PCDDs/DFs and PBDDs/DFs EMISSIONS FROM CREMATORY

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Abstract

Dioxin emission guidelines for crematories were implemented in Japan in 2000, but the effects of the new law have not yet been fully evaluated. In this study, concentrations of polychlorinated dibenzo-dioxins/dibenzo-furans (PCDDs/DFs), co-planar polychlorinated biphenyls (PCBs), and polybrominated dibenzo-p-dioxins and dibenzofurans (PBDDs/DFs) in flue gases, fly ashes, and bottom ashes (mainly bone) from several crematories were measured as a follow-up investigation. Total concentrations ($O_2$12% normalized) of PCDDs/DFs in flue gases ranged from 5.3 to 540 ng/m^3_N, and toxic equivalent concentrations ranged from 0.00018 to 11 ng-TEQ/m^3_N. Based on these measurements, the average emissions released for each body being cremated was estimated to be 1600 ng-TEQ/body. This value is about two fifths of the value recorded in 1999 which suggests that the guidelines have been effective in reducing dioxin emissions. Conversely, toxic equivalent concentrations in fly ash ranged from 0.014 to 5.0 ng-TEQ/g. Higher dioxin concentrations were observed in facilities that used a heat exchanger as a flue gas cooling device; secondary formation of PCDDs/DFs on the surface of the heat exchanger may explain this difference. In addition, PBDD/DF concentrations were measured in four crematories, but were only detected in one; in this crematory, the PBDD/DF concentration ($O_2$12% normalized) was 0.031–0.045 ng /m^3_N and was almost negligible relative to the measured concentration of PCDDs/DFs.

Introduction

In Japan in 2007, 99.9% of dead bodies were cremated and this percentage is the highest in the world. According to demographic statistics[1], the number of mortalities is increasing so it follows that the number of cremations is also increasing. For religious reasons, emissions from crematories in Japan are not regulated by the Air Pollution Control Act or the Waste Management and Public Cleansing Act. However, it is necessary to examine toxic emissions from crematories to determine their environmental impact and to take measures to reduce or monitor them if necessary.

There have been some investigations into polychlorinated dibenzo-dioxin/dibenzo-furan (PCDD/DF) emissions from crematories[2-11] but only a limited number of extensive surveys have been carried out. In the detailed survey by Takeda et al.[6], the authors measured total concentrations of PCDDs/DFs in flue gases from 27 crematories and reported values ranging from 0.074 to 29.2 ng-TEQ(WHO98)/m^3_N with an arithmetic mean value of 2.9 ng-TEQ(WHO98)/m^3_N. Based on these results, the average emission quantity per dead body cremation was estimated to be 4200 ng-TEQ(WHO98)/dead body and total emissions were estimated to be 1.8–3.8 g-TEQ(WHO98)/year based on the registered number of cremations. The dioxin emission guidelines for crematories were prepared based on the results of this investigation by Takeda et al.[6] and their data were also used for the national inventory. In Japan, measures to reduce dioxin emissions have been applied to all known dioxin sources, which has resulted in a remarkable decrease in dioxin emissions. For many sources of dioxin emissions, the emission inventory is regularly updated with the newest data. However, the data for crematories are significantly out of date; the inventory has been using emissions data obtained almost 10 years ago. While it can be broadly assumed that dioxin emissions are indeed decreasing because of the countermeasures implemented since the guidelines were enacted (such as good combustion control and installation of advanced air pollution control devices), according to the national inventory, dioxin emissions from crematories are increasing due to the increase in the number of cremations, simply because the emissions data have not been updated.

Emissions of organobromine compounds from thermal processes have been of recent public concern[12]. In crematories, emissions of these compounds might be high depending on the contents of burial accessories

that are combusted with bodies. Therefore, it is necessary to investigate the actual emissions of organobromine compounds from crematories.
In this study, PCDD/DF, co-planar polychlorinated biphenyl (PCB), and polybrominated dibenzo-p-dioxin and dibenzofuran (PBDD/DF) concentrations in flue gas, fly ash, and bottom ash (mainly bone) from several crematories were measured as a follow-up survey to evaluate the effect of the emissions guidelines and update the PCDD/DF crematory emissions data in Japan.

