



## Characteristics of Polychlorinated Dibenzop-dioxins/Dibenzofuran from Joss Paper Burned in Taiwanese Temples

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### ABSTRACT

Two types of furnaces (a mid-size furnace denoted as Furnace 1 and a small-size one denoted as Furnace 2) commonly used for burning joss paper, located in two temples in southern Taiwan, were selected for this study. With similar burning devices (semi-open systems), both furnaces can be used to burn joss paper with the aid of natural air supply through the inlets of furnace. In order to characterize peak emission of PCDD/Fs (usually on festival days) in the selected temples, the sampling campaigns were conducted during the festival period in September 2008. It was found that the mean total I-TEQ contents in the original (unburned) joss paper from Furnaces 1 and 2 were 1.41 and 4.51 ng I-TEQ/kg, respectively. The mean total I-TEQ content in the residue of burned joss paper from Furnace 1 (7.97 ng I-TEQ/kg) was approximately 15-fold that of Furnace 2 (0.490 ng I-TEQ/kg). OCDD was the most abundant congener in the joss paper and in the residues from burned joss paper. Moreover, the emission factor of total PCDD/Fs I-TEQ content of Furnace 1 (0.176 ng I-TEQ/g-feedstock) was one to two orders of magnitude higher than that of Furnace 2 (0.0203 I-TEQ/g-feedstock). This phenomenon is probably associated with the high chlorine content in the unburned joss paper of Furnace 1, and high exhaust temperature from burning joss paper in Furnace 1 (271°C) was possibly within a range that might highly promote de novo formation of PCDD/Fs. It is necessary for the investigated furnaces to install air pollution control devices in order to reduce the PCDD/F emission from joss paper burning.

**Keywords:** Polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDD/Fs); Joss paper; Temple; Metal; Congener profile.

### INTRODUCTION

Joss paper, mainly made of recycled paper and bamboo, is commonly used for ceremonial purposes in some Asian countries with Buddhist and Taoist religions. It is burned during ritual practice for deity worshipping among many families and in most temples in Taiwan. The ritual is generally practiced on the first and the fifteenth day of the Chinese lunar month. It produces non-stop heavy smoke during the long, slow, and incomplete combustion of joss paper (Lin *et al.*, 2002; Yang *et al.*, 2005; Chiang *et al.*, 2006). Pollutants such as particulate matters (PM), heavy metals, polycyclic aromatic hydrocarbons (PAHs), or others may be generated in incomplete combustion processes. High concentration and long exposure of PM from such pollutants may cause adverse human health effects (Lin *et al.*, 2008; Wang *et al.*, 2008; Xu *et al.*, 2008; Zhang *et al.*, 2008).

Joss paper burning has been found to be a significant source of toxic substances, e.g., PM (Fan and Zhang, 2001; Fang *et al.*, 2002), metals (Lau and Luk, 2001; Fang *et al.*, 2003), and PAHs (Chao *et al.*, 1997; Lin *et al.*, 2002; Lung and Hu, 2003; Fang *et al.*, 2004; Yang *et al.*, 2005; Yang *et al.*, 2007; Lin *et al.*, 2008;

Rao *et al.*, 2008). As a result, the health of people working in temples, visitors, and residents living nearby might be affected. With increasing environmental and health concerns, the public is paying more attention to toxic substances emitted from joss paper burning in temples.

Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are well known persistent organic pollutants which can be formed through combustion processes (Lee *et al.*, 2003; Lee *et al.*, 2004; Wang *et al.*, 2005; Kao *et al.*, 2006). Forest fires (Prange *et al.*, 2002), wood burning (Choudhry and Hutzinger, 1983; Wunderli *et al.*, 1996; Lemieux *et al.*, 2004), and the pyrolysis of paper fibers (Lee and Imagawa, 2006) may emit PCDD/Fs. One of our recent works has demonstrated that burning joss paper (with ~0.09% Cl) in temple furnaces is a significant source of PCDD/F emissions (Hu *et al.*, 2009). Their emission is associated with the high chloride content of paper fiber and wood. Joss paper is mainly made of materials such as recycled paper, bamboo, furniture manufacturing waste, and architectural wastes. Hence, it is possible that joss paper burning is a source of dioxin emissions.

Previous studies concerning thermal treatments of joss paper and incense focused mainly on PAHs and air pollutants (e.g., CO and PM). The PCDD/F emissions from joss paper burning have not been well studied. Therefore, the goal of this study is to characterize the PCDD/F emissions and profiles from burning joss paper at typical temples in Taiwan. Two furnaces for burning joss paper were investigated. The PCDD/F emission factors of

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the two selected furnaces are compared. In addition to PCDD/F analysis, elemental analysis was also performed for the (unburned) joss paper and the residues of burned joss paper. The results obtained provide useful information for the concerns of public health and governmental regulation about PCDD/F emission from joss paper burning.

