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Emissions of PCDD/Fs from municipal solid waste incinerators in China

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ABSTRACT

Gas emission of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) from 19 commercial municipal solid waste (MSW) incinerators in China are investigated. The emission concentrations of PCDD/Fs were 0.042–2.461 ng TEQ Nm^{-3} with an average value of 0.423 ng TEQ Nm^{-3}. The emissions of PCDD/Fs from 16 MSW incinerators were below the MEP regulation level (1.0 ng I-TEQ Nm^{-3}), while only six systems have the dioxin emission levels below the limit established by the European Union Directive of 0.1 ng I-TEQ Nm^{-3}. The emission factors of PCDD/Fs from 19 MSW incinerators were calculated to be 0.169–10.72 μg TEQ ton^{-1} MSW with an average value of 1.728 μg I-TEQ ton^{-1} MSW. The total amount of PCDD/Fs emitted from MSW incinerators to the atmosphere in China was estimated to 19.64 g TEQ $year^{-1}$ in 2006.

1. Introduction

Being the largest developing country, China generates annually 170 billion kg of municipal solid waste (MSW), accounting for 26.5% of the total production of the world (Zhang and Zhu, 2006). The lack of landfill sites for the wastes has forced some big cities to choose the incineration as an alternative technology to treat MSW. The government is planning to construct about 100 commercial-scale MSW incinerators by the year of 2010, including about seventy already existing facilities. Most of existing MSW incinerators are the stoker-type imported from abroad or made by domestic technology, and started to run after 2000. In addition, there are about 20 large scale fluidized bed incinerators of co-firing MSW with coal that have been commercially operated in China (Yan et al., 2006). However, until now there was a lack of detailed information on the dioxin emission level from individual MSW incinerator, as an important indicative of quality control of these plants, which was particularly concerned.

The formation mechanisms and emissions of PCDD/Fs from MSW incinerator have been the subject of extensive research because of serious concerns over the adverse health effects of PCDD/Fs in the environment (Buekens and Huang, 1998; Tuppurainen et al., 1998; McKay, 2002). A generally accepted conclusion is that, PCDD/Fs are mainly generated in the post-furnace by de novo synthesis or precursor synthesis, and their formation rates are primarily related to the operating condition, i.e., the flue gas temperature profile, air supply, O_2, CO, Cl, SOx and H_2O contents in flue gas, as well as carbon and metal contents in fly ash, etc. (Dickson et al., 1992; Stieglitz et al., 1993; Gullett et al., 1994; Addink and Olie, 1995a,b; Stieglitz, 1998; Chang and Huang, 2000). The de novo synthesis occurs at temperatures between 250 and 400 °C, which can produce more highly chlorinated PCDD/F congeners, and the formation rate of PCDDs is significantly lower than the PCDFs (Hagenmaier et al., 1987; Yamamoto et al., 1989). The precursor synthesis occurs at temperatures between 250 and 650 °C, which favors the formation of PCDFs, especially for the low-chlorinated ones (Nakahata and Mulholland, 2000). PCDDs is generally produced by condensation of CPhs, while the PCDFs is mainly formed through a non- or a low-chlorinated precursor followed by further chlorination reactions (Wikstrom et al., 1999). The chlorination of less chlorinated congeners produced in the gas phase (Wikstrom and Marklund, 2000) and dechlorination of highly chlorinated congeners produced by de novo synthesis (Iino et al., 2000) have been proposed as factors that might control PCDD/F isomer distributions (Ryu et al., 2006). The degree of PCDF chlorination was strongly dependent on gas-particle contact time in de novo mechanism (Ryu et al., 2003).

