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Removal of PCDD/Fs and PCBs from flue gas using a pilot gas cleaning system

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Abstract

A 100 Nm³/hr capacity pilot scale dual bag filter (DBF) system was tested on the flue gas from an actual hazardous waste incinerator (HWI), the removal efficiency of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) was also studied. The first filter collected most of the fly ash and associated chlorinated organic; then activated carbon (AC) was injected and used to collect phase chlorinated organic from the gas. Concentrations of PCDD/Fs and PCBs after the DBF system were 0.07 and 0.01 ng TEQ/Nm³, respectively, which were both far below the national emission standard. Comparing with the original single bag filter system, the PCDD/Fs concentration dropped a lot from 0.36 to 0.07 ng TEQ/Nm³. Increasing AC feeding rate enhanced their collection efficiency, yet reduced the AC utilization efficiency, and it still needs further study to select an appropriate feeding rate in the system. These results will be useful for industrial application and assist in controlling emissions of PCDD/Fs and other persistent organic pollutants from stationary sources in China.

Key words: PCDD/Fs; PCBs; dual bag filter; activated carbon
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Introduction

With the development of economy, the amount of hazardous wastes in China is increasing greatly year by year. During the decade from 2000 to 2009, the amount of hazardous waste generated increased from 8.3 to 14.3 million tons (National Bureau of Statistics of China, 2001, 2010). Most hazardous wastes are toxic, corrosive, flammable, explosive, and the pollution has the potential and hysteresis quality, so they have been one of the key and difficult problems of the global environmental protection. As incineration can realize waste harmlessness, decrease, and resource, it is considered as the most effective disposal method for hazardous waste in China (Duan et al., 2008). However, it faces the problem of huge emission of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs) and other persistent organic pollutants (POPs) in the combustion. The concentration of PCDD/Fs in stack gas from hospital waste incinerators in China was higher than the national standard of 0.5 ng TEQ/Nm³ in 2006 (Liu et al., 2006). Gao et al. (2009) investigated stack gas emission of PCDD/Fs from 14 domestic-made medical waste incinerators in China found that only two facilities exhibited emission levels below the European Union directive emission limit of 0.1 ng TEQ/Nm³, and another two plants exhibited emission levels above 10.0 ng TEQ/Nm³. According to statistics, approximately 80% of total PCDD/Fs emission (610.5 g TEQ) of waste incineration was from hazardous waste and medical waste incinerations in China in 2004 by the PCDD/Fs inventory reported in P. R. China National Implementation Plan for the Stockholm Convention on Persistent Organic Pollutants (NIP, 2007). Therefore it is necessary to control the emissions of PCDD/Fs from the hazardous waste and medical waste incinerators.

In order to reduce the emission, bag filter system with activated carbon (AC) adsorption is widely employed in industrial processes (Tejima et al., 1996; Kim et al., 2001; Karadenir et al., 2004; Chang et al., 2009; Wang et al., 2003, 2009), the removal efficiency could reach to 95%, and even higher, the PCDD/Fs concentration in the flue gas can be controlled lower than 0.5 ng TEQ/Nm³. With the increasing stringent national standard of dioxin emissions from hazardous waste incineration, the traditional single bag filter system has been difficult to meet...
it now. Increasing the amount of AC is not an effective measure that it will affect the overall removal efficiency of PCDD/Fs (Everaert and Baeyens, 2002; Li et al., 2008), and excessive AC might actually increase the potential formation of PCDD/Fs (Buekens and Huang, 1998; Chi et al., 2005). Furthermore, previous studies indicate that the utilization efficiency of activated carbon is very low, which is less than 3%, in the single bag filter system (Kim et al., 2007; Chi et al., 2008). Therefore, a new technology with high removal efficiency of PCDD/Fs and high utilization of AC is needed.

Kim et al. (2007) designed a dual bag filter (DBF) system in the municipal solid waste incinerator to improve the utilization efficiency of AC and reduce the emission of PCDD/Fs. In the system, the dioxin emission was about 0.05 ng TEQ/Nm³, with even less AC consumption (40 mg/Nm³) than the single bag filter system (100 mg/Nm³). Chi et al. (2008) also found that the DBF system with AC feeding could reduce PCDD/Fs concentration from 4.62 to (0.235 ± 0.04) ng TEQ/Nm³ in the Waltz kiln test, and the consumption of AC was reduced from 540 to 215 mg/Nm³. Lin et al. (2008) designed a DBF system in a fly ash treatment plant and found that the PCDD/F concentration in the stack flue gas after the plant was equipped with the dual bag filter system dropped enormously from 3.38–7.73 to 0.03 ng TEQ/Nm³, and the utilization efficiency of AC could change from 0.192 ng/g in the single bag filter to 0.560 ng/g in the DBF system, which improved the utilization rate two times. Therefore, the DBF system is effective to control the emission of dioxin and improve the utilization of AC, which could make up the shortcomings of the single bag system. However, few studies concerning dual bag filter system established in industrial plants in China has been published.