## MATERIALS AND METHODS

### *Description of the Selected Temples and Furnaces*

The number of registered temples in Taiwan was 11,573 in 2008 (Taiwan MOI, 2008). The scales of temples and their burning furnaces are greatly different; additionally, many shrines and altars have not been registered yet. In order to investigate PCDD/F emissions from two common joss paper burning furnaces of typical Taiwanese temples, a popular temple equipped with a mid-size furnace (denoted as Temple T) and a small-size one located in a rural area (denoted as Temple F) were investigated. Both temples are typical Buddhist-Taoist temples, located in the Pingtung County in southern Taiwan. The Temple T, built in 1984, is one of the most famous Buddhist-Taoist temples as well as a popular tourist spot. Many pilgrims and visitors frequently visit the Temple T, thus the joss papers were usually used for deity worshipping and then fed into furnace continuously from 8:00 a.m. to 7:00 p.m. daily. Built in 1999 in a rural area and equipped with a small-size furnace for burning joss paper, the Temple F has significantly fewer pilgrims than the Temple T and thus the former has much lower joss paper burning than the latter. The Temple F shows burning patterns of some shrines and altars similar to those of small-scale temples dominant in Taiwan (Taiwan MOI, 2008).

Two joss paper furnaces (denoted as Furnaces 1 and 2), located beside two temples in southern Taiwan, were selected for this study. Beside Temple T, the tetragonal Furnace 1 has a volume of  $\sim 99 \text{ m}^3$  with a stack 18 m high and 0.58 m in diameter, and the area of the joss paper feeding aperture was  $0.13 \text{ m}^2$ . Furnace 2, located beside Temple F, was hexagonal having a volume of  $\sim 75 \text{ m}^3$  with a stack 6.2 m high and 0.30 m in diameter, and the area of the joss paper feeding aperture was  $0.09 \text{ m}^2$ . With similar burning devices, both furnaces were semi-open systems, and the burning of joss paper was conducted with natural air supplied through the inlets of furnace. In order to characterize peak emission of PCDD/Fs (usually on festival days) in the selected temples, the sampling campaigns were conducted during the festival period in September 2008.

During the sampling period, the burning temperature of each furnace was measured by a gas temperature sensor (thermocouple) directly from the joss paper feeding aperture to the core position of furnace. Due to the semi-open system used by both furnaces and the change of burning temperature from varied amounts of feeding papers, the burning temperature was measured at every half hour to truly represent the actual burning temperature. The burning temperatures of Furnaces 1 and 2 were around 520 and 200°C, respectively, because the quantity of joss paper burned in the former was considerably higher than the latter.

### *Composition of Joss Paper*

There are over ten kinds of joss paper with different shapes/sizes in Taiwan. 80% of the joss paper is imported from other Asian countries. Tiangong Jin (area =  $\sim 0.10 \text{ m}^2/\text{piece}$ ) and Shou Jin (area =  $\sim 0.022 \text{ m}^2/\text{piece}$ ), the two types of joss paper most commonly used for deity worshipping in Taiwan, were selected for burning tests in this study. The joss paper was made of recycled-paper and provided from the selected temples for burning tests performed in Furnaces 1 and 2. In general, visitors/pilgrims used the joss paper supplied from the temples, but few of them might use others when they visited the temples. The total weight of burned joss paper in each complete burning

cycle was recorded. There is no restriction for the amount of joss paper that can be burned by each pilgrim/visitor; thus, the quantity of burned joss paper differed slightly with number of pilgrim in each experiment. Around 142 kg of joss paper was burned in each complete burning cycle for Furnace 1, and the sampling duration lasted for approximately 100 min each time. For Furnace 2, joss paper was intermittently fed into the furnace and around 9 kg of paper was burned in a complete cycle of 40 min. Because the pilgrims of Temple F were fewer, the quantity of paper burned in Furnace 2 was smaller.

The chemical compositions of joss paper were determined using a microwave-assisted acid-digestion method equivalent to USEPA SW846-3050b (US EPA, 1988). In this procedure, each specimen (0.05 g) was digested by a CEM digester using 10 ml  $\text{HNO}_3$  (intra-analyzed quality, J.T. Baker) assisted by a microwave system at 170°C for 20 minutes to assure complete digestion of samples. Then, the obtained suspension was filtered through an ashless Whatman #41 filter and diluted to 50 ml using ultra-pure water (specific resistance  $\geq 18.3 \text{ M}\Omega\text{cm}$ ) for metal analysis. The relative standard deviations of duplicate analyses were 1.7-3.2%, below a control level of 5%. The concentrations of the elements, including Na, Mg, Al, Ca, Mn, Fe, Ni, Cu, Zn, Sr, Cd, and Pb were analyzed by inductively coupled plasma atomic emission spectrometry (Jobin-Yvon JY-38 Plus ICP-AES). The recoveries for the metals ranged from 89% (Zn) to 111% (Pb).

