Removal of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans in Flue Gases by Venturi Scrubber and Bag Filter

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Abstract

This study investigates the individual removal efficiencies of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) by venutri scrubber and bag filter, which operated in one medical waste incinerator (MWI) and one secondary aluminum smelter (secondary ALS), respectively. Stack flue gases, effluent, and fly ash were measured for PCDD/Fs to characterize the performance of the venutri scrubber and the bag filter for reducing PCDD/F emission. The mean PCDD/F concentrations in the stack flue gases of the MWI and secondary ALS were 0.511 and 10.6 ng I-TEQ Nm^{-3} (calculated according to International Toxic Equivalency Factors [I-TEQ] and normalized to dry flue gas conditions of 273 K and 11% O_2), while concentrations in the effluent, including ash and wastewater from the venturi scrubber and fly ash from the bag filter, were 7.51 ng I-TEQ g^{-1}, 154 pg I-TEQ L^{-1}, and 5.59 ng I-TEQ g^{-1}, respectively. The average removal efficiencies of tetra-, penta-, hexa-, hepta-, and octa-PCDD/Fs by bag filter are 8.2%, 10.6%, 33.5%, 52.4%, and 59.1%, respectively. This suggests that highly chlorinated PCDD/Fs with lower vapor pressures are more easily adsorbed onto the particulate and consequently more easily removed by bag filter. The removal efficiencies of the bag filter on the total PCDD/F emission and the total PCDD/F I-TEQ emission are 37.6% and 11.2%, respectively, while those of the venturi scrubber are 46.0% and 44.5%, respectively. Although the operating conditions of the venutri scrubber and the bag filter are different, the removal efficiencies of each for PCDD/Fs is inadequate.

Keywords: PCDD/Fs, venturi scrubber, bag filter, removal efficiency

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1. Introduction

Since polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) were discovered in the flue gases and fly ash of municipal solid waste incinerators (MSWIs) in 1977 (Olie et al., 1977), they have become a serious issue in many countries because of their toxicological effects and associated adverse health implications.

The removal of PCDD/Fs in flue gases is necessary to reduce the emission of PCDD/Fs to the environment. Various equipment, some in combination and under different operating conditions, has been tested, including an electrostatic precipitator (EP), a scrubber, a bag filter, and adsorbent injection. A combination of air pollution control devices (APCDs)— a scrubber plus a bag filter with activated carbon—was determined to be "the most effective technique for PCDD/F emission control" (Buekens et al., 1998; Blumbach et al., 1994). However, most of the medical waste incinerators (MWIs) and secondary aluminum smelters (secondary ALSs) in Taiwan are equipped only with simpler APCDs (i.e., a quench chamber, venutri scrubber, and packed-bed scrubber for MWIs, and a cyclone and/or bag filter for secondary ALSs). This led to the investigation of the individual removal efficiencies of PCDD/Fs by venutri scrubber and bag filter to determine if simpler APCDs help explain why the PCDD/F emission factors of MWIs and secondary ALSs in Taiwan are 20.0 μg International Toxic Equivalency Factors (I-TEQ) ton-waste^{-1} (mean value of five MWIs) and 21.5 μg I-TEQ ton-feedstock^{-1} (mean value of four ALSs). These amounts are 208- and 224-fold more than the PCDD/F emission factor of MSWIs (0.0961 μg I-TEQ ton-waste^{-1}, mean value of 13 MSWIs), respectively (Wang et al., 2003).

This study investigates the individual removal efficiencies of PCDD/Fs by venutri scrubber and bag filter, which were equipped in one MWI and one secondary ALS, respectively. In the MWI, the stack flue gases and the effluent of the venutri scrubber, including wastewater (liquid phase) and gathered fly ash (particulate phase), are sampled. In the secondary ALS, the flue gases before and after (i.e., stack flue gases) a bag filter, as well as fly ash, are sampled. All samples are measured for PCDD/Fs to characterize the performance of the venutri scrubber and the bag filter for reducing PCDD/F emissions.

2. Material and Method

Table 1 presents basic information concerning the MWI and secondary ALS investigated here, including each feeding and the APCDs in sequence.

Sampling

Five PCDD/F samples were collected from the stack flue gas of the MWI and another five samples were simultaneously collected from the flue gases before and after the bag filter of the secondary ALS. All flue gases were sampled according to U.S. EPA Modified Method 23 (2001). This method

Table 1. Basic information concerning the medical waste incinerator (MWI) and secondary aluminum smelter (secondary ALS).

