



Characteristics of PCDD/Fs in a Particles Filtration Device with Activated Carbon Injection

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ABSTRACT

Although numerous investigations have monitored polychlorinated dibenzo-*p*-dioxin and polychlorinated dibenzofuran (PCDD/F) emissions from large municipal solid waste incinerators (MSWIs) and undertaken laboratory experiments to identify the formation mechanisms of PCDD/F, the PCDD/F profiles inside an air-pollution control device have seldom been determined. This study presents the outcome of a dioxin abatement program that injected particulate activated carbon (PAC) into an MSWI. The fly ashes collected from different locations in a bag filter were examined and the mass distribution was determined. Emissions from the stack were sampled to analyze PCDD/F content after injections of 10, 13 and 17 kg/h PAC. The concentration of PCDD congeners decreased from 117.00 to 0.32 ng/Nm³ and that of PCDF decreased from 94.84 to 0.19 ng/Nm³. The concentrations of filter cake ashes in different chambers and at different locations varied at 105.11-147.53 ng/g. Based on mass balance evaluation, the flow rate of PCDD/Fs in filter cake ash was 3.33 ± 0.50 ng/min; this value was roughly six times higher than that of fly ash in the disposal pit, indicating that filter cake ash treatment warrants considerable attention due to the policy for controlling PCDD/Fs.

Keywords: PCDD/Fs; Powder activated carbon injection; Bag filter; Filter cake ash; Mass balance.

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) were discovered in flue gases and fly ash from municipal solid waste incinerators (MSWIs) in 1977 (Olie *et al.*, 1977). Investigations revealed that PCDD/F emission in flue gases and fly ash from MSWIs were discovered by the same token in Taiwan. These PCDD/Fs are hydrophobic and do not metabolize; thus, they persist in the environment and bioaccumulate in the fatty tissues of animals and humans (US EPA, 2000).

The dioxin formation mechanism is based on the condition of source, temperature, location, and reaction type. It is normally divided into two parts. This study focuses on the bag filter to identify the temperature, source and influence. Thus, this study divided the dioxin formation mechanism into three parts. The following three mechanisms account for PCDD/F emissions from incinerators: 1) High-temperature gas phase formation (300-600°C) (Everaert and Baeyens, 2002); 2) Formation from precursors, such as chlorophenols, polychlorinated diphenyl ethers, and chlorobenzenes (Lustenhower *et al.*, 1980; Hutzinger *et al.*, 1985). For instance, chlorophenols are good surrogates for the toxicity equivalence (TEQ) of PCDD/Fs during different incineration processes (Tuppurainen *et al.*, 2000); 3) Formation by *de novo* synthesis in the low-temperature post-combustion

zone (200-400°C) through residue carbon or metal catalysts in the fly ash reaction (Dickson *et al.*, 1989; Everaert and Baeyens, 2002).

Air pollutants, such as particulate matter, heavy metals, polycyclic aromatic hydrocarbons, and dioxins generated from combustion processes adversely affect human health (Lin *et al.*, 2008; Wang *et al.*, 2008). Taiwan currently has 24 large MSWIs in operation. Due to concern regarding their adverse health effects, the government has established guidelines that regulate air pollutant emissions. For instance, Taiwan's Environmental Protection Administration (EPA) has set the dioxin emission limit in flue gas for MSWIs at 0.1 ng I-TEQ/Nm³; thus, removing PCDD/Fs from flue gases is necessary. Various combinations of air-pollution control devices (APCDs) have been examined. A dry scrubber combined with a bag filter with powder activated carbon (PAC) injected is the most effective technique for controlling PCDD/F emissions (Blumbach and Nethe, 1994; Buekens and Huang, 1998; Lee *et al.*, 2004; Wang *et al.*, 2005). Notably, PAC injection is followed by various types of APCDs to enhance the removal of PCDD/Fs, which can approach 92-99% for MSWIs (Tejima *et al.*, 1996; Dong *et al.*, 2001a; Dong *et al.*, 2001b; Abad *et al.*, 2003). After dioxins have been adsorbed onto the PAC, the PAC with fly ash is then captured by the bag filter and removed as fly ash.