The determination of moisture, ash, and combustible contents of joss paper and residues were conducted according to ASTM standard E955. For moisture analysis, 0.5 g of sample was placed in an oven at  $105 \pm 1^\circ\text{C}$  until the weight of sample was constant. Another similar sample with the same weight was heated at  $800 \pm 50^\circ\text{C}$  for 3 hours to determine its ash content. Then, the mass percents of combustibles are calculated as:  $100 - (A - B)$ , where A is the mass percent of moisture, and B is the mass percent of ash. The chlorine ( $\text{Cl}^-$ ) of unburned and residues of burned joss paper were analyzed by ion chromatography, and the analysis followed the National Environment Analysis Method of Taiwan (NIEA R205.01C/NIEA W415.52B) referred to US EPA Method 300. The moisture, ash, combustible, and chlorine content of joss paper and residues of burned joss paper from the two furnaces are listed in Table 1. Noted the high moisture content of residues in Furnace 2 is attributed to its low burning temperature.

It was found that the chlorine content of the unburned joss paper from Furnace 1 (1.10 mg/g) was about 10-fold that from Furnace 2 (0.143 mg/g) (Table 1). As mentioned before, the joss papers, commonly used for worshipping in Taiwan and provided from the temples, were manufactured from some factories in Taiwan. Due to different sources of raw materials (i.e., pulp) used by joss paper manufacturers, the compositions (e.g., moisture, ash, combustible, and chlorine content) of the selected joss paper were different. It should also be noted that the use of additives (e.g. bleach, dyes, and paint) during manufacturing was possibly related to the chlorine content in joss paper. In addition, the high ash content (81.8% and 79.0% from Furnaces 1 and 2, respectively) of residues of burned joss paper indicates that the burning of joss paper is an incomplete combustion process, which is highly related to the PCDD/F emissions.

### *PCDD/F Sampling*

The PCDD/F samples were collected from stack flue gases according to the US EPA modified Method 23 (US EPA, 1991). The sampling train adopted in this study is comparable with that specified by the US EPA modified Method 5 (US EPA, 1986). The gas-phase PCDD/Fs were collected using XAD-2 resins while the particle-phase portion was collected by quartz fiber filters (diameter = 81 mm). Prior to sampling, XAD-2 resins were spiked with PCDD/F surrogate standards pre-labeled with isotopes. Each stack flue gas sampling was performed for approximately 100 and 40 min for Furnaces 1 and 2, respectively, at the flow

**Table 1.** Compositions of joss paper and residues from burned joss paper in the two furnaces (n = 3).

Proximate Analysis (%)	Joss paper				Residues			
	Furnace 1		Furnace 2		Furnace 1		Furnace 2	
	Content	RSD (%)	Content	RSD (%)	Content	RSD (%)	Content	RSD (%)
Moisture	3.98	0.43	4.30	6.2	0.486	1.8	4.47	3.1
Ash	7.87	5.5	4.87	12	81.8	3.7	79.0	10
Combustible	88.2	1.9	90.8	0.94	17.8	7.8	4.47	2.8
Ultimate Analysis (mg/g)								
Chlorine (Cl <sup>-1</sup> )	1.10	4.5	0.143	5.3	2.18	2.3	2.08	5.5

rates of 6-10 m/s during a complete joss paper burning cycle. To ensure that the collected samples were free completion of flue gas sampling, the samples were transported to the laboratory under refrigeration with temperature less than 10°C. The selected furnaces for burning joss paper were not pre-cleaned before sampling in order to represent the typical burning situation. Nevertheless, the furnace ash collectors were pre-cleaned before each batch of burning to eliminate the possible interference from residues.

Residues of burned joss papers were collected from the ash collector installed at the bottom of each temple furnace. The ash collector was pre-cleaned before each batch of burning to eliminate possible interference from residue collection. The sampling procedure was carried out according to the Taiwan NIEA R118.02B, referred to Minnesota regulation 7035.2910 in the US. Approximately 1.2 kg of residues was collected from the temple furnace and stored in sealed containers to eliminate effects of air circulation and humidity. Samples were spread on clean aluminum foils, dried naturally, and then mixed thoroughly prior to PCDD/F analysis.

#### Analyses of PCDD/Fs

Analyses of stack flue gas samples followed the US EPA modified Method 23 (US EPA, 1991), while those for PCDD/Fs in ash followed the US EPA Modified Method 1613A (US EPA, 1994a). All of the PCDD/F analyses were performed in the Super Micro Mass Research and Technology Center of Cheng-Shiu University, certified by the Taiwan EPA for PCDD/F analysis. During 2001-2004, this certified laboratory participated in the 6th-9th Round of the International Intercalibration Study (participant code: 57) held by the Department of Chemistry of Umea University, Sweden and was in good agreement in PCDD/F analysis with qualified laboratories in the world. Essentially, nine <sup>13</sup>C<sub>12</sub>-labeled internal standards (2,3,7,8-TeCDD, 1,2,3,7,8-PeCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDD, OCDD, 2,3,7,8-TeCDF, 1,2,3,7,8-PeCDF, 1,2,3,6,7,8-HxCDF, and 1,2,3,4,6,7,8-HpCDF) were added to each sample that was extracted for 24 h. Then, the extract was concentrated and treated with sulfuric acid, followed by a series of cleanup and fraction procedures. To check recovery, the standards (<sup>13</sup>C<sub>12</sub>-1,2,3,4-TeCDD and <sup>13</sup>C<sub>12</sub>-1,2,3,7,8,9-HxCDD) were added to the sample before PCDD/F analysis during the analysis process. Recoveries of <sup>13</sup>C<sub>12</sub>-labeled PCDD/F internal standards for the tetra- through hexa-chlorinated homologues ranged 81-108%, meeting the control limits of 40-130%, while those for hepta- and octa-chlorinated homologues were between 67% and 102%, fulfilling the criteria of 25-130%.

