Removal characteristics of PCDD/Fs by the dual bag filter system of a fly ash treatment plant

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Abstract

In this study, a dual bag filter system was established to decrease the concentration of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) in the stack flue gases of a fly ash treatment plant, which retained zinc from the fly ash of electric arc furnaces. Compared to the original single bag filter system with activated carbon injection (ACI) of $40 \, \text{kg h}^{-1}$, the PCDD/F concentrations in the stack flue gas after the plant was equipped with the dual bag filter system dropped enormously from $3.38–7.73 \, \text{ng I-TEQ N m}^{-3}$ to $0.03 \, \text{ng I-TEQ N m}^{-3}$, and the total PCDD/F I-TEQ removal efficiency increased from 97.6% to 99.3% with ACI of $16 \, \text{kg h}^{-1}$, that is, about 40% of the original AC usage. The AC utilization efficiency ($0.560 \, \text{mg I-TEQ absorbed kg ACI}^{-1}$) of the dual bag filter system (AC: $16 \, \text{kg h}^{-1}$) was estimated about three times higher than that ($0.192 \, \text{mg I-TEQ absorbed kg ACI}^{-1}$) of single bag filter system with ACI ($40 \, \text{kg h}^{-1}$). The reason is that the particulate phase PCDD/Fs and some gas phase PCDD/Fs (captured by the recycled AC/ash mixture) were removed first by the first bag filter, and the rest of the gas phase PCDD/Fs were able to be adsorbed more completely by the AC/ash mixture due to a lower fly ash load, and then were removed by the second bag filter.

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1. Introduction

Electric arc furnaces (EAFs) are used to produce carbon and steel alloys from scrap material, which is often contaminated with paints, oils and coatings that contain chlorine. Because the conditions are conducive to the formation of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs), EAFs are one of the major PCDD/F emission sources in many countries [1–6]. Not only the stack flue gases but also the fly ashes, which are collected by air pollution control devices (APCDs) like bag filters, have high PCDD/F contents, which range between $0.560 \, \text{ng I-TEQ g}^{-1}$ and $1.15 \, \text{ng I-TEQ g}^{-1}$ [7–11].

The Waelz process, which was developed to obtain zinc from oxide ores initially, was commonly used to retain the zinc resources from the EAF fly ash presently [12]. High PCDD/F emissions have been a problem for all Waelz rotary-kiln installations. One Waelz rotary-kiln of Berzelius Umwelt-Service (B.U.S.) was operated in acid mode by the addition of SiO_2 as flux, and then was changed over to the basic mode. The APCDs was not changed. In acid operating mode, the PCDD/F concentration in the raw gas reached $50–1000 \, \text{ng TEQ m}^{-3}$, while in basic mode, the PCDD/F concentration drops significantly to $8–23 \, \text{ng TEQ m}^{-3}$ [13]. It is assumed that the basic substances (such as CaO) in basic mode react with HCl in the off-gas and thus reduce the chlorine supply needed for PCDD/F formation [13].

In Taiwan, EAFs discharge 170,000 tonnes year^{-1} of fly ash. In order to retain the zinc resources from the EAF fly ash, the fly ash treatment plant organized by the 13 EAF factories

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established a Waelz rotary-kiln, which treated over 70,000 tonnes of EAF dusts annually. However, extremely high PCDD/ F concentration (145 ng I-TEQ N m^{-3}) in the stack flue gases of the Waelz rotary-kiln had been observed [7]. Furthermore, the PCDD/F emission factor (560 µg I-TEQ tonnes EAF dust^{-1}) of the Waelz rotary-kiln [7] was much higher than that of other emission sources, like sinter plants (0.97 µg I-TEQ tonnes feedstock^{-1}) [14], EAFs (1.8 µg I-TEQ tonnes feedstock^{-1}) and secondary aluminum smelters (37 µg I-TEQ tonnes feedstock^{-1}) [15], and resulted in 30% of the total PCDD/F emission in Taiwan in 2004 [16].

To conform to the PCDD/F emission limits of 1.0 ng I-TEQ N m^{-3} for fly ash treatment facilities, which took effect in September 2006, the fly ash treatment plant examined in this paper proceeded with a series of improvement operations. After increasing the operational temperature of the dust settling chamber (DSC) from 480 °C to 580 °C, the PCDD/F concentrations before entering the venturi tower dropped dramatically from 1220 ng I-TEQ N m^{-3} to 394 ng I-TEQ N m^{-3} [17]. Together with injecting the activated carbon (AC) in front of the bag filter, these two measures reduced the PCDD/F concentrations in the stack flue gas from 145 ng I-TEQ N m^{-3} to 3.38–7.73 ng I-TEQ N m^{-3} [9,17]. However, even with the high AC usage (40 kg h^{-1}), the PCDD/F concentration still exceeded the limit of 1 ng I-TEQ N m^{-3}. This may have resulted from the low AC utilization efficiency (less than 3%) in the conventional activated carbon injection (ACI) for a single bag filter system [18].