Materials and Methods
Table 1 shows the configuration of 11 crematories and the sampling conditions. To evaluate the effects of the guidelines, implemented since 2000, we selected 5 crematories (Facility No.1, 2, 5, 6, and 7) that were constructed after 2000. In these particular facilities, bag filters were used as dust collectors and advanced air pollution control devices (APCDs) had been installed. Conversely, Facilities No.3, 4, 10, and 11 were not equipped with even a dust collector. All crematories had a series of one secondary combustion chamber to one main combustion chamber, and in all cases, except in Facility No.5 flue gases were cooled by air ejectors. Facilities No.1 and 5 used a heat exchanger for flue gas cooling. Natural gas and oil were used as axillary fuel in 6 and 5 of the crematories, respectively.

Table 1The configuration of 11 crematories and the sampling conditions

Facility No.	1		2		3		4		5		6	
Dust collector	Bag filter		Bag filter		-		-		Bag filter		Bag filter	
Advanced APCD	Catalyst		Activated carbon		-		-		Catalyst		Catalyst	
Flue gas cooling device	Heat exchanger + air ejecor		Air ejector		Air ejector		Air ejector		Heat exchanger + air ejecor		Air ejector	
Ventilation	Induced		Induced		Induced		Induced		Induced		Induced	
The number of secondary chambers connected to flue gas treatment line	2		2		2		1		2		2	
Fuel	Natural gas		Natural gas		Kerosene		Kerosene		Natural gas		Natural gas	
Experimental No	1	2	1	2	1	2	1	2	1	2	1	2
Cremation time	42	46	68	61	71	66	90	71	58	57	66	59
Age	81	83	69	87	84	65	74	90	64	75	91	79
Sex	female	male	male	male	female	male	female	female	female	female	female	female

Facility No.	7		8		9		10		11	
Dust collector	Bag filter		Electrostatic Precipitator		-		Screen		-	
Advanced APCD	Activated carbon		-		-		-		-	
Flue gas cooling device	Air ejector		Air ejector		Air ejector		Air ejector		Air ejector	
Ventilation	Induced		Induced		Induced		Induced		Induced	
The number of secondary chambers connected to flue gas treatment line	2		3		1		1		1	
Fuel	Kerosene		Natural gas		Kerosene		Natural gas		Kerosene	
Experimental No	1	2	1	2	1	2	1	2	1	2
Cremation time	48	68	64	45	60	63	59	64	85	64
Age	98	85	66	80	75	88	93	91	60	67
Sex	female	female	male	female	female	male	male	female	male	male

Sampling of flue gas was carried out twice for each crematory. Flue gas was sampled throughout a cremation, from ignition of the secondary burner to extinction of the main burner. During the sampling period, concentrations of dust, O_2, CO, CO_2, and NO_x were measured simultaneously. Flue gas temperature

was measured and the age and sex of the cremated bodies were recorded. Bottom ash (mainly bone) was sampled at all crematories except for Facility No.8, and fly ash was sampled at all facilities with the exception of Facilities No.1, 2, 5, 6, 7, and 9.
The concentrations of PCDDs/DFs and co-planar PCBs were measured in all sampled materials, wheareas PBDDs/DFs (tetra to octa) were only measured in samples from Facilities No.1, 2, 3, and 4. Sampling and analysis of PCDDs/DFs and co-planar PCBs were based on the Japanse standarad method, JIS K0311. Analysis of PBDD/DF concentrations followed the manual of the Ministry of Environment[13]. The notes on the specific matter such as cremation time in each crematory refer to our previous works[6]. This study used the toxicity equivalent factors (TEFs) for PCDDs/DFs and co-planar PCBs proposed by WHO/IPCS in 2006.

Results and Discussion

PCDDs/DFs and co-planar PCBs concentration

The results of the analysis of PCDD/DF and co-planar PCB concentrations in flue gas, bottom ash, and fly ash are shown in Table 2. Total concentrations ($O_2$12% normalized) of PCDDs/DFs and co-planar PCBs ranged from 4.7 to 540 ng/m^3_N, and toxic equivalent (TEQ) concentrations range from 0.00005 to 11 ng-TEQ/m^3_N. The average TEQ concentration was 0.88 ng-TEQ/m^3_N. In Facilities No.1, 2, 5, 6, 7, 8, and 9, the concentrations in both measurements were lower than 0.1 ng-TEQ/m^3_N. Facilities No.1, 2, 5, 6, and 7 were constructed since 2000, a catalytic destruction system for dioxin and NO_x had been installed in Facilities No.1, 5, and 6, and an activated carbon adsorption system had been implemented in Facilities No.2 and 7.

