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Persistent organic pollutant emissions from medical waste incinerators in China

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Abstract The huge amount of medical waste (MW) has caused a tough challenge to environmental protection in China because of its serious infectious potential. At present, incineration is the most common technology for MW disposal. Unfortunately, the medical waste incinerator (MWI) is considered one of the major sources of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs). This study was conducted to investigate the generation and the components of MW; the fingerprint of PCDD/Fs in MWI; and PCDD/F, polychlorinated biphenyl (PCB) and hexachlorobenzene concentrations in residue ash. The estimated annual production of MW was estimated to be 0.97 million tons in China in 2008; in addition, plastic and rubber accounted for 24.5% of MW contents. PCDD/F emissions from MWI could be divided into two main groups according their fingerprints, and the ratio of PCDFs/PCDDs was mostly over 1.5, with a mean value of 3.43. The toxic equivalent of PCDD/Fs was over 30 times that of the value of PCBs in the residue ash, and PCDD/F contents in fly ash accounted for approximately 67% of the total output of PCDD/Fs, which was in line with the UNEP default emission factors for MWI (class 3, 63.7%).

Keywords Medical waste · Incinerator · PCDD/F emissions · China

Introduction

Medical wastes (MW) are the hazardous refuse generated by hospitals, clinics and other related health-care institutions, and typically consist of sharp objects, chemicals and blood components. They are characterized by being toxic and infectious, and have the potential to cause health risks and environmental pollution with improper treatment and management. With the rising awareness of the importance of environmental protection, more public and scientific communities are concerned about MW disposal and proper treatment [1–3]. In many developed countries, specific rules and regulations have been implemented for MW management, and are performed highly effectively. With the fast economic development in China, many social problems and difficulties have emerged, such as environmental pollution. China has historically paid little attention to the strict management of MW. In the last decade, laws, regulations and rules, as well as standards, were gradually established and implemented at all stages of MW management, including separation, collection, packaging, storage, transport, treatment and disposal [4]; however, the actual performance of these tasks is still not perfect.

Off-site central incineration has been widely demonstrated to be a fast and commercially available technology for disposing of MW, as direct landfill has been forbidden since 2002 in China [5]. According to the National Hazardous Waste and Medical Waste Disposal Facility Construction Plan, 331 modern, high-standard, centralized incinerators were to be built by the end of 2006; in addition, on-site incineration was to be gradually abandoned [6]. However, incineration is also a controversial issue because of the potential for secondary pollution, such as emissions of HCl, NOx, heavy metals and persistent organic pollutants (POPs) [7–10]. In 2004 in China, 1.18 kg I-TEQ of PCDD/Fs was reported to have been released from medical waste incineration [11]. Therefore, it is quite essential and inevitable to understand the present

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situation of MW generation and PCDD/F emissions in thermal processes in China.

Generation and components of MW

With the stable economic development of over 7% of the GDP growth rate, the demand for and consumption of health care has also been rising significantly each year. Increasing numbers of hospitals and clinics are being built, and citizens frequently visit the health institutions. The Ministry of Health has recorded the statistical data of health consumption and construction [12]. Table 1 shows the beds in hospitals for inpatients and the visiting times of outpatients. Both beds and outpatient visiting times have markedly increased in the last decade. Meanwhile, the production of MW has also rapidly increased.

Both outpatients and inpatients generate MW during health institution visits. The waste generation rates are quite different. Based on the reported data (Table 2) [3, 13–15], the rate of inpatients in China is on average 0.58 kg/bed × day, and approximately 20–30 outpatient visits produce 1.0 kg of waste [16]. Therefore, the annual generation of MW can be calculated, as shown in Figs. 1 and 2. The yield of MW was found to increase quickly, and 0.97 million tons of MW was generated in China in 2008. Also in another report the amount of MW produced in 2002 was reported to be 0.65 million tons [17].

