



## PCDD/F Levels in the Stacks and Food of Fast Food Restaurants

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### ABSTRACT

Trace amounts of polychlorinated dibenzodioxins and dibenzofurans (PCDD/Fs) are ubiquitous in the environment. This study determined the levels of PCDD/Fs in the stacks and food of fast food restaurants. The results indicate that the dominant contribution of PCDD/Fs concentration in the stacks of fast food restaurants is PCDFs. 2,3,7,8-TeCDF concentration measured in the stacks of fast food restaurants is the highest among the seventeen investigated PCDD/F congeners, accounting for about 17.9% of the total concentration. For the PCDD/F content in food before and after frying, the OCDD concentration was the highest among the seventeen investigated PCDD/F congeners, accounting for 22.3–93.3% of the total concentration before frying and 82.6–91.8% of the total concentration after frying. The total PCDD/F concentration in the food of fast food restaurants decreases significantly after frying. The mean emission factor and emission rate for total PCDD/Fs from the stacks of fast food restaurants are 208 pg/L and 12400 pg/min, respectively.

**Keywords:** PCDD/Fs; Food; Restaurant; Stack; Emission factor; Emission rate.

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### INTRODUCTION

Polychlorinated dibenzodioxins and dibenzofurans (PCDD/Fs) are ubiquitous environmental contaminants. PCDDs and PCDFs are unwanted by-products in a variety of industrial and thermal processes. Human exposure to these compounds has been associated with adverse health outcomes (Wang *et al.*, 2009; Hu *et al.*, 2009; Chuang *et al.*, 2010a, b; Lin *et al.*, 2010; Wang *et al.*, 2010; Wu *et al.*, 2010a, b; Chen *et al.*, 2011; Chiu *et al.*, 2011; Offenberg *et al.*, 2011; Stone *et al.*, 2011; Yeh *et al.*, 2011). For the general population, food ingestion is the main route of exposure (WHO/ICPS, 1989). PCDD/PCDFs are persistent and bioaccumulate through the food chain. Many matrices contain PCDD/Fs, such as milk, eggs, meat, fishes, and animal feedstuff, have been investigated. The background levels of polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and polychlorinated biphenyls (PCBs) in these matrices can be used to estimate the typical dietary intakes for the general

population (Focant *et al.*, 2002). It is important to note that in these surveys, as well as in most reported investigations on the dietary intake of chemicals, food analyses were performed on uncooked/raw products. In an earlier study, the effect of broiling on the levels of PCDD/PCDFs and co-PCBs in ground beef (hamburger), bacon, and catfish were investigated (Schechter *et al.*, 1998). The PCDD/PCDF and co-PCB TEQ values decreased by approximately 50% for each portion after broiling.

To assess toxicologically relevant information on PCDD/Fs, the International Toxicity Equivalent (I-TEQ) values for PCDD/Fs were calculated using 17 2,3,7,8-chlorinated congeners and their International Toxicity Equivalency Factors (I-TEFs) (Kutz *et al.*, 1990). More recently, the World Health Organization (WHO) suggested modified TEFs in 1994 (TEFWHO-94), 1998 (TEFWHO-98), and 2005 (TEFWHO-05) (Ahlborg *et al.*, 1994; Van den Berg *et al.*, 1998, 2006). Although PCDD/F concentrations in many locations have been surveyed, few studies have focused on assessing the effects of PCDD/F pollutants found in homes and restaurants. In Taiwan, there are numerous fast food restaurants that cook fry food. This study determined the levels of PCDD/Fs in the stacks and food of fast food restaurants. Foods included small chicken nuggets, chicken nuggets, french fries, fish fillets, apple

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pies, and chicken fillets. The I-TEQ concentrations from the stacks and WHO-TEQ in the food are investigated in detail. The PCDD/F emission factor and emission rate from the stacks of fast food restaurants are also studied.

## EXPERIMENTAL

### *Sampling Strategy*

Three fast food restaurants in Taiwan were randomly selected for the study. The main cooking methods were deep frying and stewing. Types of cooking oil were vegetable oil, butter, and corn oil. Food oil usage rates (liters per lunch or dinner period) ranged from 11.2–16.3, with a mean of 13.4. The cooking time for lunch (or dinner) period ranged from 97–121 minutes, with a mean of 109 minutes. The mean of stack diameters (m) was 1.194. The mean stack outlet velocity was 4.12 m/s, and the mean stack inlet and outlet temperatures were 44°C and 36°C, respectively. The mean exhaust flow rate was 136.45 Nm<sup>3</sup>/min.

For stack sampling in fast food restaurants, each sample was collected on a 9–12 m high platform. The collected samples were composites of three subsamples obtained at each sampling site. We then transferred the samples to precleaned amber glass bottles and stored them at 4°C. Usually, extraction was carried out within 72 h. The PCDD/F samples were collected isokinetically from the stack flue gas of the selected incinerators according to U.S. EPA modified Method 23. The sampling train adopted in this study is comparable with that specified by U.S. EPA Modified Method 5. Prior to sampling, XAD-2 resin was spiked with PCDD/F surrogate standards pre-labeled with isotopes, including <sup>37</sup>C<sub>14</sub>-2,3,7,8-TCDD (tetrachlorodibenzo-p-dioxin), <sup>13</sup>C<sub>12</sub>-1,2,3,4,7,8-HxCDD (hexachlorinated dibenzo-p-dioxin), <sup>13</sup>C<sub>12</sub>-2,3,4,7,8-PeCDF (pentachlorinated dibenzofuran), <sup>13</sup>C<sub>12</sub>-1,2,3,4,7,8-HxCDF (hexachlorinated dibenzofuran), and <sup>13</sup>C<sub>12</sub>-1,2,3,4,7,8,9-HpCDF (heptachlorinated dibenzofuran). The recoveries of PCDD/F surrogate standards were 95–117%, and met the criteria of being within 70–130%. To ensure that the collected samples were not contaminated, one trip blank and one field blank were taken during the field sampling. Details are given in our previous work (Wang *et al.*, 2003a).

