Characterizing the Emissions of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans from Crematories and Their Impacts to the Surrounding Environment

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This study was set out to characterize PCDD/F emissions from crematories and assess their impacts on the surrounding environment. Two crematories located in southern Taiwan were investigated, including the one (C1) with no air pollution control device installed and the other (C2) installed with a bag filter. Results show the mean PCDD/F emissions (11% oxygen) from the stacks of C1 and C2 were 2.36 and 0.322 ng I-TEQ Nm^{-3}, respectively. The mean emission factors for C1 and C2 were 13.6 and 6.11 μg I-TEQ body^{-1}, respectively. The removal efficiency of the bag filter on PCDD/Fs was 55.1%. The estimated PCDD/F emission rate for all crematories in Taiwan was 0.838 g I-TEQ yr^{-1} accounting for 227% and 112% of the annual emissions from all medical waste incinerators and municipal waste incinerators, respectively. The above results indicate that PCDD/F emissions from crematories were quite significant. To assess the impact of PCDD/F emissions from a crematory to the surrounding environment, ambient air samples were collected from the downwind site of C1 with the maximum ground concentration. We found the estimated maximum ground concentration at the downwind site of C1 (= 0.521 pg I-TEQ Nm^{-3}) was much higher than that found at the background, rural area, residential area, urban area, and industrial area (= 0.006, 0.023, 0.052, 0.093, and 0.190 pg I-TEQ Nm^{-3}, respectively). The above results suggest that PCDD/F emissions from a crematory did significantly affect its surrounding environment. In conclusion, a proper control strategy should be taken immediately in order to eliminate PCDD/F emissions from crematories.

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Introduction

After polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) were discovered in the flue gases and fly ash of municipal waste incinerators in 1977 (*1*), PCDD/F emissions from various sources have become a serious issue in many countries, because of their toxicological effects and associated adverse health implications. PCDD/Fs are mainly formed during anthropogenic activities and are usually referred to as dioxins. Intensive studies have been conducted on various PCDD/F emission sources, including the waste combustion sources, chemical-industrial sources, and other thermal sources. Nevertheless, only a few studies were carried out on crematories (*2*−*7*).

In a study conducted by Fledler in Germany, PCDD/F toxic equivalent (i.e., TEQ) concentrations of ∼8 ng TEQ Nm^{-3} were measured in the stack flue gases of crematories (*2*). Takeda et al. measured PCDD/F emissions from 17 crematories in Japan found PCDD/F concentrations and their corresponding TEQ concentrations in the stack flue gases were 4.9−1200 ng Nm^{-3} (12% oxygen) and 0.064−24 ng TEQ Nm^{-3}, respectively (*5*). In the UK, a laboratory study conducted by the Warren Spring Laboratory found the mean PCDD/F TEQ concentration of 46 ng TEQ Nm^{-3} (11% oxygen) for the cremation process (*7*). The wide range of PCDD/F concentrations arising from various crematories are believed to be due to their intrinsic differences in operation conditions, air pollution control devices, and involved incinerating materials (*5*).

In the U.S. (*8*), UK (*7*), and Japan (*5*), PCDD/F emission rates for the crematory source were found as ∼9.1, 1−35, and 1.3−3.8 g TEQ yr^{-1}, respectively. In principle, the total PCDD/F emission from the crematory was relatively small as compared with that from the municipal waste incinerator. For example, a recent study conducted in Japan has indicated that the crematory emission accounted for only 0.13−0.29% of that emitted from municipal waste incinerators (*5*). But it should be noted that most crematories are equipped with a low stack and are situated in the proximity of the residential area. In particular, most of them do not adopt any air pollution control device to eliminate PCDD/F emissions from stacks. Based on these, it can be expected that PCDD/F emissions from a crematory might significantly affect its surrounding environment. Moreover, it should be noted that recently the cremation ratio has increased dramatically in Taiwan and many other countries. In Taiwan, the cremation ratio is expected to increase from 66.9% in 2000 to 85.0% in 2005. In the U.S., the cremation ratio has increased significantly from 15.2% in 1987 to 25.0% in 2000 and is expected to reach 37.0% in 2010 (*8*). In Japan, because of the encouragement of the governmental policy its current cremation ratio is as high as 99% (*4*). Based on these data, it is expected that crematories will play an important role on PCDD/F emissions not only in Taiwan area but also in many other countries.

