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PCDD/Fs EMISSION FROM THE INCINERATION OF BULKY WASTES FOCUSSING ON ELECTRICAL APPLIANCES

Jugal BHURTEL[1], Takaya HIGUCHI[2], Akiko KUBOTA[3] and Masao UKITA[4]

[1] Member of JSCE, Dr. Eng., Senior Trainer, Training Institute for Technical Instruction (TITI) (Sanothimi, Bhaktapur, Nepal)
[2] Member of JSCE, Dr. Eng., Research Associate, Department of Civil Engineering, Yamaguchi University (2-16-1 Tokiwadai, Ube 755-8611, Japan)
[3] Member of JSCE, Graduate Student, Graduate School of Science and Engineering, Yamaguchi University
[4] Member of JSCE, Dr. Eng., Professor, Department of Civil Engineering, Yamaguchi University

In order to assess PCDD/Fs emission potentials of incinerated bulky wastes, combustion experiments were carried out with refrigerators, washing machines, bed mattresses/sofas, and TVs in a small-scale batch-type combustor. High concentrations of PCDD/Fs were detected in both soot and bottom ash samples. Soot samples from refrigerators, washing machines, bed mattresses/sofas, and TVs produced PCDD/Fs concentrations of 91, 73, 52, and 12 ng-TEQ/g, respectively; while 68, 9, 14, and 8 ng-TEQ/g, respectively were detected in bottom ash. PCDD/Fs emission factors calculated from the amount and concentrations of bottom ash were 3.9, 0.27, 0.90, and 0.14 μg-TEQ/kg of the four items, respectively.

Key Words: *bulky wastes, small-scale batch-type combustors, PCDD/Fs emission*

1. INTRODUCTION

The production of electric appliances has been gradually increasing for many years in Japan. TV has the highest production rate followed by refrigerators and air conditioners. According to an estimation, a typical Japanese household possesses 2.25 pieces of TV, 1.92 air conditioners, 1.21 refrigerators, and 1.08 washing machines, etc.[1]. After a certain period of application or possibly after the acquisition of new models, most of household electric appliances are discarded and they end up in municipal solid waste (MSW) streams as bulky wastes. Nagata et al.[1] cited that the average period of household application for TVs is around 12.9 years and that of refrigerators is 10.4 years. Likewise, average periods of application for washing machines and air conditioners are 6-7 years, and 10-13 years, respectively.

Consequently, the quantity of bulky wastes such as refrigerators, washing machines, TVs, air conditioners, music systems, etc., has increased considerably in MSW streams over the years in Japan. According to estimations made by various associations of individual electric appliances as cited by Nagata et al.[1),2)], as compared to 4.64 million pieces of TV disposed of as a bulky waste in 1991, the number increased to 7.37 million in 1998. For refrigerators, washing machines, and air conditioners, they were 3.32, 3.77, and 2.02 millions in 1991 and 3.92, 4.32, and 3.92 millions in 1998, respectively. Hence, TV accounts for almost 40% of the total number of those four bulky waste items, followed by washing machines with 23%, refrigerators 21% and air conditioners 16%. The total weight of these four bulky waste items was estimated to be 723 thousand tons in 1998[1)].

After the enactment of the 'Specified Household Electrical Appliances Recycling Law' in April 2001, the above-mentioned four bulky waste items are required to be recycled through the manufacturers or dealers. However, this study was conducted in the year 2000 when there were three options of their disposal. They were recycled in cities where material recovery facilities exist and in other cities and towns they were either crushed and landfilled or landfilled directly and to a lesser extent incinerated in small-scale batch-type incinerators. The exact number of such incinerators is not available, but it is speculated that around twenty such plants are operating in Japan. Some bulky wastes incinerators are as small as 540 to 7200 kg/day capacity. Such combustion facilities are characterized by small stack heights, coupled with complete absence of mixing systems and bulk of wastes. Air pollution control is achieved mainly due to the use of secondary burner, which eliminates the soot and fly ash escaping through the stack.

Bulky wastes, such as refrigerators, washing machines, air conditioners, TVs, bed mattresses, sofas, etc.- containing high amount of various plastics and metals, including polyvinylchloride

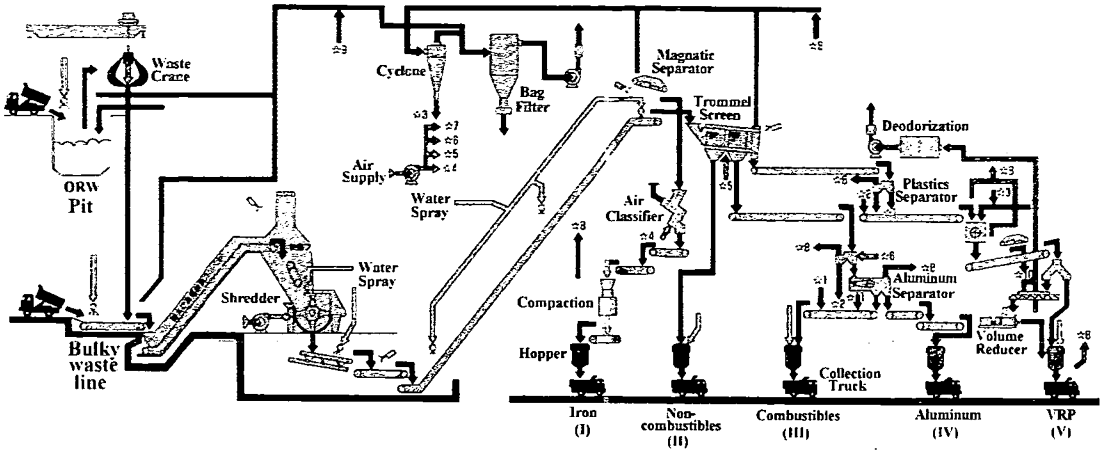

Fig. 1 Schematic diagram of bulky and other recyclable wastes recovery lines of MRF

(PVC) and copper- incinerated in such poorly regulated combustion facilities are likely to cause high emission of pollutants including polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs).