### *Analyses of PCDD/Fs*

Analyses of ambient air samples followed U.S. EPA modified Method 23 and U.S. EPA Reference Method T09A, respectively. All chemical analyses were carried out at the Super Micro Mass Research and Technology Center in Cheng Shiu University. Each collected sample was spiked with a known amount of the internal standard. After 24 h of extraction, the extract was concentrated, treated with concentrated sulfuric acid, and then subjected to a series of sample cleanup and fractionation procedures. The eluate was concentrated to ~1 mL, transferred to a vial, and then further concentrated to nearly dryness using a nitrogen stream. Prior to PCDD/F analysis, the standard solution was added to the sample to ensure recovery during the analysis process. A high-resolution gas chromatograph

(HRGC) and a high-resolution mass spectrometer (HRMS) were used for analyzing 17 PCDD/F species. The HRGC (Hewlett Packard 6970 Series gas, CA, USA) was equipped with a DB-5MS fused silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25 μm) (J&W Scientific, CA, USA) and splitless injection. Helium was employed as the carrier gas. The HRMS (Micromass Autospec Ultima, Manchester, UK) was equipped with a positive electron impact (EI+) source. The analyzer mode of selected ion monitoring (SIM) had a resolving power of 10000. The Electron energy and source temperature were 35 eV and 250°C, respectively. The toxic equivalent quantity of PCDD/Fs is given by I-TEQ = ΣXi × Ii, where I-TEQ denotes the international toxic equivalent quantity, Xi represents the concentration of PCDD/F congeners, and Ii is the international toxic equivalent factor of each PCDD/F congener (I-TEF) (NATO/CCMS, 1988).

## RESULTS AND DISCUSSION

### *Concentrations of PCDD/Fs Obtained from the Stacks of Fast Food Restaurants*

The individual concentrations of the 17 most toxic congeners of PCDD/PCDFs from the stacks of fast food restaurants are summarized in Table 1. The sum of PCDDs and PCDFs, the ratio PCDDs/PCDFs, and the TEQ for the stacks are also shown. The total PCDD/F concentrations from the stacks of fast food restaurants ranged from 87.4–94.2 pg/Nm<sup>3</sup>, with an average of 90.8 pg/Nm<sup>3</sup>. The total PCDDs/PCDFs ratios were 0.368–0.399, indicating that the main PCDD/F contribution from the stacks of fast food restaurants was PCDFs. The total I-TEQ concentrations from the stacks of fast food restaurants ranged from 8.16–8.75 pg I-TEQ/Nm<sup>3</sup>, with an average of 8.48 pg I-TEQ/Nm<sup>3</sup>. The mean total I-TEQ concentration of fast food restaurants is smaller than that of municipal solid waste incinerators (72.5 pg I-TEQ/Nm<sup>3</sup>) (Wang *et al.*, 2007), sinter plants (995–2060 pg I-TEQ/Nm<sup>3</sup> with selective catalytic reduction (SCR) and 3100 pg I-TEQ/Nm<sup>3</sup> without SCR) (Wang *et al.*, 2003b), electric arc furnaces (172 pg I-TEQ/Nm<sup>3</sup>) (Wang *et al.*, 2010), aluminum smelter plants (9020 pg I-TEQ/Nm<sup>3</sup>) (Chen *et al.*, 2004), crematories (322–2360 pg I-TEQ/Nm<sup>3</sup>) (Wang *et al.*, 2003a), and power plants (17 pg I-TEQ/Nm<sup>3</sup>) (Lin *et al.*, 2007). However, restaurants are close to daily human activity. We should thus pay more attention to PCDD/F emissions from restaurants. The TEQ PCDDs/PCDFs ratios of fast food restaurants are 0.227–0.238, indicating that the main toxicity source of PCDD/Fs was PCDFs.

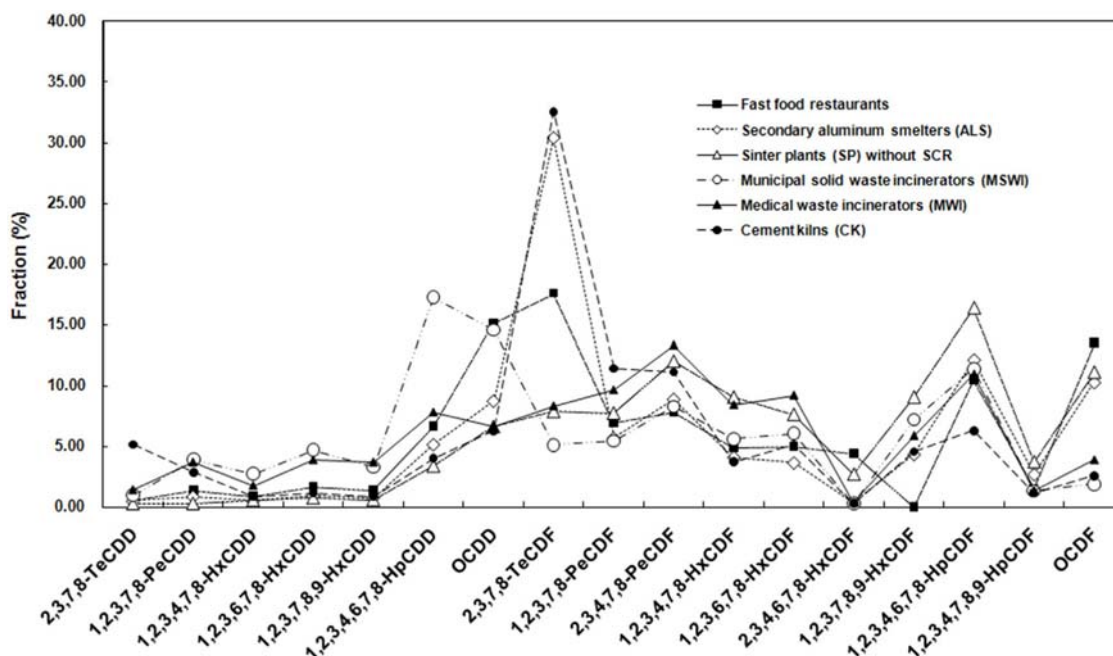
### *Congener Profiles of PCDD/Fs from the Stacks of Fast Food Restaurants*