Indeed, in addition to PCDD/Fs, PCBs, and PAHs, total suspended particles (TSP) and odor released from crematories might also cause serious problems to human health. However, the U.S. EPA has reported that there appears to be no "safe" level for dioxin exposure, and the levels of dioxin and dioxin-like chemicals found in the general U.S. population were "at or near levels associated with adverse health effects" (*9*). Subjected to both cost and manpower, only PCDD/F emissions from crematories were studied in this work. In this study, two crematories located in southern Taiwan with similar operation conditions were investigated. The congener

TABLE 1. Basic Information for the Two Investigated Crematories of C1 and C2

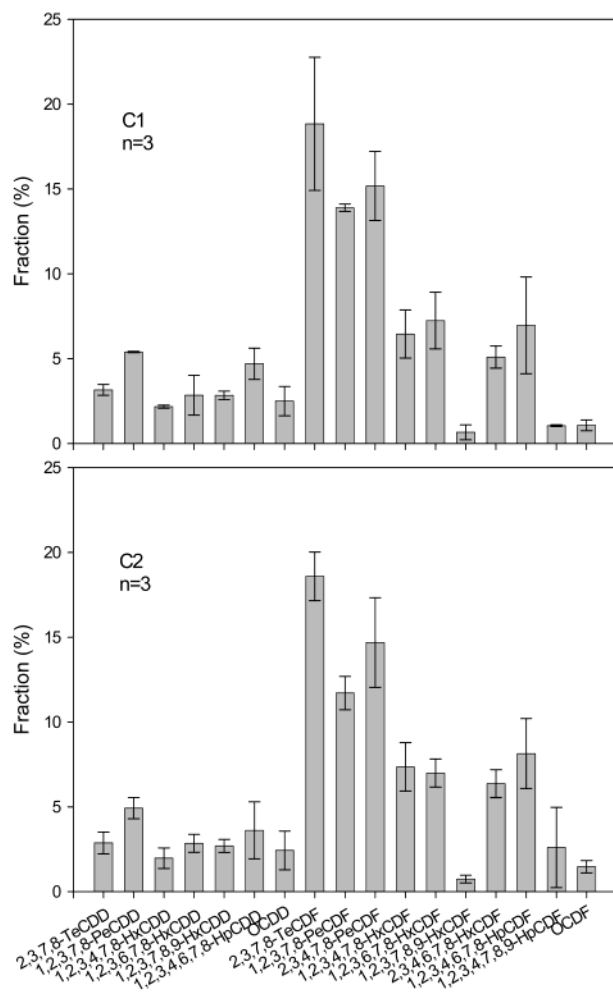| crematory	C1	C2
operational temperature of the primary combustor	730 °C	750 °C
operational temperature of the secondary combustor	620 °C	650 °C
capacity	0.5 body h^{-1}	0.5 body h^{-1}
auxiliary fuel	diesel (40 L h^{-1})	diesel (40 L h^{-1})
air pollution control devices		bag filter
temperature of the stack flue gas	300 °C	200 °C
height of the stack	5 m	6 m

FIGURE 1. Congener profiles of seventeen 2,3,7,8 chlorinated substituted PCDD/Fs containing in the stack flue gases of C1 and C2.

profiles of PCDD/Fs in the stack flue gases of these two crematories were presented and compared. PCDD/F emission factors and emission rates for both crematories were determined. Furthermore, to assess the influence of PCDD/F emissions from a crematory process to the surrounding environment, the PCDD/F concentrations in the atmosphere of the background area, rural area, residential area, urban area, and industrial sites were measured and were compared with the results that were obtained from the vicinity of a crematory.