Thus, the main focus of this study is to evaluate the PCDD/Fs emission potentials of some bulky wastes incinerated in small-scale batch-type combustors and to assess the role of such poorly regulated combustors in overall dioxin emission scenario. Additionally, the outcome of the study will also be helpful in assessing the PCDD/Fs emission potential of the above-mentioned electrical appliances burned in uncontrolled combustion conditions, as in case of inadvertent fires of buildings, illegal open burnings, etc.

The study consists of two parts. In the first part, material compositions of three bulky waste items: refrigerators, washing machines and TVs, were determined in an experiment conducted at a material recovery facility. The experiments were especially focused on the determination of the amounts and types of plastics in bulky wastes items in order to understand their PCDD/Fs emission potentials. In the second part, bulky wastes were combusted in an existing batch-type small-scale incineration facility and PCDD/Fs concentrations were measured in soot and bottom ash collected after their combustion. The items were chosen on the basis of their relative abundance in bulky waste fraction of MSW.

2. MATERIALS AND METHODS

(1) Determination of the composition of bulky wastes

Investigations on the composition of bulky wastes were carried out at a local Material Recovery Facility (MRF). The schematic diagram of bulky and other recyclable wastes (ORW) recovery lines of the MRF is shown in **Fig. 1**.

Three bulky waste items, viz. refrigerators, washing machines, and TVs were selected for the experiment. Different and mostly old models of 20 pieces of refrigerators, 50 washing machines, and 50 TVs (of which 19 were wooden framed) were separately placed in the bulky refuse line of the MRF and passed through various processes such as rotary hammer, magnetic separators, rotary drum (trommel) screen, plastic materials separators (air classification), aluminum separators, and thermal plastic volume reducers. The MRF outputs segregated by the above processes are divided into five categories: iron (ferrous metals), aluminum, combustible wastes (CW), non-combustible wastes (NCW), and volume-reduced plastics (VRP). The VRPs are soft and light plastics separated in plastic substance separators by air classification and subsequently their volume is reduced thermally to make them compact for landfilling.

However, these outputs of MRF are very broad, contain various mixed waste items and do not provide complete information on the composition of input waste. Hence, further separation of each output category into various material fractions is necessary to know a detailed composition of each bulky waste item. Therefore, representative samples for further analysis were taken from the collection trucks of output categories. Each output was divided into four equal portions of manageable sizes and representative samples were taken from one of the portions for further separation into various fractions. Since the recycling of one bulky waste item results in five output categories as depicted in **Fig. 1**, the total number of samples was 15. Sizes of refrigerators, washing machines, and TVs used in

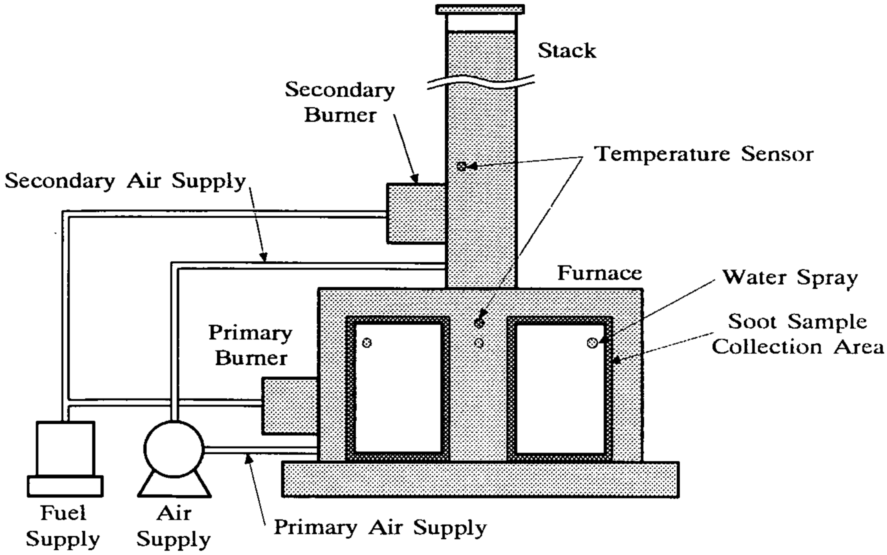

Fig. 2 Schematic diagram of full-scale incineration plant used for the combustion experiments

Table 1 Number of bulky wastes input and size of output samples (excluding motors/compressors)

		Refri.	WM	TV
Weight distribution of samples	weight (kg)	numbers		
	10-20		14	21
	20-30		25	12
	30-40	1	11	14
	40-50	1		3
	50-60	5		
	60-70	10		
	70-80	2		
	80-90	1		
Total numbers		20	50	50
Total input weight (kg)		1232	1158	1130
Total sample weight (kg)		16.3	22.4	28.5

the experiments as well as the total input weights and amounts of samples taken for further analysis are depicted in **Table 1**. Since water is sprayed during the separation and crushing of bulky waste items in the MRF, the samples were dried at ambient temperature approximately for one week and the dry weight was measured before being manually separated into various material fractions.