	MWI	Secondary ALS
Operation Type	Intermittent (8 hrs per day)	Intermittent (one batch time: 2-3 hrs)
Feeding Waste/Material	Infectious and Pathological Waste (160 kg hr^{-1})	Aluminum Scrap (5.29 ton hr^{-1}) Silicon (0.310 ton hr^{-1}) Aluminum Ingot (3.2 ton hr^{-1}) Aluminum Liquid (6.5 ton hr^{-1})
Auxiliary Fuel	Commercial Diesel (80 L hr^{-1})	No. 6 Heavy Oil (467 L hr^{-1})
Air Pollution Control Devices in Sequence (Operation Temperature)	Quench Chamber (60 °C) Venturi Scrubber (45 °C) Packed-Bed Scrubber (40 °C)	Bag Filter (110~140 °C)

can be used to determine PCDD/F emission from municipal waste combustors. Calibration standards were selected for regulated emission levels for municipal waste combustors. A sampling train adopted in this study is comparable with that specified by U.S. EPA Modified Method 5 (2001). The principle of this method is that a sample of the flue gas is withdrawn isokinetically from an emissions unit and particulate matter is collected by a series of impingers followed by a filter. The weight of the particulate matter is determined gravimetrically after removing uncombined water from the impinger solution, and washing the probe/glassware and filter. The company certified by the Taiwan EPA to sample PCDD/Fs in stack flue gas performed the samplings. Prior to sampling, XAD-2 resin was spiked with PCDD/F surrogate standards pre-labeled with isotopes. Each stack flue gas sampling lasted for ~3 h. To ensure that the collected samples were contamination-free, one trip blank and one field blank were taken while the field sampling was conducted (Wang et al., 2003). Effluent from the venturi scrubber and fly ash from the bag filter were simultaneously collected every 30 minutes during stack flue gas sampling, and conformed to U.S. EPA Method 8280B (1998)– Revision 2 January 1998. This method is appropriate for the detection and quantitative measurement of PCDD/Fs in water (at part-per-trillion concentrations), soil, fly ash, and chemical waste samples, including still bottoms, fuel oil, and sludge matrices.

Analysis of PCDD/Fs

Effluent samples were filtered by pretreated glass-fiber filters to separate the wastewater (liquid phase) and gathered fly ash (particulate phase). The fly ash retained on the filter was freeze-dried to remove water content. For wastewater analysis, stable isotopically labeled analogs of 15 of the 2,3,7,8-substituted PCDD/Fs were spiked into a 1 L sample, and the sample was extracted with methylene chloride in a separatory funnel. All fly ash and wastewater samples were analyzed for

Table 2. PCDD/F concentrations and their corresponding relative standard deviation (RSD) in the (stack) flue gases of the medical waste incinerator (MWI) and secondary aluminum smelter (secondary ALS).

PCDD/Fs	MWI		Secondary ALS			
	Stack flue gases (n=5)		Stack flue gases (n=5)		Flue gases before bag filter (n=5)	
	Mean	RSD (%)	Mean	RSD (%)	Mean	RSD (%)
PCDDs	1.57	68.0	10.9	41.5	22.8	56.5
PCDFs	5.58	74.5	88.7	47.7	148	63.6
PCDFs/PCDDs Ratio	3.43	20.3	7.25	31.9	5.4	34.0
Total (ng Nm^{-3})	7.14	72.8	99.6	46.7	171	60.8
PCDDs I-TEQ (ng I-TEQ Nm^{-3})	0.0539	64.9	1.35	63.6	1.86	53.3
PCDFs I-TEQ (ng I-TEQ Nm^{-3})	0.457	73.9	9.22	54.4	10.4	52.8
PCDFs/PCDDs I-TEQ Ratio	8.51	21.5	7.80	29.5	5.77	18.6
Total I-TEQ (ng I-TEQ Nm^{-3})	0.511	72.8	10.6	55.5	12.3	52.5