Taiwan's government has set regulations for ashes generated by MSWIs; roughly 2 × 10⁶ tons of incinerator residue is produced annually. Fly ash, including filter cakes, fabric filters and fly ash pits, has the highest dioxin concentration produced by MSWIs (Huang and Buekens, 1995; Lin *et al.*, 2008). Although PAC injection technology can effectively decrease the flue gas dioxin concentration, PAC injection transfers the dioxin in gas

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phase to a particle phase, which increases total dioxin emissions (including those in fly ash and flue gas) from MSWIs (Chang and Lin, 2001; Giugliano *et al.*, 2002). The memory effect increases the dioxin concentration in flue gas after PAC injection, i.e., the dioxin or precursor desorbs slowly to flue gas and increases the dioxin concentration in the stack, and reduces removal efficiency for PAC injection to a level lower than that expected (Chang and Lin, 2001). In other studies, together with injecting PAC into the front of the bag filter, these two measures reduce PCDD/F concentrations in the stack flue gas from 145 ng I-TEQ/Nm³ to 3.38-7.73 ng I-TEQ/Nm³. Even with high PAC usage (40 kg/h), the PCDD/F concentration in flue gas still exceeds the regulatory limit of 1 ng I-TEQ/Nm³ (factory process). This may be due to low PAC utilization efficiency (< 3%) in conventional activated carbon injection for a single bag filter system (Chi *et al.*, 2007; Kim *et al.*, 2007; Li *et al.*, 2007).

Due to its simple engineering and high efficiency, PAC injection has become a popular retrofit technology for reducing dioxin emissions at most MSWIs. The PAC injection has been widely used with different APCDs for dioxin removal. However, few studies have focused on obtaining optimal dioxin control with PAC injection and a filter for MSWIs. In this study, different amounts of PAC were injected and the PCDD/F concentrations in stack flue gases were measured to investigate control of PCDD/F emissions. Additionally, the PCDD/F content in the bag filter ashes at different locations was determined.

EXPERIMENTAL APPROACH

The MSWI used in this study, which has been operating in southern Taiwan for 7 years, has two incineration units, with a total capacity of 900 tons of waste daily. The incinerator operating temperature is 850-1050°C. Each incinerator is equipped with a semidry scrubber (230-250°C) and bag filter (160-180°C) for controlling gaseous and particulate emissions. The PAC is injected between the semidry scrubber and bag filter. Mean flue gas generation rate was 95.11 KNm³/hr. The rate at which fly ash and cake ash were generated was estimated at 785.63 kg/h. Fig. 1 shows the operational parameters, a flow diagram, and sampling points in the MSWI.

Flue gas was sampled at the bag filter inlet and across every process unit of the flue gas cleaning system (PAC injection and bag filter). Sampling was performed during operation with PAC injection of 10, 13 and 17 kg/h and the performance of PAC in reducing PCDD/F emissions was evaluated.

Fly ashes were collected from the storage pit and bag filters. The bag filter cake ashes were sampled in chambers D, H and J. The bag filter, an APCD, has 10 chambers, each with 150 glass fiber filters (6 m × 12 cm i.d.) coated with Teflon. The samples from the D, H and J chambers were taken from random filter cakes in the bag filter. Every sample collect filter cakes over 10 filters. Fig. 1 presents a diagram of this process.