TABLE 2. Mean PCDD/Fs Emission Factors for the Two Investigated Crematories of C1 and C2

	C1 ($n = 3$)		C2 ($n = 3$)	
PCDD/Fs	mean g body^{-1}	RSD %	mean g body^{-1}	RSD %
2,3,7,8-TeCDD	2.26	74	0.969	61
1,2,3,7,8-PeCDD	4.01	80	1.70	46
1,2,3,4,7,8-HxCDD	1.59	78	0.738	31
1,2,3,6,7,8-HxCDD	1.76	48	0.894	53
1,2,3,7,8,9-HxCDD	2.19	87	0.964	52
1,2,3,4,6,7,8-HpCDD	3.78	93	0.977	82
OCDD	2.12	101	0.552	43
2,3,7,8-TeCDF	12.9	66	6.39	48
1,2,3,7,8-PeCDF	10.3	80	3.71	46
2,3,4,7,8-PeCDF	10.7	71	4.82	45
1,2,3,4,7,8-HxCDF	5.24	94	2.73	49
1,2,3,6,7,8-HxCDF	5.90	95	2.34	42
1,2,3,7,8,9-HxCDF	0.358	19	0.242	66
2,3,4,6,7,8-HxCDF	4.00	89	2.19	44
1,2,3,4,6,7,8-HpCDF	6.05	104	3.06	55
1,2,3,4,7,8,9-HpCDF	0.766	77	0.490	65
OCDF	0.698	58	0.495	60
total PCDD/Fs (μg body^{-1})	74.6	81	33.3	43
total I-TEQ (μg I-TEQ body^{-1})	13.6	75	6.11	45

Material and Methods

Investigating PCDD/F Emissions from the Cremation Process. Two crematories (denoted as C1 and C2) located in southern Taiwan were selected in this study. Both crematories had the same capacity (= 0.5 body h^{-1}) and were equipped with two combustion chambers (i.e., a primary and a secondary combustion chamber) operated under very similar combustion conditions (= 730 °C and 620 °C and 750 °C and 650 °C in the primary and secondary combustion chambers for C1 and C2, respectively). Both crematories used diesel as their auxiliary fuel with the same feeding rate specified at 40 L h^{-1}. C1 was not equipped with any air pollution control device, but C2 was facilitated with one bag filter with an automatic shaking control unit. Both crematories were equipped with a low stack (stack height = 5 and 6 m for C1 and C2, respectively). Basic information for these two crematories is described in more detail in Table 1.

Three PCDD/F samples were collected from the stack flue gas for each of the two selected crematories according to the U.S. EPA modified Method 23. The sampling train adopted in this study is comparable with that specified by the U.S. EPA Modified Method 5. Prior to sampling, XAD-2 resin was spiked with PCDD/F surrogate standards prelabeled with isotopes. The sampling time for each stack flue gas sample was ~2.5 h. To ensure the free contamination of the collected samples, one trip blank and one field blank were also taken when the field sampling was conducted.

Assessing PCDD/F Emissions from the Crematory to the Surrounding Environment. To assess the influence of PCDD/F emissions from a crematory on the surrounding environment, four ambient air samples were collected from two sampling sites at the vicinity of C1. The above two sampling sites were known with the maximum ground concentrations of C1 determined by using the Industrial Source Complex Short-Term Model (ISCST3). Yet, it is true that the accuracies regarding the use of ISCST3 on estimating maximum ground PCDD/F concentrations might be affected by the deposition of particle-phase PCDD/Fs and the decay of PCDD/Fs in the air due to photolysis (10). In this study, because C1 was known with a low stack and hence the duration of emitted PCDD/Fs transported from the stack to the ground level could be quite short. Based on this, we assumed both the deposition of particle-phase PCDD/Fs and photolysis of PCDD/Fs during the transportation period were

TABLE 3. PCDD/F Emission Factors for the Crematory Obtained from Different Studies

emission factors	reference	annotation
13.6 μg I-TEQ body^{-1}	this study	none of APCD
6.11 μg I-TEQ body^{-1}	this study	bag filter as its APCD
2.4−80 μg I-TEQ body^{-1}	(7)	
6 μg I-TEQ body^{-1}	(3)	
28 μg I-TEQ body^{-1}	(10)	derived from one crematory in Germany
70−80 μg I-TEQ body^{-1}	(10)	derived from two crematories in UK
0.5 μg I-TEQ body^{-1}	(10)	derived from one crematory in U.S.
9.2 μg I-TEQ body^{-1} (regarding ND as 0)		
11 μg I-TEQ body^{-1} (regarding ND as the half value of the detection limit)	(4)	derived from 10 crematories in Japan
3.97 μg I-TEQ body^{-1} (arithmetical mean)	(5)	derived from 17 crematories in Japan
1.83 μg I-TEQ body^{-1} (geometric mean)		