A high resolution gas chromatography with a mass spectrometer (HRGC/MS) was used to determine the concentrations of seventeen individual PCDD/Fs. The HRGC (Hewlett Packard 6970 Series gas, CA) was equipped with a DB-5 fused silica capillary column (L = 60 m, ID = 0.25 mm, and film thickness = 0.25 μm) and splitless injection (J&W Scientific, CA, USA). The oven temperature was programmed as follows:

initial temperature at 150°C (held for 1 min), increasing to 220°C at 30°C/min (held for 12 min), then to 240°C at 1.5°C/min (held for 5 min), and finally to 310°C at 1.5°C/min (held for 20 min). Helium was used as the carrier gas. The HRMS (Micromass Autospec Ultima, Manchester, UK) was equipped with a positive electron impact (EI+) source. The analyzer mode was set to ion monitoring with resolving power at 10,000. The electron energy and the source temperature were set at 35 eV and 250°C, respectively. Method detection limits (MDLs) for the seventeen individual PCDD/Fs were determined via the standard deviations obtained from the analyses of matrix-spiked samples. The MDLs of PCDD/Fs for unburned joss paper, residues of burned joss paper, and flue gas were 0.281-8.65, 0.233-3.63, and 0.271-12.7 pg, respectively. For the precision and reproducibility of analysis, the recoveries of PCDD/F precision and recovery (PAR) stock standards for each sample batch were 42-115%.

## RESULTS AND DISCUSSION

#### PCDD/Fs in the (Original) Unburned Joss Paper

Tables 2 and 3 list the mean PCDD/F content in the unburned joss paper and the residues of burned joss paper after burning from Furnaces 1 and 2, respectively. To assess toxicologically relevant information on PCDD/Fs, the I-TEQ values for PCDD/Fs were calculated using the contents of seventeen 2,3,7,8-chlorinated substitutes and their Toxicity Equivalency Factors (TEFs). The TEF value adopted in this study is referred to the "International" scheme (NATO/CCMS, 1998) because it reflects the toxic potency of each tested PCDD/F compound relative to the 2,3,7,8-TCDD congener. The means of the total PCDD/F and total I-TEQ contents (summation of seventeen individual PCDD/Fs and TEQs) in the unburned joss paper from Furnace 1 were 233 ng/kg and 1.41 ng I-TEQ/kg, respectively, lower than those from Furnace 2 (2043 ng/kg and 4.51 ng I-TEQ/kg, respectively). The PCDF/PCDD ratios in the unburned joss paper from Furnaces 1 and 2 were 0.0977 and 0.143, respectively, indicating that significantly more PCDDs were produced than PCDFs by the precursor reaction from burning joss paper. The high PCDD/F content in the unburned joss paper indicated that PCDD/Fs were possibly generated as by-products in manufacturing chlorine-bleached paper.

The congener profiles of the seventeen PCDD/Fs were selected to characterize the dominant species. Each selected congener was normalized by dividing its content by the sum of the seventeen PCDD/F congeners. The congener profiles of PCDD/Fs in the unburned joss paper and the residues of burned joss paper from Furnaces 1 and 2 are shown in Fig. 1. Among the PCDD/F species (Furnace 1 with a total content of 233 ng/kg and Furnace 2 with a total content of 2043 ng/kg), OCDD, OCDF, and 1,2,3,4,6,7,8-HpCDD were the top three dominant congeners in the unburned joss paper, implying that the higher chlorine substituted PCDD/Fs were predominant in the unburned joss paper.

**Table 2.** Mean PCDD/F contents (ng/kg-feedstock) in the joss paper and the residue from burned joss paper in Furnace 1, respectively (n = 3).

