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Abstract: A 1000 Nm³/h capacity pilot scale dual baghouse filter system was tested on flue gas and fly ash from a municipal solid waste incinerator, and the removal efficiency of dioxins, heavy metals and HCl was studied. Activated carbon was injected at the inlet of the first baghouse filter to remove the gas phase dioxins and heavy metals, and baking soda was injected at the inlet of the second baghouse filter to remove HCl. Concentrations of dioxins at the outlet of the first and second baghouse were 0.034 and 0.011 ng TEQ/Nm³, respectively, which were both far below the national emission standard. The particulate matter concentration was 0.85 mg/m³, and the heavy metals leaching concentration of the fly ash from the second baghouse filter was lower than the hazardous waste identification standard. HCl concentration was almost zero at the outlet of the second baghouse filter when the optimal equivalence ratio of baking soda to HCl was 1.6. In addition, the estimated fly ash yield was 2.35% of the incinerated solid waste for the dual baghouse filter system, which was significantly lower than 3.5% as the traditional semidry scrubber + single baghouse filter.

Keywords: dioxins; HCl; dual bag; removal; heavy metals; pilot study



Incineration has been a major approach for reducing the amount of municipal solid waste in China [1]. A total of 136 million tons of solid waste was incinerated, and more than 500 incineration plants were run in 2021 in China [2]. One of the key issues in the development of municipal solid waste incinerators (MSWI) lies in the disposal technology of flue gas pollutants. Due to the complex composition of solid wastes, after incineration, the flue gas usually contains a large amount of pollutants, such as dioxins, heavy metals, HCl, etc., which are extremely toxic to the environment and human health [3,4]. The daily emission limits for dust, SO₂, HCl and dioxins are 20, 80, 50 mg/m³ and 0.1 ng TEQ/m³. Therefore, a series of air pollution control devices have been utilized in waste incineration plants, such as semi-dry scrubbers, activated carbon injection coupled with baghouse filters, wet scrubbers and catalytic systems to remove pollutants from flue gas to meet the national emission standards [5–7].

Hung et al. indicated that when the start-up temperature of the incinerator was below 850 °C, dioxin emissions increased [8]. In waste incinerator stack gas, dioxin emissions vary. Zhu et al. investigated 57 stack gas samples from six MSWIs and the PCDD/Fs emissions were in the range of 0.007 to 0.059 ng TEQ/Nm³, respectively [9]. However, Qiu et al. studied the PCDD/Fs emissions from MSWIs in northern China and found that the dioxins showed a large variation from 0.016 to 0.29 ng TEQ/m³ [10], some of which exceeds the national standard. Ni et al. investigated 19 municipal waste incinerator flue gas dioxin analysis results which showed that 16% of the data exceeded 1.0 ng TEQ/Nm³, 68% of the results exceeded 0.1 ng TEQ/Nm³, and the emission factor was 0.169–10.7 μ g TEQ/t [11]. In solid waste incineration air pollution control systems, bag filters with activated carbon



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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). injection are widely employed. The removal efficiency for PCDD/Fs can be higher than 95% [12–14]. In recent years, the traditional single bag filter system has struggled to meet the increasing stringent national standards of dioxin emissions from waste incineration. To fully reduce the emissions from the pollutants, a wet scrubber system was adopted in some new waste incinerator plants [15]. However, the memory effect of the wet scrubber can enhance the TEQ concentration owing to an increase in the total mass concentration of PCDD/Fs and the proportion of low-chlorinated congeners [16]. PCDD/Fs and heavy metals produced by waste incineration are not easy to degrade [17], and their contributions to environmental pollution and harmful effects to humans have become a hot topic of continuous concern [18,19]. Furthermore, to reduce the acid gas emissions, lime was injected excessively into the semidry scrubber [20,21], leading to the production of high amounts of fly ash. Shemwell et al. found that the reaction between lime and HCl would stop to a certain extent, which reduced the HCl removal efficiency and lime utilization rate [22]. Therefore, a new technology with high removal efficiency for PCDD/Fs and acid gas is required.