Analyses of stack flue gas and fly ash samples followed the US EPA Modified Method 23 and Modified Method 1613, respectively. All chemical analyses were conducted at the Super Micro Mass Research and Technology Center, Cheng-Shiu University, an accredited laboratory in Taiwan for analyzing PCDD/Fs. Prior to analysis, each sample was spiked with a known amount of the ¹³C₁₂-labeled internal standard to the extraction thimble. After extraction for 24 h in a Soxhlet extractor, the sample extract was concentrated and then treated with concentrated sulfuric acid. A series of sample cleanup and fractionation procedures followed. The eluate was concentrated to approximately 1mL and transferred into a vial. The concentrate was further concentrated to near dryness using a nitrogen stream. Prior to analysis, the standard solution for recovery checking was added to the sample.

A high-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) was used for PCDD/F analyses. The HRGC (Hewlett Packard 6970 Series Gas, Agilent, CA, USA) was equipped with a DB-5MS fused silica capillary column ($L = 60\text{m}$, $ID = 0.25\text{ mm}$, film thickness = 0.25 μm) (J & W Scientific, CA, USA) with splitless injection. The HRMS (Micromass Autospec Ultima, Manchester, UK) was equipped with a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring (SIM) was used with a resolving power of 10,000. Details of analyses can be found in Wang *et al.* (2007).

To evaluate the partitioning of the PCDD/F concentration for each sample from the bag filter system, the results obtained for

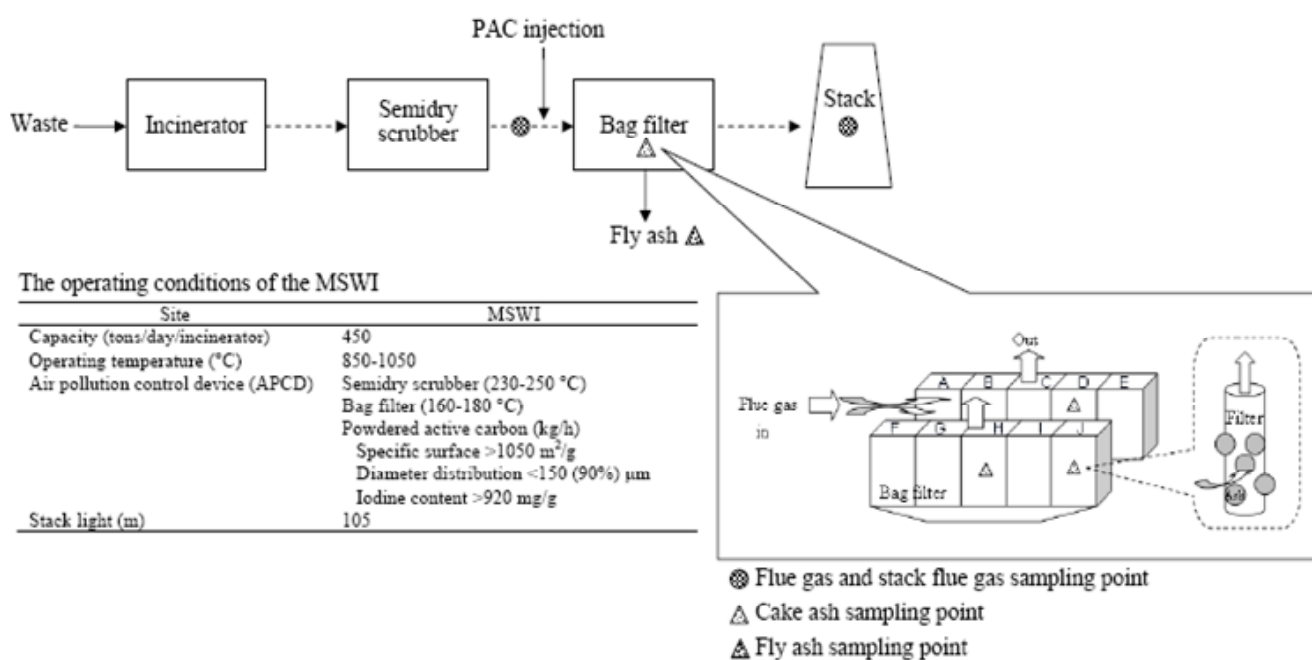


Fig. 1. Flow diagram and sampling points of the MSWI.

