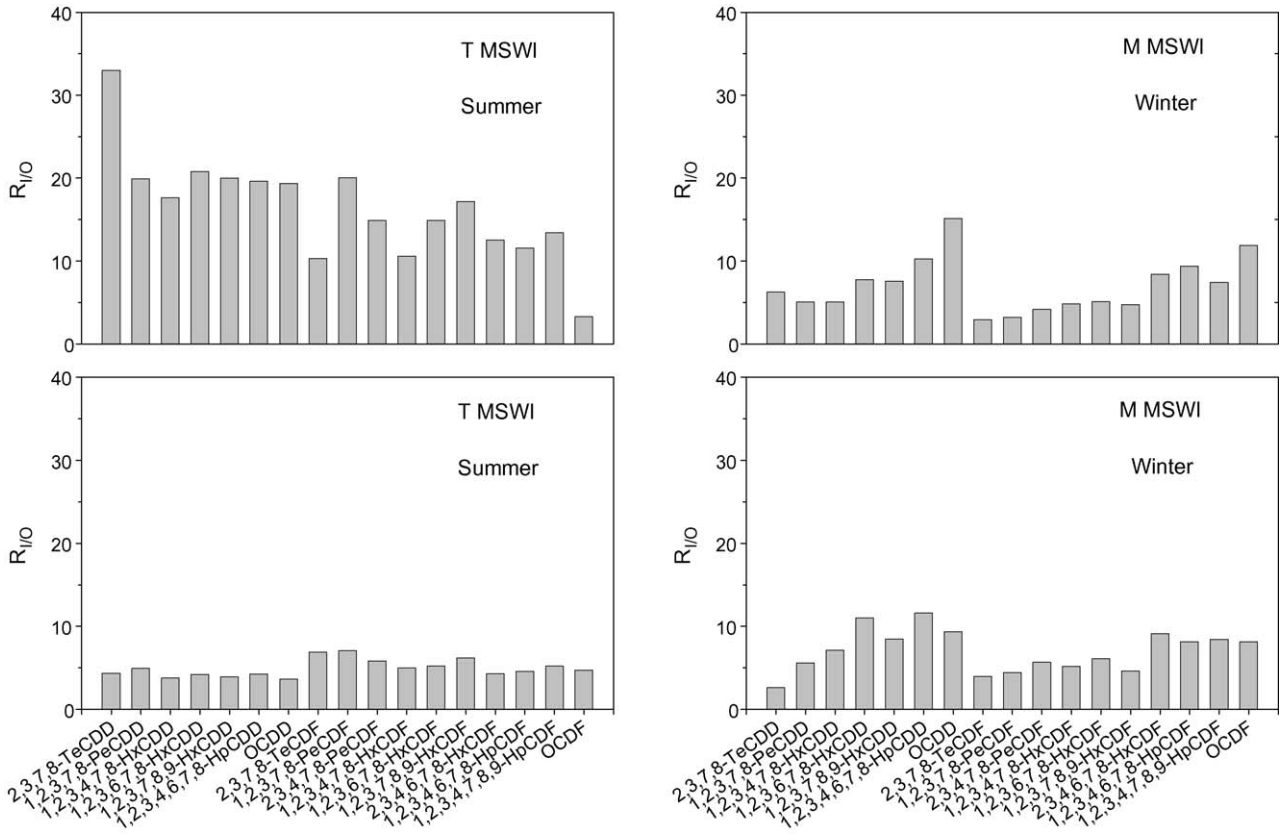

Fig. 7. I/O ratio for each PCDD/F congener ($R_{I/O}$) during summer and winter for two MSWIs.

17 congeners from both the ambient air and serum samples. In fact, the congener profiles of PCDD/Fs in the surrounding environment also exhibited these four dominant congeners (Figs. 3 and 4).

As previously mentioned in the outdoor air, PCDFs were the primary distributors of toxicity for PCDD/Fs due to the fact that the ratios of PCDDs/PCDFs (I-TEQ or WHO-TEQ) at all sampling sites were less than unity. This scenario was also true in the workplace air. The PCDDs/PCDFs ratios in the workplace air, however, were somewhat different from those in the outdoor air. In the outdoor air (Tables 2 and 3), the means of the PCDDs/PCDFs ratio (0.586, 0.845, 0.794, and 0.696) were all less than unity during summer and winter for both T and M MSWIs, while in the workplace air (Tables 6 and 7), the mean of the PCDDs/PCDFs ratio was less than unity only during winter for T MSWI (0.657 in Table 6). They were higher than unity in other circumstances (1.05, 1.44, and 1.11).