A pilot plant equipped with a dual bag filter system with 2000 N m^3 h^{-1} capacity has been evaluated with regard to its PCDD/F removal efficiency for the flue gases of an actual municipal solid waste incinerator [18]. The PCDD/F concentration in the flue gases after being treated by the dual bag filter system could reach 0.05 ng I-TEQ N m^{-3}, with even less AC consumption than with the single bag filter system [18]. However, few studies concerning dual bag filter systems established on commercial plants had been published.

In this study, a dual bag filter system was established to reduce the PCDD/F concentration in the stack flue gases of the Waelz rotary-kiln. The flue gases before and after the first bag filter (FBF) and in the stack were sampled simultaneously to evaluate the removal characteristics of the FBF, second bag filter (SBF) and the whole dual bag filter system, respectively. The PCDD/F contents in the discharged ashes of the DSC, cyclone, FBF and SBF were also measured to obtain the PCDD/F distributions in the Waelz process with a dual bag filter system. Furthermore, the AC utilization efficiency of the dual bag filter system was compared with that of the single bag filter system.

2. Experimental

2.1. Basic information concerning the EAF fly ash treatment plant

The EAF fly ash treatment plant (capacity: 70,000 tonnes year^{-1}), which was established in 1999 to recover zinc oxide via the Waelz process, is depicted in Fig. 1. Due to the EAF fly ashes (chlorine content: 5%) were collected from different factories [7], the pellet process was designed to make the raw materials size uniformly. The feeding raw materials contain fly ash pellets (60–70%), coke (10–20%) and sand (10–20%) and the fly ash pellets (13.2 tonnes h^{-1}) were made of EAF fly ash (12 tonnes h^{-1}) and fine coke (1.2 tonnes h^{-1}). The Waelz process was conducted in a high temperature rotary-kiln (1200 °C) that was 40 m long and had a 3.6 m outer diameter. The higher side of the rotary-kiln was raw material feed (20.6 tonnes h^{-1}) and the lower side was slag discharge (11 tonnes h^{-1}). The slag
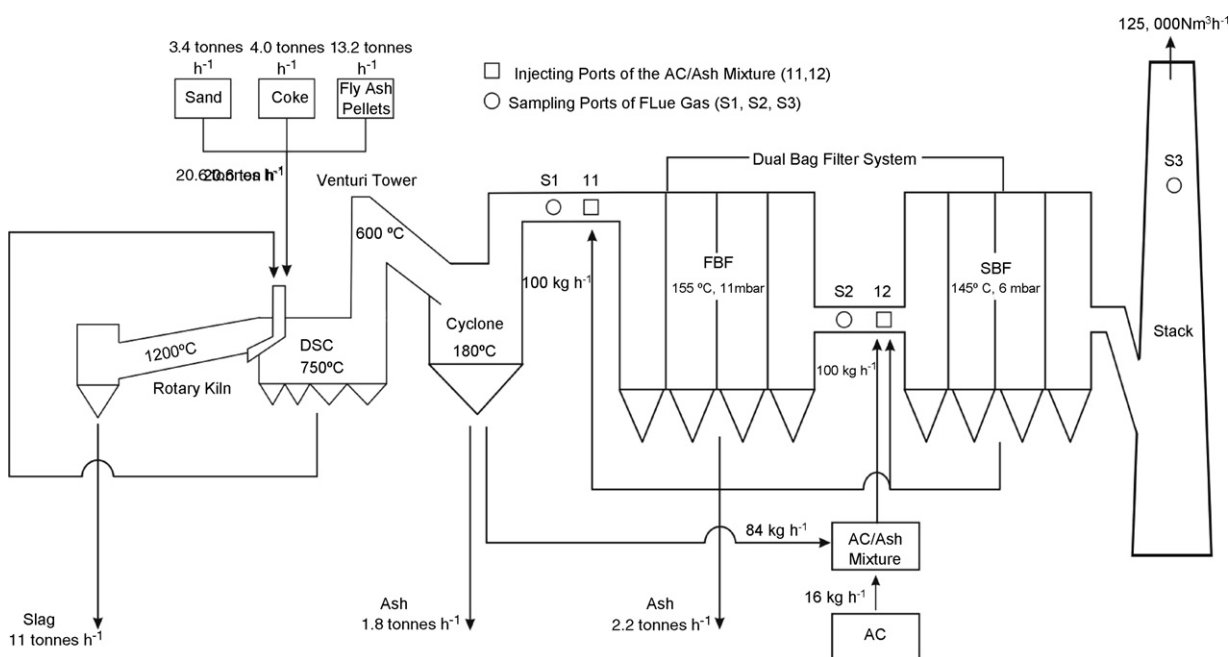

Fig. 1. The EAF fly ash treatment plant and its APCDs.

was used as road sub-base. The APCDs were DSC (750 °C), venturi tower (600 °C), cyclone (180 °C) and dual bag filter system, which were composed of FBF (155 °C), ACI and SBF (145 °C) in series. The discharged fly ashes from DSC were recycled to the rotary-kiln feed, while the fly ash gathered from the cyclone (1.8 tonnes h^{-1}) and FBF (2.2 tonnes h^{-1}) were used to recover the zinc. In order to avoid ignitions of the AC due to the high oxygen concentration (16%) in the flue gas, the AC (16 kg h^{-1}) was mixed with cyclone fly ash (84 kg h^{-1}) first and then the AC/ash mixture (100 kg h^{-1}) was injected prior to the SBF (I2). The SBF ashes, which were not emitted out of the system, were recycled back to the front of the SBF and some (100 kg h^{-1}) were injected before the FBF (I1).