PCDD/F species	Joss paper		Residue	
	Mean	RSD (%)	Mean	RSD (%)
2,3,7,8-TeCDD	0.146	13	1.03	10
1,2,3,7,8-PeCDD	0.155	4.6	0.692	18
1,2,3,4,7,8-HxCDD	0.136	24	BD	--
1,2,3,6,7,8-HxCDD	0.777	22	0.264	23
1,2,3,7,8,9-HxCDD	0.419	27	BD	--
1,2,3,4,6,7,8-HpCDD	11.5	18	1.67	6.8
OCDD	199	18	15.8	12
2,3,7,8-TeCDF	3.55	15	20.9	13
1,2,3,7,8-PeCDF	0.312	18	7.14	9.5
2,3,4,7,8-PeCDF	0.451	20	7.05	11
1,2,3,4,7,8-HxCDF	0.345	22	1.57	12
1,2,3,6,7,8-HxCDF	0.278	24	1.93	10
1,2,3,7,8,9-HxCDF	0.352	36	1.62	13
2,3,4,6,7,8-HxCDF	BD	--	BD	--
1,2,3,4,6,7,8-HpCDF	2.59	31	3.29	13
1,2,3,4,7,8,9-HpCDF	0.309	46	0.12	173
OCDF	12.6	15	15.7	13
PCDDs	212	18	19.5	10
PCDFs	20.7	17	59.3	10
PCDFs/PCDDs	0.0977	9.0	3.06	10
Total PCDD/Fs	233	18	78.8	8.5
PCDDs (ng I-TEQ/kg-feedstock)	0.671	15	1.43	12
PCDFs (ng I-TEQ/kg-feedstock)	0.735	18	6.53	11
PCDFs/PCDDs	1.10	6.3	4.56	11
Total PCDD/Fs (ng I-TEQ/kg-feedstock)	1.41	16	7.97	11

BD: below detection limit.

#### PCDD/Fs in the Residues from Burned Joss Paper

After the joss paper was burned, the mean total-PCDD/Fs in the residues of burned joss paper from Furnaces 1 and 2 were 78.8 and 19.4 ng/kg, respectively, while those of total I-TEQ contents were 7.97 and 0.490 ng I-TEQ/kg, respectively (Tables 2 and 3). Although the total I-TEQ content in the residue of burned joss paper from Furnace 1 was approximately 15-fold that of Furnace 2, both were below the regulated hazardous industrial waste standard in Taiwan (1000 ng I-TEQ/kg) (Taiwan EPA, 2001). In addition, the total PCDFs I-TEQ concentration was higher than the total PCDD I-TEQ level in each furnace (Tables 2 and 3), revealing that PCDFs were the main contributor to total PCDD/F I-TEQ concentration in the residues.

The most abundant congener was OCDD (Fig. 1). It was found that PCDD concentrations increased with increasing number of chlorinated substitutes in Furnaces 1 and 2, whereas the concentration of PCDFs was elevated for the lower chlorinated congeners (tetra- and penta-) except for the OCDF congeners. These results are very similar to those of some previous studies, in which OCDD and OCDF dominated in PCDD/Fs in bottom ash from burning native and waste woods (Oehme and Müller, 1995; Wunderli *et al.*, 2000).

It was found that the I-TEQ content in residues of burned joss paper from Furnace 1 (7.97 ng I-TEQ/kg) was 4-fold that of unburned joss paper (1.41 ng I-TEQ/kg). However, an opposite trend was observed for I-TEQ content of residues and unburned joss paper (0.490 and 4.51 ng I-TEQ/kg) for Furnace the TEQ of PCDD/Fs increased after the joss paper was burned in Furnace 1. As mentioned before, additives (e.g., chlorine bleach, dyes, and

paint) in joss paper are likely responsible for differences in PCDD/F content. The chlorine in joss paper and the temperature of burning were possibly related to the increase of I-TEQ of PCDD/Fs from burning joss paper in Furnace 1. However, further research is warranted to investigate PCDD/F formation/reduction mechanisms when joss paper is burned in temple furnaces.

#### PCDD/F Concentrations in the Flue Gas

The mean PCDD/F concentrations of individual congeners in the flue gases of the two furnaces are shown in Table 4. The mean total-PCDD/F levels in the flue gases of Furnaces 1 and 2 were 27.1 and 2.14 ng/Nm<sup>3</sup>, respectively, while their corresponding total I-TEQ contents were 4.13 and 0.166 ng I-TEQ/Nm<sup>3</sup>, respectively. The PCDD concentrations increased with increasing number of chlorinated substitutes; thus, OCDD was the dominant PCDD/F in the flue gas. The total I-TEQ level of Furnace 1 was one to two orders of magnitude higher than that of Furnace 2. This phenomenon may be due to effects of chlorine in the unburned joss paper (Table 1) and the burning temperature on PCDD/F formation. Note that the chlorine content in the unburned joss paper of Furnace 1 was approximately 10-fold that of Furnace 2. The burning temperature of Furnace 1 (520°C) was higher than that of Furnace 2 (200°C), but still lower than those (e.g., 850°C) of some municipal solid waste incinerators, indicating that the partial PCDD/Fs in original joss paper might not be destroyed after burning. Additionally, the exhaust temperature for burning joss paper from Furnace 1 (271°C) was possibly within a range that might highly promote de novo synthesis of PCDD/Fs, resulting in high PCDD/F concentrations

**Table 3.** Mean PCDD/F contents (ng/kg-feedstock) in the joss paper and the residue from burned joss paper in Furnace 2, respectively (n = 3).