Fig. 1 shows the congener profiles of 17 PCDD/F species from the stacks of fast food restaurants. The 2,3,7,8-TeCDF concentration was the highest among the seventeen investigated PCDD/F congeners, accounting for about 17.3–17.8% of the total concentration, with an average of 17.6%. OCDD was the second dominant among the seventeen investigated PCDD/F congeners, accounting

**Table 1.** Mean concentrations of PCDD/Fs from the stacks of fast food restaurants.

PCDD/Fs	Range	Average	RSD (%)	I-TEF
2,3,7,8-TeCDD	0.502–0.573	0.541	4.99	1
1,2,3,7,8-PeCDD	1.12–1.32	1.25	4.08	0.5
1,2,3,4,7,8-HxCDD	0.818–0.877	0.832	3.00	0.1
1,2,3,6,7,8-HxCDD	1.42–1.64	1.51	5.30	0.1
1,2,3,7,8,9-HxCDD	1.15–1.29	1.24	4.60	0.1
1,2,3,4,6,7,8-HpCDD	5.84–6.52	6.05	4.61	0.01
OCDD	12.4–14.8	13.7	6.41	0.001
2,3,7,8-TeCDF	15.1–16.6	16.0	3.64	0.1
1,2,3,7,8-PeCDF	6.08–6.57	6.32	3.05	0.05
2,3,4,7,8-PeCDF	6.83–7.34	7.09	2.91	0.5
1,2,3,4,7,8-HxCDF	4.27–4.69	4.47	4.03	0.1
1,2,3,6,7,8-HxCDF	4.20–4.99	4.56	6.36	0.1
2,3,4,6,7,8-HxCDF	3.72–4.24	4.03	5.21	0.1
1,2,3,7,8,9-HxCDF	ND <sup>a</sup>	NA <sup>a</sup>	NA	0.1
1,2,3,4,6,7,8-HpCDF	9.12–10.3	9.55	4.44	0.01
1,2,3,4,7,8,9-HpCDF	1.32–1.47	1.39	5.04	0.01
OCDF	12.0–12.8	12.3	2.59	0.001
PCDDs	23.5–26.9	25.2	4.75	
PCDFs	63.9–67.3	65.6	2.32	
PCDDs/PCDFs	0.368–0.399	0.383		
Total PCDD/Fs	87.4–94.2	90.8	2.83	
PCDDs pg I-TEQ/Nm <sup>3</sup>	1.53–1.68	1.598	3.94	
PCDFs pg I-TEQ/Nm <sup>3</sup>	6.62–7.07	6.885	2.70	
PCDDs/PCDFs	0.227–0.238	0.23		
Total PCDD/Fs (pg I-TEQ/Nm <sup>3</sup> )	8.16–8.75	8.483	2.88	

<sup>a</sup> ND: Not detectable; NA: Not available.



**Fig. 1.** Congener profiles of 17 PCDD/F species from the stacks of fast food restaurants and other sources.

for 14.2–15.8% of the total concentration, with an average of 15.1%. OCDF was the third dominant among the seventeen investigated PCDD/F congeners, accounting for 13.2–13.9% of the total concentration, with a mean value of 13.5%. Among the seventeen investigated PCDD/F

congeners from the stacks of fast food restaurants, 2,3,7,8-TeCDF, OCDD, and OCDF were the three dominant PCDD/Fs, accounting for about 46.6% of the total concentration. Fig. 1 also shows the congener profiles of 17 PCDD/F species for various sources (Wang *et al.*,

2003b; Chen *et al.*, 2004; Kao *et al.*, 2010). For fast food restaurants, secondary aluminum smelters (ALS), and cement kilns (CK), the 2,3,7,8-TeCDF emission rate was the highest among the seventeen investigated PCDD/F congeners, accounting for 17.6, 30.4, and 32.6% of the total concentrations, respectively. However, the highest among the seventeen investigated PCDD/F congeners in sinter plants (SP) without SCR was 1,2,3,4,6,7,8-HpCDF, accounting for 16.4% of the total concentration. 1,2,3,4,6,7,8-HpCDD was the highest among the seventeen investigated PCDD/F congeners in municipal solid waste incinerators (MSWI), accounting for 17.3% of the total concentration, and 2,3,4,7,8-PeCDF was the highest among the seventeen investigated PCDD/F congeners in medical waste incinerators (MWI), accounting for 13.3% of the total concentration.

#### **PCDD/F Content in the Food of Fast Food Restaurants before Frying**

The individual content of the 17 most toxic congeners of PCDD/PCDFs from the food of fast food restaurants before frying is summarized in Table 2. The total PCDD/F concentrations in the food of fast food restaurants before frying were 13.91, 7.11, 25.29, 27.00, 11.74, and 14.95 pg/g-l.w.(lipid) for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively. Fish fillets had the highest PCDD/F content, and chicken nuggets had the lowest. The ratios of

PCDDs/PCDFs were 5.34–23.45 for small chicken nuggets, chicken nuggets, french fries, apple pies, and chicken fillets, indicating that the main contribution of their PCDD/F content before frying was PCDDs. However, the ratio of PCDDs/PCDFs concentrations was 0.85 for fish fillets, indicating that the main contribution of PCDD/Fs for fish fillets before frying was PCDFs.

The total WHO-TEQ in the food of fast food restaurants before frying were 0.1184–2.4940 pg WHO/g-l.w.(lipid). The WHO PCDDs/PCDFs ratios for small chicken nuggets, french fries, and apple pie were 1.42, 1.13, and 1.096, respectively, indicating that their main toxicity source of WHO-TEQ before frying was PCDDs. However, the WHO PCDDs/PCDFs ratio for chicken fillets was 0.775, indicating that its main toxicity source of WHO-TEQ before frying was PCDFs.