In this study, a 100 Nm³/hr capacity pilot scale DBF system was used to study the removal characteristics of PCDD/Fs and PCBs from flue gas of an actual hazardous waste incinerator (HWI) (20 tons/day). The test was operated in the condition with different AC feeding rates (50 and 250 mg/Nm³), and the AC utilization efficiency of different AC feeding rates was also investigated.

1 Materials and methods

1.1 DBF system in hazardous waste incinerator

The DBF system was designed at the flue bypass of a HWI with 20 tons/day capacity. The average value of total exhaust gas was 8000 Nm³/hr. As shown in Fig. 1, the system consists of three major parts, waste pretreatment system, incineration system, and flue gas treatment system. Waste pretreatment system includes waste pretreatment and feed system; incineration system includes rotary kiln and the second combustion chamber; flue gas treatment system includes quenching tower, and air pollution control devices.

The DBF system was composed of two bag filter connected in series as shown in Fig. 1. Hydrated lime was sprayed into the entrance of the first bag filter (FBF) to remove acid gases when the acid gases concentration, such as SO₂, HCl, etc, in the flue gas was high, and they could be
removed together with fly ash by filtration. In the FBF, fly ash and particle fraction of dioxin attached were removed by filtration, and the filtering layer on the first bag surface formed by fly ash could absorb a certain amount of gaseous dioxin. The other gaseous dioxin escaped from the first bag filter enters the second bag filter (SBF), where AC was injected to absorb the rest gaseous dioxin.

AC can be gathered on the bottom of the SBF, and then be injected into the second bag again to form a recycle system of AC, which could reduce AC consumption. The recycle system of AC was shown in Fig. 1. First, we shut off controller 3 and controller 2, open controller 1. Then we cleaned the FBF dust once, closed controller 1 and open controller 2 after about twenty minutes. Last, we closed controller 2 and open controller 1 after about one minute. And we repeated the process again until we finished a sampling. After sampling, we opened controller 3 to discharge AC totally. Three sampling ports (S1, S2, and S3) were set in the DBF system (Fig. 1). For sampling, flue gas passed through an ice-cooled impingers containing high purity water and toluene, respectively, then a filtering device containing PUF.

Table 1 shows design parameters of the DBF system. During testing, the volume of flue gas was controlled through the DBF about 100 Nm$^3$/hr. The inlet temperature of the FBF and SBF was 180 and 160°C, respectively. The outlet temperature of the SBF was 140°C. The gas velocity of both filters was 0.35 m/sec. Filtration area of both bag filters were 2.72 m$^2$. AC was uniform feed by packing auger system, and the feed rates were 50 and 250 mg/sec of both filters was 0.35 m/sec.

### 2 Results and discussion

#### 2.1 PCDD/Fs emission of the HWI

The normal PCDD/Fs concentration in the flue gas at stack of the HWI is about 0.36 ng TEQ/Nm$^3$, which is below the national standard of 0.5 ng TEQ/Nm$^3$. Figure 2 illustrates the PCDD/F TEQ concentration profiles in the flue gas (normal) and reveals PCDDs contributed the most TEQ values, especially 2,3,4,7,8-PeCDF, which was similar to previous studies (Chang et al., 2002; Karadenir et al., 2004; Gao et al., 2009; Ren et al., 2011). It might be due to the high amount of chlorine content in the waste. The chlorine content in the waste played an important role in determining the congener profiles and formation mechanisms of PCDD/Fs in stack gas. When the amount of chlorine content was high, PCDD/Fs would be formed through de novo synthesis, and more PCDFs would be formed than PCDDs (Wilhelm et al., 2001; Wang et al., 2003; Li et al., 2008). The amount of chlorine content of hazardous waste and hospital waste was usually higher than that of municipal waste due to high content of polyvinyl chloride (PVC). Therefore the de novo synthesis was dominant in the incineration over precursor formation and gave high content of PCDDs.

Although PCDD/Fs concentration in stack flue gas of the HWI is below the national standard, there were still some ways to control the emission to a lower level as previous studies (Kim et al., 2007; Chi et al., 2008; Lin et al., 2008). If we improve the removal efficiency of bag filter system with AC, we would control PCDD/Fs emission close to 0.1 ng TEQ/Nm$^3$, and even lower.