PCDD/F species	Joss paper		Residue	
	Mean	RSD (%)	Mean	RSD (%)
2,3,7,8-TeCDD	0.0630	2.2	0.0460	93
1,2,3,7,8-PeCDD	0.104	15	0.106	92
1,2,3,4,7,8-HxCDD	0.268	12	0.0330	173
1,2,3,6,7,8-HxCDD	2.87	28	0.178	62
1,2,3,7,8,9-HxCDD	1.08	22	0.151	89
1,2,3,4,6,7,8-HpCDD	119	32	1.84	50
OCDD	1663	17	12.8	58
2,3,7,8-TeCDF	1.89	5.0	0.574	98
1,2,3,7,8-PeCDF	0.148	2.5	0.228	126
2,3,4,7,8-PeCDF	0.194	4.3	0.366	109
1,2,3,4,7,8-HxCDF	1.43	12	0.201	91
1,2,3,6,7,8-HxCDF	0.425	10	0.191	92
1,2,3,7,8,9-HxCDF	0.585	14	0.228	95
2,3,4,6,7,8-HxCDF	0.00875	173	BD	--
1,2,3,4,6,7,8-HpCDF	33.2	16	0.629	40
1,2,3,4,7,8,9-HpCDF	3.11	13	0.134	53
OCDF	215	20	1.65	68
PCDDs	1787	18	15.2	53
PCDFs	256	19	4.20	45
PCDFs/PCDDs	0.143	2.6	0.277	50
Total PCDD/Fs	2043	18	19.4	47
PCDDs (ng I-TEQ/kg-feedstock)	3.39	22	0.167	79
PCDFs (ng I-TEQ/kg-feedstock)	1.12	12	0.323	102
PCDFs/PCDDs	0.330	11	1.94	60
Total PCDD/Fs (ng I-TEQ/kg-feedstock)	4.51	20	0.490	91

BD: below detection limit

in the flue gases of Furnace 1.

As shown in Table 4, the concentrations of total PCDF and total PCDF I-TEQ in the flue gas of Furnace 1 were both higher than those of total PCDD and total PCDDs I-TEQ, respectively, indicating that PCDFs were the main contributor to total PCDD/F and total PCDD/F I-TEQ for the Furnace 1. A possible reason is that the de novo synthesis of PCDD/Fs might occur during the cooling down process and that PCDFs were superior to PCDDs in the PCDD/F formation. However, the PCDF/PCDD ratio in the flue gas of Furnace 2 was 0.743, which is less than 1 (Table 4). This result differed from that of Furnace 1, revealing that more PCDDs were produced than PCDFs from precursor reactions in the burning process. It is interesting to note that although PCDDs accounted for a major fraction of total-PCDD/F levels, the total PCDD I-TEQ content was lower than that of total PCDF I-TEQ content, which is due to the different toxicity equivalency factor values of PCDD/F congeners.

PCDD/F emission factors of flue gas are based on the sampling volumes and weights of feeding samples. The emission factors of both total PCDD/F and total PCDD/F I-TEQ contents in the flue gas of Furnace 1 were 1.16 ng/g-feedstock and 0.176 ng I-TEQ/g-feedstock, respectively, while those of Furnace 2 were 0.271 ng/g-feedstock and 0.0203 ng I-TEQ/g-feedstock, respectively. These levels are two to three orders of magnitude higher than those of industrial wood combustors in the US (US EPA, 2001) and also one to two orders of magnitude higher than those of forest fires in the US (Gullett and Touati, 2003).

Furthermore, the above results also show that about 93% of total-PCDD/F was found in the flue gas; the remaining 7% was

absorbed on the surface of bottom ashes. It has been reported that transition metals such as Cu, Fe, and Zn (having a positive correlation with PCDD/F concentration) can catalyze the formation of PCDD/Fs during combustion processes; among these metals, copper makes a significant contribution to the PCDD/F formation (Olie et al., 1998). Therefore, the presence of copper (37.2 mg/g) and other minor metals (Fe and Zn) in the unburned joss paper from Furnaces 1 and 2 might act as catalysts that enhanced the formation of PCDD/Fs through surface catalysis. Due to the persistence and toxicity of PCDD/Fs, air pollution control devices (e.g., bag filters and wet scrubbers) should be installed to minimize the impact of PCDD/F emission from joss paper burning in temples.

#### **Metal Compositions**

The mean contents of twelve main metals in unburned joss paper and in residues of burned joss paper are listed in Tables 5 and 6. Al (2%) and Ca (1.5%) were dominant in the joss paper from Furnace 1, whereas Ca (3.5%) and Na (0.2%) were the most predominant metals in the joss paper from 1 Furnace 2. The mass contents of metals in joss paper were calculated according to total weight of joss paper used, and those of residues from burned joss paper were based on the weight of burned joss paper multiplied by ash contents of 7.87% and 4.87% for Furnaces 1 and 2, respectively. By this way, the increase or decrease of metal mass percentage in Tables 5 and 6 was obtained. (The "decrease (-)" means the content of metal in the residue of burned joss paper was lower than that of unburned one). After heating, some metals were partially vaporized, so the contents of elements (i.e., Ni and