Many factors influence the composition of MW, including function, scale, season and management. Therefore, it is very difficult to exactly classify the components of MW. We reviewed the surveys of MW composition by different researchers in different cities (Table 3). Regarding the average composition of MW shown in Table 3, the major components are fibrous material, plastic and rubber, which have a high caloric value (10,000–16,000 kj/kg) and chlorine content (4.5–7.5%) compared to municipal solid waste [19, 20].

Table 1 Beds in hospitals and outpatient visits in China [12]

Year	2003	2004	2005	2006	2007	2008
Beds in health institution (million)	3.16	3.27	3.37	3.51	3.7	4.04
Annual visit times of outpatients (billion)	2.10	2.20	2.30	2.45	2.43	3.53

Table 2 Generation rate of medical waste by inpatients (kg/bed day) [3, 13–15, 18]

City	Changsha	Nanjing	Wuhan	Hangzhou	Urumqi	Average
Yield	0.60	0.68	0.57	0.48	0.58	0.58

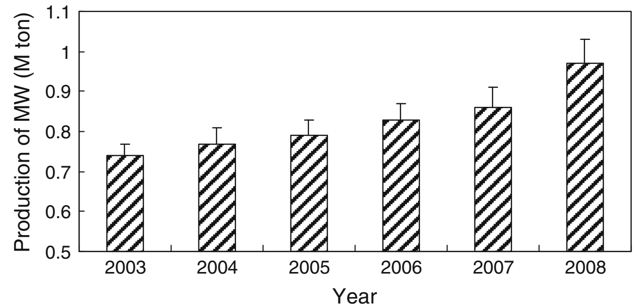

Fig. 1 Annual production of MW in China (Mton)

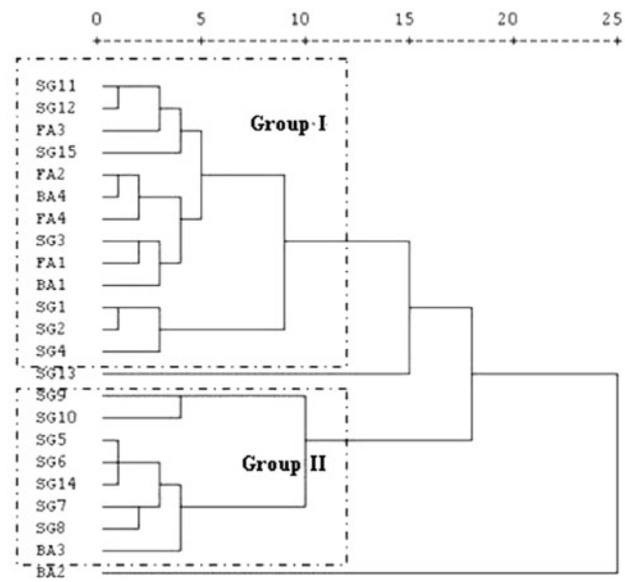

Fig. 2 HCA of PCDD/F homologue in the MWI samples collected (*SG* stack gas samples, *FA* fly ash samples, *BA* bottom ash samples)

Table 3 Composition of MW in China [14, 21–23]

City	Components percentage (%)				
	Plastic and rubber	Fibrous material	Organic tissue and others	Glass	Metal and others
Changsha	23	12	6	55	4
Beijing	20.1	23.3	26.0	12.0	18.6
Zhengzhou	29.4	26.6	25.1	16.5	2.4
Bazhong	17.4	62.7	5.3	11.9	2.7
Huzhou	47.5	36.9	7.1	5.6	2.9
Shanghai	32.4	22.8	9.3	14.9	20.6
Average	28.3	30.7	13.1	19.3	8.5

PCDD/F emissions from MWI

Materials and methods

MWI is among the major sources of PCDD/Fs in the environment [24]. We conducted the determination of PCDD/Fs in stack gas and PCDD/Fs, PCBs and HxCBz in residue ash collected from several waste incinerators. Brief information about the MWI as well as the sample numbers is shown in Table 4. Open burning of MW is defined as MWI8. Hazardous waste was co-combusted with medical waste in HWI9.