(2) Determination of plastics composition

To determine the composition of plastics materials, we first separated plastics fractions from all MRF output categories. We tried to produce maximum possible fractions using physical properties such as weight, hardness, appearance, color, and so on. The composition of separated plastics was then determined with visual observation as well as using a plastic material identification equipment (PLID-3, DKK.TOA Corporation), which produces absorption spectrums of near infrared ray when light is reflected from the plastic surface. Plastics are then identified from the spectrums produced. The working principle of similar identification equipment is given elsewhere[3]. Seven categories of plastics were identified in our experiments: polypropylene (PP), polystyrene (PS), polyethylene (PE), polyvinylchloride (PVC), polyurethane (PUR) foam, (Acrylonitrile Butadiene Styrene) ABS, and those, which could not be identified clearly, were categorized as others.

(3) Full-scale combustion experiments

There are different kinds of small-capacity batch-type incinerators being used in Japan. Some are equipped with primary and secondary burners, whereas some have only one burner to control the emission of flue gas. In this study, experiments were conducted in one of the existing full-scale bulky waste incineration plants with an average daily maximum waste combustion capacity of 2.5 tons. The plant, illustrated in **Fig. 2**, consists of both primary and secondary burners, air and fuel supply systems, water spray system, around 12m high centrally located stack, and a 20m^3 capacity furnace. Primary and secondary air supply systems are installed below the primary and secondary burners, respectively. There are two thermocouples. One is placed inside the furnace 30cm below the top of the inner wall and another above the secondary burner. Three automatic water sprays are located at equal distances along the length of the furnace as depicted in **Fig. 2**. During the combustion, secondary burner

Table 2 Details of bulky wastes applied in combustion experiments

Run	Item	Size (cm)	Number	Average weight (kg)	Total weight (kg)
I	Refrigerators				
	Large	170×63×53	15	50	
	Medium	120×50×53	2	40	875
	Small	110×46×45	3	15	
II	Washing Machines				
	Mechanical	71×80×44	28	20	
	Automatic (steel)	81×56×56	3	40	1080
	Automatic (plastics)	81×51×51	16	25	
III	Televisions				
	Large	-	6	70	
	Medium	-	60	24	2560
	Small	-	70	10	
IV	Spring mattress				
	Double bed	197×135×16	6	25	
	Single bed	197×98×16	6	15	
	Sofa				
	Large	-	6	40	554
	Small	-	7	7	
	Cushion	-	5	5	

operating at around 970°C is switched on initially and the primary burner starts its operation after 15 minutes. Once self-ignition starts in the furnace, auxiliary fuel supply of the first burner is terminated and it resumes automatically when the temperature inside the furnace becomes lower than 400°C but not later than four hours after ignition. All burner systems are switched off after four hours of operation. Heavy oil is used as the auxiliary fuel.

Refrigerators, washing machines, TVs, and spring bed mattresses and sofas of varying sizes and models were incinerated in the first, second, third, and fourth runs, respectively. Motors from washing machines, compressors and CFC gas from refrigerators were removed and the CRTs of TVs were broken prior to combustion. **Table 2** shows the details of each run. Twenty large, medium and small-sized refrigerators with a total weight of 875 kg were incinerated in the first run. To facilitate a complete combustion of internal portions, doors and other parts of refrigerators were removed. The combustion experiment in the second run was carried out with 47 mechanical and automatic washing machines (total weight- approx.1 ton). In the third run, 136 pieces of various sizes and models of TVs with a total weight of approximately 2.5 tons were incinerated in the plant, among them at least 19 TVs had wooden frames. Finally, different sizes of bed mattresses and sofas with the total weight of 554 kg were combusted. After the completion of combustion experiments, the auxiliary fuel supply through the burners was terminated after four hours of operation and water was sprayed many times to reduce the temperature inside the furnace using the automatic water sprinklers. Samples were collected next day morning when the furnace was already cool. However, in the first run, when refrigerators were incinerated, automatic water sprinklers were switched off due to technical problem and water was sprayed manually after the combustion next day morning. Temperatures were monitored continuously for the first five to six hours of combustion and then it was done occasionally.

After the completion of combustion, soot samples attached on outer surfaces of the furnace doorframe, as shown in **Fig. 2**, were collected using soft metal brush. The surfaces were properly cleaned with water and hard metal brush before each combustion experiment to minimize the effect of previous combustion. After the completion of combustion, bottom residues were collected and the volume was measured using plastic buckets of known capacity. Subsequently, the densities of the residues were measured to calculate the total weight. Large sized metal pieces included in residues were removed to obtain the actual bottom ash.