Table2 The results of the analysis of PCDD/DF and co-planar PCB concentrations in flue gas, bottom ash, and fly ash

Crematory		Cremation time (min)	Gas temperature (°C)	Dry gas volume (m^3_N/ h)	PCDDs/DFs+co-PCBs						Dust [a] (g / m^3_N)	CO [a] (ppm)	NOx [a] (ppm)	O_2 (%)
					Flue gas [a]		Bottom ash		Fly ash					
					ng / m^3_N	ng TEQ / m^3_N [b]	ng / g	ng TEQ / g [b]	ng / g	ng TEQ / g [b]				
No.1	1	42	150	9,210	16	0.096	0.020	0.0000033	520	13	<0.003	<20	100	17.9
	2	46			4.7	0.000053					<0.009	<30	50	20.0
No.2	1	68	110	14,600	9.0	0.0059	0.012	0.00000035	71	1.4	0.028	120	84	19.7
	2	61			18	0.012					<0.005	35	120	19.2
No.3	1	71	220	6,210	75	1.1	0.18	0.0033			0.17	260	84	18.4
	2	66			41	0.62					0.14	360	130	18.6
No.4	1	90	580	3,250	54	1.0	0.044	0.00000062			0.041	250	91	15.0
	2	71			5.6	0.084					0.064	64	99	15.2
No.5	1	58	140	9,160	12	0.011	2.40	0.042	310	5.0	<0.008	26	86	19.8
	2	57	150	7,780	5.3	0.00018					<0.006	56	110	19.4
No.6	1	66	86	13,900	12	0.0036	0.12	0.00055	9.1	0.10	<0.007	<4	97	19.6
	2	59			17	0.010					<0.006	<3	120	19.5
No.7	1	48	88	14,200	14	0.025	0.029	0	37	0.61	0.005	<20	120	19.1
	2	68			5.5	0.0037					0.005	33	100	19.1
No.8	1	64	170	11,200	25	0.062					0.032	<8	110	19.0
	2	45	140	9,880	14	0.059					0.032	32	100	19.3
No.9	1	60	290	4,560	9.4	0.089	0.12	0.00067			0.13	<30	110	18.1
	2	63	300	3,590	10	0.10					0.16	<10	130	17.9
No.10	1	59	250	3,700	540	11	0.02	0.00000035	1.1	0.014	0.15	180	84	17.4
	2	64	230	4,860	86	1.7					0.10	<40	93	18.1
No.11	1	84	400	4,130	84	1.6	0.31	0.0044			0.19	270	82	15.8
	2	64			85	1.7					0.17	77	110	16.6
Maximum		90	580	14,600	540	11	2.40	0.042	520	13	0.064	360	130	20.0
Minimum		42	86	3,250	4.7	0.000053	0.012	0	1.10	0.014	<0.003	<3	82	15.0
Median		64	170	7,780	15	0.073	0.080	0.00028	54	1.0	0.032	34	100	18.8
Arithmetic mea		62	240	8,020	52	0.88	0.32	0.0051	160	3.4	0.066	88	100	18.3
Geometric mear		61	220	7,040	21	0.55	0.081	0.000073	40	0.65	0.028	42	99	18.2

a PCDDs/DFs+co-PCBs, dust, CO and NO_x concentrations were normalized by 12% of O_2.
b The TEQ concentrations were calculated using WHO2006-TEF.

Conversely, at least one measurement in each of Facilities No.3, 4, 10, and 11 was higher than 1 ng-TEQ /m^3_N. These facilities were constructed before 2000 and, with the exception of Facility No.10, did not use any dust collectors. Although Facility No.10 was equipped with a simple screen, the dust concentration was not sufficiently low for effective dust removal. Specifically, the first sampling at Facility No.10 recorded a total dioxin concentration of 11 ng-TEQ/m^3_N, which exceeds the value set in the guidelines (5 ng-TEQ/m^3_N for existing facility). Based on the concentrations of other gases, the combustion conditions during these sampling periods were not inferior to those during the first measurement at Facility No.11. One possible