#### 2.2 PCDD/Fs removal

The total concentrations of PCDD/Fs at different sampling ports and removal efficiencies of PCDD/Fs of the FBF, SBF, and DBF system were shown in Table 2. The removal rates of PCDD/Fs were calculated from the total concentrations of PCDD/Fs of the raw flue gas and treated flue gas.

Table 3 shows the results of the chemical and physical properties of AC used in the experiment. The characteristics of AC are very important in the adsorption process. The type of AC is HC 767, produced by Zhejiang Hangmu Timber Industrial Co., Ltd. AC Branch. The characteristics of AC pore before adsorption were tested by Quantochrome Autosobe. Particle size distribution of AC was measured by Malvern particle size analyzer, it was found that AC particle size was mainly distributed between 10–100 μm. The average pore radius of AC used in our test was 3.164 nm, and the BET surface area of AC was 1291.66 m$^2$/g. The specific surface area of AC was calculated using the Quantochrome Autosobe data.

### 2.3 Sample analysis

The cleanup procedure of PCDD/Fs and PCBs samples were according to the US EPA method 1613 (for PCDD/Fs samples) and 1618 (for PCBs samples), respectively. Samples were analyzed by means of high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS JEOL, JMS-800D) with a DB-5MS (60 m length × 0.25 mm ID × 0.25 μm film) column. The mean recoveries of standards for PCDD/Fs and PCBs range from 67% to 131% and 40% to 110%, respectively. The recoveries are all within acceptable 25% to 150% range.

The concentrations were normalized to dry air, 11% O$_2$, 1.01 × 10$^5$ kPa and 237 K. In this study, the total TEQ concentration of PCDD/Fs is calculated using international toxic equivalence factors (I-TEFs), while the TEQ of PCBs is calculated using the World Health Organization (WHO)-TEFs (van den Berg et al., 2006).

#### 2.3.1 TEQs of PCDD/Fs samples

The PCDD/Fs were analyzed using the HRGC/HRMS JEOL, JMS-800D) with a DB-5MS (60 m length × 0.25 mm ID × 0.25 μm film) column. The mean recoveries of standards for PCDD/Fs are all within acceptable 25% to 150% range.
SBF and DBF system with different AC feeding rates are shown in Table 2.

When AC feeding rate was 50 mg/Nm$^3$, concentration of PCDD/Fs in the flue gas after the DBF system was 0.07 ng TEQ/Nm$^3$, which was much lower than the national standard of 0.5 ng TEQ/Nm$^3$, and the removal efficiency of PCDD/Fs was 97.0%. Comparing with the PCDD/Fs emission of the hazardous waste incinerator, the concentration had dropped nearly an order from 0.36 to 0.07 ng TEQ/Nm$^3$, the control effect was obvious. When we changed AC feeding rate to 250 mg/Nm$^3$, the concentration of PCDD/Fs in the flue gas after the DBF system was 0.07 ng TEQ/Nm$^3$ with a higher removal efficiency of 97.2%, which had a little better effect comparing with the feeding rate of 50 mg/Nm$^3$. We also find that the PCDD/Fs concentration after FBF system were 0.16 and 0.31 ng TEQ/Nm$^3$ without AC injection, which were lower than the emission of the single bag filter system of 0.36 ng TEQ/Nm$^3$. It might be attributed to the fact that the formation of PCDD/Fs in the post-combustion zone of the actual combustion process (Huang and Buekens, 1995). Further study is needed to investigate the formation potential and removal mechanisms of PCDD/Fs in flue gas.

As shown in Table 2, the PCDD/Fs removal efficiency of the FBF system with different feeding rates were both higher than that of the SBF system. Similar result was also reported in literature (Lin et al., 2008). Because most of the solid phase and a amount of gaseous PCDD/Fs were adhered to the surface of fly ash and removed by filtration, and the concentration of dioxins second bag filter was far below the first one, and it was more difficult to decrease to a lower one. The removal efficiency of PCDD/Fs of the DBF system increased about 0.2%, from 97.0% to 97.2%, when AC feeding rate increased from 50 to 250 mg/Nm$^3$. So increasing AC feeding rate could improve the removal efficiency in some certain, due to there are more pore sites for gas phase PCDD/Fs adsorption. However, excessive AC injection rate would reduce the usage of the AC, increase the total PCDD/Fs discharge and might actually increase the potential of PCDD/Fs formation (Buekens and Huang, 1998; Everaert and Baeyens, 2002; Chi et al., 2005; Li et al., 2008). Therefore, we should select the reasonable AC feeding rate in practical application and avoid waste of AC.