PCDD/Fs by using U.S. EPA modified Method 1613B, while the analysis of all flue gas samples conformed to U.S. EPA Modified Method 23B (2001). All chemical analyses were conducted by an accredited laboratory, the Super Micro Mass Research and Technology Center at Cheng Shiu University, which is certified by the Taiwan EPA for analyzing PCDD/Fs. Each collected sample was spiked with a known amount of the internal standard prior to PCDD/F analysis. The extract was concentrated, treated with concentrated sulfuric acid, and followed by a series of sample cleanup and fractionation procedures. Prior to analysis, the standard solution was added to ensure recovery during the analysis process. A high-resolution gas chomatograph/high-resolution mass spectrometer (HRGC/HRMS) was used for PCDD/F analyses. The HRGC (Hewlett Packard 6970 Series, CA, USA) was equipped with a DB-5 fused silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25 μm) (J&W Scientific, CA, USA) and splitless injection. The HRMS (Micromass Autospec Ultima, Manchester, UK) mass spectrometer was equipped with a positive electron impact (EI+) source. The analyzer mode of selected ion monitoring (SIM) was used with resolving power at 10,000. The electron energy and source temperature were specified at 35 eV and 250 °C, respectively (Wang et al., 2003).

3. Results and Discussion

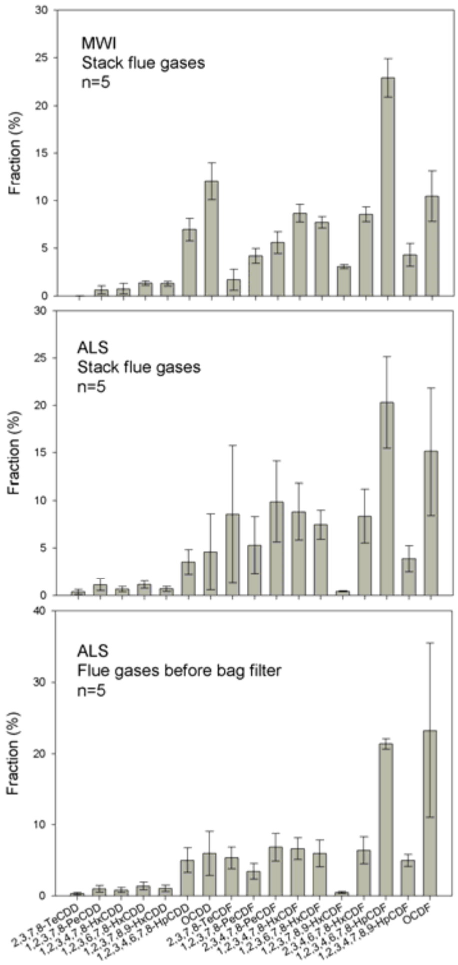

Figure 1. PCDD/F congener profiles in the flue gases of the medical waste incinerator (MWI) and secondary aluminum smelter (secondary ALS).

Characteristics of PCDD/F in the stack flue gases of the MWI and the secondary ALS

Table 2 lists the mean PCDD/F concentrations in the stack flue gases of the MWI and secondary ALS, which are 0.511 ng I-TEQ Nm^{-3} (calculated according to International Toxic Equivalency Factors and normalized to the dry flue gas conditions of 273 K and 11% O_2) [RSD: 72.8%] and 10.6 ng I-TEQ Nm^{-3} (RSD: 55.5%), respectively. In Taiwan, the regulation of PCDD/F emission from newly established electric arc furnaces (EAFs) is 0.5 ng I-TEQ Nm^{-3}; however, there is still no regulation for secondary ALSs. The mean PCDD/F concentration (10.6 ng I-TEQ Nm^{-3}) of the secondary ALS obtained in this study is 21 times higher than 0.5 ng I-TEQ Nm^{-3}. In flue gases before the bag filter of the secondary ALS, the mean PCDD/F concentration is 12.3 ng I-TEQ Nm^{-3}.

PCDD/F emission from most combustion processes are detected as a mixture of 75 PCDD and 135 PCDF congeners. The mixture can be translated into profiles, which represent the distribution of individual PCDD/Fs. These profiles may serve as a signature or fingerprint of the types of PCDD/Fs associated with particular incinerators and APCDs, and are usually referred to as congener profiles. Figure 1 shows the congener profiles of the 17 2,3,7,8 chlorinated substituted PCDD/Fs (mean±SD)

Table 3. PCDD/F concentrations (contents) in the effluent from venturi scrubber and fly ash from bag filter.