The stack gas samplings were carried out according to USEPA method 23 by isostack sampler (M5, KNJ Engineering, South Korea). The particle phase was collected using a glass fiber filter, and the gas phase was collected using XAD-2 resin. The fly ash was collected in the exit of the bag house and bottom ash in the exit of the slag remover. The preparation (extraction and purification) of samples and the instrument analysis of PCDD/Fs and PCBs were performed as in our previous studies [10, 25]. The analysis of PCBs and PCDD/Fs was done by HRGC/HRMS on a 6890 Series gas chromatograph (Agilent, USA) and coupled to a JMS-800D mass spectrometer (JEOL, Japan). The HxBz analysis was conducted in the GC-ECD (Agilent 6890N GC). The pretreatment of HxCBz analysis was according to the state method of HJ/T 74-2001 and GB 7492-87 in China, consisting of extraction and a clean-up procedure (H_2SO_4 treatment, multi-layer silica gel column and Florisil).

PCDD/F homologue pattern in the samples collected from MWI

Hongcai Gao [9] measured the stack gas emissions of PCDD/Fs from MWI in China. The concentration ranged from 0.08 to 31.6 ng I-TEQ Nm^{-3}, with a median value of 0.41 ng I-TEQ Nm^{-3} and an average concentration of 4.22 ng I-TEQ Nm^{-3}. In our research, the level of PCDD/F emissions ranged from 0.32 to 16.43 ng I-TEQ Nm^{-3}, with a median of 4.69 ng I-TEQ Nm^{-3} and a mean of 5.85 ng I-TEQ Nm^{-3}. According to our research, much work and many improvements need to be carried out in order to meet the present emission limits (0.5 ng I-TEQ Nm^{-3}) and the more stringent regulations coming in the future (0.1 ng I-TEQ Nm^{-3}). PCDD/F homologue patterns as the fingerprint of PCDD/Fs are the characteristic emissions from different sources. The degree of chlorination of PCDD/Fs and the ratio of PCDFs to PCDDs are shown in Table 5. The ratio of PCDFs to PCDDs was over 1.5 in most of the samples, and 3.43 on average. That the ratio of PCDFs/PCDDs is higher than 1.0 is a typical combustion fingerprint of PCDD/Fs. Also, de novo synthesis was indicated to be the predominant mechanism of PCDD/F formation [26]. The degree of chlorination of PCDD/Fs was also calculated and ranged from 4.90 to 6.11, being 5.50 on average. Compared to the PCDD/F emissions from municipal solid waste incinerators (6.68, an average of 12 units) [27], the degree of chlorination was not directly increased by the higher content of chlorine in waste.

Hierarchical cluster analysis (HCA) was used to evaluate the similarities and differences of PCDD/F homologue patterns in the samples collected from MWI. According to

Table 4 Basic information about MWI and the samples

No.	Facility type	Air pollution control	Stack gas	Fly ash	Bottom ash
MWI1	Shaft kiln + SCC	SDS + AC + FB	SG1, SG2	FA1	BA1
MWI2	Rotary kiln + SCC	SDS + AC + FB	SG3, SG4	FA2	BA2
MWI3	Pyrolysis boil + SCC	SDS + AC + FB	SG5, SG6	FA3	–
MWI4	Rotary kiln + SCC	SDS + AC + FB	SG7, SG8	–	–
MWI5	Pyrolysis	None	SG9, SG10	–	–
MWI6	Rotary kiln + SCC	DS + AC + FB	SG11, SG12	–	–
MWI7	Pyrolysis boiler	WS	SG13	–	–
MWI8	Open burning	–	–	–	BA3
HWI9	Rotary kiln + SCC	DS + AC + FB + WS	SG14, SG15	FA4	BA4