#### **Congener Profiles of PCDD/Fs in the Food of Fast Food Restaurants before Frying**

Fig. 2 shows the congener profiles of 17 PCDD/F species from various raw food at fast food restaurants. For all of the food before frying, the OCDD concentration was the highest among the seventeen investigated PCDD/F congeners, accounting for about 88.4, 78.3, 93.3, 22.3, 76.9, and 88% of the total content for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively. 1,2,3,4,6,7,8-HpCDD was the second dominant among the seventeen investigated PCDD/F

**Table 2.** PCDD/F content in the food before frying (pg/g-l.w.(lipid)).

PCDD/Fs	Small chicken nuggets	Chicken nuggets	French fries	Fish fillets	Apple pies	Chicken fillets
2,3,7,8-TeCDD	0.0159	0.0130	0.0176	0.256	0.0345	0.00780
1,2,3,7,8-PeCDD	0.0305	0.0404	0.0513	0.744	0.101	0.0227
1,2,3,4,7,8-HxCDD	0.0333	0.0327	0.0444	0.644	0.0870	0.0197
1,2,3,6,7,8-HxCDD	0.0659	0.0927	0.0469	0.681	0.0920	0.0557
1,2,3,7,8,9-HxCDD	0.0491	0.0598	0.0760	1.10	0.149	0.0617
1,2,3,4,6,7,8-HpCDD	0.701	0.522	0.415	2.93	0.396	0.733
OCDD	12.3	5.57	23.6	6.01	9.03	13.2
2,3,7,8-TeCDF	0.0649	0.080	0.0644	1.31	0.104	0.0806
1,2,3,7,8-PeCDF	0.0452	0.105	0.0495	0.719	0.0970	0.0455
2,3,4,7,8-PeCDF	0.0443	0.0857	0.0620	0.900	0.122	0.0717
1,2,3,4,7,8-HxCDF	0.0399	0.0850	0.0648	0.941	0.127	0.0576
1,2,3,6,7,8-HxCDF	0.0398	0.0549	0.105	1.53	0.206	0.0466
2,3,4,6,7,8-HxCDF	0.0586	0.0419	0.155	2.24	0.303	0.0685
1,2,3,7,8,9-HxCDF	0.0267	0.0184	0.0704	1.02	0.138	0.0312
1,2,3,4,6,7,8-HpCDF	0.113	0.0917	0.105	1.52	0.205	0.112
1,2,3,4,7,8,9-HpCDF	0.0411	0.0284	0.108	1.57	0.213	0.0480
OCDF	0.248	0.196	0.251	2.87	0.339	0.332
PCDDs	13.19	6.33	24.26	12.37	9.89	14.06
PCDFs	0.72	0.79	1.03	14.62	1.85	0.89
PCDDs/PCDFs	18.26	8.03	23.45	0.85	5.34	15.71
Total PCDD/Fs	13.91	7.11	25.29	27.00	11.74	14.95
PCDDs pg WHO/g-l.w.(lipid)	0.0695	0.0777	0.0921	1.2729	0.1727	0.0529
PCDFs pg WHO/g-l.w.(lipid)	0.0489	0.0774	0.0816	1.2211	0.1576	0.0682
PCDDs/PCDFs	1.420	1.004	1.130	1.042	1.096	0.775
Total PCDD/Fs (pg WHO/g-l.w.(lipid))	0.1184	0.1551	0.1737	2.4940	0.3302	0.1211

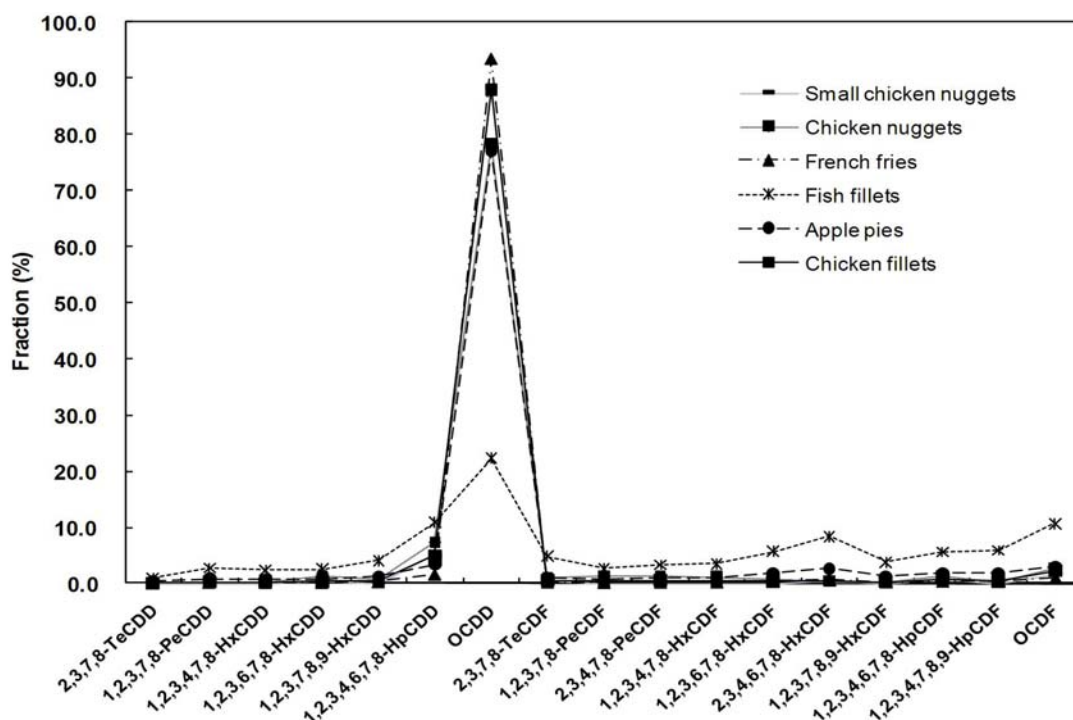


Fig. 2. Congener profiles of 17 PCDD/F species from various types of raw food at fast food restaurants.

congeners, accounting for about 5.0, 7.3, 1.6, 10.9, 3.4, and 4.9% of the total content for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively. OCDF was the third dominant among the seventeen investigated PCDD/F congeners, accounting for about 1.8, 2.8, 1.0, 10.6, 2.9, and 2.2% of the total content for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively.