2.2. PCDD/F sampling

All stack flue gas samplings and chemical analyses in this study were carried out by our accredited laboratory for PCDD/F stack flue gas samplings and analyses in Taiwan. Three sampling ports (see Fig. 1) were situated in the inlet of FBF (S1), the inlet of SBF (S2) and the stack (S3). The flue gas samplings from these three sampling ports were concurrent. The sampling time of each flue gas sample was 2 h, and a total of three samples were collected from each sampling port. The PCDD/F samples were collected isokinetically from the flue gases according to US EPA Modified Method 23. The sampling train adopted in this study is comparable with that specified by US EPA Modified Method 5. The flue gas samplings were performed on transect of the duct for each sampling port. Each 1 sampling axis and 24 positions on the axis was performed for the inlet of FBF and the inlet of SBF, while 2 intersect axes and 12 positions on the axes was performed for the stack. The recoveries of PCDD/F surrogate standards were 89–115%, and met the criteria within 70–130%. Although there were DSC, venture tower and cyclone equipped prior to the FBF, the particulate concentration in the flue gases before the FBF still reached 913.5 mg N m^{-3} [7]. To prevent the higher pressure drop occurring in the glass fiber filter, which was caused by the high particulate concentration, the glass fiber filter in the sampling train for sampling flue gases in the inlet of FBF (S1) was replaced periodically during sampling. To ensure the free contamination of the collected samples, one trip blank and one field blank were also taken during the field sampling. The details are similar to these given in our previous work [19].

In addition to flue gas samples, the discharged fly ashes from DSC, cyclone, FBF and SBF were collected to measure their PCDD/F contents. To obtain good representative samples, the fly ashes were sampled four times during the flue gas sampling period at intervals of half an hour and combined to a composite fly ash sample. Three composite fly ash samples were measured for PCDD/F for each device.

2.3. Analyses of PCDD/Fs

Analyses of stack flue gas and fly ash samples followed the US EPA Modified Method 23 and Modified Method 1613, respectively. Prior to analysis, each collected sample was spiked with a known amount of the $^{13}C_{12}$-labeled internal standard to

Table 1
PCDD/F concentrations in the flue gases before the FBF, before the SBF and in the stack

Sampling pores	Inlet of FBF		Inlet of SBF		Stack	
	Total PCDD/Fs (ng N m^{-3})	Total I-TEQ (ng I-TEQ N m^{-3})	Total PCDD/Fs (ng N m^{-3})	Total I-TEQ (ng I-TEQ N m^{-3})	Total PCDD/Fs (ng N m^{-3})	Total I-TEQ (ng I-TEQ N m^{-3})
PCDD/F concentrations	35.8	2.88	1.35	0.104	0.434	0.03
	51.7	4.12	2.62	0.234	0.381	0.03
	77.0	5.95	2.84	0.230	0.419	0.03
Mean	54.8	4.32	2.27	0.189	0.412	0.03
R.S.D. (%)	37.9	35.7	35.4	39.2	6.6	0

the extraction thimble. After being extracted for 24 h in a Soxhlet extractor, the extract was concentrated, treated with concentrated sulfuric acid, and then followed by a series of sample cleanup and fractionation procedures. The eluate was concentrated to approximately 1 mL and transferred to a vial. The concentrate was further concentrated to near dryness, using a stream of nitrogen. Prior to analysis, the standard solution for recovery checking was added to the sample. The recoveries of PCDD/F internal standards for the tetra-chlorinated homologues through hexa-chlorinated homologues were between 67% and 99%, and met the criteria within 40–130%, while that for the hepta- and octa-chlorinated homologues were between 54% and 103%, and met the criteria within 25–130%.

A high-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) was used for PCDD/Fs analyses. The HRGC (Hewlett Packard 6970 Series gas, CA, USA) was equipped with a DB-5MS fused silica capillary column ($L = 60$ m, ID = 0.25 mm, film thickness = 0.25 μm) (J & W Scientific, CA, USA), and with a 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 resolving power at 10,000. Details of the analytical procedures are given in our previous work [19].