SCC secondary combustion chamber, *SDS* semi-dry scrubber, *AC* active carbon, *FB* fabric bag, *DS* dry scrubber, *WS* wet scrubber

Table 5 Chlorination degree of PCDD/Fs and the ratio of PCDFs to PCDDs

		MWI1	MWI2	MWI3	MWI4	MWI5	MWI6	MWI7	MWI8	HWI9
Stack gas	PCDFs:PCDDs	4.90	3.84	5.44	2.88	1.68	2.32	3.67	–	3.48
	Chlorination	5.83	5.82	4.90	4.92	4.96	5.25	6.39	–	5.04
Fly ash	PCDFs:PCDDs	4.62	2.61	3.88	–	–	–	–	–	1.49
	Chlorination	5.64	5.52	5.33	–	–	–	–	–	5.80
Bottom ash	PCDFs:PCDDs	3.29	0.68	–	–	–	–	–	7.91	2.21
	Chlorination	5.69	6.11	–	–	–	–	–	5.16	5.67

the analysis results, the samples were assembled into two main groups (group I and II). The PCDD/F fingerprint of these two main groups is depicted in Fig. 3. In group I, percentages of TeCDFs to HxCDFs were similar, and the portion of PCDFs gradually decreased with the increasing degree of chlorination in group II.

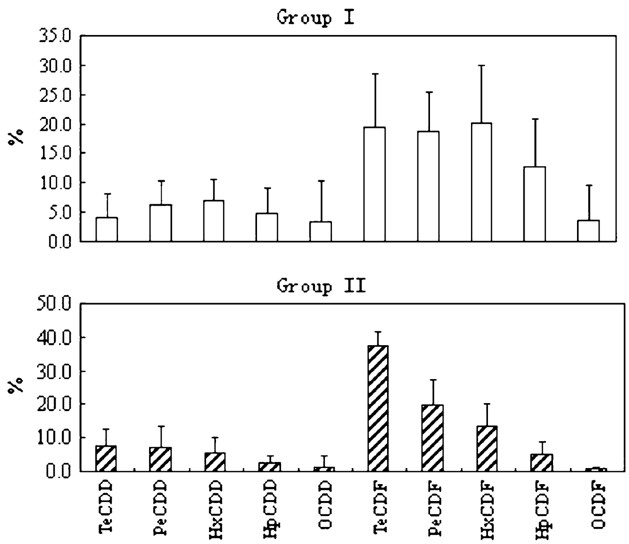

Fig. 3 PCDD/F fingerprint of the two main groups

POP concentrations in residue ash

In China, there are no precise regulations for the limits of PCDD/Fs in ash. Also the environmental protection agencies do not monitor the PCDD/F concentrations in residue ash. Actually, a quantity of organic pollutants existed in ash. Therefore, the measurement of POPs in residue ash was conducted in this study, and the results are shown in Tables 6 and 7. The concentration of PCDD/Fs ranged from 0.16 to 7.31 ng WHO-TEQ g^{-1} in fly ash, 3.95×10^{-3} to 0.17 ng WHO-TEQ g^{-1} in bottom ash of incinerators and 4.04 ng WHO-TEQ g^{-1} in ash from open burning, respectively. In addition, the concentration of PCBs ranged from 12.1×10^{-3} to 0.39 ng WHO-TEQ g^{-1} in fly ash, 0.08×10^{-3}–3.00×10^{-3} in bottom ash of incinerators and 0.94 ng WHO-TEQ g^{-1} in ash from open burning. The TEQ of PCBs was so tiny in fly ash that it was only approximately 3% compared to the value of PCDD/Fs. Also, there was a quantity of HxCBz in fly ash and bottom ash. More attention needs to be paid to fly ash treatment to prevent secondary pollution.

