THE EMISSION AND FORMATION OF PCDDs/PCDFs IN FLUE GAS SAMPLES FROM CREMATORIES

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Abstract

The concentrations of polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in flue gas samples emitted from 6 crematories using diesel oil were measured. The sampling and analytical processes are based on HJ 77.2-2008. The total concentrations of PCDDs/PCDFs in these samples ranged from 56.1 to 578 ng/m^3 (273.15K, 101.325kPa, at 11% O_2 normalized), while their TEQ values were 0.96~8.1 ng I-TEQ/m^3 with the similar congener profiles. The 17 types of 2,3,7,8-substituted isomers were detected in all flue gas samples, and 2,3,4,7,8-P_5CDF was the most important contributor to the TEQ concentrations. The total concentration of PCDFs was higher than that of PCDDs, and the concentration of T_4CDFs was the highest in PCDFs. The *de novo* synthesis was dominant in PCDDs/PCDFs formation. The emission factor calculated in this research was between 16.7 and 95.0 μg TEQ/body with an average value of 34.3 μg TEQ/body.

Introduction

Dioxins and furans, more precisely polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDDs/PCDFs) are two groups of persistent and toxicologically significant trace organic contaminants that are produced enintentionally[1]. PCDDs/PCDFs are formed as unintentional by-products in certain processes and activities. The ten Main Source Categories are listed in Standardized Toolkit for Identification and Quantification of Dioxin and Furan Releases (Toolkit)[2].Crematory, as one of the special process, is already included in the Toolkit.

Funeral rite usually depends on religion and custom of a country. Cremation is a common practice in China to destroy human bodies by burning. According to the statistical report published by Ministry of Civil Affairs of the People's Republic of China[3], up to the end of 2007, there have been 4,838 crematories in the whole country, and the total population of the cremated bodies was up to 4,421,100, accounting for 48.4% of the dead human bodies. In many economic developed regions in China, the demand for cremation has been on the rise. Recently, PCDDs/PCDFs emissions from crematories have become a serious problem in China.

The essential components for crematory are the charging of the coffin and the corpse, the main combustion chamber, and where applicable the secondary chamber. In some advanced crematories, appropriate air pollution control system could be equipped. At last, the flue gas generated during cremation is discharged to the

atmosphere via the stack. In China, Most furnaces are fired using diesel oil. In this research, concentrations of PCDDs and PCDFs in flue gas samples from 6 crematories using diesel oil were measured. The emission and formation of PCDDs/PCDFs from crematories were discussed, and the emission factor for the crematories was estimated.

Materials and Methods

Sampling activities

6 crematories using diesel oil at three different sites in China were selected. In terms of the different structures of the main combustion chamber, all of the crematories were classified into two types, flat crematory and trolley crematory. In this research, 3 flat crematories and 3 trolley crematories were tested. The workflow for those crematories was similar. The corpse and coffin were sent into the main combustion chamber and heated by the igniter, air and diesel oil were added during the burning period in order to maintain steady combustion. All the facilities had no air pollution control systems, and finally, gases left through the stacks. Only in crematory A, C, and F, a secondary combustion chamber was adopted.

Flue gas was sampled through each crematory. For cremation of one dead body, sampling started from igniting the main combustion chamber and finished when all the cremation procedures were over. In consideration of the detection limit and other influence factor, total length of sampling time needed to be persisted during the whole process of cremating 2 to 4 dead bodies, and 3 samples were sampled from each crematory. Moreover, age, weight and sex of the dead bodies and burial accessories in the coffin were recorded. A total of 18 samples were collected. The sampling and analysis processes are based on HJ 77.2-2008.