explanation for this difference may be that *de novo* synthesis occurred on the surface of the flue gas duct or on the simple screen.
These results emphasize the importance of efficient dust removal, using a bag filter or electrostatic precipitator, in preventing the emission of dioxins. However, dioxin concentrations varied even among new facilities. The structure of Facility No.9 was of the old type and similar to that of Facility No.11, but concentrations of dioxins in emissions were low. In other words, while the effect of including an APCD in the crematory structure is significant, combustion conditions are also important because PCDD/DF concentrations are influenced by many factors such as the dead body and burial accessories.
TEQ levels in bottom ash were very low and ranged from 0.00000035 to 0.042 ng-TEQ/g. Conversely, TEQ levels in fly ash collected from dust collectors ranged from 0.014 to 15 ng-TEQ/g. The concentrations in fly ash in Facilities No.1 and 5 exceeded the regulations on TEQ concentrations in solid waste incineration fly ash (3 ng-TEQ/g). According to Table 2, the CO concentrations in both facilities were relatively low, which suggests that the combustion conditions were sufficient. Because the heat exchanger is used as a gas cooling device in these facilities, secondary formation may have occurred.
In this study, fewer males were cremated than females, but age and sex had no influence on the total concentration of PCDDs/DFs.

PBDDs/DFs concentration
The PBDD/DF concentrations measured in flue gas, bottom ash, and fly ash are shown in Table 3. As indicated, PBDDs/DFs were not detected in three crematories. In Facility No.4, PBDD/DF concentrations ($O_2$12% normalized) were 0.031–0.045 ng/m^3_N, which is 1/100–1/2000 of the measured concentration of PCDDs/DFs in the same flue gas. The most significant contributing homologues were tetrabromodibenzofurans (TeBDFs).

Table3 The PBDD/DF concentrations measured in flue gas, bottom ash, and fly ash

Crematory		Cremation time (min)	Gas temperature (°C)	Dry gas volume (m^3_N / h)	PBDDs/DFs		
					Flue gas [a]	Bottom ash	Fly ash
					ng / m^3_N	ng / g	ng / g
No.1	1	42	150	9,210	N.D.	N.D.	0.072
	2	46			N.D.		
No.2	1	68	110	14,600	N.D.	N.D.	0.068
	2	61			N.D.		
No.3	1	71	220	6,210	N.D.	N.D.	
	2	66			N.D.		
No.4	1	90	580	3,250	0.045	0.0038	
	2	71			0.031		
Maximum		90	580	14,600	0.045	0.0038	0.072
Minimum		42	110	3,250	N.D.	N.D.	0.068
Median		67	185	7,710	0.038		0.070
Arithmetic mean		64	270	8,320	0.038		0.070
Geometric mean		63	210	7,220	0.037		0.070

Watanabe and Sakai[12] reviewed the environmental release and behavior of brominated flame retardants where PBDDs/DFs and PCDDs/DFs were introduced into municipal solid waste incinerator (MSWI) flue gas. According to this report[12], the average concentrations of PCDDs/DFs and PBDDs/DFs (tetra to hexa) from 75 incineration plants were 770 ng/m^3_N and 4.0 ng/m^3_N respectively. Measured PBDDs/DFs concentrations from the crematories in this study were much lower than these values for MSWIs, and the ratio of PBDDs/DFs to PCDDs/DFs was equal to or lower than that in MSWI flue gas. PBDDs/DFs were also detected in bottom ash only in Facility No.4 and were found in concentrations less than one tenth of those of PCDDs/DFs. PBDDs/DFs were detected in fly ash in both Facilities No.1 and 2, but were found in concentrations much lower than those for PCDDs/DFs. Although more data should be gathered to make more robust conclusions, and the detection limit for PBDDs/DFs is not excellent, it is assumed here that crematory emissions of PBDDs/DFs are negligible relative to those for PCDDs/DFs.

Total emission from all crematories in Japan

The total emissions from all active crematories in Japan were estimated using the following equations:

Total emissions (ng-TEQ/year) = Emission quantity (ng-TEQ/body) × the number of cremations (bodies/ year) (1)

Emission quantity (ng-TEQ/body) = TEQ concentration (ng-TEQ/m^3_N) × dry gas volume (m^3_N/h) × cremation period (h) / the number of cremations (bodies) (2)

The emission quantity, shown in Table 4, ranged from 0.041 to 16000 ng-TEQ/body. There was roughly a 0.4 million-fold difference in emission quantity between crematories. The arithmetic and geometric means were 1600 and 110 ng-TEQ/body, respectively. When we converted the emission quantity recorded in 1999[6] to one based on the TEFs proposed by WHO/IPCS in 2006, the arithmetic and geometric means were 4200 and 1900 ng-TEQ/body, respectively. Thus, the ratios of arithmetic and geometric mean dioxin emissions in 2007 to those in 1999 were roughly 2/5 and 1/20, respectively.