3. Results and discussion

3.1. PCDD/F concentrations in the flue gases

The mean PCDD/F concentrations in the flue gas before the FBF (S1), before the SBF (S2) and in the stack (S3) are listed in Table 1, and were 54.8 ng N m^{-3}, 2.27 ng N m^{-3} and 0.412 ng N m^{-3}, respectively. The corresponding mean I-TEQ concentrations were 4.32 ng I-TEQ N m^{-3}, 0.189 ng I-TEQ N m^{-3} and 0.03 ng I-TEQ N m^{-3}, respectively. Compared to the original single bag filter system with ACI of 40 kg h^{-1}, the PCDD/F concentrations in the stack flue gas dropped enormously from 3.38–7.73 ng I-TEQ N m^{-3} [9,17], which had slightly higher particulate phase than gas phase PCDD/F concentration [17], to 0.03 ng I-TEQ N m^{-3}, which is much lower than the PCDD/F regulation of 1 ng I-TEQ N m^{-3}. Fig. 2 illustrated the PCDD/F I-TEQ concentration profiles in the flue gas before the FBF (S1), before the SBF (S2) and in the stack (S3), and revealed that the I-TEQ values were more contributed by PCDFs, especially by 2,3,4,7,8-PeCDF, than by PCDDs.

3.2. PCDD/F removal efficiencies of the dual bag filter system

The total PCDD/F removal efficiencies of the whole dual bag filter system, FBF and SBF are listed in Table 2, and were 99.2%, 95.8%, 79.5% and 99.3%, 95.6%, 82.3% on mass and toxicity bases, respectively. The calculated PCDD/F removal efficiency (97.6%) [17], of the original single bag filter system with ACI of 40 kg h^{-1} was comparable to that (96.6–98.9%) [20] of the combination of APCDs, that is, a dry scrubber plus a bag filter with ACI. However, the PCDD/F removal efficiency (97.6%) of
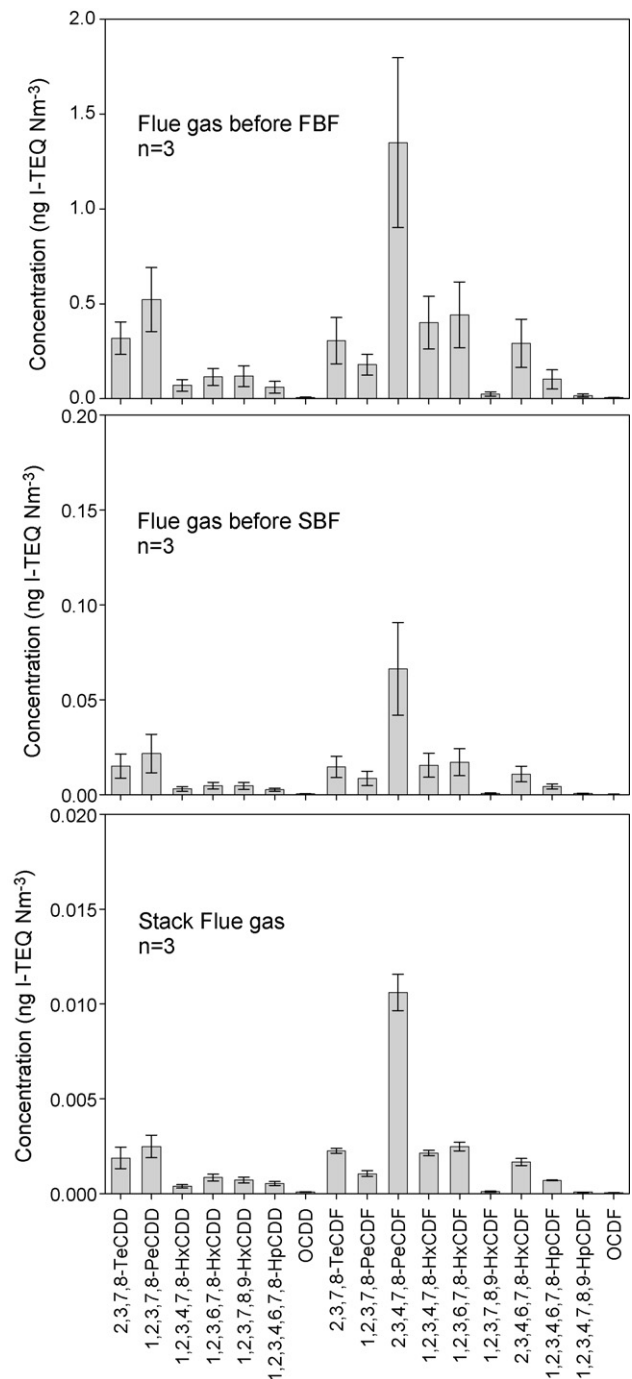

Fig. 2. PCDD/F I-TEQ concentrations in the flue gases.