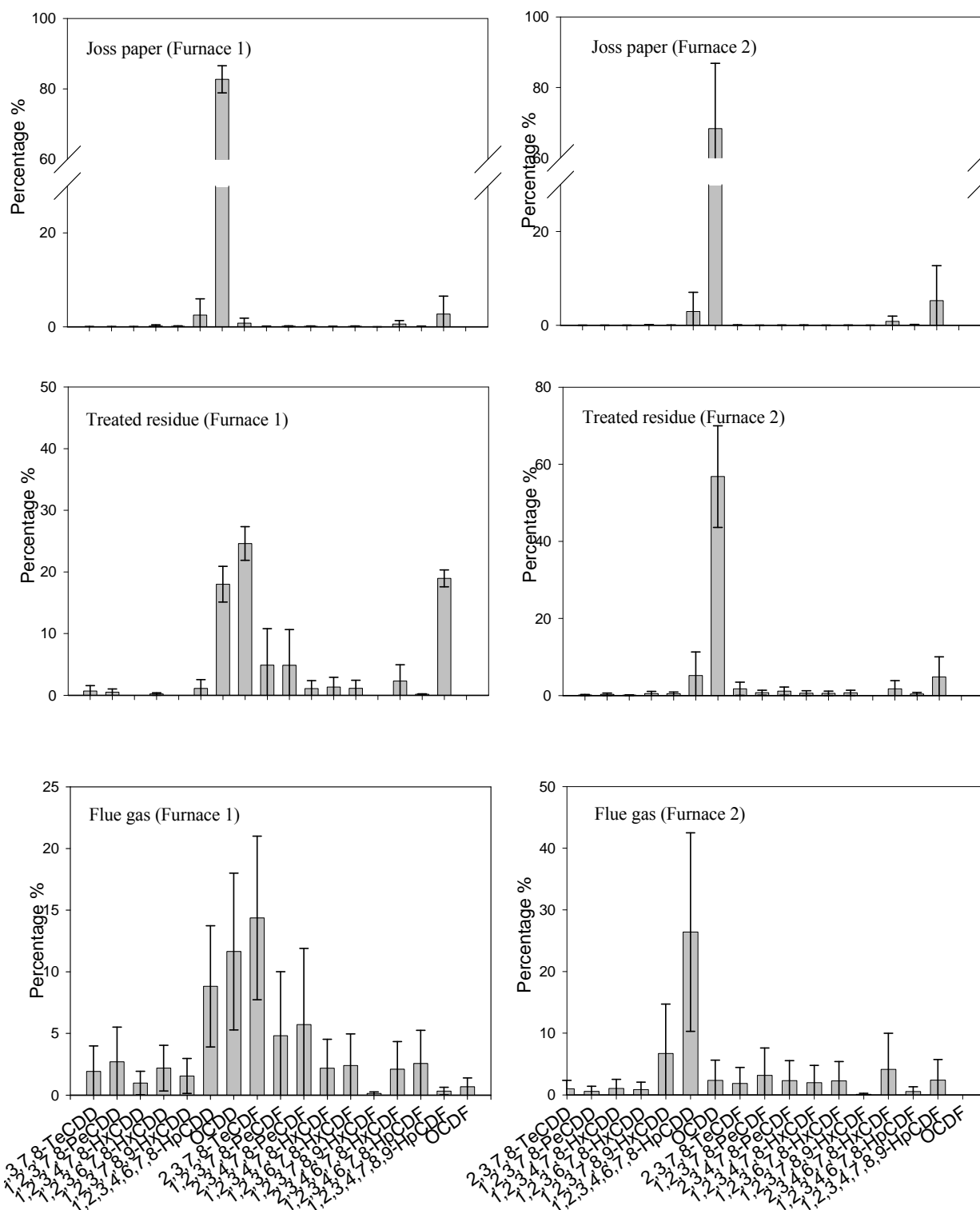


Fig. 1. PCDD/F congener profiles of joss paper, residues, and flue gases collected from Furnaces 1 and 2.

Ca) in the residue of burned joss paper were lowered by 6% to 94% (based on mass); however, those of Mn, Fe, Cu, and Zn increased. Interestingly, some metals (such as Na, Mg, Al, Sr, and Pb) exhibited opposite variation in mass in the two furnaces. This finding is probably due to the different burning conditions (e.g., temperature and oxygen) of these two furnaces.

## CONCLUSIONS

The mean total I-TEQ contents in the (unburned) joss paper from Furnaces 1 and 2 were 1.41 and 4.51 ng I-TEQ/kg, respectively. The high PCDD/F contents in the joss paper indicate that PCDD/Fs were possibly generated as by-products in the

manufacture of chlorine-bleached paper. After joss paper was burned, the mean total I-TEQ contents in the residue of burned joss paper from Furnaces 1 and 2 were 7.97 and 0.490 ng I-TEQ/kg, respectively. OCDD was the most abundant congener in the joss paper and in the residues from burned joss paper. Moreover, the total I-TEQ level of Furnace 1 (4.13 ng I-TEQ/Nm<sup>3</sup>) was one to two orders of magnitude higher than that of Furnace 2 (0.166 ng I-TEQ/Nm<sup>3</sup>). Due to the persistence and

toxicity of PCDD/Fs, air pollution control devices (e.g., bag filters and wet scrubbers) should be installed for the investigated furnaces to control the PCDD/F emission from joss paper burning.