Taking above mentioned aspects into consideration, flue gas emitted from waste incineration must be cleaned before discharge into the atmosphere. Toward this aim, numerous municipal, regional, and national governments have taken stringent measures on the emission levels of PCDD/Fs from MSW incinerators. In recent years, dioxin emissions from MSW incinerators have been reduced to levels <0.1 ng TEQ Nm^{-3}. For better understanding PCDD/F emission levels from MSW incinerators, several research groups have compared and analyzed the discharge data of PCDD/Fs in their corresponding countries (Fabrellas et al., 2001; Abad et al., 2006; Watanabe et al., 2007; Choi et al., 2008). However, the information on PCDD/Fs emission from MSW incinerators in China is still insufficient although some dioxin emission data have been reported (Yan et al., 2006; Bie et al., 2007).

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In this study, we present the emission data of PCDD/Fs and compare the pattern of PCDD/Fs in stack gas of 19 MSW incinerators in China. Using principal component analysis (PCA) and hierarchical cluster analysis (HCA), we analyze the possible grouping of similar emission and demonstrate the abundant PCDD/F congeners, as well as characteristic profiles, in the corresponding groupings.

2. Experiments

2.1. Sample collection

Stack gas samples were collected from MSW incinerators in China, and the informations on incinerators are given in Table 1.

According to US EPA method 23a, stack gas samples were collected with isostack sampler (TCR TECORA, Italy). The sampling box consisted of glass fiber filter (Donglu Environmental Technology Co. Ltd., Tianjin), resin (amberlite XAD-2, Supleco) and 5 impingers (2 water, 2 empties and 1 silica gel). The $^{13}C_{12}$-labelled EDF-4054 with 100 ng mL^{-1} in nonane as sampling standard was spiked to XAD-2 resin before sampling of stack gas.

The $^{13}C_{12}$-labelled EDF-4053 was applied as extraction standard, and EDF-4055 was applied as recovery standard for PCDD/Fs, respectively.

2.2. Sample extraction and analysis

Stack gas samples were transferred into the Soxhlet which was spiked with a mixture of $^{13}C_{12}$-labelled PCDD/Fs internal standards, and extracted with toluene for 24 h. They were then repeatedly washed with sulphuric acid until there was no color. The clean-up procedure for PCDD/Fs analysis was performed with two columns, namely multi-layer silica gel column and basic alumina column.

All labelled standard solution for sampling and analysis of PCDD/Fs were purchased from Cambridge Isotope Laboratory (Andove, MA, USA). Silica gel and basic alumina were purchased from Merck (Germany) and ICN Medical (Germany), respectively. Anhydrous sodium sulfate (Aldrich, reagent grade) was rinsed with hexane and then dried. All pesticide residue solvents were obtained from J.T. Baker (USA).

The purified extract was analyzed using a high resolution mass spectrometer (Micromass, UK) interfaced with a Hewlett-Packard (Palo Alto, CA, USA) 6890 Plus gas chromatograph. The capillary GC column Rtx-2330 (made by Restec) (60 m × 0.25 mm × 0.1 μm) were used to analysis of PCDD/Fs (Ni et al., 2005). Samples were injected in a splitless mode at an injector temperature of 280 °C and at an initial column temperature of 90 °C. After 1.5 min, the temperature was programmed at 25 °C min^{-1} to 180 °C, then at 3 °C min^{-1} up to 260 °C and held for 25 min. The ion source was operated at 260 °C, the electron energy was 37 eV, ionization current was 0.5 mA, ion accelerating voltage was 8 kV and the mass spectrometer was tuned to a mass resolution of 10000 under positive EI conditions. And all data were obtained in the selected ion monitoring (SIM) mode. Toxic equivalents values for PCDD/Fs were calculated by using international-toxicity equivalency factor (I-TEF). All data is normalized by 11% O_2 content.