PCDD/Fs	Effluent from Venturi scrubber		Fly ash from Bag filter
	Ash (n=3) ($ng\ g^{-1}$)	Wastewater (n=3) ($pg\ L^{-1}$)	(n=3) ($ng\ g^{-1}$)
PCDDs	30.0	514	10.9
PCDFs	81.5	1,340	144
PCDFs/PCDDs Ratio	2.71	2.6	13.2
Total	111	1,850	155
PCDDs I-TEQ	1.23	39.1	0.609
PCDFs I-TEQ	6.28	115	4.98
PCDFs/PCDDs I-TEQ Ratio	5.11	2.94	8.20
Total I-TEQ	7.5	154	5.59

detected from the stack flue gases of the MWI and the secondary ALS. The congener profiles are quite similar to those obtained from U.S. EPA (2000). That the congener profiles in the flue gases before and after the bag filter show little change reveals that the removal mechanism of PCDD/Fs by bag filter should be physical (i.e., adsorption and desorption) reactions.

Characteristics of PCDD/F in the effluent and fly ash

Table 3 lists the mean PCDD/F concentrations (contents) in the effluent, including ash and wastewater from venturi scrubber and fly ash from bag filter, which are 7.51 ng I-TEQ g^{-1}, 154 pg I-TEQ L^{-1}, and 5.59 ng I-TEQ g^{-1}, respectively. Both the concentration of effluent and fly ash exceed the environmental quality standard (soil: 1 ng I-TEQ g^{-1}; water: 1 pg I-TEQ L^{-1}) and effluent standard (effluent: 10 pg I-TEQ L^{-1}) of the Japan Ministry of the Environment (2002). The disposal of the wastewater and fly ash needs to be carefully conducted to prevent secondary environmental pollution. The high PCDFs/PCDDs ratio (=13.2) of fly ash from bag filter reveals that de novo synthesis not only occurs but also dominates in the post-combustion zone of the secondary ALS.

Removal of PCDD/F by venturi scrubber

Table 4 lists PCDD/F emission rates from the effluent and stack flue gas and the removal efficiency of the venturi scrubber. The removal efficiency is estimated by mass balance as follows:

Removal efficiency (%) = A/(A+B)*100%

where: A = PCDD/F emission rates from the effluent, including ash and wastewater (μg/hr); and B = PCDD/F emission rates from stack flue gases.

Table 4 reveals that PCDD/F emission rates from the effluent (ash and wastewater) and stack flue gas are 2.86, 1.54, and 5.48 μg I-TEQ hr^{-1}, which comprise 28.9%, 15.6%, and 55.5% of the total

Table 4. PCDD/F emission rates (μg hr^{-1}) and the removal efficiency (%) of venturi scrubber.

PCDD/Fs	Effluent from venturi scrubber		Stack flue gases (μg hr^{-1})	PCDD/F removal efficiency of venturi scrubber (%)
	Fly ash (μg hr^{-1})	Wastewater (μg hr^{-1})		
2,3,7,8-TeCDD	0.0540	0.0904	0.149	49.2
1,2,3,7,8-PeCDD	0.359	0.345	0.494	58.7
1,2,3,4,7,8-HxCDD	0.492	0.248	0.553	57.2
1,2,3,6,7,8-HxCDD	0.812	0.346	0.930	55.4
1,2,3,7,8,9-HxCDD	0.660	0.480	0.868	56.8
1,2,3,4,6,7,8-HpCDD	3.78	1.88	4.81	54.0
OCDD	6.60	1.75	8.62	49.2
2,3,7,8-TeCDF	0.928	0.286	1.37	47.0
1,2,3,7,8-PeCDF	1.20	0.689	3.16	37.5
2,3,4,7,8-PeCDF	2.14	1.14	4.32	43.1
1,2,3,4,7,8-HxCDF	3.21	1.10	6.58	39.6
1,2,3,6,7,8-HxCDF	2.80	1.14	5.85	40.3
1,2,3,7,8,9-HxCDF	3.42	1.62	2.21	69.6
2,3,4,6,7,8-HxCDF	0.956	0.780	6.59	20.9
1,2,3,4,6,7,8-HpCDF	10.2	3.49	17.4	44.2
1,2,3,4,7,8,9-HpCDF	1.42	1.56	2.81	51.5
OCDF	6.08	1.54	8.17	48.3
PCDDs	12.8	5.14	16.4	52.1
PCDFs	32.4	13.3	58.4	43.9
Total	45.1	18.5	74.8	46.0
PCDDs I-TEQ	0.474	0.391	0.688	55.7
PCDFs I-TEQ	2.38	1.15	4.79	42.4
Total I-TEQ	2.86	1.54	5.48	44.5

I-TEQ emission rates, respectively. The removal efficiencies of the venturi scrubber on the total PCDD/F emission and the total PCDD/F I-TEQ emission are 46.0% and 44.5%, respectively. Different results were reported on the removal efficiency of PCDD/F by wet scrubber. Vogg et al. (1994) indicated the wet scrubber as a potential PCDD/F source and Kim et al. (2001) reported that the whole congeners of PCDD/F were enriched in the wet scrubber by representing removal

Table 5. PCDD/F flow rates (emission rate) and the PCDD/F removal efficiency of bag filter.