the PCDD/Fs formed were further evaluated using mass balance calculations. Mass fluxes of PCDD/Fs in the flue gas around the bag filter were obtained using concentrations and flow rate measurements, whereas the fluxes in the bag filter cake ash and fly ash were calculated based on the PCDD/F content in collected fly ash samples and their corresponding production rates. The mass balance was based on the concentration of PCDD/Fs, including flue gas before the bag filter (no PAC), stack flue gas, cake ash, and fly ash. At the time of sampling, flue gas production rate was 95.11 KNm³/hr, sampling volume was 2.19 Nm³, and fly ash and cake ash amounts were 10.0195 g and 3 g over 120 min.

RESULTS AND DISCUSSION

PCDD/Fs Desorbed in Flue Gases with PAC Injection

Table 1 shows the concentrations of total PCDD and PCDF congeners when the PAC injection rate increased from 10 to 17 kg/h in flue gas, and the values for desorbed flue gas relative to PAC injection. The degree of chlorination of both PCDDs and PCDFs decreased markedly as the PAC injection amount increased, indicating that chlorination become increasingly important. Total PCDD congeners decreased from 117 to 0.32 ng/Nm³, and total PCDF congeners decreased from 94.84 to 0.19 ng/Nm³. Huang and Buekens (1995), who reviewed research regarding the mechanisms of PCDD/F formation, concluded that “de novo synthesis” can produce PCDD/Fs with a PCDF/PCDD ratio > 1, while precursor formation produces PCDD/Fs with a PCDF/PCDD ratio << 1. In this study, the PCDD/PCDF ratio was 1.25-13.93 and the toxic equivalence was 0.32-0.65, demonstrating the significance of the PCDD/F concentrations as the precursor mechanism. The toxic equivalence is the *de novo* mechanism for PCDD/F formation.

Experimental results show that the maximum PCDD/F concentration and toxic equivalence were for a PAC injection of 10 kg/h, and the minimum PCDD/F concentration and toxic equivalence were for a PAC injection of 17 kg/h. However, the PCDD/F concentrations in flue gas were markedly decreased with PAC injection. The PAC injection was followed by various types of APCDs to enhance removal of PCDD/Fs, which approached 92-99% (Tejima *et al.*, 1996; Dong *et al.*, 2001a; Dong *et al.*, 2001b; Abad *et al.*, 2003). However, PAC injection only helps adsorb PCDD/Fs when it does not decompose PCDD/Fs. To meet the PCDD/F emissions standard of 0.1 ng I-TEQ/Nm³, the amount of PAC injected is typically excessive; thus, it is a costly treatment process.

Table 1. Mean PCDD/F content in flue gas by injection PAC.

Compound	Content (ng/Nm ³)			
	(i) no PAC ^a	PAC injection and across to the bag filter (kg/h PAC)		
		(ii) 10	(iii) 13	(iv) 17
Concentration				
Total PCDDs	117.00	17.88	4.42	0.32
Total PCDFs	94.84	1.10	0.60	0.19
PCDD/PCDF Ratio	1.25	13.93	7.39	1.65
Total PCDD/Fs	211.84	18.97	5.01	0.50
I-TEQ				
Total PCDDs	2.93	0.05	0.02	0.01
Total PCDFs	9.05	0.08	0.05	0.02
PCDD/PCDF Ratio	0.32	0.65	0.49	0.39
Total I-TEQ	11.97	0.13	0.07	0.03

In addition to the quantity of PCDD/PCDFs desorbed, PAC injection also impacts the congener profile of desorbed PCDD/PCDF species found in adsorption traps. Fig. 2 shows data for desorbed PCDD and PCDF congener group profiles. The desorbed PCDD/PCDF species trapped in stack flue gas were mainly HpCDD/OCDD-HpCDF/OCDF (Lee *et al.*, 2003; Kao *et al.*, 2006), resulting from the dechlorination of PCDDs/PCDFs, and following similar trends relative to PAC injection as the total PCDD/F content. There are different influences in the HpCDD and OCDD. Experimental results indicate that I-TEQ decreased markedly when PAC was injected of 17 kg/h. Everaert *et al.* (2003) and Li *et al.* (2008) indicate that increasing the quantity of activated carbon has a limited effect on the overall PCDD/F removal efficiency.