Kim et al. designed a dual baghouse system to improve the utilization efficiency of activated carbon and reduce the dioxins emissions [23]. Less activated carbon injection (40 mg/m^3) was realized in the dual bag system, which is much lower than the single baghouse system. Chi et al. also found that the dual bag filter system could reduce PCDD/Fs emission to 0.235 ng TEQ/m³ [24]. Therefore, the dual bag filter system is effective to control the dioxin emissions and improve the utilization of activated carbon [25]. However, the above studies only concentrated on the dioxins emission, disregarding acid gas removal and fly ash production.

In this study, a 1000 Nm³/h capacity pilot dual baghouse filter system was used to study the removal characteristics of PCDD/Fs, heavy metals and acidic gas. Activated carbon was injected at the inlet of the first baghouse filter to remove the gas phase dioxins and heavy metals, and baking soda was injected at the inlet of the second baghouse filter to remove HCl. In addition, the fly ash production rate was calculated and compared to that of the traditional single baghouse system.

2. Materials and Methods

2.1. Dual Baghouse System

The dual baghouse pilot system was built in a solid waste incineration plant, which has a total treatment capacity of 300 t/d.

The incinerator was equipped with a grate furnace, and the furnace temperature was kept above 850 °C to prevent dioxin formation. The air pollution control devices for this incinerator include a selective non-catalytic reduction system, a semidry scrubber, activated carbon injection coupled with a baghouse filter, and a selective catalytic reduction tower. As shown in Figure 1, the flue gas was elicited from the main flue and the flue gas flow was designed to be 1000 Nm³/h. The model of baghouse was a TMC56 pulse bag filter. The bag material was PTFE and the bag size was $\varphi 130 \times 1000$ mm. There were 56 bags in a baghouse and the total filtration area was 22 m². To completely remove the dioxins, heavy metals, acid gas and particulate matters, activated carbon and baking soda were fed at the inlets of the first and second bags, respectively.



Figure 1. The structure of the test bench of the light and large double bag system (the red stars means the sampling points).

The designed cross-section pressure of the pilot typhoon track was between -300 and -700 pa, and the actual operation data are usually between -450 and -600 pa.

2.2. Methods of Collecting Samples

The flue gas and fly ash samples were both collected from the pilot system. For flue gas samples, dioxins were sampled at the inlet of the first baghouse, the inlet of the second baghouse and the outlet of the second baghouse in the pilot system using a dioxin sampler (model KNJ23, KNJ, Asan-si, Korea). To study the effect of activated carbon injection, the flue gas at the three points were sampled with and without activated carbon injection. The feeding speed for activated carbon was 0.5 kg/h. The sampling process was described in detail in our previous research [26]. After each flue gas sampling process, the glass sampling equipment was rinsed with acetone and toluene, respectively, to avoid contamination. In addition, the HCl concentration at the outlet of the secondary baghouse was monitored and the feeding speed of baking soda was set as 0.2, 0.3, 0.4, 0.5 kg/h, respectively. For the fly ash from the first baghouse and the second baghouse was collected.

The activated carbon and baking soda used in this study were from Honeycomb Activated Carbon Company Ltd. In Ningbo, Zhejiang Province, China and Rongfeng Chemical Company Ltd. in Shouguang, Shandong Province, China. The moisture content in activated carbon was 9.24%, and the ash content was 6.48%. The proportion of particles with a particle size less than 0.074 mm was 93.6%. The specific surface area of the activated carbon was 1556 m²/g and the adsorption pore volume was 0.8606 mL/g. The particle diameter of the baking soda was 0.15–0.18 mm.

2.3. Characterization and Analysis

The dioxins in the flue gas and fly ash samples were pretreated according to the US EPA method 1613 (Tetra-through Octa-Chlorinated Dioxins and Furans by Isotope Dilution HRGC/HRMS). Through pretreatment, other organic impurities and heavy metals in the sample can be removed. Unlike flue gas samples, fly ash samples need to be soaked in 2 mol/L hydrogen chloride solution with a liquid–solid ratio of 40 mL/g for 4 h. After the flue gas samples were concentrated, the pollutants were removed by an acidic silica gel column and alumina column. After the pretreatment was completed, the concentrate was removed with nitrogen. ¹³C labeled standard compounds were added to the samples. Finally, the sample was analyzed by high resolution gas chromatography/high resolution mass spectrometry (HRGC/HRMS, JMS-800D, JEOL, Tokyo, Japan) with DB-5MS column (60 m × 0.25 mm × 0.25 mm). The pretreatment and the detection were described in detail in our previous research [27].