#### PCDD/F Content in the Food of Fast Food Restaurants after Frying

Table 3 summarizes the individual concentrations of the 17 most toxic congeners of PCDD/PCDFs, the sum of PCDDs and PCDFs, the ratio of PCDDs/PCDFs, the total I-TEQ (given in pg I-TEQ/Nm<sup>3</sup>), and the total WHO-TEQ (given in pg WHO-TEQ/Nm<sup>3</sup>) in samples of cooked food. The total PCDD/F concentrations in the food of fast food restaurants after frying were 11.68, 6.20, 12.36, 11.54, 10.16, and 11.43 pg/g-l.w.(lipid) for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively. French fries had the highest PCDD/F content, and chicken nuggets had the lowest. The ratios of PCDDs/PCDFs were 10.58–23.71 for all the food after frying, indicating that the main contribution of PCDD/Fs for all the food after frying was PCDDs.

The total WHO-TEQ in the food of fast food restaurants after frying were 0.0704–0.1473 pg WHO/g-l.w.(lipid). The WHO PCDDs/PCDFs ratios for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets were 1.274, 1.176, 1.206, 1.148, 1.228, and 1.100, respectively, indicating that their main toxicity source of WHO-TEQ after frying was PCDDs.

#### Congener Profiles of PCDD/Fs in the Food of Fast Food Restaurants after Frying

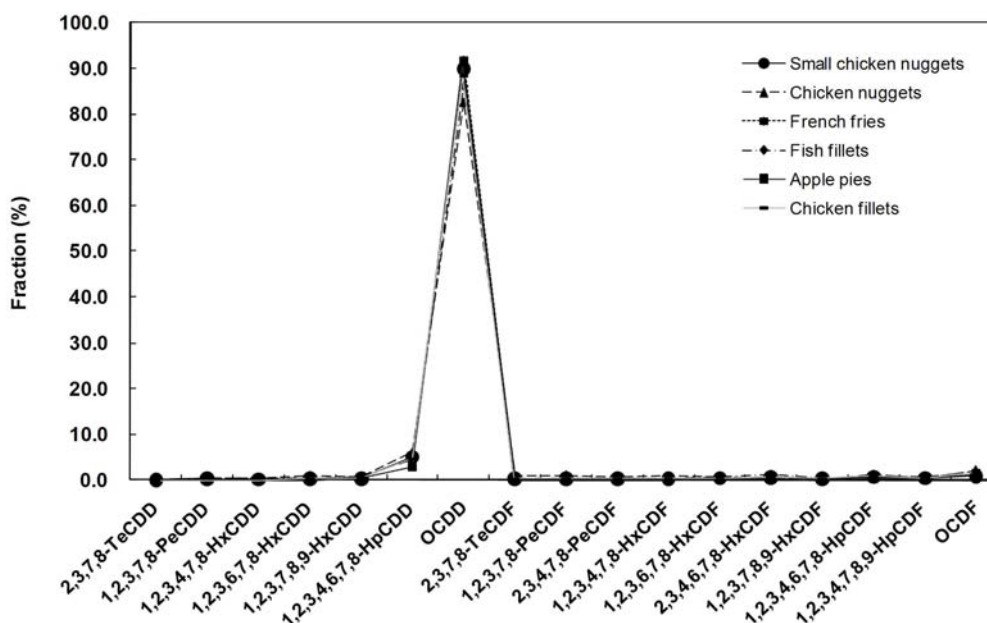
Fig. 3 shows the congener profiles of 17 PCDD/F species from various cooked food at fast food restaurants. Similar to food at fast food restaurants before frying, the OCDD concentration was the highest among the seventeen investigated PCDD/F congeners for all food after frying, accounting for about 89.9, 82.6, 91.8, 86.1, 91.4, and 88.2% of the total content for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively. 1,2,3,4,6,7,8-HpCDD was the second dominant among the seventeen investigated PCDD/F congeners, accounting for about 5.1, 6.1, 2.9, 4.6, 2.9, and 4.6% of the total content for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively. OCDF was the third dominant among the seventeen investigated PCDD/F congeners, accounting for about 0.8, 2.2, 1.1, 1.8, 1.5, and 1.7% of the total content for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively.

#### Comparison of PCDD/F Content and Congener Profiles of PCDD/Fs before and after Frying in the Food of Fast Food Restaurants

From Table 2 and Table 3, all six types of food have smaller total PCDD/F concentrations after frying. The total PCDD/F concentrations for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively, after frying were 84.0, 87.1, 48.9, 42.8, 86.5, and 76.5% of those before frying. It is likely that a certain amount of fat became attached to the food after frying, significantly decreasing the PCDD/F content.