3. Results and discussions

3.1. TEQ Contribution of PCDD/Fs in stack gas

The gas emission levels of PCDD/Fs from the 19 waste incineration facilities in China were investigated, and the analyzed data are presented in Fig. 1. The emissions of PCDD/Fs ranged from 0.042 to 2.461 ng TEQ Nm^{-3} with an average value of 0.423 ng TEQ Nm^{-3}. The dioxin emission levels of 16 MSW incinerators are below 1.0 ng TEQ Nm^{-3}, which is the emission limit regulated by Ministry of Environmental Protection of Peoples Republic of China (MEP), while only six systems have the dioxin emission levels below the emission limit (0.1 ng TEQ Nm^{-3}) regulated by other developed countries such as Germany. Although higher emission levels from MSW incinerators in Korea were also observed in an early report (Oh et al., 1999), the emission levels in most reports (Fabrellas et al., 2001; Ulrich et al., 2004) are less than 0.1 ng TEQ Nm^{-3}, which, to a certain degree, suggests that the MSW incineration techniques in China should be further improved.

The gas emissions of PCDD/Fs from three China-made MSW incinerators built by the Weining Co. Ltd. in 2002 (M10), 2004 (M13), 2006 (M12) was compared. These three incineration facilities are the stoker-type, composed of grate furnace, boiler, cyclone precipitator, semi-dry scrubber and fabric filter. In order to reduce dioxin emission, this company has been made efforts to improve the effectiveness of air pollution control system to reduce dioxin emission by modifying design and adding active carbon injection device. The emission levels of PCDD/Fs in stack gases were shown in Fig. 2. The average emission concentration of PCDD/Fs was 0.377 ng TEQ Nm^{-3} in 2002. It is found that the high-

Table 1
Status of the investigated waste incineration facilities.

	Facility type	Stack temperature (°C)	Capacity (t d^{-1})	Sample times	APCD combination
M01	Grate incinerator	160	225	4	Semi-dry scrubber + activated carbon + fabric filter
M02	Fluidized bed incinerator	165	385	5	Semi-dry scrubber + activated carbon + fabric filter
M03	Fluidized bed incinerator	156	500	4	Semi-dry scrubber + activated carbon + fabric filter
M04	Fluidized bed incinerator	165	200	6	Semi-dry scrubber + fabric filter
M05	Grate incinerator	156	150	4	Semi-dry scrubber + fabric filter
M06	Fluidized bed incinerator	185	225	6	Semi-dry scrubber + activated carbon + fabric filter
M07	Grate incinerator	150	500	3	Semi-dry scrubber + activated carbon + fabric filter
M08	Grate incinerator	180	385	4	Semi-dry scrubber + fabric filter
M09	Grate incinerator	175	385	3	Semi-dry scrubber + fabric filter
M10	Grate incinerator	170	250	7	Semi-dry scrubber + activated carbon + fabric filter
M11	Grate incinerator	175	500	3	Semi-dry scrubber + activated carbon + fabric filter
M12	Grate incinerator	175	385	4	Semi-dry scrubber + activated carbon + fabric filter
M13	Grate incinerator	180	385	8	Semi-dry scrubber + fabric filter
M14	Grate incinerator	180	225	3	Semi-dry scrubber + fabric filter
M15	Fluidized bed incinerator	165	400	3	Semi-dry scrubber + activated carbon + fabric filter
M16	Grate incinerator	170	200	6	Semi-dry scrubber + fabric filter
M17	Grate incinerator	170	500	4	Semi-dry scrubber + activated carbon + fabric filter
M18	Fluidized bed incinerator	165	250	6	Semi-dry scrubber + activated carbon + fabric filter
M19	Grate incinerator	175	400	3	Semi-dry scrubber + activated carbon + fabric filter

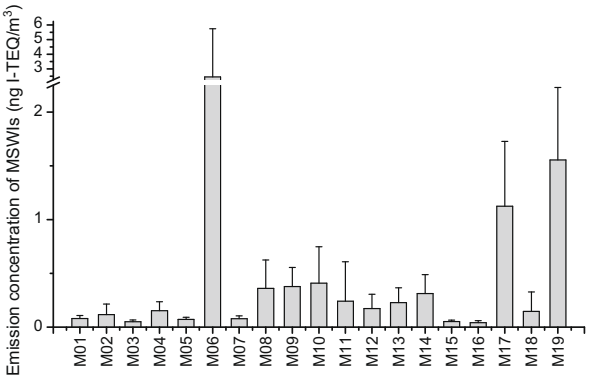

Fig. 1. The emission levels of PCDD/Fs from 19 MSW incinerators in China.