(4) Sample extraction and purification for PCDD/Fs analysis

Around 10 grams of soot, and dried and finely grind bottom ash samples were taken for PCDD/Fs analysis. Sample pretreatment and clean up were performed according to the 'Standard Method of Dioxins Measurement and Analysis of Samples from Waste Treatment' prepared by the Ministry of Health and Welfare, Japan in 1997. In brief, in the beginning, the sample is pretreated with 2M of hydrochloric acid for about one hour. After acid pretreatment, the sample is filtered and the liquid phase undergoes different steps of separation. The solid phase is extracted in accelerated solvent extractor (ASE). The clean up consisted of combined silica and aluminum oxide columns. Other procedures were performed as described previously[4]. Tetra through octa PCDD/Fs concentrations were determined using a medium resolution mass spectrometry and GC system. Instrumental conditions of GC-MS are presented elsewhere[4]. Co-planar PCBs were not measured in this study.

3. RESULTS AND DISCUSSION

(1) Composition of bulky wastes

The material outputs of MRF from bulky wastes are depicted in **Table 3**. Since water is used during various processes of separation in the MRF, the total material output of all bulky wastes was always greater than the input amount. The water content was estimated to be 5.2% to 6.8% of the total output weights. It should be noted that the input and output weights do not include the weights of compressors and motors in refrigerators and washing machines, which were removed before the wastes were placed in the recycling line. Similarly, the output weight does not include negligible amounts of dust and other finer particles accumulated in the bag filter and under the conveyer belts.

As **Table 3** shows, iron, combustible, and non-combustible fractions are the dominant outgoing waste categories. As expected, iron fraction occupies the maximum weight in refrigerator and washing machines. On the contrary, in TVs non-combustible wastes occupy more than half of the weight followed by combustibles and iron is only a small fraction. In all items, VRPs and aluminum are minor fractions ranging from 1-5% of the total output weight.

However, as mentioned earlier, output categories of MRF do not show a detailed composition of bulky wastes. For example, all outputs contained considerable amount of plastics, copper, and other such materials. Therefore, representative samples were taken from each output categories of MRF and further manual separation of samples was carried out in the laboratory.

Plastics were separated as hard, soft, polyurethane, styrene foam, and sponge; and metals as iron, aluminum, and copper. Others included rubber, integrated circuits (ICs), bag filter dust, and other finer particles, which could not be separated further. Detailed composition of each bulky waste is given in **Table 4**. Iron and plastics are predominant

Table 3 MRF outputs of bulky wastes (excluding compressors and motors)

MRF outputs	Refri.		WM		TV	
	kg	%	kg	%	kg	%
Non-combustible wastes (NCW)	250	19	200	16	670	58
Combustible wastes (CW)	400	31	470	38	320	28
Iron	570	44	530	43	150	13
Aluminum	10	1	9	1	10	1
VRP	70	5	23	2	13	1
Total output weight	1300	100	1232	100	1163	100
Water content (%)	5.2		6.0		6.8	

fractions in refrigerators and washing machines. The amount of glass is the largest fraction in TVs and that of other bulky wastes is negligible. Copper content of refrigerator is 5.8% and that of TVs and washing machines are around 2.2 and 1.2%, respectively. The plastic, iron, and fiber contents of spring bed mattresses are 10, 55, and 35%, respectively[5]. Among the plastics, the percentage of hard plastics is the biggest fraction in all bulky wastes.

(2) Composition of plastics in bulky waste items

Composition of plastics in electrical appliances, as depicted in **Table 5,** indicates high content of PP, PS and ABS. The predominant plastics in washing machines and TVs are PP and PS, respectively. Refrigerator consists of a considerable amount of ABS and PUR foam (32 and 20%, respectively). The PVC contents of refrigerators, washing machines, and TVs are approximately 9, 3, and 2% of the total amount of plastics in each of them, respectively. Hence, the overall PVC contents in an average refrigerator, washing machine and TV are approximately 4, 1.3, and 0.5%, respectively.

(3) Bulky wastes combustion

In all experiments, as shown in **Fig. 3**, combustion temperature increased steadily but remained below 400°C during the entire period of active combustion, which means that the first burner had to work throughout the combustion period. Especially in case of mattresses/sofas and TVs it shows unusually low combustion temperatures. This could be due to several factors. The first thermocouple in the furnace **(Fig. 2)** is located on the wall surface and not at the center of the furnace, and it is too high from the region where actual combustion takes place (e.g., furnace floor and area near the primary burner). Hence, the temperature profiles depicted in **Fig. 3** might not represent the actual burning condition. They could be useful only for relative comparisons among four experiments and they should not be used for the assessment of bulky

Table 4 Detailed composition of bulky wastes (excluding the weight of motors/compressors) (wt.%)