The PCDD/F TEQ concentration profiles in the flue gas (S1, S2 and S3) with different feeding rates at three sampling sites are shown in Fig. 2. The PCDD/F TEQ

\[
\text{Removal efficiency: } R = \frac{(\text{concentration of PCDD/Fs in inlet} - \text{concentration of PCDD/Fs in outlet})}{\text{concentration of PCDD/Fs in inlet}} \times 100%.
\]

S1, S2, S3 are sampling ports.

### Table 2

<table>
<thead>
<tr>
<th>AC feeding rate (mg/Nm$^3$)</th>
<th>Concentration of PCDD/Fs (ng TEQ/Nm$^3$)</th>
<th>Removal efficiency of PCDD/Fs (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>S1</td>
<td>S2</td>
</tr>
<tr>
<td>50</td>
<td>2.28</td>
<td>0.16</td>
</tr>
<tr>
<td>250</td>
<td>2.46</td>
<td>0.31</td>
</tr>
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</table>

Removal efficiency: $R_n (n = \text{BF, SBF, total}) = [(\text{concentration of PCDD/Fs in inlet} - \text{concentration of PCDD/Fs in outlet})/\text{concentration of PCDD/Fs in inlet}] \times 100%$. S1, S2, S3 are sampling ports.
concentration profiles in the flue gas before the FBF system were similar between the conditions, and PCDD/Fs contributed the most TEQ values, especially 2,3,4,7,8-PeCDF and HxCDF. The result was similar to previous studies (Kim et al., 2007; Lin et al., 2008). It might due to the high amount of chlorine content in the waste and the de novo synthesis in the incineration (Wilhelm et al., 2001; Wang et al., 2003). The distribution of PCDD/Fs changed a little after flue gas flowed through the FBF, and remained the same after flue gas flowed through the SBF. PCDFs still contributed the most TEQ values, especially 2,3,4,7,8-PeCDF. When we increased AC feeding rate from 50 to 250 mg/Nm³, the distribution of PCDD/Fs in the flue gas after the SBF did not change. So the AC injection did not affect the distribution of PCDD/Fs much in the flue gas. We can conclude that the DBF system had a greater ability to control PCDD/F emission without changing the distribution of PCDD/Fs comparing with the single bag system of the HWI.

Figure 3 shows the removal efficiencies of PCDD/F congeners of bag filter systems with different feeding rates. The removal efficiencies of PCDD/F congeners of the FBF system were nearly similar, but become irregular of the SBF system. We can explain that the particulate phase PCDD/Fs were the main object to be removed in the FBF system, the removal mechanism of the particulate phase PCDD/Fs depended on the filtration of the bag filter, so the removal efficiency was nearly similar. The removal mechanism of the SBF system was much more complicated, it depends on gas-solid phase proportion of PCDD/F congeners, the adsorption capacity of AC to PCDD/F congeners, and some other factors. It might be due to the complicated removal mechanism and process, the removal efficiency of PCDD/F congeners of the SBF system become irregular. Although some previous studies (Chi et al., 2008; Lin et al., 2008) found that the highly chlorinated PCDD/Fs with low vapor pressure so easily adhered to the particles that the removal efficiencies of PCDD/F congeners from the SBF system decreased with the increase in the chlorinated substitution numbers, there exists many potential influencing factors in our system, including materials, incinerator and operation conditions, and it needs a further study on the removal mechanism of the SBF system.

2.3 PCBs removal

Under the same experimental conditions, we analyzed the removal characteristic of PCBs in the DBF system (Table 3).

When AC feeding rate was 50 mg/Nm³, concentration of PCBs in the flue gas changed from 60.6 to 10.9 pg TEQ/Nm³, and the removal efficiency of PCBs of the DBF system was 82.0%, which was lower than the removal efficiency of PCDD/Fs. When we changed AC feeding rate to 250 mg/Nm³, concentration of PCBs in the flue gas changed from 57.5 to 9.5 pg TEQ/Nm³, with the removal efficiency of 83.5%, which was a little higher than the feeding rate of 50 mg/Nm³ was. We could find that the removal efficiency of PCBs was much lower than PCDD/Fs. It might be attributed to the low concentration of PCBs, and it is difficult to reduce it to a lower level. The second possible cause is that the amount of AC is not sufficient. As shown in Table 3, the removal efficiency increased from 39.4% to 46.0% after we increased the AC feeding rate, so it may need much more AC to absorb PCBs in the system.