er temperature (>230 °C) of boiler outlet tended to result in a larger production of PCDD/Fs (Zhang et al., 2008). After two years, the temperature of the outlet of Air Heater was decreased to below 200 °C by increasing a group of economizer before the air heater, and thus the average emission concentration was decreased to 0.228 ng TEQ Nm^{-3}. In 2006, the activated carbon injection device was upgraded and 0.173 ng TEQ Nm^{-3} of emission concentration was achieved.

3.2. Congener profiles of PCDD/Fs in various MSW incinerators

Principal component analysis (PCA) and hierarchical cluster analysis (HCA) have been widely applied to analyze the congener profiles of PCDD/Fs in various environmental media and the original sources of various PCDD/F congeners (Cheng et al., 2003; Lee et al., 2007; Choi et al., 2008). To better understand the congener profiles of dioxins in stack gases form various MSW incinerators, principal component analysis (PCA) and hierarchical cluster analysis (HCA) were used to evaluate the possible groupings of similar emissions and the dominant congeners in a defined grouping, as well as to illustrate the characteristic profiles for such groupings within the resulting data. PCA and HCA were performed on normalized and standardized PCDD/Fs congener data using SPSS 11.0. An agglomerative type clustering was performed for HCA. The non-linear iterative partial least squares algorithm and "nearest neighbor" linking with Euclidean distances was performed for PCA.

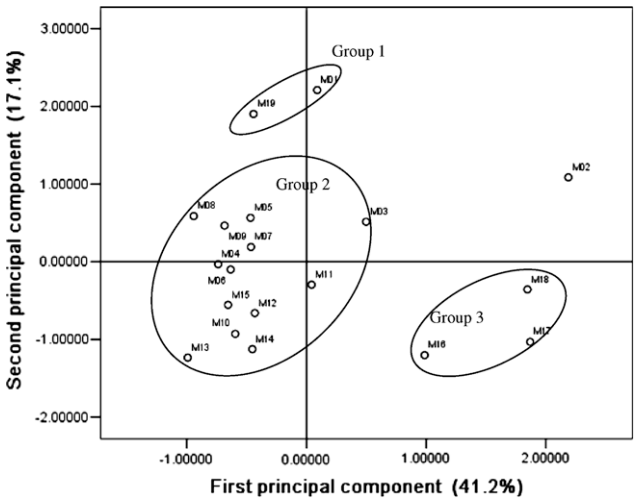

Fig. 3. Scope plot of principal component analysis for relative contributions of the 17 toxic dioxin congeners in the stack gases from MSW incinerators.

The scope plot of principal component analysis for relative contributions of the 17 toxic dioxin congeners is shown in Fig. 3, and the similarities and differences in the dioxin patterns are shown in Fig. 4. Based on the four components extracted from PCA, PC1 and PC2 accounts for 58.3% of the total variance. These two multivariate pattern comparison results indicated three groups (groups 1, 2, 3) for the all congener patterns in stack gases from MSW incinerators.

As shown in Fig. 5, OCDD or HpCDD were the highest in the pattern of PCDDs, while 1,2,3,4,6,7,8-HpCDF was the highest in the pattern of PCDF for all samples. The proportions of 2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF and 1,2,3,6,7,8-HxCDF were similar for groups 1 and 2, which suggested these compounds can be formed from the same precursors by the chlorination reactions of lower chlorinated-CDF homologues (Wikstrom et al., 1999). The ratios of PCDFs to PCDDs for all gas samples were more than 1, implied the de novo synthesis is always dominant (Everaert and Baeyens, 2002).