TEQ of dioxins is calculated according to the following formula:

$$\Gamma EQ = \Sigma[C_i] \times TEF_i \tag{1}$$

 C_i : the concentration of a certain class of dioxins, ng/m³; TEF_i: the corresponding toxic equivalent factor.

Sampling of particulate matter is based on the national environmental protection standard HJ 836-2017 of the People's Republic of China (determination of low concentration particulate matter in exhaust gas from fixed pollution sources by gravimetric method). The collection instrument is Laoying smoke and dust tester 3012H (Laoying Environmental Technology Co., Ltd., Qingdao, China). According to the method mentioned in the standard, the filter tubes and other materials used in the experiment were dried and weighed. The sampling process was conducted in strict accordance with the methods mentioned in the standard. Three samples were collected at each location and the average value was taken as the final result.

The leaching concentrations of heavy metals in fly ash were analyzed according to the national standard HJ/T 299-2007 (Solid waste-extraction procedure for leaching toxicity-sulphuric acid and nitric acid method). HCl concentration in the flue gas was monitored by FTIR (DX4000, Gasmet, Vantaa, Finland), and the average value of sampling time was taken as the final result.

3. Results and Discussion

3.1. Effects on Dioxin Emissions

The fly ash was sampled from the first baghouse without activated carbon injection, the first baghouse with activated carbon injection and the second baghouse with activated carbon injection. The toxic equivalent quantity (TEQ) of 17 2,3,7,8-substituted PCDD/Fs for the fly ash sampled from the three locations was analyzed, as shown in Figures 2 and 3.



Figure 2. Dioxin TEQ concentrations from the fly ash samples.

It can be seen from the figure that after activated carbon injection, the toxic equivalent of dioxins was significantly reduced, which was shown as the first baghouse fly ash > the first baghouse fly ash with activated carbon injection > the second baghouse fly ash with activated carbon injection > the second baghouse fly ash with activated carbon injection. The PCDD/Fs TEQs of the fly ash from the three sampling points were 48.1, 26.2 and 1.7 ng TEQ/kg, respectively. After activated carbon injection, the TEQ of dioxin in the first baghouse fly ash was only 54.47% of that without injection. The PCDD/Fs concentration in the second baghouse fly ash was significantly lower than that of the first baghouse fly ash, which was only 6.49%, and some PCDD/F isomers had dropped to an undetectable level. The data before and after analysis showed that the toxicity equivalent of dioxins in the ash sample after feeding was the lowest. It was 3.53%

lower than of that before feeding, and the total removal efficiency of dioxins exceeded 96%. This shows that activated carbon feeding and two-stage bag filters have an obvious effect on reducing dioxin content in fly ash.



Figure 3. A total of 17 PCDD/Fs TEQ in fly ash: first baghouse without activated carbon injection, first baghouse with activated carbon injection and second baghouse with activated carbon injection.

Based on the analysis of dioxins in fly ash, sampling and analysis of dioxins in gas samples were carried out at five points: inlet of the first baghouse, inlet and outlet of the second baghouse with and without activated carbon injection. The results are shown in Figure 4.

In the absence of activated carbon injection, the PCDD/Fs concentration at the inlet of the secondary baghouse was much lower than that at the inlet of the first baghouse, which proved the high efficiency of the baghouse filter. Moreover, the dioxin level at the outlet of the secondary baghouse was further reduced. Taking 2,3,4,7,8-PeCDF as an example, the PCDD/Fs TEQ at the inlet of the first baghouse was 0.1947 ng/Nm³, which was higher than the emission standard. The PCDD/Fs concentrations at the inlet and outlet of the second baghouse decreased to 0.042 and 0.004 ng/Nm³, respectively, only 21.57% and 2.26% of that at the inlet of the first baghouse. Through analyzing the 17 toxic PCDD/F isomers, it can be seen that the removal rate of dioxins for all isomers exceeds 90% except for 2,3,7,8-TCDD, for which the removal rate is only 72.41%. Furthermore, 1,2,3,4,7,8,9-HpCDF has the highest removal efficiency of 98%. This shows that without activated carbon injection, the dual baghouse system has obvious effect on reducing dioxin levels in flue gas.