PCDD/F Content in the Bag Filter Ashes from Different Locations

Generally, PAC injected into the bag filter can adsorb PCDD/F pollutants in the flue gas. The PAC injection and removal efficiencies are significant. To quantify the residues on the bag filter, PCDD/F concentrations on filter ashes from different locations were measured. Table 2 shows the mean PCDD/F contents in bag filter ashes obtained from the fly ash storage pit and filter cake ashes from different chambers. The concentration of total PCDD/Fs in fly ash was 7.58 ng/g, and the PCDD/PCDF ratio was 1.39. The concentrations of filter cake ashes in different chambers and locations were 105.11-147.53 ng/g, and the PCDD/PCDF ratio was 8.67-25.06. A number of different mechanisms influence the PCDD/PCDF content in ashes when flue gas passes across the bag filter. The amount of PACs injected may cause changes in the characteristics of the fly ash matrix (i.e., specific area and active sites). Comparing the filter ash and particulate phase of stack flue gas indicates that PAC injection technology can reduce flue gas PCDD/F concentrations; however, the technology increases the total amount of PCDD/Fs discharged (including those in fly ash and flue gas) from MSWIs. Activated carbon provides the basic organic material and catalytic surface for PCDD/F formation.

The possibility of a homogeneous gas phase reaction was also investigated. Fig. 3 shows the data for the PCDD and PCDF congener groups. The main congener profiles of the concentration are HpCDD and OCDD. The I-TEQ are PeCDF and HxCDF in fly ash, and HxCDD and PeCDF in filter cake ash. These analytical results show that samples from different chamber of bag filters have different concentrations and ratios. The congener profiles and the PCDD/PCDF ratio of flue gas and ashes also differed.

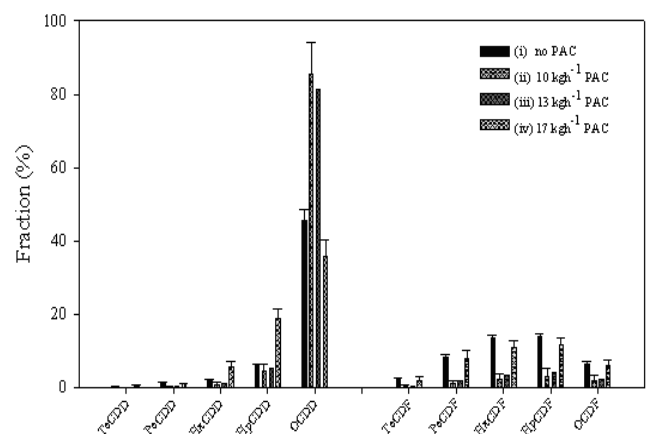
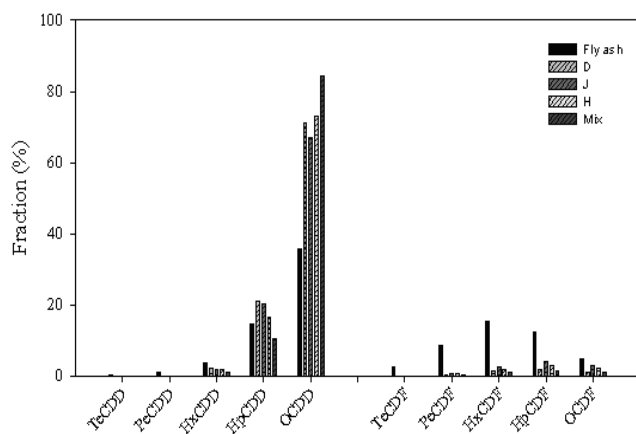


Fig. 2. Impact of PAC on PCDD and PCDF formation from PAC injection (i), (ii), (iii), and (iv).