Items	Fractions	Plastics: HP	SP	PUR	StyF	Sponge	Al	Fe	Cu	Glass	Wood	Others: Rubber	IC	Others	Dust
		%													
Refrigerators	NCW	6.25	0.06	4.07	0.10	0.08	0.28	1.31	3.05	0.00	0.00	0.15	0.04	0.73	3.12
	CW	22.38	0.10	2.38	0.44	0.09	0.82	0.43	2.13	0.00	0.00	0.62	0.14	1.16	0.08
	Iron	0.91	0.00	0.01	0.00	0.01	0.00	42.28	0.54	0.00	0.00	0.04	0.00	0.03	0.04
	Al	0.04	0.00	0.01	0.01	0.00	0.61	0.00	0.06	0.00	0.00	0.02	0.00	0.02	0.00
	VRP	1.63	0.08	2.73	0.39	0.13	0.05	0.00	0.03	0.00	0.00	0.00	0.00	0.14	0.19
	Total			**41.9**			**1.8**	**44.0**	**5.8**	**0.0**	**0.0**		**6.5**		
Washing machines	NCW	8.16	0.09	0.05	0.01	0.11	1.95	2.40	0.40	0.61	0.00	0.11	0.24	0.22	1.89
	CW	33.03	1.28	0.00	0.00	0.00	1.23	0.62	0.43	0.00	0.00	0.67	0.55	0.26	0.09
	Iron	0.86	0.01	0.00	0.00	0.00	0.00	41.32	0.32	0.00	0.00	0.23	0.00	0.15	0.12
	Al	0.12	0.00	0.00	0.00	0.00	0.60	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00
	VRP	1.24	0.11	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.03	0.02	0.05	0.42
	Total			**45.1**			**3.8**	**44.3**	**1.2**	**0.6**	**0.0**		**5.1**		
TV	NCW	5.51	0.04	0.00	0.00	0.01	1.05	1.35	0.35	32.49	3.89	0.10	2.19	0.03	10.59
	CW	14.71	0.04	0.00	0.00	0.00	0.70	1.40	1.53	1.76	5.63	0.01	1.69	0.01	0.04
	Iron	0.30	0.00	0.00	0.00	0.00	0.00	9.08	0.32	0.00	0.01	0.00	0.14	3.04	0.01
	Al	0.02	0.00	0.00	0.00	0.00	0.68	0.13	0.01	0.00	0.00	0.00	0.02	0.00	0.00
	VRP	0.23	0.04	0.00	0.00	0.00	0.00	0.00	0.02	0.01	0.62	0.00	0.02	0.02	0.15
	Total			**20.9**			**2.4**	**12.0**	**2.2**	**34.3**	**10.2**		**18.1**		

HP- hard plastics, SP- soft plastics, PUR- polyurethane, StyF- Styrene foam, IC- integrated circuit.

Table 5 Composition of plastics in bulky wastes (wt.%)

Plastics	Refrigerators	WM	TV
PP	19.6	90.4	4.5
PS	12.1	2.5	78.9
ABS	31.8	1.4	7.8
PVC	9.3	2.8	2.1
PUR	20.3	0.1	-
PE	1.9	0.5	1.3
Others	5.0	2.2	5.5

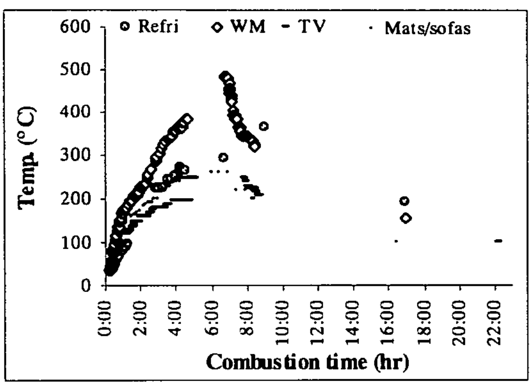

Fig. 3 Change in temperatures near furnace wall

wastes incinerators. Change in temperatures as indicated by the secondary thermocouple were in the range of 900 to1000°C in all experiments, which is more realistic because a complete combustion of all four items was observed as suggested by physical appearances and reduced volumes. Relatively higher temperature was achieved during the combustion of refrigerators and washing machines due to higher content of plastics. In case of TVs and mattresses/sofas, the temperature achieved was lower, which might be due to low plastics and high amount of glass in TVs and the use of flame-retardants in TVs and mattresses. Hence, more auxiliary fuel was needed for their combustion. Usually temperature inside the furnace was in the range of 80 to 100°C, when combusted bulky wastes were removed from furnace and samples were collected in the next day morning. However, since automatic water sprinklers were not used in refrigerator combustion, the furnace temperature in the first run remained at around 200°C even after the completion of the run.

(4) Total PCDD/Fs and TEQ values of samples

Total PCDD/Fs and toxicity equivalent (WHO-TEQ)[6] values of all samples are presented in **Fig. 4**. In general, as expected, soot samples produced higher concentrations of PCDD/Fs as compared to bottom ash samples. High TEQ values were detected in soot samples of refrigerators, washing machines, and bed mattresses/sofas. Maximum TEQ of 91 ng/g was detected in refrigerators soot sample, while minimum was detected in TVs soot with 12 ng-TEQ/g. Similarly, 73 and 52 ng-TEQ/g were detected in soot samples of washing machines and mattresses/sofas.

Unlike municipal solid waste bottom ash, where PCDD/Fs concentrations are negligible as compared to fly ash, the bottom ash from bulky waste combustion is found to contain a considerable amount of PCDD/Fs. As compared to PCDD/Fs concentrations in the bottom ash of washing machines, mattresses/sofas and TVs (9, 14, and 8 ng-TEQ/g, respectively), significantly higher concentrations were detected in refrigerator bottom ash, which produced almost 68 ng/g of TEQ. One possible explanation of generally higher concentration of PCDD/Fs in all bottom ash samples might be linked to the burning condition itself. Due to poor air supply system inside the furnace, all soot and fly ash produced during the combustion could not possibly escape the furnace via stack. Instead, a part of them might have remained on the furnace surfaces and floor. Hence, the presence of soot particles in bottom ash, which led to the detection of higher PCDD/Fs concentrations in bottom ash samples, cannot be ruled out. In case of refrigerator sample, another possible reason of higher PCDD/Fs concentration in samples might be related to high contents of both PVC and copper in incinerated refrigerators. A correlation between material content and PCDD/Fs emission is shown in Section **3. (6)**.