PCDD/Fs	Flue gases before bag filter ($\mu g\ hr^{-1}$)	Stack flue gases ($\mu g\ hr^{-1}$)	Fly ash ($\mu g\ hr^{-1}$)	PCDD/F removal efficiency of bag filter (%)
2,3,7,8-TeCDD	24.4	21.0	0.551	13.9
1,2,3,7,8-PeCDD	83.9	63.8	2.98	24.0
1,2,3,4,7,8-HxCDD	68.9	35.1	2.76	49.1
1,2,3,6,7,8-HxCDD	114	62.0	4.78	45.5
1,2,3,7,8,9-HxCDD	87.0	35.9	3.58	58.7
1,2,3,4,6,7,8-HpCDD	403	164	20.1	59.2
OCDD	412	190	25.6	54.0
2,3,7,8-TeCDF	469	516	12.8	2.42
1,2,3,7,8-PeCDF	281	308	12.2	3.80
2,3,4,7,8-PeCDF	576	553	25.3	4.01
1,2,3,4,7,8-HxCDF	520	483	41.5	7.16
1,2,3,6,7,8-HxCDF	472	396	33.0	16.1
1,2,3,7,8,9-HxCDF	33.2	21.3	2.73	36.0
2,3,4,6,7,8-HxCDF	507	396	31.2	21.8
1,2,3,4,6,7,8-HpCDF	1770	985	138	44.4
1,2,3,4,7,8,9-HpCDF	389	180	41.0	53.6
OCDF	2010	719	460	64.2
PCDDs	1190	572	64.8	52.1
PCDFs	7030	4560	855	35.1
Total	8220	5130	858	37.6
PCDDs I-TEQ	97.8	68.1	3.62	30.4
PCDFs I-TEQ	526	486	29.6	7.66
Total I-TEQ	624	554	31.0	11.2

efficiencies of -25% to -5731%. But Wevers et al. (1991) reported 71% of removal efficiencies of PCDD/F by wet scrubber, which is comparable to the result obtained in this study.

Removal of PCDD/F by bag filter

Table 5 lists the PCDD/F mass flow (emission) rates from the flue gases before and after the bag filter, and from fly ash, which are 8220, 5130, and 858 $\mu g\ hr^{-1}$, respectively. The removal efficiency of the bag filter is also shown in Table 5 and is determined by the following equations, (in which A = flue gases before the bag filter, B = flue gases after the bag filter, and C = fly ash):

Table 6. Removal efficiency of PCDD/Fs in different combination of air pollution control devices.

Combination of air pollution control devices	PCDD/F concentration and Removal efficiency	Reference
MSWI: Spray dryer absorber (140 °C) and Bag Filter	Spray lime mixed with 3-5 kg of activated carbon per hour PCDD/F in (a) untreated gas: 2.894 ng-TEQ/Nm^3, (b) purified gas: 0.033 ng-TEQ/Nm^3. Removal: 98.9%	Kim et al. 2001
MSWI: Spray dryer absorber (160 °C) and Bag Filter	Spray 40-60 kg of Sorbalit per hour PCDD/F in (a) untreated gas: 1.232 ng-TEQ/Nm^3, (b) purified gas: 0.029 ng-TEQ/Nm^3. Removal: 97.7%	
MSWI (capacity: 450 ton/day-incinerator) is equipped with cyclones, dry lime sorbent injection systems (DSI) and fabric filters (FF) as APCDs.	With activated carbon injection 115 kg/day (a) PCDD/F concentration before APCD: 47.5 ng/Nm^3, (b) PCDD/F concentration after APCD: 1.63 ng/Nm^3. Removal: 96.6%	Chang et al., 2002
MSWI is equipped with cyclones, dry sorbent injection and fabric filters.	With activated carbon injection 120 kg/day PCDD/F concentration in the stack flue gases: 0.71 ng/Nm^3. Removal: 98.9% With activated carbon injection 100 kg/day PCDD/F concentration in the stack flue gases: 0.89 ng/Nm^3. Removal: 98.6%	Chang et al., 2001
MWI: Quench Chamber (60 °C), Venturi Scrubber (45 °C), Packed-Bed Scrubber (40 °C)	PCDD/F concentration in the stack flue gases: 7.14 ng/Nm^3. Removal efficiency (%) of venturi scrubber: 46%	This study
Secondary ALS: Bag Filter (110~140 °C)	(a) PCDD/F concentration before bag filter: 171 ng/Nm^3, (b) PCDD/F concentration after bag filter: 99.6 ng/Nm^3. Removal efficiency of bag filter: 37.6%	This study