negligible. In this study, the dispersion parameters (such as atmospheric stability and mixing height) adopted in ISCST3 were determined based on the hourly meteorological data. According to the data obtained from the local weather bureau, we found that the prevailing winds were NW and N with their average wind speeds of 4.2 m s^{-1} and 2.4 m s^{-1}, respectively. Based on this, two sampling sites situated at the downwind sites of C1 with distances 80 m (SE) and 65 m (E) away from the stack were then determined in this study. For comparisons, five sampling sites were also selected for collecting ambient air samples during the same time. The first sampling site, the Keng-Ting National Park ($n = 2$), was situated at the southern end of Taiwan. This site was selected because it was far away from all possible pollution sources and hence its PCDD/F concentration could be regarded as the background level. The second sampling site was located at the Taitung county ($n = 4$), the least industrialized area in Taiwan, and hence was thought to be representative for the rural area. The other thee sampling sites were selected from the residential area ($n = 2$), urban area ($n = 4$), and industrial area ($n = 4$) of the same city as where C1 was located (i.e., the Kauhsiung city, the most industrialized area in Taiwan).

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 $\sim$0.225 m^3 min^{-1}. Each sample was collected continuously on three consecutive days (sampling volume = $\sim$972 m^3). The PS-1 sampler was equipped with a quartz-fiber filter for sampling particle-phase PCDD/Fs and followed by a glass cartridge for sampling gas-phase PCDD/Fs, respectively. A known amount of surrogate standard was spiked to the glass cartridge in the laboratory prior to the field sampling being conducted.

Sample Analysis. Analyses of stack flue gas and ambient air samples followed the U.S. EPA modified method 23 and EPA Reference Method T09A, respectively. All chemical analyses were carried out by the Super Micro Mass Research and Technology Center in Cheng Shiu Institute of Technology—the only accredited laboratory in Taiwan for PCDD/F analyses. 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 $\sim$1 mL, then transferred to a vial, and then further concentrated to nearly dryness by using a nitrogen stream. Prior to PCDD/F analysis, the standard solution was added to the sample to ensure the recovery during the analysis process.

Two high-resolution gas chomatographs/high-resolution mass spectrometers (HGC/HMS) were used for PCDD/Fs analyses (one for analyzing stack flue gas samples and the other for ambient air samples). The HGC (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) and with a splitless injection. The oven temperature program was set according to the following: begin at 150 °C (held for 1 min), then increase at 30 °C min^{-1} to 220 °C (held for 12 min), then increase at 1.5 °C min^{-1} to 240 °C (held for 5 min), and finally increase at 1.5 °C min^{-1} to 310 °C (held for 20 min). Helium was used as the carrier gas. The HMS (Micromass Autospec Ultima, Manchester, UK) mass spectrometer was equipped with a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring (SIM) was used with resolving power at 10 000. The electron energy and source temperature were specified at 35 eV and 250 °C, respectively.

Results and Discussion

Characteristics of PCDD/F Emissions from Crematories. The congener profiles of the 2,3,7,8-substituted PCDD/Fs were selected as the signatures of the crematory emissions. Each selected congener was normalized by reference to the total weight of all 2,3,7,8-congeners. Figure 1 shows the congener profiles of the seventeen 2,3,7,8 chlorinated substituted PCDD/Fs (mean±SD) detected from the stack flue gases of C1 and C2. The top three congeners for both crematories were 2,3,7,8-TeCDF, 2,3,4,7,8-PeCDF, and 1,2,3,7,8-PeCDF. The above results were quite similar to the congener profiles obtained from 10 crematories in Japan (4). Indeed, the involved incinerating materials (such as the weight of the dead body, sex, type of coffin, and other accompanied funeral materials, etc.) in crematories of the above-mentioned study might be different for us. However, the above-mentioned study has concluded that the incinerating materials might have a very limited effect on the congener profiles (4). The results obtained from this study further support the plausibility of the above inference.