All toxic congeners were detected in samples. Very high dominance of 2,3,4,7,8-PeCDF was observed, which on average, made a contribution of 36% to the total TEQ values of samples. It was followed by 1,2,3,4,7,8-HxCDF with an average percentage contribution of 14%.

Total concentration of PCDD/Fs homologues also exhibited similar general trend as found in TEQ values. Maximum total PCDD/Fs of 3340 ng/g was detected in refrigerator soot sample and the minimum of 340 ng/g was obtained in the bottom ash of TVs. In soot samples, the dioxins to furan ratio ranged from 0.22 in mattresses/sofas sample to 0.31 in refrigerators. However, the soot sample of TV produced a higher than one ratio of 1.03. While in bottom ash, the ratio varied from 0.22 to 0.57. These kinds of dioxin to furan ratio are consistent with the results seen in MSW, hazardous waste incinerators, open burnings and various other combustion sources[4),7)].

The homologues profiles of each sample illustrated in **Fig. 5** indicate a high content of lower chlorinated PCDFs isomers. In all soot samples, the total concentration of TCDFs is much higher than that of other homologues. The next largest homologues are PeCDFs. In bottom ash, the homologue profiles are dominated by HxCDDs and HxCDFs, as well as PeCDDs and TCDFs. The

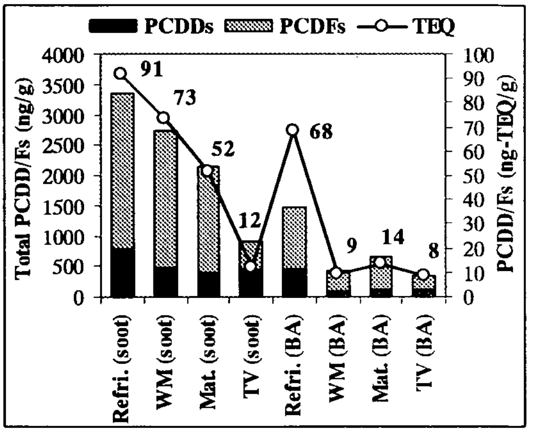

Fig. 4 PCDD/Fs and TEQ values of the samples

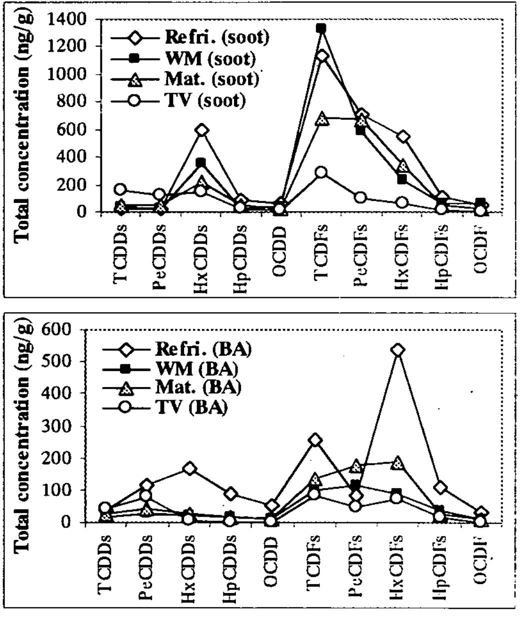

Fig. 5 Distribution of PCDD/Fs homologues in samples

PCDFs homologue profiles obtained in this study are not different from what was commonly found in other combustion sources[7),8),9)]. However, profiles of PCDD/Fs in soot samples are rare and existing literature generally focuses only on air samples. In one available example, Ruokojarvi et al.[10)] reported to find similar kind of profiles in soot samples collected in a simulated house fire experiment.

(5) Calculation of PCDD/Fs emission factors

The total amount of bottom residues obtained after the combustion varied widely. The highest amount of approximately 290kg or almost 11% of the initial weight was obtained in case of TVs, followed by refrigerator with 63kg or 7% of the input weight. Washing machines produced only 4% of residues, and mattresses/sofas generated 8% approximately. However, the bottom residues of washing machines and refrigerators contained a considerable amount of large metals, whereas that from TVs contained almost 86% of big glass pieces. Hence, such big unburned articles, which could not be crushed and ground by a pestle, were removed from the residues to obtain the actual amount of bottom ash as depicted in **Table 6**. This actual amount is then used for PCDD/Fs measurement.

Estimated emissions of PCDD/Fs per unit weight of bulky waste are calculated based on the concentrations of PCDD/Fs in bottom ash and total amount of bottom ash produced from the combustion. The PCDD/Fs concentrations in soot could not be utilized for soot-born emission factors calculation, because it was not feasible to reasonably estimate the actual amount of soot produced in the combustion. Besides, depending on the combustion condition, soot production varies widely, and, hence, emission factors in soot may not be useful as universal values. However, high concentrations of PCDD/Fs in soot could be an indicator of high health risks of incineration plant operators and the contamination of surrounding environment.