With activated carbon injection, the dioxin level at the inlet of the secondary bag was further reduced compared to that without activated carbon injection, e.g., the concentration of 2,3,4,7,8-PeCDF decreased to 21.85% before feeding, and other PCDD/F isomers were also greatly reduced. The lowest PCDD/F TEQ value was found at the outlet of secondary baghouse, and the toxic equivalent of some dioxins, such as OCDD and OCDF were reduced to nearly undetectable levels. This is due to the activated carbon that was sprayed at the inlet of the first-stage bag, which adsorbs most of the dioxins in the flue gas and particulate matter after filtration. This results in lower dioxin concentrations in the flue gas compared to that without activated carbon at the inlet of the second bag. By comparing the PCDD/F concentrations at the outlet of the secondary baghouse after activated carbon feeding with that at the inlet of the first baghouse without feeding, it can be concluded that the removal efficiency of dioxin types exceeds 95% except for 2,3,7,8-TCDD. This proves



that activated carbon feeding can further improve the dioxin removal ability of the double bag system.

Figure 4. PCDD/Fs TEQ in the flue gas (a) inlet of the first baghouse, (b) inlet and (c) outlet of the second baghouse without activated carbon injection, (d) inlet and (e) outlet of the second baghouse with activated carbon injection.

The PCDD/Fs concentrations at the inlet of the first baghouse filter, the inlet and outlet of the second baghouse with activated carbon injection are presented in Figure 5.



Figure 5. PCDD/Fs concentrations the inlet of the first baghouse filter, the inlet and outlet of the second baghouse with activated carbon injection.

As shown in Figure 5, the PCDD/Fs concentration at the inlet of the first baghouse was 0.481 ng/Nm³, while the PCDD/Fs concentrations at the inlet and outlet of the second baghouse was 0.034 and 0.011 ng/Nm³, respectively, which were 7.11% and 2.2% of the initial concentration, and the final removal rate of the total dioxins was 98%. After the dual baghouse system treatment, the dioxins concentration in flue gas is significantly lower than the national standard, which meets the ultra-low emission requirements. This demonstrates that the dual bag system coupled with activated carbon injection has an obvious effect on the removal of dioxins in waste incineration flue gas.

3.2. Effects on Heavy Metal Emissions

Since heavy metals in flue gas are usually enriched in particles, the particle concentration at each position of the dual-bag system is detected to measure the filtering effect of the dual-bag on particles. Particulate matter samples were collected at the inlet of the first baghouse and the inlet and outlet of the second baghouse respectively for 2 h. The average value in the sampling time was taken, as shown in Table 1 below.

Table 1. Sampling results of particles in three locations.

Location	Particle Concentration/mg⋅m ⁻³	
Inlet of the first baghouse	32.7	
Inlet of the second baghouse	3.91	
Outlet of the second baghouse	0.85	

The concentration of particulate matter at the outlet of the first baghouse was significantly reduced to 3.91 mg/m^3 , only about 12% of the initial concentration. Furthermore, the second baghouse further intercepted the particulate matter in flue gas, reducing its concentration to 0.85 mg/m^3 . Efficient filtration of particulate matter by the dual bag system means that heavy metals in flue gas are also settled, which makes the content of heavy metals in flue gas far below or meeting the emission standard.

Because the heavy metal content in flue gas is difficult to measure directly, heavy metal analysis of fly ash filtered by bag filters was carried out to determine the effect of the dual bag system on heavy metal removal in flue gas. Due to the actual working environment, the dual bag system still needs to be fed, and the feeding will change the fly ash composition and affect the analysis of heavy metal content. Therefore, it is necessary to sample and analyze the fly ash before and after activated carbon injection to measure the influence of heavy metal removal.