Table 2. Mean PCDD/F content in ashes.

Compound	Content (ng/g)				
	Fly ash	Cake ash			
		D	J	H	Mix ^a
Concentration					
Total PCDDs	4.23	99.56	132.17	135.60	127.87
Total PCDFs	3.35	5.55	15.25	11.94	5.10
Ratio PCDD/PCDF	1.39	17.92	8.67	11.36	25.06
Total PCDD/Fs	7.58	105.11	147.42	147.53	132.98
I-TEQ					
Total PCDDs	0.11	0.60	0.81	0.75	0.46
Total PCDFs	0.35	0.36	0.75	0.68	0.32
Ratio PCDD/PCDF	0.32	1.68	1.07	1.09	1.44
Total I-TEQ	0.46	0.96	1.56	1.43	0.78

^a Mix cake ash containing different chambers of D, J, and H.

**Fig. 3.** Homologues of PCDD and PCDF in ashes.

PCDD/Fs Mass Balance in the Bag Filter

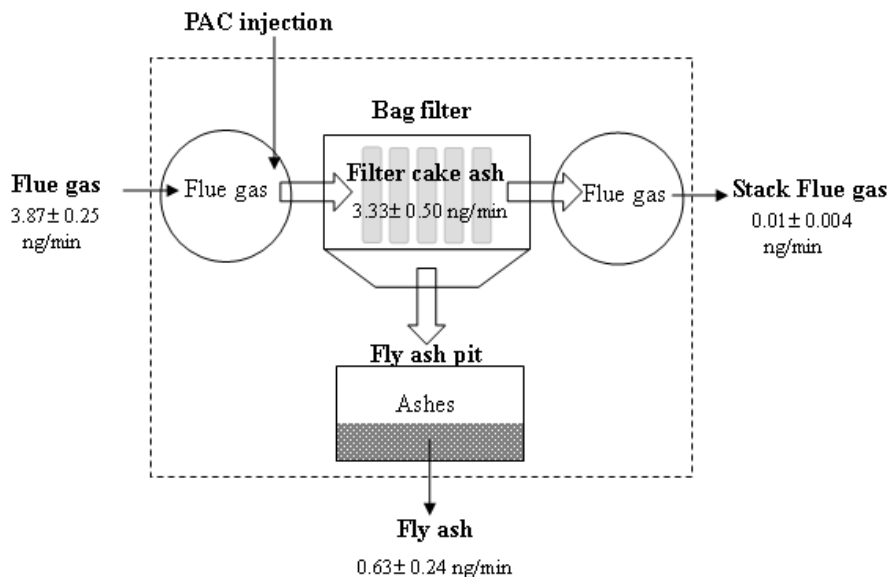
To determine the content and distribution of PCDD/Fs in the bag filter system, flue gas and ashes were sampled from different

locations, and the mass balance of PCDD/Fs in the bag filter was evaluated.

A detailed evaluation of experimental results is based on mass balance calculations (Fig. 4). Mass fluxes of PCDD/Fs in flue gas around the PAC injection and bag filter sites were obtained from actual concentrations and flow rate measurements, while the fluxes in solid and liquid residues were calculated using their PCDD/F content and corresponding mass production rates. In flue gas without PAC injected before the bag filter section, the PCDD/F flow rate was 3.87 ± 0.25 ng/min. After the bag filter, the PCDD/F flow rate reduced to 0.01 ± 0.004 ng/min and the fly ash was 0.63 ± 0.24 ng/min. These findings indicate that stack gas levels constitute a minor contribution to total PCDD/Fs emitted by the MSWI; thus, complying with the regulatory limit of 0.1 ng I-TEQ/Nm³. The highest concentration of PCDD/Fs was found in fly ash responsible for PAC injection, and the PCDD/Fs shifted to solid phase. The PCDD/F levels of the filter cake ash flow rate were 3.33 ± 0.50 ng/min, with a concentration roughly six-fold higher than that in fly ash. Surprisingly, the most significant contribution to the total PCDD/Fs released was filter cake ash (83.90%), followed by fly ash (15.87%); the contribution of stack flue gas was almost negligible. These analytical results have important implications for PCDD/Fs in filter cake ash.