The emission factors were calculated using the following formula:

$$E = \frac{C_{BA} \times BA}{W_{BW}} \qquad (1)$$

where,

- E = estimated emissions per unit mass of bulky wastes incineration (ng-TEQ/kg)
- C_{BA} = concentrations of PCDD/Fs in bottom ash samples (ng-TEQ/g)
- BA = total weight of bottom ash (g)
- W_{BW} = total wt. of incinerated bulky wastes (kg)

The calculated emission factors are depicted in **Table 6**. One kg of refrigerator produced almost 4 μg PCDD/Fs TEQ, followed by mattresses/sofas with around 1 μg TEQ. As compared to these two items, emissions from washing machines and TVs were only 0.3 and 0.1 μg TEQs, respectively. Hence, the average emissions of PCDD/Fs from a single refrigerator, washing machine, TV and mattress/sofa are found to be 170, 6.3, 2.5, and 17 μg TEQs, respectively. These factors indicate considerably high PCDD/Fs emission potentials of bulky wastes.

Table 6 Estimation of total PCDD/Fs emission in bottom ash

Bulky wastes	Bottom residues (dry)		TEQ values		Emission factors (TEQ)	
	Coarse residues	BA	Conc.	Total	by wt.[a]	by item[b]
	kg		ng/g	mg	μg/kg	μg /piece
Refri.	13	50	68	3.4	3.9	170
WM	12	33	8.9	0.30	0.27	6.3
TV	248	41	8.5	0.35	0.14	2.5
Mats.	7	37	14	0.50	0.90	17

a Divided by total weight of combusted bulky wastes;
b Divided by number of combusted bulky wastes

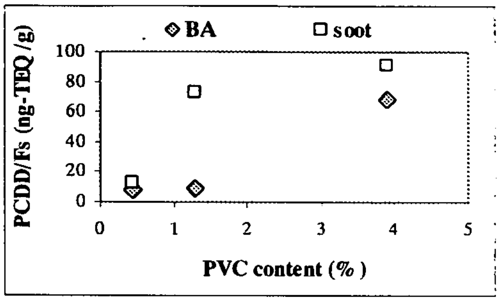

Fig. 6 Relationship between PVC contents and PCDD/Fs concentrations

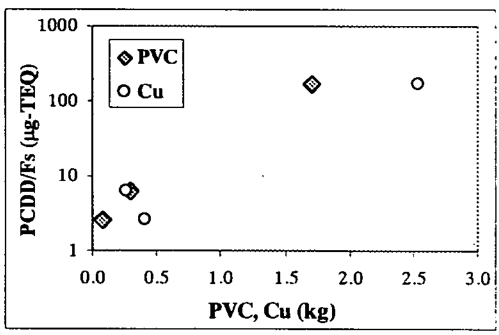

Fig. 7 Material contents vs. PCDD/Fs emissions from the combustion of one piece of bulky waste

(6) Relationship between material contents of bulky wastes and PCDD/Fs

As stated earlier, an average refrigerator, washing machine and TV contain approximately 4, 1.3, and 0.5% of PVC, respectively. With the increase in PVC content the emissions of PCDD/Fs also increased in both soot and bottom ash samples as **Fig. 6** indicates. Mattresses/sofas are not included in the analysis due to the lack of original material composition data. Similarly, the relationship between copper and PVC in each piece of bulky waste with the emission of PCDD/Fs from its combustion is shown in **Fig. 7**. These figures suggest that an increase in PCDD/Fs emission may be associated with increases in PVC and Cu contents of bulky wastes. It supports our earlier speculation that higher amount of PVC in refrigerators as compared to washing machines and TVs might be responsible for higher PCDD/Fs concentrations in its combustion samples. Many researches have already established that copper acts as a catalyst for PCDD/Fs formation during the combustion, while PVC has been found to act as a chlorine source[4), 11)]. In our previous lab-scale study, we demonstrated that the combustion of various plastics together with PVC can produce significant amount of PCDD/Fs in conditions generally found in small incinerators[4)]. Tatsuichi et al.[11)] have shown that dioxins concentrations in bottom ash increases exponentially with the increase in PVC contents in samples. A similar trend can be found in **Fig. 6** and **Fig. 7** as well. Although these observations are consistent with currently available theories of PCDD/Fs formation from wastes combustion, more conclusive explanation on the relationship between wastes composition and PCDD/Fs emissions are not possible from this study due to limited data points.

(7) Significance of the study

This study indicates that small-scale batch-type combustors may have a significant pollution potential. Lemieux et al.[7)] postulated that backyard burning of wastes in barrels represents the "worst-case-scenario" for PCDD/Fs formation that combines various factors, such as poor mixing, low combustion temperatures, oxygen-starved conditions, presence of HCl and/or chlorine and catalysts (e.g. Cu). The batch-type bulky wastes combustor employed in this study also appears to fit the above conditions. Although in such facilities bulky wastes are mixed together for combustion and not separately burned as done in this study, the mixing ratio depends solely on the experience of the plant operator and the availability of wastes items on a given day. Hence, it is possible that the mixing ratio frequently remains poor. Furthermore, inadequate air supply systems, large sizes and bulk of wastes ensure an incomplete combustion of wastes and a possibility of high emissions of pollutants. Furthermore, the concentration of PCDD/Fs in bottom ash is much higher than that usually found in municipal solid waste incineration plants bottom ash. Such high pollutant concentrations make it highly hazardous materials and should be handled with proper care to prevent leaching to underground water and protect the plant operators' health.