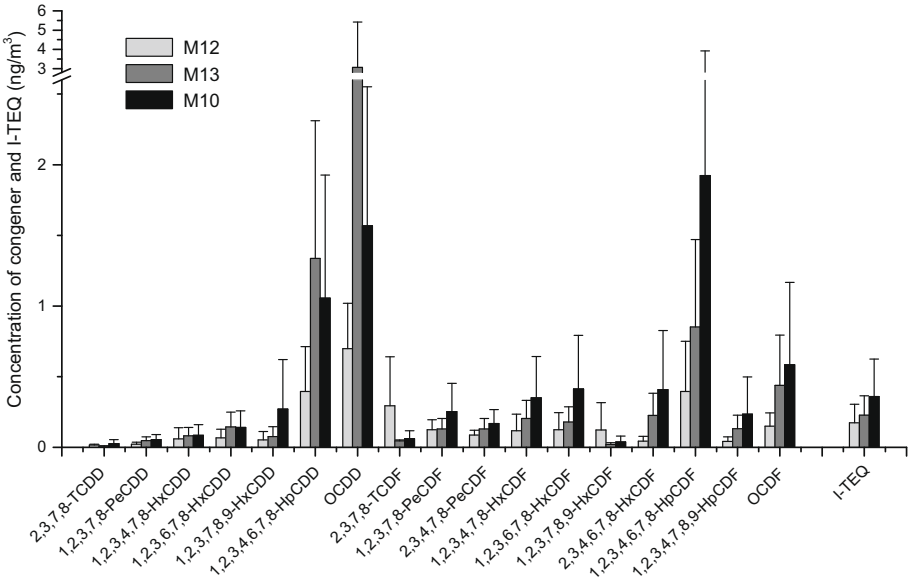

Fig. 2. Emission levels of PCDD/Fs from three home made MSW incinerators built by Weining Co. Ltd.

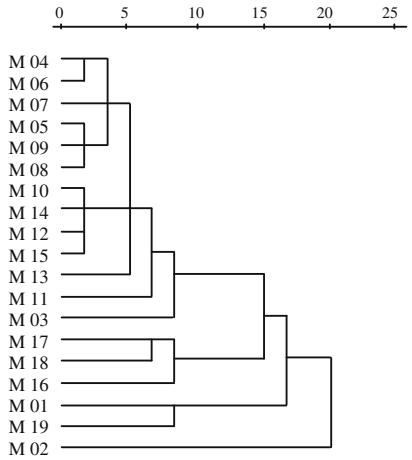

Fig. 4. HCA dendrogram using "nearest neighbor" linking with Euclidean distances for normalization.