the single bag filter system with ACI of 40 kg h^{-1} was lower than that (99.3%) of the dual bag filter system with ACI of 16 kg h^{-1}, that is, about 40% of the original AC usage. We extrapolated that the reason is that the particulate phase PCDD/Fs and some gas phase PCDD/Fs (captured by the recycled AC/ash mixture) were removed first by the FBF, and the rest of the gas phase PCDD/Fs were able to be adsorbed more completely by AC/ash mixture due to a lower fly ash load and then were removed by the SBF. The higher PCDD/F removal efficiency of the FBF than that of the SBF may be attributed to the greater difficulty

Table 2
PCDD/F removal efficiencies of the whole dual bag filter system, FBF and SBF

	Dual bag filter system		FBF		SBF	
	Mean (%)	S.D. (%)	Mean (%)	S.D. (%)	Mean (%)	S.D. (%)
Total PCDD/Fs	99.2	0.3	95.8	0.8	79.5	10.1
Total I-TEQ	99.3	0.3	95.6	1.1	82.3	11.1
PCDD						
TCDD	99.3	0.4	95.3	1.7	83.7	13.7
PeCDD	99.5	0.3	95.9	1.9	84.4	13.2
HxCDD	99.2	0.5	96.0	0.9	80.1	14.1
HpCDD	98.8	0.8	95.4	1.1	76.4	11.3
OCDD	98.6	0.2	95.3	1.3	68.6	11.5
PCDF						
TCDF	99.1	0.4	95.1	1.3	82.1	9.3
PeCDF	99.3	0.3	95.3	1.3	82.9	11.2
HxCDF	99.4	0.3	96.2	0.9	82.5	10.7
HpCDF	99.2	0.5	95.6	1.0	82.4	5.9
OCDF	99.3	0.1	96.6	1.8	74.0	15.6
Total PCDDs	98.9	0.5	95.6	0.7	74.7	11.6
Total PCDFs	99.3	0.3	95.9	0.8	81.9	9.0

S.D.: standard deviation.

in absorbing the gas phase PCDD/Fs with the AC/ash mixture than in removing the particulate phase PCDD/Fs with the bag filter.

The removal efficiencies of the FBF among the different PCDD/F homologues were similar, because the particulate phase PCDD/Fs were the main object to be removed. However, the PCDD/F removal mechanism of the SBF using ACI is mostly due to gas phase adsorption. Because the low chlorinated PCDD/F congeners with higher vapor pressure have more mass fraction in the gaseous phase, but the highly chlorinated PCDD/F congeners are primarily associated with particulates, the removal efficiencies of gas phase PCDD/Fs were higher than particle-bound PCDD/Fs for ACI [21,22]. This point can explain that the PCDD/F removal efficiencies of the SBF decreased with the increase in the chlorinated substitution numbers, and that of HpCDD, OCDD and OCDF were below 80%. Li et al. [16] observed the similar results that adding more amounts of AC (ACI of 20 kg h^{-1}, 40 kg h^{-1} and 50 kg h^{-1}) did enhance the PCDD/F removal efficiencies, and the reduction fractions of low chlorinated PCDD/F congeners were much higher than those of highly chlorinated PCDD/F congeners. Particularly, a lower amount of ACI (20 kg h^{-1}), not only cannot remove highly chlorinated PCDD/Fs, but also the carbon surface of the AC can act as a precursor for the formation promotion of highly chlorinated PCDD/F congeners.

3.3. PCDD/F contents in the discharged ashes of the different APCDs

The mean PCDD/F contents and their standard deviation (S.D.) in the discharged ashes of DSC, cyclone, FBF and SBF were shown in Table 3 and were 6.86 ± 0.842 ng I-TEQ g^{-1}, 0.444 ± 0.254 ng I-TEQ g^{-1}, 4.83 ± 0.253 ng I-TEQ g^{-1} and 51.1 ± 5.51 ng I-TEQ g^{-1}, respectively. Compared with previ-

ous studies [7,9], the reported PCDD/F contents in DSC ashes were close, and in the range of 4.17 ng I-TEQ g^{-1} to 6.86 ng I-TEQ g^{-1}. The PCDD/F contents in cyclone ashes were also similar to earlier studies, and the range was from 0.444 ng I-TEQ g^{-1} to 0.76 ng I-TEQ g^{-1}.

The PCDFs/PCDDs ratios in the discharged ashes of DSC, cyclone, FBF and SBF were also shown in Table 3. The PCDFs are more dominant in the FBF and SBF ashes (PCDFs/PCDDs ratio = 2.49–2.79) than in the cyclone ashes (1.06) and DSC ashes (0.6). Huang and Buekens [23] reviewed much research regarding the formation mechanisms of PCDD/F and concluded that the "de novo synthesis" can produce PCDD/Fs with the characteristic of "PCDFs/PCDDs ratio >1", while the "precursor formation" produces PCDD/Fs with "PCDFs/PCDDs ratio ≪ 1". Everaert and Baeyens [24] also indicate that precursor formation occurs mainly at temperatures ranged at 300–600 °C, with the typical PCDFs/PCDDs ratio <1. Consequently, the increasing PCDFs/PCDDs ratio reveals that de novo synthesis occurs during the cooling down process of the flue gas temperature.