3.3. Comparisons of PCDD/Fs in the ambient air of surrounding environment and workplace

Comparing the total PCDD/F concentrations in T MSWI (Tables 2 and 6), the mean in the workplace air was 13 times (38.4/2.90) higher than that in the outdoor air during summer; during winter, it was about 5 times (3.91/0.864) higher. Regarding total PCDD/F WHO-TEQ concentrations, the mean in the workplace air was 15 times (2.42/0.163) higher than that in the outdoor air during summer; during winter, it was about 5 times (0.320/0.0615) higher.

On the other hand, comparing the total PCDD/F concentrations in M MSWI (Tables 3 and 7), the mean in the workplace air was 10 times (14.6/1.51) higher than that in the outdoor air during summer; during winter, it was about 8 times (2.71/0.330) higher. Speaking of total PCDD/F WHO-TEQ concentrations, the mean in the workplace air was five times (0.415/0.0835) higher than that in the outdoor air during summer; during winter, it was about six times (0.134/0.0230) higher.

It is summarized that the total PCDD/F concentrations in the workplace air were 5–13 times higher than that in the outdoor air; the PCDD/Fs WHO-TEQ concentrations in the workplace air were 5–15 higher than that in the outdoor air. In addition to total PCDD/F concentration, the individual congener concentrations also exhibit a similar phenomena. Fig. 7 shows the I/O ratio ($R_{I/O}$, the mean concentration in the workplace air divided by that in the outdoor air) of each congener of PCDD/Fs during summer and winter for T and M MSWIs. It is very clear that the $R_{I/O}$ is higher than unity for all the 17 congeners during two seasons for two MSWIs. In particular, this phenomenon is more significant during summer for T MSWI, where the $R_{I/O}$ ranged between 10 and 20 for most PCDD/F congeners.

However, the above results of this study reveal the fact that workplace environments are much worse than outdoors in T and M MSWIs. As we mentioned previously, there was much attention paid to the PCDD/F impact on the surrounding environment,

but only little research associated with the dioxin influence on the workplace. Therefore, further research should be conducted on the exposure of laborers to PCDD/Fs emitted in the workplace air.

4. Conclusions

(1) The total PCDD/F and the total PCDD/F WHO-TEQ concentrations in the workplace air were 5–13 and 5–15 times higher than those in the outdoor air, respectively. Obviously, it is worthwhile to explore more on health risk assessment for exposure of PCDD/Fs emitted from MSWIs, particularly in the workplace air.
(2) Mean total PCDD/F I-TEQ concentrations in the outdoor air ranged between 0.0216 and 0.155 pg I-TEQ/Nm^3 (or ranged between 0.0230 and 0.163 pg WHO-TEQ/Nm^3) and averaged 0.0783 pg I-TEQ/Nm^3 (or 0.0828 pg WHO-TEQ/Nm^3) during two seasons for two MSWIs, which were 6.5-fold higher than that of a remote site (0.0119 pg I-TEQ/Nm^3 or 0.0132 pg WHO-TEQ/Nm^3) in Taiwan. However, the above outdoor air concentration levels in the MSWI were still much lower than the air quality limitation of PCDD/Fs (0.6 pg I-TEQ/Nm^3) in Japan.
(3) PCDFs were the primary toxicity distributors for PCDD/Fs in the outdoor air; owing to the ratios of PCDDs/PCDFs (I-TEQ) at all sampling sites ranging from 0.181 to 0.492 (ranged from 0.238 to 0.651 for WHO-TEQ) and were less than unity.
(4) The OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD and 1,2,3,4,6,7,8-HpCDF were the four dominant species in both workplace and outdoor air near MSWIs.
(5) By spraying water on and wetting both the fly and bottom ashes, the mean total PCDD/F WHO-TEQ concentration in the workplace air was reduced 86.8% in the T MSWI. The above results indicate that an appropriate improving action did inhibit the fugitive emission of PCDD/Fs and reduce the health risk of workers during work handling ashes in MSWIs.

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