**Table 3.** PCDD/F content in the food after frying (pg/g-l.w.(lipid)).

PCDD/Fs	Small chicken nuggets	Chicken nuggets	French fries	Fish fillets	Apple pies	Chicken fillets
2,3,7,8-TeCDD	0.00771	0.0105	0.00846	0.0149	0.00687	0.00903
1,2,3,7,8-PeCDD	0.0225	0.0294	0.0246	0.0434	0.0200	0.0263
1,2,3,4,7,8-HxCDD	0.0194	0.0198	0.0213	0.0376	0.0173	0.0228
1,2,3,6,7,8-HxCDD	0.0356	0.0583	0.0236	0.0397	0.0183	0.0415
1,2,3,7,8,9-HxCDD	0.0333	0.0478	0.0450	0.0644	0.0447	0.0461
1,2,3,4,6,7,8-HpCDD	0.591	0.377	0.361	0.525	0.295	0.531
OCDD	10.5	5.12	11.3	9.94	9.28	10.1
2,3,7,8-TeCDF	0.0270	0.0570	0.0254	0.0475	0.0206	0.0361
1,2,3,7,8-PeCDF	0.0235	0.0651	0.0238	0.0419	0.0193	0.0336
2,3,4,7,8-PeCDF	0.0272	0.0503	0.0298	0.0525	0.0242	0.0413
1,2,3,4,7,8-HxCDF	0.0291	0.0511	0.0311	0.0549	0.0255	0.0351
1,2,3,6,7,8-HxCDF	0.0460	0.0356	0.0505	0.0890	0.0410	0.0539
2,3,4,6,7,8-HxCDF	0.0677	0.0316	0.0743	0.131	0.0603	0.0793
1,2,3,7,8,9-HxCDF	0.0308	0.0144	0.0338	0.0596	0.0275	0.0361
1,2,3,4,6,7,8-HpCDF	0.0764	0.0739	0.0710	0.111	0.0606	0.0963
1,2,3,4,7,8,9-HpCDF	0.0475	0.0222	0.0521	0.0918	0.0423	0.0556
OCDF	0.0975	0.134	0.142	0.203	0.150	0.199
PCDDs	11.21	5.66	11.83	10.66	9.69	10.76
PCDFs	0.47	0.54	0.53	0.88	0.47	0.67
PCDDs/PCDFs	23.71	10.58	22.17	12.09	20.56	16.16
Total PCDD/Fs	11.68	6.20	12.36	11.54	10.16	11.43
PCDDs pg WHO/g-l.w.(lipid)	0.0460	0.0569	0.0468	0.0787	0.0388	0.0527
PCDFs pg WHO/g-l.w.(lipid)	0.0361	0.0483	0.0388	0.0686	0.0316	0.0479
PCDDs/PCDFs	1.274	1.176	1.206	1.148	1.228	1.100
Total PCDD/Fs (pg WHO/g-l.w.(lipid))	0.0820	0.1052	0.0857	0.1473	0.0704	0.1006

**Fig. 3.** Congener profiles of 17 PCDD/F species from various types of cooked food at fast food restaurants.

Moreover, the total WHO-TEQ PCDD/F concentrations for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively, after frying were 69.2, 67.8, 49.3, 5.90, 21.3, and 83.1% of those before frying.

Few studies investigated 17 kinds of PCDD/F congener before and after frying at fast food restaurants. In our study, OCDD, 1,2,3,4,6,7,8-HpCDD, and OCDF were the three dominant congeners before and after frying (Fig. 2 and Fig. 3). The OCDD congeners for small chicken nuggets,

chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively, after frying were 85.4, 92, 48, 165, 103, and 76.7% of those before frying; the 1,2,3,4,6,7,8-HpCDD congeners for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively, after frying were 84.3, 72.1, 86.9, 17.9, 74.5, and 72.4% of those before frying; the OCDF congeners for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively, after frying were 39.2, 68.1, 56.5, 7.10, 44.2, and 59.8% of those before frying. Although the OCDD congeners for fish fillets after frying were 1.65 times larger than those before frying, the other 16 congeners for fish fillet after frying were 3.6–17.9% of those before frying. Although the OCDD congeners for apple pies after frying were 1.028 times larger than those before frying, the other 16 congeners for apple pies after frying were 19.9–74.5% of those before frying. Accordingly, the sum of 17 PCDD/F species from six kinds of cooked food at fast food restaurants is smaller than that of raw food.

For small chicken nuggets, the seven kinds of PCDD congeners (2,3,7,8-TeCDDs, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDDs, 1,2,3,6,7,8-HxCDDs, 1,2,3,7,8,9-HxCDDs, 1,2,3,4,6,7,8-HpCDDs, OCDD) after frying were 48.3, 73.6, 58.5, 54.0, 67.8, 84.3, and 85.4% of those before frying. The total PCDD content after frying was 85% of that before frying. The ten kinds of PCDF congener after frying were 39–115.6% of those before frying. The total PCDF content after frying was 65.5% of that before frying. For total PCDDs and PCDFs of the six types of food, the total PCDD concentrations after frying for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively, were 85.0, 89.5, 48.8, 86.2, 98.0, and 76.6% of those before frying. The total PCDF concentrations after frying for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively, were 65.5, 67.9, 51.6, 6.0, 25.4, and 74.5% of those before frying. Our findings imply that there was a notable decrease in the total PCDD and PCDF concentrations after frying.

Table 4 summarizes the fat content and WHO-TEF of

the six types of food before and after frying in fast food restaurants. All six types of food exhibit larger fat content after frying, indicating that some cooking oil is present in the food after frying. Small chicken nuggets, chicken nuggets, apple pies, and chicken fillets exhibit smaller WHO-TEF values after frying. The WHO-TEF values of small chicken nuggets and chicken fillets meet the criterion of the Department of Health (DOH) in Taiwan (Taiwan DOH, 2009).

#### PCDD/Fs Emission Factor and Emission Rate

PCDD/F samples collected from the stacks of fast food restaurants were associated with different food oil consumption rates, cooking times, stack outlet velocities, and stack diameters. Thus, the PCDD/Fs emission factor was calculated according to the following equation (Li et al., 2003):

$$EF_{\text{PCDD/Fs}} = (\text{PCDD/Fs concentration} \times [(1/4) \times \pi \times d^2] \times v \times 60 \times t \times 10^{-3} / CR_{\text{food-oil}}) \quad (1)$$

where  $d$ ,  $v$ ,  $t$ , and  $CR_{\text{food-oil}}$  are, respectively, the stack diameter (in meters), stack outlet velocity (in meters per second), cooking time (in minutes per lunch period or dinner period), and food oil consumption rate (in liters of food oil per lunch or dinner period). Table 5 summarizes the PCDD/F emission factor (pg per liter of used cooking oil) at fast food restaurants. The emission factors for PCDDs and PCDFs, respectively, from the stacks of fast food restaurants ranged from 54.0–61.7 pg/L and 147–155 pg/L, with an average of 57.8 pg/L and 151 pg/L. The emission factors for total PCDD/Fs from the stacks of fast food restaurants ranged from 201–216 pg/L, with an average of 208 pg/L.