Firstly, the fly ash from the first baghouse without activated carbon feeding was collected. Then, under the same working conditions, the bag system was fed. At the inlet of the first-level bag, activated carbon was sprayed at a feeding rate of 1 kg/h, and baking soda was sprayed at the inlet of the second-level bag at a feeding rate of 0.3 kg/h. The ash from the first-level bag and second-level bag were collected after feeding. ICPOES tests were performed on the three samples above, and the content of heavy metals and national identification standards were compared. The results are shown in Table 2.

Element	Primary Bag Ash/mg∙kg ⁻¹	Primary Bag Ash (Feeding)/mg∙kg ^{−1}	Secondary Bag Ash (Feed)/mg∙kg ⁻¹	Removal Efficiency/%	Identification Standard/mg∙kg ⁻¹
Cr	8.5	6.7	1.7	80.00	15
Ni	2.6	1.5	0.3	88.46	5
Cu	295.5	8.7	2.4	99.19	100
Zn	505.6	78.2	9.7	98.08	100
As	21.7	6.0	2.1	90.32	5
Se	2.0	0.9	0.5	75.00	1
Cd	23.2	0.5	0.2	99.14	1
Ba	0.5	2.4	0.2	60.00	100
Pb	108.3	5.6	0.1	99.91	5
Hg	4.0	0.06	0.1	97.50	0.1

Table 2. Test results of heavy metals in three ash samples.

The results showed that the content of heavy metals in the three samples were far below the national identification standard. After feeding, the heavy metal content of the first-grade bag dust decreased significantly compared with that before feeding. It was speculated that the activated carbon was filtered and settled with particulate matter, and the total amount of bag dust increased, which reduced the relative content of heavy metals. After feeding, the heavy metal content of secondary bagged ash decreased or remained stable compared with that of primary bagged ash. Cr, Zn, Pb and other elements decreased significantly, while Hg increased slightly. The main reason is that the primary bag has completed the basic filtration of flue gas, and the content of particulate matter in the flue gas reaching the secondary bag is low. At the same time, the unreacted baking soda at the inlet of the secondary bag increases the total amount of ash in the secondary bag, which further reduces the relative content of heavy metals. The increase of Hg content by 0.04 mg/m^3 was small and lower than the identification standard, which was speculated to be caused by the test error. According to the statistics of removal efficiency, the removal efficiencies of Cu, Zn, Cd, Pb and Hg exceeded 95%, and the removal efficiencies of Cr, Ni and As exceeded 80%.

According to the analysis above, it is concluded that the double bag system has obvious effect on reducing heavy metals in flue gas, which is significantly lower than the national emission standards and can achieve ultra-low emissions. At the same time, activated carbon and baking soda feeding can effectively reduce the relative content of heavy metals in bag fly ash, which is lower than the hazardous waste standard, and has a great effect on the subsequent burial or resource utilization of fly ash.

3.3. Effects on HCl Emissions

According to the reaction formula of HCl and baking soda:

$$HCl + NaHCO_3 = NaCl + CO_2 + H_2O$$
 (2)

The best equivalent ratio of HCl to baking soda is 1, that is, 1 mol HCl corresponds to 84 g baking soda. However, in the actual industrial process, baking soda cannot completely

react with HCl, and some baking soda will be filtered by bags. Therefore, the use of baking soda should be increased in the industrial process.

According to the monitoring data, the average flue gas flow rate of the experimental platform of the dual bag system was 1400 m³/h, the initial average HCl concentration was 35 ppm, and the average flue gas temperature was 175.5 °C. According to the gas volume formula and reaction formula, the spraying amount of baking soda was 0.183 kg/h under the ideal condition of complete reaction. Considering that the actual dosage of baking soda is higher than the theoretical dosage, four groups of gradient experiments with baking soda dosage of 0.2, 0.3, 0.4 and 0.5 kg/h were established. The baking soda was sprayed at the inlet of the secondary bag. The HCl content in the flue gas after the secondary bag was measured and the results are shown in Figure 6.



Figure 6. Effect of baking soda addition rate on HCl concentration in flue gas.