Generally, an APCD combined with PAC injection can control PCDD/F emissions. Comparing the input (flue gas) and output (filter cake ash, fly ash, and stack flue gas), 2.78% of PCDD/Fs was produced. Many studies have indicated that fly ash (including filter cakes, fabric filter, and fly ash pit) contains the highest dioxin emissions from MSWIs (Huang and Buekens, 1995; Lin *et al.*, 2008). The partitions were mainly on filter ash (about 83.9%). Relatively in fly ash, filter ash is comparatively steady (38.05% and 14.99%, respectively).

Although the operating temperature of the bag filter was only 160-180°C, which is below the favorable range of 250-400°C for PCDD/F formation, PCDD/Fs were captured by the active carbon in the bag filter. Furthermore, the duration fly ash remained in the bag filter was longer than that for other incinerator units, thereby increasing the amount of PCDD/Fs downstream. Similar trends were observed by Giugliano *et al.* (2002), Abad *et al.* (2003) and Wevers and De Fr'e (1998). The PAC injection system was located between the semidry scrubber and bag filter. Additionally, the PAC injection system is a batch system; the bag filter was

**Fig. 4.** PCDD/Fs mass balance across the PAC injection and bag filter.

purged for 30 sec every 30 minutes. This purging involved passing air through the bag filter to remove dust, and then coating it with activated cake adsorbents to remove dioxins and other noxious pollutants downstream. Most filter ashes dropped from the filter into the fly ash pit. Mistakes in the sampling techniques may have altered experimental results. Notably, obtaining a sample in the bunker in which 50% of incineration occurs is difficult. Since wastes cannot be mixed perfectly, samples may not be completely representative. The experimental results suggest that the treatment should be studied extensively to control PCDD/F formation in fly ash.

CONCLUSIONS

For PAC injection of 10, 13, and 17 kg/h, the total concentration of PCDD congeners decreased from 117 to 0.32 ng/Nm³; the total concentration of PCDF congeners decreased from 94.84 to 0.19 ng/Nm³; and the PCDD/PCDF ratio was 1.25-13.93. Experimental results indicate that the PCDD/F concentration and toxic equivalence were minimal at a PAC injection of 17 kg/h.

Additionally, filter cake ash concentrations in samples obtained from different chambers and locations in the bag filter were 105.11-147.53 ng/g, and the PCDD/PCDF ratio was 8.67-25.06. A number of different mechanisms influence the PCDD/PCDF content in ashes when flue gas passes across the bag filter.

The PCDD/F flow rate in the flue gas without PAC injection before the bag filter section was 3.87 ± 0.25 ng/min via mass balance calculations. After the bag filter, the PCDD/F flow rates reduced to 0.01 ± 0.004 ng/min and the fly ash was 0.63 ± 0.24 ng/min. The highest concentration of PCDD/Fs was in the fly ash responsible for PAC injection, and the PCDD/Fs shifted to solid phase. The PCDD/F concentration of the filter cake ash flow rate was 3.33 ± 0.50 ng/min, roughly 6-fold higher than that in fly ash. Compared with fly ash, filter ash was comparatively steady (38.05% and 14.99%, respectively). These experimental results suggest that the filter cake ash and filter treatment should be investigated extensively to control PCDD/F formation in fly ash.

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