Analysis processes

Treated the filter with HCl(2mol/L) and rinsed the filter and glass collection components with reagent water and acetone immediately. Then put the filter and XAD-2 resin into a desiccator to be dried at room temperature for about 5 days. The liquid portion was extracted by Dichloromethane. The labelled compound stock solution (EPA-1613LCS，Wellington Laboratories) was spiked into the XAD-2 resin and after that, the pollutants collected on the XAD-2 resin and the filter were extracted by Soxhlet with toluene for 19h. The two parts of the extracts were concentrated and combined as one sample, and then purification was carried out. Firstly, the sample extract was treated by H_2SO_4. Secondly, multi-layer silica gel column and active-carbon impregnated silica gel column were used for further cleanup, and then, almost all the solvent in the sample was removed by a stream of nitrogen. Finally, 5uL internal standard spiking solution (EPA-1613ISS，Wellington Laboratories) and 45uL decane were added into the sample.

PCDDs/PCDFs were identified and quantified by HRGC-HRMS (Agilent 6890N/ Waters Autospec Ultimate NT) with a DB-5MS column (60m×0.25mm×0.25μm).

Quality assurance/quality control (QA/QC) system was strictly carried out during the whole sampling and analysis processes. All the recovery standards in each samples varied within acceptable limit from the method.

Results and Discussion

The concentrations of PCDDs/PCDFs are shown in Table 1. Total concentrations of PCDDs/PCDFs in these samples ranged from 56.1 to 578 ng/m^3 (273.15K, 101.325kPa, at 11% O_2 normalized), while their TEQ values were 0.96~8.1 ng I-TEQ/m^3.Compared with the emission from crematories in other countries, the concentration of PCDDs/PCDFs in this research maintained the basically same level as that in Japan, and lower than that in Thailand (Table 2).

Total TEQ concentrations of PCDDs/PCDFs for flat crematories ranged from 2.3~6.4 ng I-TEQ/m^3, and those of trolley crematories ranged from 0.96~8.1 ng I-TEQ/m^3. In comparison with the TEQ concentrations for each crematory, which type of crematories has obvious advantage in dioxin pollution control could not been concluded. Differences in the concentration caused by age, weight and sex of the dead bodies could not be identified.

Table 1: Concentrations of PCDDs/PCDFs in flue gas samples

Crematory	Sample ID	Total concentrations (PCDDs+PCDFs) (ng/m^3)	TEQ concentrations (ng I-TEQ/m^3)	Average (ng I-TEQ/m^3)
F-A	Flat-A-1	230	4.3	5.1
	Flat-A-2	410	6.4	
	Flat-A-3	359	4.7	
F-B	Flat-B-1	304	4.0	3.7
	Flat-B-2	366	4.7	
	Flat-B-3	174	2.3	
F-C	Flat-C-1	230	4.4	3.3
	Flat-C-2	171	3.2	
	Flat-C-3	127	2.3	
T-D	Trolley-D-1	56.1	7.9	3.4
	Trolley-D-2	72.3	0.96	
	Trolley-D-3	304	1.2	
T-E	Trolley-E-1	274	4.1	6.0
	Trolley-E-2	578	8.1	
	Trolley-E-3	434	5.9	

Crematory	Sample ID	Total concentrations (PCDDs+PCDFs) (ng/m^3)	TEQ concentrations (ng I-TEQ/m^3)	Average (ng I-TEQ/m^3)
T-F	Trolley-F-1	361	4.8	2.9
	Trolley-F-2	79.3	1.6	
	Trolley-F-3	133	2.4	

Table 2: Comparison of PCDDs/PCDFs in flue gas samples from crematories in China and in other countries

Country	Year	TEQ concentrations(ng I-TEQ/m^3)	References
China	2008	0.96~8.1（273.15K，101.325kPa，11% O_2 normalized）	In this research
Japan	1999	0.0099~6.5（273.15K，101.325kPa，12% O_2 normalized）	4
Thailand	1997-2000	6.3~28.6（273.15K，101.325kPa，11% O_2 normalized）	5

Contaminant fingerprinting identifies chemicals and other substances in the environment, as well as the sources of those materials[6]. Dioxin/Furan fingerprints can be displayed graphically by plotting the relative abundances of the dioxin and furan congeners. Fig 1 shows the congener profiles for flue gas emissions from crematories. It can be seen from Fig.1 that the congener profiles in flue gas samples from those 6 crematories were similar. The 17 types of 2,3,7,8-substituted isomers were detected in all flue gas samples, and 2,3,4,7,8-P_5CDF was the most important contributor to total TEQ concentration, its concentration rate ranged from 37% to 50%.