The previous study [7] concluded that the higher PCDD/F content of DSC ash was attributed to the de novo synthesis, because their DSC operating temperature was about 467 °C, and the PCDFs/PCDDs ratio in the flue gas of DSC outlet was greater than 1. However, in this study, because the operating temperature was at 750 °C, and the PCDFs/PCDDs ratio of the DSC ashes was less than 1, the PCDD/F formation could be through precursor formation. With sufficient retention time and precursor formation, the PCDD/F contents in DSC ashes were higher than that in cyclone ashes, even higher than that in FBF ashes.

The PCDD/F contents (5.24 ng I-TEQ g^{-1}) in the bag filter ashes of the single bag filter system with ACI of 40 kg h^{-1} [9] was about five times higher than that without ACI (0.97 ng I-TEQ g^{-1}) [7], because of the gas phase PCDD/F adsorption by

Table 3
PCDD/F content (mean ± S.D.) in the discharged ashes of the different APCDs

		DSC	Cyclone	FBF	SBF	Reference
Dual bag filter system with ACI (16 kg h^{-1})	PCDDs (ng g^{-1})	153 ± 20.9	6.51 ± 5.12	18.9 ± 1.78	118 ± 26.9	This study
	PCDFs (ng g^{-1})	90.3 ± 4.25	6.50 ± 4.35	46.8 ± 1.81	327 ± 51.4	
	PCDFs/PCDDs ratio	0.60 ± 0.0527	1.06 ± 0.119	2.49 ± 0.155	2.79 ± 0.261	
	Total PCDD/Fs (ng g^{-1})	243 ± 25.1	13.0 ± 9.47	65.7 ± 3.54	445 ± 76.8	
	Total I-TEQ (ng I-TEQ g^{-1})	6.86 ± 0.842	0.444 ± 0.254	4.83 ± 0.253	51.1 ± 5.51	
Single bag filter system without ACI	Total I-TEQ (ng I-TEQ g^{-1})	4.17	0.76	0.97	–	[7]
Single bag filter system with ACI (40 kg h^{-1})	Total I-TEQ (ng I-TEQ g^{-1})	4.82	0.746	5.24	–	[9]

AC. The extremely elevated PCDD/F contents were observed in the SBF fly ashes (51.1 ng I-TEQ g^{-1}) of the dual bag filter system, which was about 10 and 50 times higher than that of the single bag filter system with ACI and without ACI, respectively.

The emission factor and the I-TEQ contribution fraction of the different output media of the fly ash treatment plant are shown in Table 4. The total emission factors of the fly ash treatment plant were 0.840 mg I-TEQ tonnes EAF fly ash^{-1}, 1.12 mg I-TEQ tonnes EAF fly ash^{-1} and 0.955 mg I-TEQ tonnes EAF fly ash^{-1} for single bag filter system without ACI, single bag filter system with ACI and dual bag filter system, respectively. Although the fly ash treatment plant had preceded a series of operation improvements, the total PCDD/F emission based on the EAF fly ash feeding was still maintained at the same level, revealing that the measures adopted merely transferred PCDD/Fs from the gas stream to fly ash.

3.4. PCDD/F contribution fractions of the different output media

The PCDD/F I-TEQ contribution fractions in the discharged ashes of cyclone, bag filter and in the stack flue gases (see Table 4) were about 20.4%, 13.0% and 66.7%, respectively for the single bag filter system without ACI [7]. Most of the PCDD/Fs generated were emitted via stack flue gases. After adopting ACI to control PCDD/Fs, the PCDD/F I-TEQ contribution fractions of the bag filter ash increased enormously to 84.1%, while that of the stack flue gas decreased dramatically to 3.79% [9]. ACI resulted in most of the PCDD/Fs being adsorbed and retained with the bag filters. After changing to the

dual bag filter system, the higher AC utilization efficiency made the PCDD/F I-TEQ contribution fractions in the stack flue gas further decrease to 0.0307%, even less than that of the slag.

The rough estimates of the AC utilization efficiency of the single bag filter system (AC: 40 kg h^{-1}) and the dual bag filter system (AC: 16 kg h^{-1}) were calculated as follows:

The AC utilization efficiency (mg I-TEQ absorbed kg ACI^{-1})

$$= \frac{(A - B)C}{D}$$

where A is the PCDD/F I-TEQ content in the bag filter ash of the single bag filter system with ACI or the dual bag filter system, B the PCDD/F I-TEQ content in the bag filter ash of the single bag filter system without ACI, C the fly ash discharging rate of the single bag filter system with ACI or the dual bag filter system, D is the AC feeding rates of the single or dual bag filter system.

So the AC utilization efficiency of the single bag filter system (AC: 40 kg h^{-1}) and the dual bag filter system (AC: 16 kg h^{-1}) were 0.192 mg I-TEQ absorbed kg ACI^{-1} and 0.560 mg I-TEQ absorbed kg ACI^{-1}, respectively. The AC utilization efficiency of the dual bag filter system (AC: 16 kg h^{-1}) was about 3 (=0.560/0.192) times higher than that of single bag filter system with ACI (40 kg h^{-1}).