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**Table 4.** Mean PCDD/F concentrations (ng/Nm<sup>3</sup>) in the flue gases of the two furnaces (n = 3).

PCDD/Fs	Furnace 1		Furnace 2	
	Mean (ng/Nm <sup>3</sup> )	RSD (%)	Mean (ng/Nm <sup>3</sup> )	RSD (%)
2,3,7,8-TeCDD	0.915	50	0.0195	27
1,2,3,7,8-PeCDD	1.27	56	0.0416	21
1,2,3,4,7,8-HxCDD	0.448	69	0.0249	10
1,2,3,6,7,8-HxCDD	0.948	93	0.0446	4.2
1,2,3,7,8,9-HxCDD	0.692	79	0.0365	6.5
1,2,3,4,6,7,8-HpCDD	3.33	101	0.265	16
OCDD	4.37	72	0.809	25
2,3,7,8-TeCDF	5.16	52	0.0999	26
1,2,3,7,8-PeCDF	2.30	50	0.0789	22
2,3,4,7,8-PeCDF	2.73	50	0.135	24
1,2,3,4,7,8-HxCDF	1.04	50	0.0986	17
1,2,3,6,7,8-HxCDF	1.14	51	0.0847	17
1,2,3,7,8,9-HxCDF	0.0619	62	0.0959	16
2,3,4,6,7,8-HxCDF	0.999	52	0.00425	30
1,2,3,4,6,7,8-HpCDF	1.21	55	0.177	13
1,2,3,4,7,8,9-HpCDF	0.146	76	0.0231	15
OCDF	0.319	56	0.102	21
Total PCDDs	12.0	76	1.24	20
Total PCDFs	15.1	51	0.899	17
PCDFs/PCDDs	1.44	--	0.743	--
Total PCDD/Fs	27.1	59	2.14	13
Total PCDDs (ng I-TEQ/Nm <sup>3</sup> )	1.80	55	0.0543	18
Total PCDFs (ng I-TEQ/ Nm <sup>3</sup> )	2.33	50	0.112	22
PCDFs/PCDDs	1.33	--	2.05	--
Total PCDD/Fs (ng I-TEQ/Nm <sup>3</sup> )	4.13	52	0.166	21

**Table 5.** Mean metal content (mg/kg) of the joss paper and the residue from burned joss paper in Furnace 1 (n = 3).

	Joss paper		Residue		Increase (+) <sup>b</sup> or decrease (-) <sup>b</sup> (%)
	Mean (mg/kg)	RSD <sup>c</sup> (%)	Mean (mg/kg)	RSD <sup>c</sup> (%)	
Na	2187	37	11400	13	-58
Mg	761	15	12600	0.8	+32
Al	20167	7.8	104333	3.4	-59
Ca	15100	8.0	177000	1.5	-6.2
Mn	16.1	3.4	260	1.3	+29
Fe	176	31	5687	2.6	+158
Ni	3.48	22	18.1	5.5	-58
Cu	37.2	11	846	3.3	+82
Zn	27.2	6.3	409	4.4	+20
Sr	21.7	9.3	283	4.0	+4.1
Cd	BD <sup>a</sup>	--	0.271	5.9	--
Pb	BD <sup>a</sup>	--	45.5	3.8	--

<sup>a</sup>Below detection limit; <sup>b</sup>Based on mass; <sup>c</sup>RSD = relative standard deviation.

**Table 6.** Mean metal content (mg/kg) of the joss papers and the residue from burned joss paper in Furnace 2 (n = 3).

	<i>Joss paper</i>		<i>Residue</i>		<i>Increase (+)<sup>b</sup> or decrease (-)<sup>b</sup> (%)</i>
	Mean (mg/kg)	RSD <sup>c</sup> (%)	Mean (mg/kg)	RSD <sup>c</sup> (%)	
Na	1902	54	159120	4.9	+318
Mg	1447	41	14312	2.1	-51
Al	740	67	29307	4.6	+98
Ca	35020	84	101449	19	-86
Mn	BD <sup>a</sup>	--	574	7.7	--
Fe	384	37	9527	41	+24
Ni	BD <sup>a</sup>	--	BD <sup>a</sup>	--	--
Cu	BD <sup>a</sup>	--	835	12	--
Zn	18.0	34	732	87	+103
Sr	35.8	61	210	9.7	-71
Cd	BD <sup>a</sup>	--	BD <sup>a</sup>	--	--
Pb	13.3	68	16.5	20	-94

<sup>a</sup>Below detection limit; <sup>b</sup>Based on mass; <sup>c</sup>RSD = relative standard deviation.

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