The study may provide some useful information on the source of PCDD/Fs during accidental fires of residential and commercial buildings. Such fires are under investigation in many countries as one of the major source of dioxin emission. Thomas and Spiro as cited by Carroll[12)] estimated the contribution of building fires in the US at 50 g-TEQ/year, while in Germany the estimation is around 81 g-TEQ (covering accidental fires of vehicles as well) and in Denmark it ranges from 0.5-20 g-TEQ as air emission and 1-30 g-TEQ in residues[13)]. Many researchers found high concentrations of PCDD/Fs in soot and ash samples obtained from real structural fires[12)]. Nevertheless, the origin of dioxins in such fires is investigated only focusing on the estimation of PVC materials burnt. From this study it can be speculated that electric appliances, as well as materials like mattresses and sofas may have a greater role in pollutants emissions during fires. High PCDD/Fs concentrations in soot samples indicate that even at partial burning conditions of accidental fires probably a considerable amount of dioxins is produced from these appliances. However, the results obtained in this study alone are not enough to estimate PCDD/Fs emission from fires involving electric appliances. Based on the complex set of variables that affect emissions of pollutants from accidental fires, further studies involving real fires samples would be warranted.

4. CONCLUSIONS

Emissions of PCDD/Fs from the combustion of bulky wastes, such as refrigerators, washing machines, TVs, and bed mattresses and sofas were investigated in this study. High concentrations of PCDD/Fs in soot samples were obtained from their combustion in a batch-type small-scale incineration facility. 91,73, 52, and 12 ng-TEQ/g of PCDD/Fs were detected in soot samples from the combustion of refrigerators, washing machines, bed mattresses/sofas, and TVs, respectively. Similarly, 68, 9, 14, and 8 ng-TEQ/g of PCDD/Fs were detected in their bottom ash samples. These values are much higher than generally found in MSW incineration bottom ash.

Emission factors were calculated from the total amount of bottom ash and concentrations of PCDD/Fs in them. 3.9, 0.27, 0.90, and 0.14 μg-TEQ were found to be emitted from the combustion of one kg of refrigerators, washing machines, bed mattresses/sofas, and TVs, respectively. It was observed that with the increase in PVC contents and copper in incinerated bulky wastes, the PCDD/Fs concentrations in soot and bottom ash also increased.

High amount of numerous types of plastics, including PVC, in all bulky wastes, and particularly in refrigerators and washing machines, coupled with poor burning conditions in the incinerator employed in this study may have a combined effect in producing higher amounts of PCDD/Fs in samples.

These results indicate high emission potential of bulky waste incineration. Although since April 2001, four major bulky wastes- refrigerators, washing machines, TVs and air conditioners- have been recycled through manufacturers, other bulky wastes like mattresses, sofas, and numerous small and medium sized household electrical appliances will continue to be incinerated in limited number of incinerators. More preventive measures should be implemented in plants designated for their combustion. Due to the application of secondary burner, the concentration of PCDD/Fs is generally low in flue gas emitted from such plants. However, as demonstrated here, bottom ash and soot may contain considerable concentrations of PCDD/Fs. High pollutants concentrations in soot necessitates long hours operations of secondary burners to ensure complete destruction of soot, gas, and fly ash-attached pollutants. Unusually high PCDD/Fs concentration in bottom ash makes it highly hazardous material and sites where they were previously landfilled should be reassessed. However, for a complete assessment of such small-scale combustors, measurement of PCDD/Fs in samples obtained from the incineration of mixed bulky wastes, including all kinds of appliances that are brought to the plants for combustion, is necessary.

Accidental structural fires have been on focus as a possible source of dioxins. High PCDD/Fs concentrations in soot obtained in this study may provide some information on the source of dioxins during such fires. Further studies involving samples from real fire cases will lead to more conclusive evidence.

Finally, due to extensive use of brominated flame retardants in various appliances, it is likely that the amount of dioxins from their combustion will increase significantly if brominated dioxins are included in the analysis.

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家電製品を中心とした粗大ごみの焼却に伴う PCDD/Fsの発生

Jugal BHURTEL・樋口隆哉・久保田彰子・浮田正夫

粗大ごみの焼却による PCDD/Fs 発生ポテンシャルを評価するために，小型バッチ式焼却炉で冷蔵庫，洗濯機，ベッド・マットレス・ソファ類およびテレビの焼却実験を行った．その結果，すすおよび焼却灰ともに高濃度の PCDD/Fs が検出され，冷蔵庫，洗濯機，ベッド・マットレス・ソファ類およびテレビのそれぞれについて，すすでは 91，73，52，12 ng-TEQ/g，焼却灰では 68，9，14，8 ng-TEQ/g となった．焼却灰の量と濃度から PCDD/Fs の発生原単位を求めたところ，それぞれの品目で 3.9，0.27，0.90，0.14 μg-TEQ/kg-焼却物となった．