PCDD/F Emissions from Crematories. Table 2 shows the mean emission factors for C1 (no air pollution control device was installed) and C2 (equipped with a bag filter) were 74.6 and 33.3 μg body^{-1} (in terms of total PCDD/F emissions) and 13.6 and 6.11 μg I-TEQ body^{-1} (in terms of total I-TEQ emissions), respectively. It is known that both C1 and C2 had quite comparable operation conditions (see Table 1). Therefore, it was assumed that both crematories might result in similar PCDD/F emissions during the cremation process. Based on this, the removal efficiency of the bag filter could be determined according to the following equation

$$\text{removal efficiency } (\eta; \%) = (A - B)/A \times 100\%$$

where A and B were the mean emission factors of C1 and C2, respectively. Based on this, it can be found that the removal efficiencies of the bag filter on the total PCDD/F emission and the total PCDD/F I-TEQ emission were 55.4% (= (74.6−33.3)/74.6) and 55.1% (= (13.6−6.11)/13.6), respectively. The
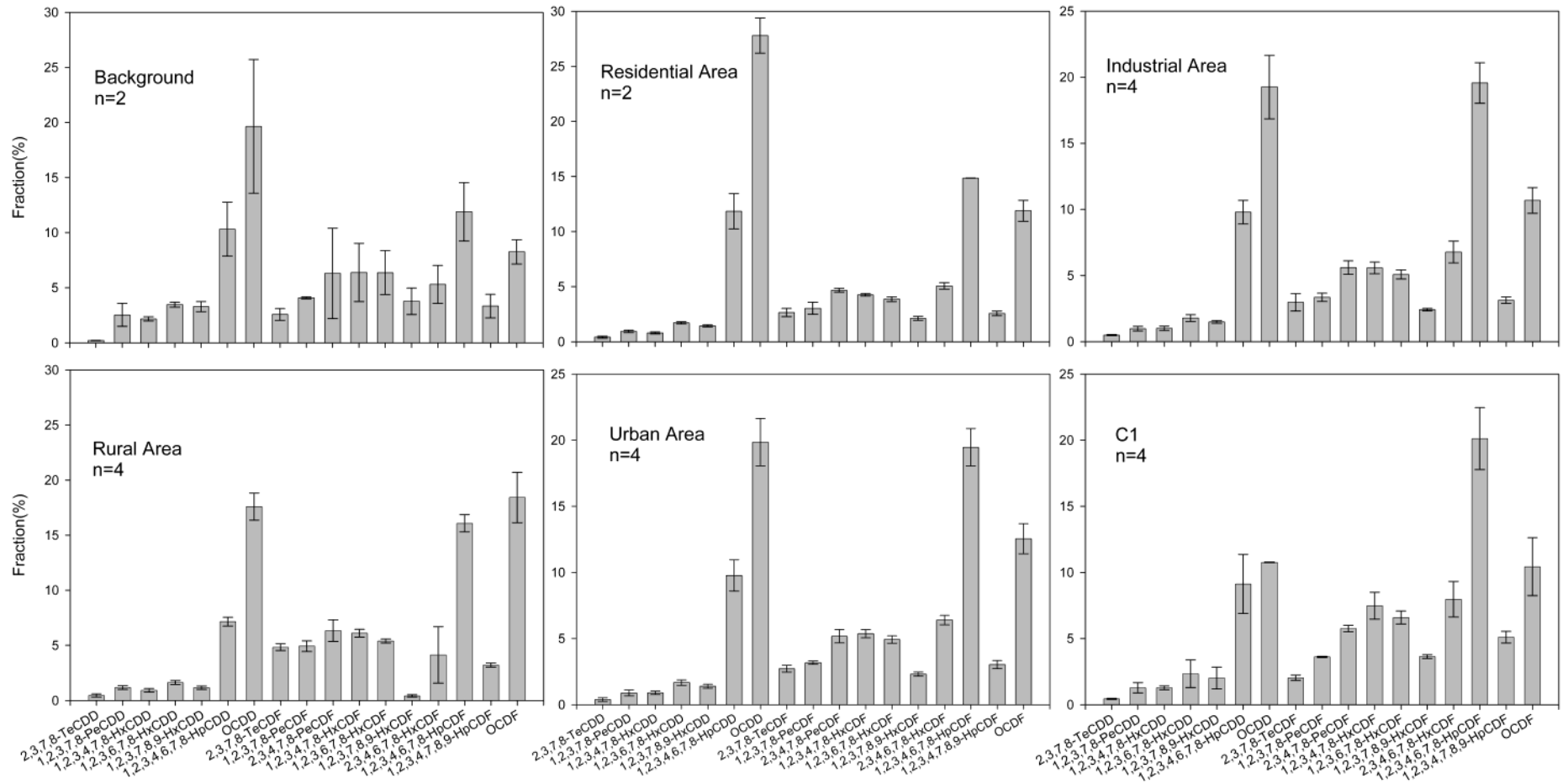
FIGURE 2. Congener profiles of seventeen 2,3,7,8 chlorinated substituted PCDD/Fs in ambient air.