The PCDD/Fs emission rate was calculated using:

$$ER_{\text{PCDD/Fs}} = (\text{PCDD/Fs concentration}) \times V \quad (2)$$

where  $V$  ( $\text{Nm}^3/\text{min}$ ) is the exhaust flow rate from the stack of fast food restaurants. Table 6 summarizes the PCDD/F emission rate (pg per minute) at fast food restaurants. The emission rates for PCDDs and PCDFs, respectively, from

**Table 4.** Fat content and WHO-TEF of raw and cooked food in fast food restaurants.

Food		Fat content (%)	WHO-TEF	Standard of Taiwan DOH <sup>a</sup>
Small chicken nuggets	raw	15.26	1.81	2
	cooked	16.39	1.34	2
Chicken nuggets	raw	19.76	3.06	2
	cooked	26.4	2.78	2
French fries	raw	6.57	1.14	---
	cooked	16.52	1.41	---
Fish fillets	raw	0.53	1.32	---
	cooked	9.32	1.37	---
Apple pies	raw	3.94	1.30	---
	cooked	12.66	0.89	---
Chicken fillets	raw	11.75	1.42	2
	cooked	13.3	1.34	2

<sup>a</sup> The regulated standard of Taiwan Department of Health (DOH).

**Table 5.** PCDD/F emission factor (pg per liter of used cooking oil) at fast food restaurants.

PCDD/Fs	Range	Average	STD
2,3,7,8-TeCDD	1.15–1.32	1.24	0.062
1,2,3,7,8-PeCDD	2.75–3.04	2.87	0.117
1,2,3,4,7,8-HxCDD	1.88–2.01	1.91	0.057
1,2,3,6,7,8-HxCDD	3.27–3.77	3.46	0.184
1,2,3,7,8,9-HxCDD	2.64–2.96	2.86	0.130
1,2,3,4,6,7,8-HpCDD	13.4–15.0	13.9	0.641
OCDD	28.5–34.0	31.5	2.015
2,3,7,8-TeCDF	34.6–38.1	36.7	1.338
1,2,3,7,8-PeCDF	13.9–15.1	14.5	0.443
2,3,4,7,8-PeCDF	15.7–16.9	16.3	0.472
1,2,3,4,7,8-HxCDF	9.81–10.8	10.3	0.413
1,2,3,6,7,8-HxCDF	9.63–11.4	10.5	0.666
2,3,4,6,7,8-HxCDF	8.54–9.73	9.24	0.482
1,2,3,7,8,9-HxCDF	ND	NA	NA
1,2,3,4,6,7,8-HpCDF	20.9–23.5	21.9	0.974
1,2,3,4,7,8,9-HpCDF	3.03–3.38	3.19	0.160
OCDF	27.5–29.4	28.1	0.730
PCDDs	54.0–61.7	57.8	2.744
PCDFs	147–155	151	3.494
Total PCDD/Fs	201–216	208	5.901

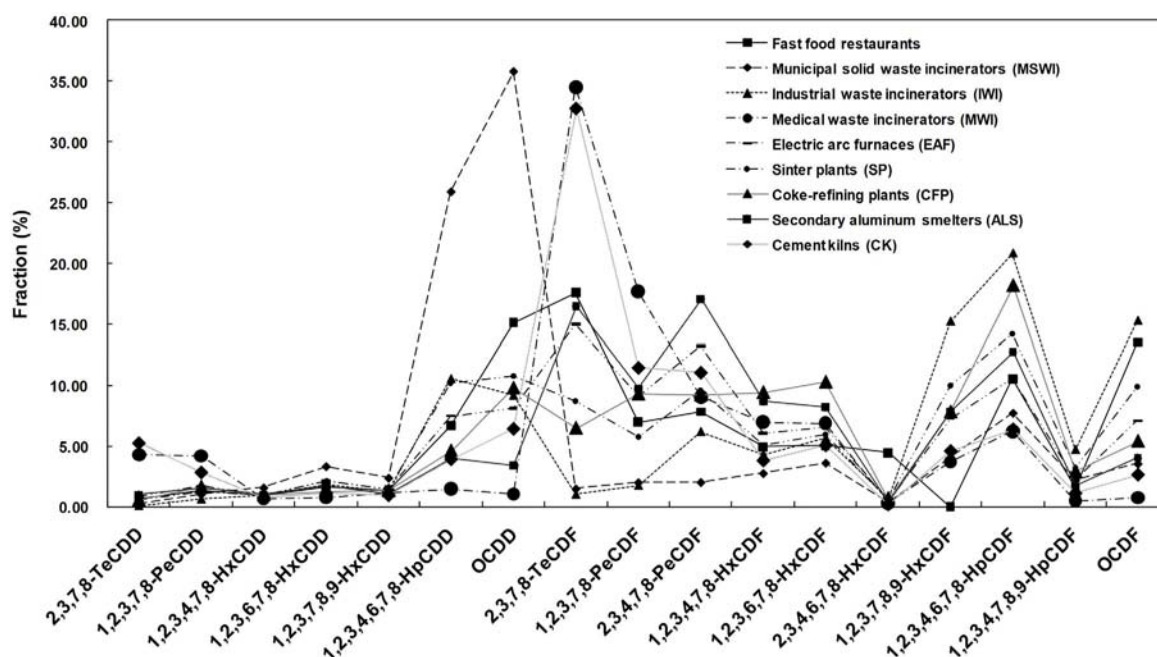
**Table 6.** PCDD/F emission rate (pg/min) at fast food restaurants.