3.5. PCDD/F congener profiles in the fly ashes and flue gases

The congener profiles of the seventeen 2,3,7,8-chlorinated substituted PCDD/Fs (mean ± S.D.) detected from the fly ashes and flue gases are shown in Figs. 3 and 4, respectively.

Table 4
The emission factors and the I-TEQ contribution fractions of the output media

		Slag	Cyclone	FBF	Stack	Total	Reference
Emission factors mg I-TEQ ton-EAF fly ash^{-1}	Single bag filter system without ACI	–	0.171	0.109	0.560	0.840	[7]
	Single bag filter system with ACI (40 kg h^{-1})	0.00173	0.134	0.944	0.0425	1.12	[9]
	Dual bag filter system with ACI (16 kg h^{-1})	0.00226[a]	0.0666	0.886	0.000293	0.955	This study
I-TEQ contribution (%)	Single bag filter system without ACI	–	20.4	13.0	66.7	100	[7]
	Single bag filter system with ACI (40 kg h^{-1})	0.154	11.9	84.1	3.79	100	[9]
	Dual bag filter system with ACI (16 kg h^{-1})	0.237	6.98	92.8	0.0307	100	This study

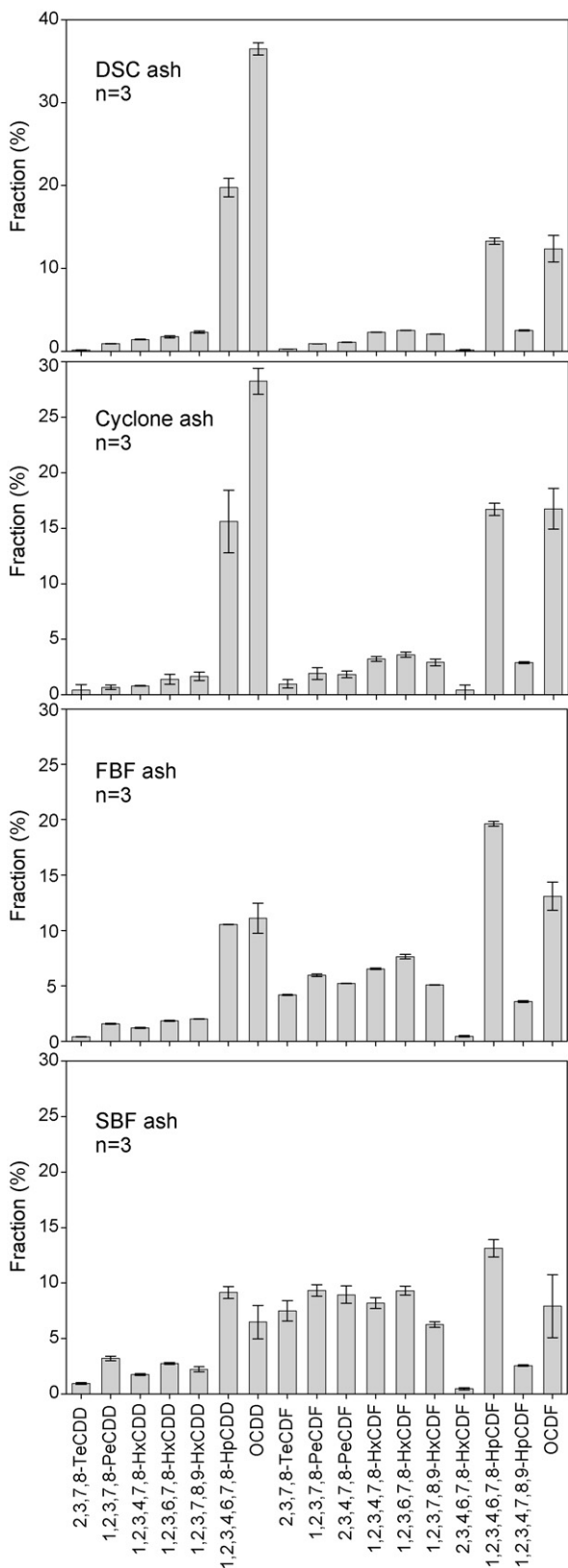[a] Calculated according to the PCDD/F content obtained by Li et al. [9].