If A < B + C, that is, PCDD/Fs are generated in the bag filter,
Removal efficiency (%) = C/(B+C)*100%.
If A > B + C, (i.e., PCDD/Fs are declined in the bag filter),
Removal efficiency (%) = (A-B)/A*100%.

The removal efficiency of PCDD/Fs by bag filter apparently increases with the degree of chlorination. The average removal efficiencies of tetra-, penta-, hexa-, hepta-, and octa-PCDD/Fs are 8.2%, 10.6%, 33.5%, 52.4%, and 59.1%, respectively. It suggests that highly chlorinated PCDD/Fs with lower vapor pressures are more easily adsorbed onto the particulate and consequently more easily removed by bag filter. The typical operating temperature of bag filter (in this study, 110~140 °C) makes it unsuitable for removing semi-volatile pollutants, such as PCDD/Fs and PAHs.

The removal efficiencies of the bag filter on the total PCDD/F emission and the total PCDD/F I-TEQ emission are 37.6% and 11.2%, respectively. This result is lower than the 55.4% (mass) and 55.1% (toxicity) of Wang et al. (2003), and the 45% (mass) and 64% (toxicity) of Giugliano et al. (2002).

Comparing the Removal Efficiency of PCDD/Fs by Other Combinations of APCDs

The removal efficiency of PCDD/Fs in several different APCD combinations and their PCDD/F concentrations before and after APCDs are listed in Table 6. Kim et al. (2001) reported that the combination of a spray dryer absorber and a bag filter had a removal efficiency of 99%. Chang et al. (2001) also reported that the combination of cyclones, dry sorbent injection, and fabric filters had a removal efficiency of 99%, suggesting that a scrubber plus a bag filter with activated carbon may be regarded as "the most effective technique for PCDD/F emission control". Compared with the venutri scrubber and bag filter investigated in this study, this combination of spray dryer absorber and bag filter is measurably more efficient than either the venutri scrubber or the bag filter used alone. Thus, the individual removal efficiency of the venutri scrubber and bag filter on PCDD/F emission is inadequate.

4. Conclusions

The mean PCDD/F concentrations in the stack flue gases of the MWI and secondary ALS are 0.511 and 10.6 ng I-TEQ Nm^{-3}, which is 21 (=10.6/0.5) times higher than 0.5 ng I-TEQ Nm^{-3}, the regulation of PCDD/F emission from new established EAFs. The mean PCDD/F concentrations (contents) in the effluent, including ash and wastewater from a venturi scrubber and fly ash from a bag filter, are 7.51 ng I-TEQ g^{-1}, 154 pg I-TEQ L^{-1}, and 5.59 ng I-TEQ g^{-1}, respectively. Both the concentration of effluent and fly ash exceed the environmental quality standard and effluent standard. The average removal efficiencies of tetra-, penta-, hexa-, hepta-, and octa-PCDD/Fs by bag filter are 8.2%, 10.6%, 33.5%, 52.4%, and 59.1%, respectively. It suggests that highly chlorinated PCDD/Fs with lower vapor pressures are more easily adsorbed onto the particulate and consequently more easily removed by bag filter. The removal efficiencies of the bag filter on the total PCDD/F emission and the total PCDD/F I-TEQ emission are 37.6% and 11.2%, while that of venturi scrubber are 46.0% and 44.5%, respectively. Kim et al. (2001) reported that the combination of spray dryer absorber and bag filter had a removal efficiency of 99% for PCDD/Fs, which may allow it to be regarded as "the most effective technique for PCDD/F emission control". The combination of spray dryer absorber and a bag filter is measurably more efficient than either the venutri scrubber or the bag filter being used alone. Although the operating conditions of the venutri scrubber and the bag filter are different, the removal efficiency of PCDD/Fs for each is inadequate.

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