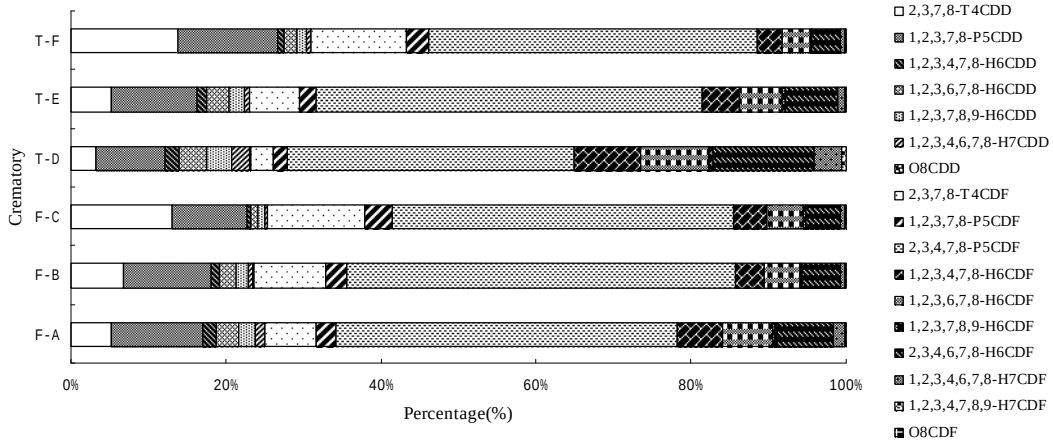

Fig.1 Congener profiles of 2,3,7,8-substituted PCDDs/PCDFs in flue gas samples

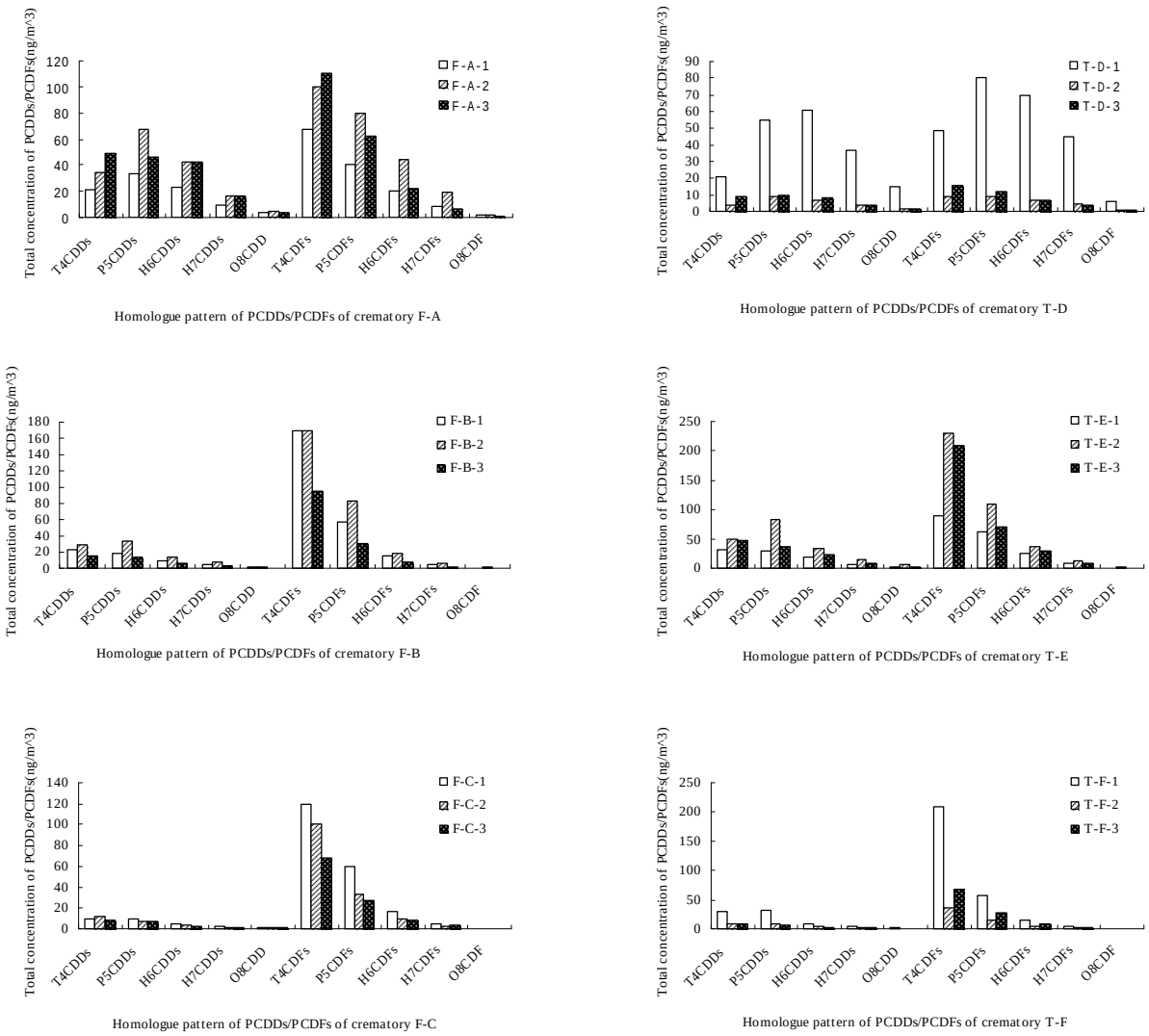

Fig.2 Homologue patterns of PCDDs/PCDFs in flue gas samples

The homologue patterns of PCDDs/PCDFs in flue gas samples were shown respectively in Fig.2.The PCDF homologue profiles were quite similar expected T-D-1, having a trend that the concentration of T_4CDFs was the highest in the homologue patterns and then the proportions of homologues became lower with the number of the chlorine atoms increased. In contrast, PCDD homologue patterns were inconsistent. Two kinds of patterns were identified, one has the highest concentration in T_4CDDs and the other has a mountain shape. Those conclusions are in agreement with the emission characteristics from crematories in Japan[4].

The homologue profiles in T-D-1 flue gas samples were different form others, especially in PCDF homologue profiles. The fluctuation in combustion conditions and operational state, including a great many burial accessories may influence PCDDs/PCDFs formation and emission from crematories.

The ratios of PCDFs to PCDDs for all the flue gas samples were more than 1, implied the *de novo* synthesis is always dominant in PCDDs/PCDFs formation[7].

Emission factors to air for all crematories were calculated using the following equation:

Emission factor (μg TEQ/body) = TEQ concentration(ng TEQ/m^3)×dry standard gas volume(m^3/h)× cremation period(h/body)/1000

Emission factor level calculated in this research was between 16.7 and 95.0 μg TEQ/body with an average value of 34.3 μg TEQ/body. And the emission factor recommended for poor combustion conditions crematory in Toolkit 2005 is 90 μg TEQ/body. As a result, the crematories should be considered to be an important emission source and the application of best available techniques and best environmental practices (BAT/BEP) technology in this industry should be an effective measure to reduce and eliminate formation of PCDDs/PCDFs.

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