These mean values were multiplied by 1,193,967, the number of bodies cremated in 2007, and the total emissions were estimated to be 0.13–1.9 g-TEQ/year, expressed as the range of the arithmetical mean from a geometric mean. This is equal to 0.04–0.7% of the present PCDD/DF inventory in Japan.

Table4 The Emission quantity of PCDDs/DFs and Co-planar PCBs

Creamtory		Sampling time min	TEQ concentration ng-TEQ/m^3_N	Dry gas volume m^3_N/h	Emission quantity ng-TEQ/dead body
No.1	1	42	0.10	3200	210
	2	46	0.000053	1000	0.041
No.2	1	68	0.0059	2100	14
	2	61	0.012	2900	35
No.3	1	71	1.1	1800	2300
	2	66	0.62	1700	1100
No.4	1	90	1.0	2200	3400
	2	71	0.084	2100	210
No.5	1	58	0.011	1200	13
	2	57	0.00018	1400	0.24
No.6	1	66	0.0036	2200	8.6
	2	59	0.010	2300	23
No.7	1	48	0.025	3000	60
	2	68	0.0037	3000	13
No.8	1	64	0.062	2500	170
	2	45	0.059	1900	83
No.9	1	60	0.089	1500	130
	2	63	0.10	1200	130
No.10	1	59	11	1500	16000
	2	64	1.7	1600	2800
No.11	1	84	1.6	2400	5300
	2	64	1.7	2000	3700
Max					16000
Minumum					0.041
Median					130
Arithmetic mean					1600
Geometric mean					110

The United States Environmental Protection Agency reported that emissions of dioxins from crematories in Japan were 0.27 g-ITEQ/year in 2000[14]. The proportional contribution of dioxins from crematories to total dioxin emissions was 0.02%. Conversely, according to the European Union dioxin inventory, crematories released dioxins at the rate of 9–19 g-TEQ/year and contributed 0.2–0.8% of the total dioxin emissions in

the EU[15]. This suggests that crematory-derived dioxin emissions in Japan have been reduced to the same level as that in the US and the EU despite the much higher percentage of cremations in Japan (99.9% compared to 20–40% in the US and EU) and the fact that the absolute number of cremated bodies in Japan is more than 1.5 times that in the US and EU. Thus, we conclude that the dioxin emission guidelines for crematories in Japan have been successful and are justified. Also, the installation of advanced APCDs has been shown to exert a significant influence on the reduction of PCDD/DF emissions.

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References
1. The Ministry of Health, Labour and Welfare, 2008. Population Survey Report.
2. The Working Group of Subcommittee Air/Technology of the Federal Government Federal States Pollution Control Committee, Germany, 1994. Determination of requirements to limit emissions of dioxins and furan, 127.
3. Hutzinger O. and Fiedler H., *Chemosphere* 1993; 27: 121.
4. Eguchi S., Takeda N. and Sakai S., *Organohalogen Compounds*1996 ; 27: 127.
5.Takeda N., Takaoka M., Fujiwara T., Takeyama H. and Eguchi S., Chemosphere 2000 ; 40: 575.
6. Takeda N., Takaoka M., Fujiwara T., Takeyama H. and Eguchi S., Chemosphere 2001 ; 43: 763.
7. Kim DH., Kim YK., Kim SJ. and Ok G., *Organohalogen Compounds*2003 ; 63: 9.
8. Xu Z.,Wedman E., Bucher R., Petzoldt O., Sasamoto T. And Wilken M., *Organohalogen Compounds*2003; 63: 216.
9. Francois F., Bernaert P. and Baert R., *Organohalogen Compounds* 2003; 63: 252.
10. Livolsi B., Labrousse S., Baron P. and Fiani E., *Organohalogen Compounds* 2006 ; 68: 848.
11.Santarsiero A., Trevisan G., Cappiello G., Formenton G. and Dell'Andrea E., *Microchem. J.* 2005 ;79: 299-306.
12. Watanabe I. and Sakai S., *Environ. International* 2003;29: 665.
13. The Ministry of Environment, Draft manual on analysis method for PBDDs/DFs, 2007.
14.National Center for Environmental Assessment Office of Research and Development: U.S. Environmental Protection Agency An Inventory of Sources and Environmental Releases of Dioxin-Like Compounds in the United States for the Years 1987, 1995, and 2000, 2006 ;1-37.
15.North Rhine Westphalia State Environment Agency on behalf of the European Commission, Directorate General for Environment (DG ENV), Ulrich Quass, Michael Fermann, Günter Bröker: The European Dioxin Emission Inventory Stage II, 2000 ; 1 : 41.