The congener pattern in group 1 is characterized by 2,3,7,8-substituted PCDFs (load factor is 75.4%) with a high proportion of 1,2,3,4,6,7,8-HpCDF (24.7 ± 1.7%) and 1,2,3,4,6,7,8-HpCDD (12.5 ± 0.9%). The congener pattern in group 1 was similar to the results (H1 and H2) reported by Wang and coworkers, who investigated the gas emission of PCDD/Fs from a medical waste incinerator and regarded the chlorine content in the waste played an important role in determining the congener profiles of PCDD/Fs in stack gas (Wang et al., 2003). In the group 2, the contributions of 1,2,3,4,6,7,8-HpCDD, OCDD and 1,2,3,4,6,7,8-HpCDD to the total amount of PCDD/Fs were 14.3 ± 1.6%, 27.2 ± 5.4% and 15.8 ± 2.7%, respectively, and the total contribution of 2,3,7,8-substitued PCDFs was 51.6%. Most samples in the study belonged to group 2, and the dioxin congener patterns seemed to be not affected by the type of incinerators and the design of air pollution control devices (APCDs). The dioxin congener pattern in group 2 was similar to the results (M2 and M3) reported by Wang et al. (2003) and Bie et al. (2007). In group 3, the proportion of PCDFs to the total amount of PCDD/Fs was 76.7% with a high proportion of 2,3,7,8-TCDF (19.9 ± 7.6%), 1,2,3,7,8-PeCDF (13.9 ± 3.2%), 2,3,4,7,8-PeCDF (10.5 ± 0.8%) and 1,2,3,4,6,7,8- HpCDF (11.3 ± 1.9%), and PCDF isomer distributions indicated that low-chlorinated dibenzofurans was more than the high-chlorinated dibenzofurans. This result was different from other published emission data of MSW incinerators (Wang et al., 2003; Abad et al., 2006; Choi et al., 2008). MSW incinerator M02 excluded from groups 1–3 was characterized by 2,3,7,8-TCDF (12.0 ± 1.7%), 1,2,3,7,8-PeCDF (9.6 ± 8.9%), 2,3,4,7,8-PeCDF (11.4 ± 3.2%), 1,2,3,4,7,8-PeCDF (10.3 ± 5.0%), 1,2,3,6,7,8-PeCDF (9.2 ± 3.3%), and 1,2,3,4,6,7,8- HpCDF (11.0 ± 1.6%), and the ratio of PCDDs to PCDFs for it was 3.0.