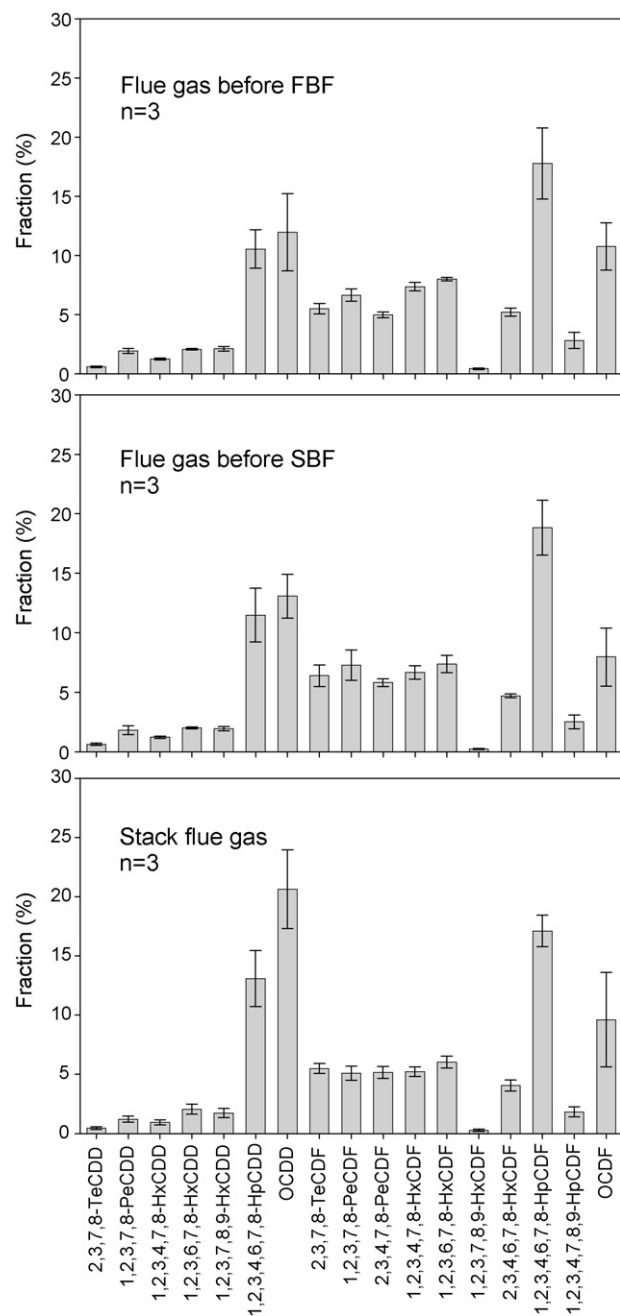Fig. 3. PCDD/F congener profiles detected from the fly ashes.

Fig. 4. PCDD/F congener profiles detected from the flue gases.

The PCDD/F congener profiles in the flue gases before (S1) and after FBF (S2) were similar, although fly ashes were already removed. Because the percentages of gas phase and particulate phase PCDD/F in the flue gases before the bag filter were very similar [17], this shows that the congener profiles of the gas phase and particulate phase PCDD/Fs in the flue gases were alike.

Comparing the congener profiles of the cyclone, FBF and SBF ashes with each other, the fractions of the low chlorinated substituted PCDD/Fs in these three different air pollution control ashes increased with the flue gas flow direction, revealing the occurrence of de novo synthesis and gas phase PCDD/F adsorption by AC, which had higher removal efficiency on the

lower chlorinated substituted congeners. The PCDD/F congener profiles in the flue gas altered slightly after passing through the SBF (S3). The fraction of the low chlorinated substituted PCDD/Fs decreased while that of the high chlorinated substituted PCDD/Fs increased. The PCDD/F congener profile in the stack flue gas was influenced by the gas phase PCDD/F adsorption of the SBF using ACI, because the low chlorinated PCDD/F congeners with higher vapor pressure have more mass fraction in the gaseous phase.

4. Conclusions

For the fly ash treatment plant, the mean PCDD/F I-TEQ concentrations in the flue gas before the FBF (S1), before the SBF (S2) and in the stack (S3) were $4.32\,\text{ng I-TEQ N m}^{-3}$, $0.189\,\text{ng I-TEQ N m}^{-3}$ and $0.03\,\text{ng I-TEQ N m}^{-3}$, respectively. Compared with the original single bag filter system with ACI of $40\,\text{kg h}^{-1}$, the PCDD/F concentrations in the stack flue gas after the plant was equipped with the dual bag filter system dropped enormously from $3.38–7.73\,\text{ng I-TEQ N m}^{-3}$ to $0.03\,\text{ng I-TEQ N m}^{-3}$, and the PCDD/F I-TEQ contribution fractions in the stack flue gas further decreased to 0.0307% of the total PCDD/F I-TEQ emission, even less than that of the slag. The total PCDD/F I-TEQ removal efficiencies of the whole dual bag filter system, FBF and SBF, were 99.3%, 95.6% and 82.3%, respectively. For the SBF, the PCDD/F removal efficiencies decreased with the increase in the chlorinated substitution numbers, and that of HpCDD, OCDD and OCDF were below 80%. The reason is that the PCDD/F removal mechanism of the SBF using ACI is mostly due to gas phase adsorption, and the low chlorinated PCDD/F congeners with higher vapor pressure have more mass fraction in the gaseous phase.

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