TABLE 4. Mean PCDD/F Concentrations Found in Ambient Air of the Background, Rural Area, Residential Area, Urban Area, Industrial Area, and the Vicinity of C1

PCDD/Fs	background (n = 2) mean pg Nm^{-3}	RSD %	rural area (n = 4) mean pg Nm^{-3}	RSD %	residential area (n = 2) mean pg Nm^{-3}	RSD %	urban area (n = 4) mean pg Nm^{-3}	RSD %	industrial area (n = 4) mean pg Nm^{-3}	RSD %	C1 (n = 4) mean pg Nm^{-3}	RSD %
2,3,7,8-TeCDD	0.000	47	0.002	38	0.004	11	0.005	20	0.013	36	0.029	26
1,2,3,7,8-PeCDD	0.002	4	0.004	13	0.008	3	0.013	24	0.027	40	0.088	62
1,2,3,4,7,8-HxCDD	0.002	29	0.003	27	0.007	2	0.013	30	0.027	42	0.085	46
1,2,3,6,7,8-HxCDD	0.002	31	0.005	24	0.015	4	0.024	28	0.049	43	0.166	75
1,2,3,7,8,9-HxCDD	0.002	24	0.004	26	0.012	3	0.020	27	0.041	41	0.142	71
1,2,3,4,6,7,8-HpCDD	0.007	14	0.023	19	0.101	23	0.142	29	0.269	45	0.626	58
OCDD	0.013	7	0.057	19	0.237	16	0.283	18	0.516	41	0.708	36
2,3,7,8-TeCDF	0.002	56	0.016	17	0.022	4	0.039	10	0.076	18	0.136	45
1,2,3,7,8-PeCDF	0.003	39	0.016	16	0.026	8	0.045	16	0.088	31	0.237	35
2,3,4,7,8-PeCDF	0.005	91	0.020	20	0.040	6	0.074	18	0.150	36	0.381	40
1,2,3,4,7,8-HxCDF	0.005	73	0.020	13	0.036	7	0.077	19	0.151	40	0.480	23
1,2,3,6,7,8-HxCDF	0.005	65	0.017	14	0.033	5	0.070	18	0.138	41	0.428	29
1,2,3,7,8,9-HxCDF	0.003	6	0.001	35	0.018	2	0.033	14	0.067	45	0.241	39
2,3,4,6,7,8-HxCDF	0.004	66	0.014	67	0.043	4	0.092	20	0.188	48	0.540	51
1,2,3,4,6,7,8-HpCDF	0.009	57	0.051	11	0.126	10	0.279	20	0.538	45	1.30	25
1,2,3,4,7,8,9-HpCDF	0.002	5	0.010	12	0.022	18	0.043	21	0.085	43	0.330	27
OCDF	0.006	24	0.058	5	0.100	2	0.178	12	0.288	41	0.660	15
total PCDD/Fs	0.070	37	0.320	14	0.849	10	1.43	17	2.71	41	6.57	36
PCDDs	0.028	13	0.097	19	0.383	16	0.500	21	0.941	41	1.84	51
PCDFs	0.042	53	0.223	13	0.466	5	0.930	16	1.77	41	4.73	30
PCDDs/PCDFs ratio	0.742	42	0.432	10	0.819	11	0.536	9	0.535	8	0.377	23
total I-TEQ (pg I-TEQ/ Nm^3)	0.006	58	0.023	17	0.050	4	0.093	16	0.190	38	0.521	41

above results were similar to the results that obtained by Giugliano et al. (*11*). In their study, they measured PCDD/F concentrations at both inlet and outlet of the fabric filter of a municipal solid waste incinerator. Although the removal efficiency on total particulates was as high as >99.9%, removal efficiencies on total PCDD/F emissions and total PCDD/F I-TEQ emissions were ~45% and ~64%, respectively. Based on this, it is concluded that the removal efficiency of the bag filter on PCDD/F emissions was inadequate.