Distribution of PCDD/Fs output in MWI

Pollutants from incinerators can be emitted into the environment via several routes. The three major routes include fly ash, stack gas and bottom ash. In order to understand the

Table 6 PCDD/F concentrations in residue ash (ng g^{-1})

	FA1	FA2	FA3	FA4	BA1	BA2	BA3	BA4
2378TeCDD	0.23	0.64	27.5×10^{-3}	3.47×10^{-3}	3.54×10^{-3}	1.31×10^{-3}	0.14	n.d.
12378PeCDD	1.02	1.33	0.13	22.5×10^{-3}	5.76×10^{-3}	9.43×10^{-3}	0.43	0.48×10^{-3}
123478HxCDD	0.77	0.81	0.10	21.9×10^{-3}	12.0×10^{-3}	12.3×10^{-3}	0.24	0.53×10^{-3}
123678HxCDD	0.92	1.44	0.21	54.3×10^{-3}	17.2×10^{-3}	26.9×10^{-3}	0.31	1.01×10^{-3}
123789HxCDD	1.94	0.65	0.35	64.7×10^{-3}	35.2×10^{-3}	10.0×10^{-3}	0.57	1.36×10^{-3}
1234678HpCDD	4.97	8.12	0.82	0.34	0.10	0.22	1.42	4.98×10^{-3}
12346789OCDD	4.93	9.08	0.73	0.62	0.26	0.32	1.46	9.19×10^{-3}
2378TeCDF	1.65	4.42	0.33	34.4×10^{-3}	25.9×10^{-3}	8.95×10^{-3}	1.34	1.02×10^{-3}
12378PeCDF	3.59	4.69	0.58	64.7×10^{-3}	47.1×10^{-3}	12.8×10^{-3}	2.19	1.62×10^{-3}
23478PeCDF	5.56	7.57	1.16	0.13	0.23	32.5×10^{-3}	3.90	3.44×10^{-3}
123478HxCDF	7.06	5.57	1.01	0.12	96.5×10^{-3}	35.2×10^{-3}	3.03	3.11×10^{-3}
123678HxCDF	6.84	6.00	0.25	0.13	0.11	36.4×10^{-3}	2.92	3.06×10^{-3}
234678HxCDF	7.27	8.57	1.01	0.15	0.12	62.5×10^{-3}	3.68	3.73×10^{-3}
123789HxCDF	1.50	2.34	0.25	38.6×10^{-3}	26.7×10^{-3}	14.1×10^{-3}	0.94	1.08×10^{-3}
1234678HpCDF	22.9	21.8	2.77	0.54	0.44	0.23	8.17	11.9×10^{-3}
1234789HpCDF	2.92	3.32	0.35	60.0×10^{-3}	50.3×10^{-3}	22.2×10^{-3}	0.88	1.49×10^{-3}
12346789OCDF	6.31	10.5	0.76	0.26	0.37	0.13	1.78	7.51×10^{-3}
Total of 17 congeners	80.3	96.9	10.9	2.64	1.94	1.18	33.4	55.5×10^{-3}
WHO-TEQ	7.31	9.31	1.16	0.16	0.17	53.0×10^{-3}	4.04	3.95×10^{-3}

n.d. not detected

Table 7 PCB and HxCBz concentrations in residue ash (ng g^{-1})