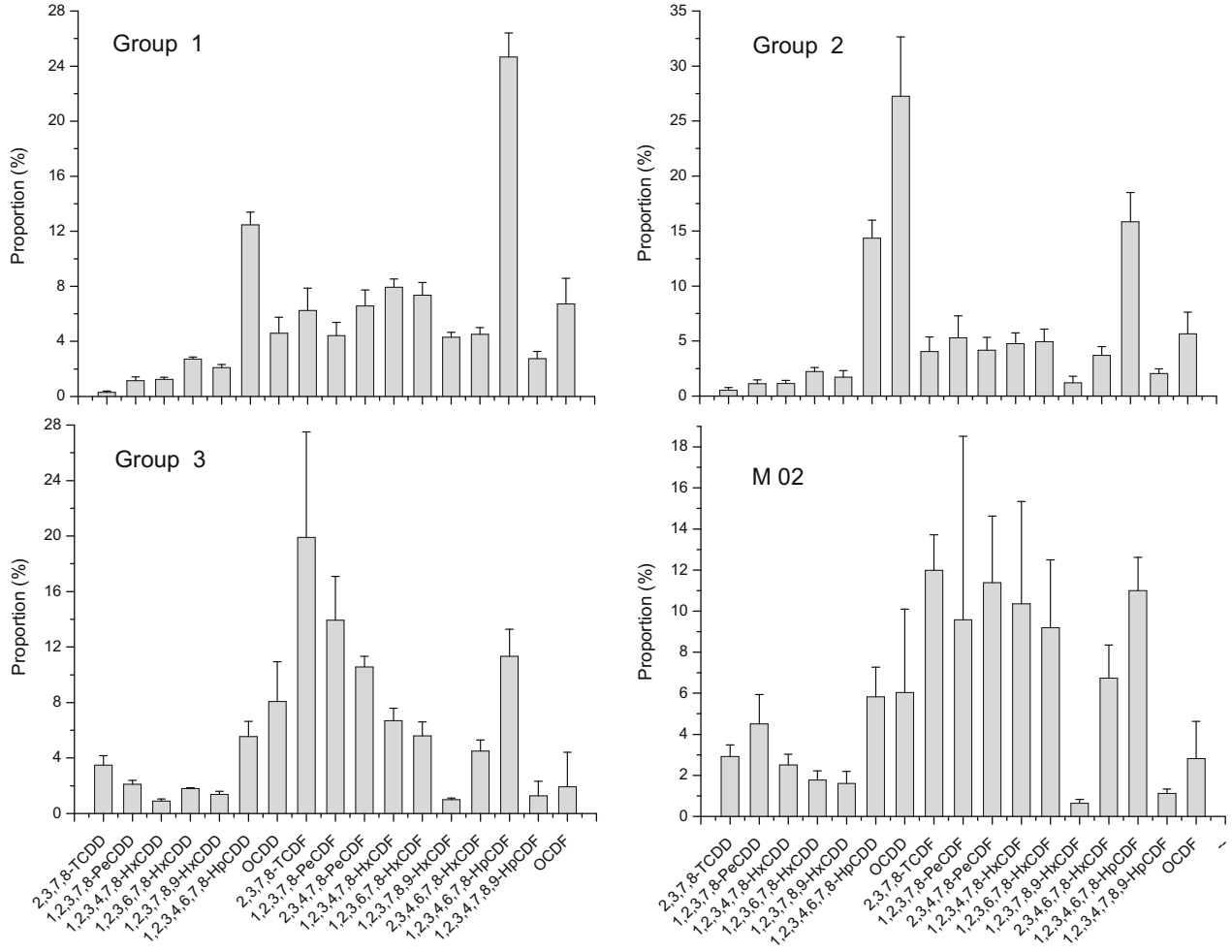

Fig. 5. Representative congener patterns of PCDD/Fs in each group of municipal solid waste incinerator.

Table 2
Mean value of dioxin emission factors (μg ton^{-1} MSW) and relative standard deviations (RSD, %) of the 19 MSW incinerators.