Table 3 shows total PCDD/F I-TEQ emission factors of crematories reported by other research. It can be found that the emission factors found in this study were quite comparable with that found by Takeda et al. (*4*). However, the wide range of PCDD/F emission factors found in other studies indicating that to generalize a universal emission factor for the cremation process might be not possible at this stage. Obviously, this could be due to the intrinsic differences in the types of combustion chamber, the operating conditions, and the types of air pollution control devices among various crematories.

Currently, a total of 32 crematories have been established in the Taiwan area. All crematories are facilitated with a low stack. Half of them were equipped with no air pollution control device, and the rest of them were only equipped with either a bag filter or a cyclone. According to statistical data provided by the Ministry of the Interior, there were ~85 000 cremations (cremation ratio = 66.9%) in 2000. By directly adopting the emission factors of C1 and C2 (assuming C1 and C2 are representative to those crematories installed without and with air pollution control devices, respectively), this study yielded the total PCDD/F I-TEQ emission rate for all crematories was ~0.838 g I-TEQ yr^{-1}. Comparing the above results with other emission sources, we found the total emission from crematories accounted for ~227% and 112% of the emissions from medical waste incinerators (= 0.369 g I-TEQ yr^{-1}) (*12*) and municipal waste incinerators (= 0.750 g I-TEQ yr^{-1}) (*13*), respectively. Unlike what was found in Japan (i.e., crematory emissions accounted for only 0.13–0.29% of I-TEQ yr^{-1} of that emitted from municipal waste incinerators) (*5*), crematories in Taiwan did play a much more important role in PCDD/F emissions.

Significance of PCDD/F Emissions from Crematories on the Surrounding Environment. Figure 2 shows the congener profiles of PCDD/Fs of the background, rural area, residential area, urban area, industrial area, and the vicinity of C1, respectively. All six categories show that the most abundant congeners in the atmosphere were 1,2,3,4,-6,7,8- HpCDD, OCDD, 1,2,3,4,6,7,8- HpCDF, and OCDF, which were consistent with those found in other studies (*14–17*). Table 4 lists the mean PCDD/F concentrations for the above six categories as 0.070, 0.320, 0.849, 1.43, 2.71, and 6.57 pg Nm^{-3}, respectively, and the corresponding I-TEQ concentrations were 0.006, 0.023, 0.052, 0.093, 0.190, and 0.521 pg I-TEQ Nm^{-3}, respectively. In principle, the results obtained from this study (except for the concentration of the vicinity of C1) are similar to that found in Germany (e.g., rural area = 0.025–0.070 pg I-TEQ Nm^{-3}; urban area = 0.070–0.350 pg I-TEQ Nm^{-3}) (*18*). In this study, the mean I-TEQ concentration in the vicinity of C1 was ~86.8, 22.6, 10.0, 5.6, and 2.7 times higher than that of the background, rural area, residential area, urban area, and industrial area, respectively. The high I-TEG concentration found in the vicinity of C1 might be because the involved crematory (i.e., C1) had a low stack and was installed with no air pollution control devices.

Yet, it is true that PCDD/F emissions obtained from this study were on a time-weighted-average basis. It did not provide real-time variations on PCDD/F emissions. However, in this study we did find the contents of N_2, O_2, and CO_2 in stack flue gases of C1 and C2 during the sampling period were quite stable (N_2, O_2, and CO_2 concentrations = ~80.6%, 14.6%, 4.80% for C1 and = ~80.2%, 15.8%, and 3.90% for C2, respectively). At this stage, whether PCDD/F emissions were also as stable as the above compounds warrants the need for further investigation. Nevertheless, the results obtained from this study do indicate that the impact of PCDD/F emissions from crematories to the surrounding environment was quite significant. Therefore, it is concluded that a proper control strategy should be taken immediately in order to eliminate PCDD/F emissions from crematory sources.

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