PCDD/Fs	Range	Average	STD
2,3,7,8-TeCDD	68.6–78.2	73.8	3.66
1,2,3,7,8-PeCDD	164–180	170	6.93
1,2,3,4,7,8-HxCDD	112–120	114	3.40
1,2,3,6,7,8-HxCDD	194–224	206	10.9
1,2,3,7,8,9-HxCDD	157–176	170	7.74
1,2,3,4,6,7,8-HpCDD	796–890	826	38.1
OCDD	1696–2021	1875	119.8
2,3,7,8-TeCDF	2058–2263	2179	79.5
1,2,3,7,8-PeCDF	829–896	863	26.3
2,3,4,7,8-PeCDF	932–1002	967	28.0
1,2,3,4,7,8-HxCDF	583–639	610	24.6
1,2,3,6,7,8-HxCDF	572–680	622	39.6
2,3,4,6,7,8-HxCDF	507–578	550	28.6
1,2,3,7,8,9-HxCDF	ND	NA	NA
1,2,3,4,6,7,8-HpCDF	1245–1399	1303	57.9
1,2,3,4,7,8,9-HpCDF	180–201	190	9.52
OCDF	1634–1748	1673	43.4
PCDDs	3208–3669	3434	163.1
PCDFs	8715–9186	8957	207.7
Total PCDD/Fs	11923–12855	12391	351

the stacks of fast food restaurants ranged from 3208–3669 pg/min and 8715–9186 pg/min, with an average of 3434 pg/min and 8957 pg/min. The emission rate for total PCDD/Fs from the stacks of fast food restaurants ranged from 11900–12900 pg/min, with an average of 12400 pg/min.

Fig. 4 summarizes the profiles of 17 PCDD/Fs species of the emission rate for fast food restaurants and other various sources (Kao *et al.*, 2007). The PCDD/F emission rates (pg/min) at various resources are summarized in

Table 7. For fast food restaurants, medical waste incinerators (MWI), electric arc furnaces (EAF), and cement kilns (CK), the 2,3,7,8-TeCDF emission rate was the highest among the seventeen investigated PCDD/F congeners, accounting for 17.6, 34.5, 15.0, and 32.7% of the total emission rates, respectively. However, for industrial waste incinerators (IWI), sinter plants (SP), and coke-refining plants (CFP), the 1,2,3,4,6,7,8-HpCDF emission rate was the highest among the seventeen investigated PCDD/F congeners, accounting for 20.8, 14.2, and 18.2% of the

**Fig. 4.** Congener profiles of 17 PCDD/F species of the emission rate for various sources.



**Table 7.** PCDD/F emission rate (pg/min) for various sources.

Emission	Fast food restaurants	Municipal solid waste incinerators (MSWI)	Industrial waste Incinerators (IWI)	Medical waste Incinerators (MWI)	Electric arc Furnaces (EAF)	Sinter Plants (SP)	Coke-refining Plants (CFP)	Secondary aluminum smelters (ALS)	Cement kilns (CK)
2,3,7,8-TeCDD	73.8	4900	8820	10400	106000	32486	19200	222000	87600
1,2,3,7,8-PeCDD	171	22000	60800	10100	226000	79681	56200	339600	47200
1,2,3,4,7,8-HxCDD	114	33700	88500	1620	120000	60974	34100	219600	12400
1,2,3,6,7,8-HxCDD	206	71300	179000	1910	282000	133607	44000	379200	18500
1,2,3,7,8,9-HxCDD	170	51000	117000	2810	193000	90928	45000	231000	15700
1,2,3,4,6,7,8-HpCDD	826	554000	1020000	3520	991000	642581	158000	906000	65400
OCDD	1880	766000	889000	2510	1080000	683234	336000	780000	107000
2,3,7,8-TeCDF	2180	32000	97800	84000	1990000	551009	222600	3804000	548000
1,2,3,7,8-PeCDF	863	43000	170300	43000	1200000	364260	318600	2244000	191000
2,3,4,7,8-PeCDF	967	43400	599900	22000	1750000	604943	316800	3942000	185000
1,2,3,4,7,8-HxCDF	610	59200	418700	16980	801000	323835	323400	1998000	63600
1,2,3,6,7,8-HxCDF	622	77500	548000	16700	873000	378885	352800	1890000	84600
2,3,4,6,7,8-HxCDF	550	7020	58100	720	59200	31259	24060	93000	3110
1,2,3,7,8,9-HxCDF	0	87500	1477000	9000	963000	633983	268200	1800000	76800
1,2,3,4,6,7,8-HpCDF	1300	165300	2019000	15100	1403000	900555	624000	2940000	106800
1,2,3,4,7,8,9-HpCDF	190	46700	457000	1190	292000	199341	99600	381600	18400
OCDF	1670	75960	1482000	1760	940000	625107	185400	930000	44100
PCDDs	3430	1503000	2365000	32900	2990000	1723489	693000	3077000	354000
PCDFs	8960	637500	7330000	210000	10300000	4613177	2735000	20020000	1322000
Total PCDD/Fs	12400	2140000	9690000	243000	13270000	6336665	3429000	23100000	1676000

total emission rates, respectively. OCDD was the highest among the seventeen investigated PCDD/F congeners in municipal solid waste incinerators (MSWI), accounting for 35.8% of the total emission rate.

## CONCLUSIONS

In this study, we determined the PCDD/F levels in the stacks and food of fast food restaurants. The results indicate that the total I-TEQ concentrations from the stacks of fast food restaurants ranged from 8.16–8.75 pg I-TEQ/Nm<sup>3</sup>, with an average of 8.483 pg I-TEQ/Nm<sup>3</sup>. The 2,3,7,8-TeCDF concentration was the highest among the seventeen investigated PCDD/F congeners, accounting for about 17.3–17.8% of the total concentration. The total PCDD/F concentrations for small chicken nuggets, chicken nuggets, french fries, fish fillets, apple pies, and chicken fillets, respectively, after frying were 84.0, 87.1, 48.9, 42.8, 86.5, and 76.5% of those before frying, indicating that frying significantly decreases PCDD/F content. For all food before and after frying, the OCDD concentration was the highest among the seventeen investigated PCDD/F congeners. The PCDD/Fs emission factor was evaluated using food oil consumption rates, cooking times, stack outlet velocities, and stack diameters. The PCDD/F emission rate was estimated from the exhaust flow rate. The main emission factor and emission rate of PCDD/Fs from the stacks of fast food restaurants was PCDFs. This study provides essential information for conducting health and environmental impact studies of stacks and food at fast food restaurants.

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