PCDD/Fs	M01	M02	M03	M04	M05	M06	M07	M08	M09	M10
2,3,7,8-TCDD	0.014	0.112	0.017	0.051	0.510	0.028	0.041	0.128	0.194	0.145
1,2,3,7,8-PeCDD	0.051	0.203	0.038	0.135	1.802	0.082	0.065	0.288	0.440	0.447
1,2,3,4,7,8-HxCDD	0.054	0.104	0.022	0.138	1.981	0.079	0.058	0.445	0.503	0.366
1,2,3,6,7,8-HxCDD	0.095	0.077	0.048	0.230	3.188	0.148	0.123	0.738	1.046	0.779
1,2,3,7,8,9-HxCDD	0.080	0.076	0.040	0.201	2.519	0.092	0.085	1.419	0.728	0.454
1,2,3,4,6,7,8-HpCDD	0.341	0.211	0.189	1.759	20.66	0.881	0.645	5.535	6.479	5.310
OCDD	0.319	0.157	0.261	3.420	28.98	1.803	1.239	8.225	7.730	10.57
2,3,7,8-TCDF	0.063	0.429	0.349	0.206	3.988	0.096	0.272	0.323	0.760	0.824
1,2,3,7,8-PeCDF	0.187	0.188	0.141	0.383	4.993	0.224	0.266	1.325	2.378	1.960
2,3,4,7,8-PeCDF	0.209	0.506	0.171	0.317	8.826	0.277	0.220	0.884	0.975	1.232
1,2,3,4,7,8-HxCDF	0.276	0.513	0.176	0.576	7.938	0.280	0.232	1.837	1.824	1.231
1,2,3,6,7,8-HxCDF	0.295	0.429	0.148	0.490	9.882	0.299	0.278	2.164	1.906	1.678
1,2,3,7,8,9-HxCDF	0.021	0.029	0.125	0.059	0.711	0.240	0.024	0.197	0.144	0.117
2,3,4,6,7,8-HxCDF	0.312	0.294	0.008	0.514	11.01	0.199	0.225	2.144	1.238	1.211
1,2,3,4,6,7,8-HpCDF	0.900	0.442	0.326	1.938	24.90	1.575	0.721	10.10	5.539	4.981
1,2,3,4,7,8,9-HpCDF	0.096	0.041	0.041	0.323	4.386	0.122	0.130	1.235	0.786	0.647
OCDF	0.232	0.069	0.119	0.000	9.041	0.259	0.602	3.067	2.084	2.485
TEQ	0.286	0.678	0.226	0.581	10.72	0.390	0.343	1.887	1.973	1.871
	M11	M12	M13	M14	M15	M16	M17	M18	M19	
2,3,7,8-TCDD	0.123	0.084	0.032	0.165	0.024	0.054	0.673	0.176	0.091	
1,2,3,7,8-PeCDD	0.233	0.120	0.176	0.267	0.053	0.023	0.537	0.141	0.428	
1,2,3,4,7,8-HxCDD	0.200	0.319	0.302	0.239	0.057	0.013	0.253	0.045	0.546	
1,2,3,6,7,8-HxCDD	0.401	0.366	0.534	0.418	0.135	0.021	0.493	0.109	1.611	
1,2,3,7,8,9-HxCDD	0.263	0.286	0.283	0.257	0.070	0.017	0.500	0.073	1.148	
1,2,3,4,6,7,8-HpCDD	1.513	2.139	4.974	3.723	0.769	0.081	1.686	0.219	9.172	
OCDD	3.027	3.776	11.39	9.685	1.590	0.158	1.910	0.198	0.099	
2,3,7,8-TCDF	0.696	1.591	0.171	0.519	0.120	0.287	7.606	0.560	6.024	
1,2,3,7,8-PeCDF	1.560	0.671	0.485	2.992	0.247	0.112	4.975	1.006	1.811	
2,3,4,7,8-PeCDF	1.052	0.465	0.483	0.885	0.209	0.081	3.946	0.646	4.190	
1,2,3,4,7,8-HxCDF	0.787	0.634	0.761	1.541	0.217	0.057	3.833	0.364	4.780	
1,2,3,6,7,8-HxCDF	1.021	0.672	0.667	1.918	0.214	0.068	1.602	0.384	3.719	
1,2,3,7,8,9-HxCDF	0.058	0.663	0.072	0.098	0.019	0.005	0.874	0.022	4.832	
2,3,4,6,7,8-HxCDF	0.610	0.231	0.837	0.978	0.252	0.077	0.589	0.278	0.233	
1,2,3,4,6,7,8-HpCDF	1.134	2.140	3.170	5.001	0.811	0.221	2.785	0.455	14.72	
1,2,3,4,7,8,9-HpCDF	0.142	0.222	0.489	0.626	0.089	0.020	0.299	0.055	1.715	
OCDF	0.413	0.807	1.634	1.381	0.363	0.056	0.618	0.069	4.165	
TEQ	1.279	0.935	0.848	1.592	0.294	0.169	4.789	0.811	5.040	

3.3. Estimation of dioxin emission factors from MSWIs

The emission factors of PCDD/Fs to the atmosphere from 19 MSW incinerators, were calculated to be 0.169–10.72 μg TEQ ton^{-1} MSW with an average 1.728 μg TEQ ton^{-1} MSW (Table 2). These emission factors were based on the total weights of MSW, the flow rates of dry stack gas, and the gas emission concentrations of PCDD/Fs. The average emission factor was larger than other reported data (Fabrellas et al., 2001; Chen, 2004; Choi et al., 2008). The total emission of PCDD/Fs from 19 MSW incinerators was calculated to be 3.247 g TEQ year^{-1} according to an operational time of 300 d year^{-1}, and 24 h d^{-1} for each MSW incinerator. The dioxin emission factors varied largely according to the different MSW incinerators. The home made MSW incinerators had the higher dioxin emission factors than the ones imported from abroad did. In 2006, the activity levels of MSW incinerators were 11.38 million tons in China (Committee for Treatment of Urban Domestic Refuse of CAEP, 2008). Using the average emission factor 1.728 μg I-TEQ ton^{-1} MSW, the annual PCDD/F emission amount were calculated to be 19.64 g year^{-1} from MSW incinerators in China.

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