The PCB removal efficiency of the FBF system with different feeding rates were both higher than that of the SBF system (Table 3), which were the same with the removal efficiency of PCDD/Fs. We could explain the phenomenon that most of the solid-phase and part of gas-phase PCBs that adhered to the fly ash were removed by filtration.

Figure 4 shows the PCB concentration profiles with different feeding rates at three sampling sites. The distribution of PCB congeners in the inlet of the FBF system was similar between the conditions, and PCB 77, 118 and 105 were the main congeners, account for nearly 90%. The distribution of PCB congeners changed little after the flue gas flowed through the FBF and SBF system, and it still remained the same when we increased AC feeding rate from 50 to 250 mg/Nm³. The phenomenon was the same with that of PCDD/Fs, and the DBF system with AC feeding rate 250 mg/Nm³ had more PCA removed than AC feeding rate 50 mg/Nm³.
injection did not change the distribution of PCB congeners much. We also found that PCB 126 and 169 contributed the most TEQ values, account for more than 99%. The result was similar to previous studies (Kim et al., 2004; Chi et al., 2008; Choi et al., 2008; Guo et al., 2009), due to their TEF of 0.1 and 0.01 (Luthardt et al., 2002; Aries et al., 2004; Abad et al., 2006).

Figure 5 shows the removal efficiencies of PCB congeners of bag filter systems with different feeding rates. The removal efficiencies of PCB congeners of the FBF system were different and irregular, but were nearly similar of the SBF system, which was the same with the removal efficiency of PCDD/Fs. We could explain it that all PCB congeners were mostly distributed in vapor-phase after flue gas flowed through the FBF system. Chang et al. observed the similar result that the ratio of vapor-phase PCBs to solid-phase PCBs is about 3 for all twelve PCB congeners (Chang et al., 2009; Chi et al., 2008). So the low removal efficiencies of PCB 118, 114, 105 and 157 in the FBF system might be attributed to most of them were gas-phase in the flue gas, and few adhered to the fly ash. The removal efficiencies of PCB congeners of the SBF system improved a lot when AC feeding rate increased from 50 to 250 mg/Nm$^3$, but the distribution of removal efficiencies were nearly the same. We could consider increasing AC feeding rate to improve the removal efficiency of PCBs, but the reasonable amount needs further study.

2.4 AC utilization efficiency

As AC was just injected before the SBF system, we estimated AC utilization efficiency ($E$) by comparing PCDD/Fs and PCBs concentration changed before and after the SBF system, which was calculated as follows:

$$E = \frac{(S_a - S_b)}{R} \tag{1}$$

where, $S_a$ was the PCDD/Fs or PCBs concentration after the SBF, $S_b$ was the PCDD/Fs or PCBs concentration before the SBF, and $R$ was the feeding rate. When AC feeding rate changed from 50 to 250 mg/Nm$^3$, the AC utilization efficiency of removing PCDD/Fs dropped from 1.68 to 0.96 ng TEQ/g, and the AC utilization efficiency of removing PCBs dropped from 0.14 to 0.03 ng TEQ/g. The utilization efficiencies of them dropped about 42.9% and 78.5%, respectively.

The AC feeding rate changed from 50 to 250 mg/Nm$^3$, PCDD/Fs and PCBs removal efficiency increased, but the AC utilization efficiency decreased greatly. We can find that, in a certain range, increasing the AC feeding rate can improve the removal efficiency of PCDD/Fs and PCBs, but it also can reduce AC utilization efficiency greatly in the DBF system.

3 Conclusions

According to the results of the experiment, the PCDD/Fs and PCBs concentration after the DBF system were 0.07 and 0.01 ng TEQ/Nm$^3$, respectively, which were both far below the national emission regulation and the removal efficiency of them were about 97% and 83%, respectively. Comparing with the original single bag filter system, the PCDD/Fs concentration dropped a lot from 0.36 to 0.07 ng TEQ/Nm$^3$. The removal efficiency of PCDD/Fs and PCBs in the DBF system increased 0.4% and 1.7% when AC feeding rate increased from 50 to 250 mg/Nm$^3$, while the AC utilization efficiency dropped 43.1% and 77.5%. In a certain range, increasing the AC feeding rate can improve the removal efficiency to remove dioxin-like compounds, but it also can reduce the AC utilization efficiency greatly in the DBF system. We conclude that there is no need to increase the AC feeding rate to reduce emission of PCDD/Fs in the actually combustion process, excessive AC injection will lead to a waste of resource, and it needs further study to decide a reasonable AC injection rate to control the emission of PCBs. The results of this article will be useful for industrial application and assist in controlling emissions of PCDD/Fs and other POPs from stationary sources in China.
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