	FA1	FA2	FA3	FA4	BA1	BA2	BA3	BA4
TeCB(#77)	1.26	3.76	0.54	93.5×10^{-3}	45.9×10^{-3}	26.9×10^{-3}	11.9	3.55×10^{-3}
TeCB(#81)	0.35	2.04	0.14	57.6×10^{-3}	6.08×10^{-3}	13.7×10^{-3}	1.53	0.65×10^{-3}
PeCB(#105)	0.39	0.55	0.26	74.0×10^{-3}	15.5×10^{-3}	16.3×10^{-3}	3.51	1.36×10^{-3}
PeCB(#114)	0.15	0.17	87.7×10^{-3}	35.2×10^{-3}	5.25×10^{-3}	6.6×10^{-3}	1.93	0.15×10^{-3}
PeCB(#118)	0.52	0.46	0.31	58.9×10^{-3}	17.2×10^{-3}	16.0×10^{-3}	2.16	2.55×10^{-3}
PeCB(#123)	92.9×10^{-3}	0.10	55.1×10^{-3}	16.9×10^{-3}	2.68×10^{-3}	1.55×10^{-3}	0.28	0.46×10^{-3}
PeCB(#126)	1.17	3.66	0.41	0.11	17.6×10^{-3}	28.2×10^{-3}	9.09	0.81×10^{-3}
HxCB(#156)	0.48	0.67	0.22	53.3×10^{-3}	7.15×10^{-3}	14.9×10^{-3}	2.74	0.22×10^{-3}
HxCB(#157)	0.29	0.59	0.11	54.1×10^{-3}	4.30×10^{-3}	10.1×10^{-3}	1.39	0.07×10^{-3}
HxCB(#167)	0.22	0.26	0.11	22.7×10^{-3}	2.95×10^{-3}	5.23×10^{-3}	0.74	n.d.
HxCB(#169)	0.52	2.25	0.13	54.1×10^{-3}	4.88×10^{-3}	15.0×10^{-3}	2.15	n.d.
HpCB(#189)	0.44	1.07	0.16	56.2×10^{-3}	6.67×10^{-3}	19.3×10^{-3}	1.41	0.17×10^{-3}
Σco-PCBs	5.88	15.6	2.52	0.69	0.14	0.17	38.8	9.99×10^{-3}
WHO-TEQ	0.12	0.39	42.1×10^{-3}	12.1×10^{-3}	1.82×10^{-3}	3.00×10^{-3}	0.94	0.08×10^{-3}
HxCBz	74.8	5.05	6.20	8.36	1.08	1.78	87.5	0.45

n.d. not detected

Table 8 Distribution of PCDD/F output in MWI (WHO-TEQ)

	Output	Dioxin concentration	Mass/volume flow per ton of waste	Dioxin flow (μg-TE/ton of waste)	Percent of total output (%)
MWI1	Stack gas	5.03 ng Nm^{-3}	15,000 Nm^3	75.5	22.4%
	Fly ash	7.31 ng g^{-1}	30 kg	219.3	65.0%
	Bottom ash	0.17 ng g^{-1}	250 kg	42.5	12.6%
MWI2	Stack gas	7.5 ng Nm^{-3}	15,000 Nm^3	112.5	27.6%
	Fly ash	9.31 ng g^{-1}	30 kg	279.3	69.1%
	Bottom ash	53.0×10^{-3} ng g^{-1}	250 kg	13.3	3.3%
HWI9	Stack gas	0.44 ng Nm^{-3}	10,000 Nm^3	4.4	44.9%
	Fly ash	0.16 ng g^{-1}	30 kg	4.8	49.0%
	Bottom ash	3.95×10^{-3} ng g^{-1}	150 kg	0.6	6.1%

portion of pollutant emissions in different substances, an assessment was conducted. The analyzed results are presented in Table 8. The results demonstrated that fly ash dominated in the output of dioxin, accounting for 67% of the total dioxin output on average. The distribution of PCDD/Fs in different substances was consistent with the UNEP default emission factors for MWI (class 3). On the other hand, the distribution of PCDD/Fs in MWI was similar to the results for MSWI [28]. Consequently, disposing of residue ash using an appropriate method is also essential.

Conclusion

In this study, the annual generation of MW in China was estimated. The amount of MW observed was huge, and it is increasing significantly. The characteristic PCDD/F emissions from MWI were investigated, and the organic contaminants in residue ash were also measured. It is essential to pay more attention to the disposal of fly ash and to implement much stricter regulations to prevent secondary pollution.

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