It can be seen from the graph that with the increase of baking soda addition rate, the HCl content in flue gas decreased significantly, and after baking soda, the addition rate reached 0.3 kg/h, HCl was no longer detected in the flue gas after secondary bagging. It can be preliminarily concluded that for the dual bag system test bench, the optimal dosage of baking soda is 0.3 kg/h under relatively stable working conditions, that is, the optimal equivalence ratio of baking soda to HCl is 1.6.

In the actual situation, the types and components of incineration waste and incineration conditions will be different, resulting in a large difference in HCl content in flue gas. Usually, the difference between different batches of garbage is relatively large, and the fluctuation of the same batch of garbage is relatively small. In the case of uniform batch waste incineration, if the incineration conditions remain unchanged, the HCl concentration maintains a small fluctuation, which can be regarded as constant concentration, and the amount of baking soda is calculated according to the average concentration.

3.4. Effects on Fly Ash Yield

Based on the data from the pilot plant and main flue, the fly ash yield under the conditions of quicklime and soda was preliminarily estimated, and the actual effect of small soda on reducing fly ash yield was clarified. According to the manufacturer and the sampling results, the relevant data of the main flue and the pilot station were calculated, as shown in Table 3. The above data may have certain fluctuations according to the actual working conditions. In the estimation process, the maximum value is adopted for calculation, while the differences caused by different conditions such as structure and flow velocity are ignored.

Position	Index	Numerical Value
Main flue collector	Total ash design Design fly ash yield Limestone feeding amount Fume gas volume	8000–9000 kg/day 2.5–3.5% 3500 kg/day 76,000 Nm ³ /h
Pilot test bench Baking soda feeding amount Fume gas volume		7.2 kg/day 1000 Nm ³ /h

Table 3. Data on fly ash yield calculation of light main flue and pilot station.

In the process of flue gas diversion between the main flue and the pilot plant, the content change of gaseous components can be ignored, so the proportion of acid gas in the flue gas of the main flue and the pilot plant is the same by default. When the feed rate of baking soda reached 0.3 kg/h, the acidic gas in the tail gas of the pilot plant was basically completely removed. Since the flue gas volume of the main flue was 76 times that of the pilot plant, the daily consumption of baking soda in the main flue was set to be 547.2 kg when the double bag system was adopted. The daily ash yield of the original main flue was 9000 kg, the fly ash yield was 3.5%, and the raw lime feeding amount was 3500 kg. When the raw lime was replaced by baking soda, the reaction loss and excessive baking soda were ignored. The daily ash yield of the main flue was reduced to 6047.2 kg, and the fly ash yield was 2.35%. It can be seen that after the replacement of lime with baking soda, the fly ash yield decreased by 1.15% compared with the set value of 3.5%, and was less than the minimum value of 2.5%.

According to the above numerical analysis, it can be concluded that the use of the dual bag system and soda instead of lime can reduce the fly ash production rate and save the cost of fly ash disposal.

4. Conclusions

The preliminary experiment on the removal effect of dioxin, heavy metals and HCl by the dual-bag system was completed in a pilot scale, and the change of fly ash yield was estimated. The results showed that compared with the traditional single bag system, the dual bag system coupled with activated carbon and baking soda injection could effectively improve the removal efficiency of the pollutants and reduce the fly ash yield of the system. For dioxins and HCl in flue gas, the highest removal efficiencies were 98% and 100%, respectively. Dioxins and heavy metals in fly ash also declined significantly, far lower than the national emission standards. The estimated fly ash yield of 2.35% is significantly lower than the set value of 3.5%, and is 2.5% lower than the minimum design value of the main flue. It can be concluded that the double bag system has a remarkable effect on the removal of pollutants from waste incineration flue gas and reduction of fly ash yield, which can meet the national emission standards and industry needs. The removal effect of the system on pollutants is better than that of the wet method and catalytic tower mentioned in the introduction.

Based on the above experiments and preliminary results, future research directions may focus on the following aspects: determining the influence of activated carbon and baking soda with different properties on pollutant removal; the continuous removal effect of the system on pollutants under long-term working conditions; and whether changes in waste types and working conditions have an impact on pollutant removal. Through further experiments, the pollutant removal capacity of the dual-bag system may be measured more accurately.

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Abbreviations

MSWI	Municipal solid waste incineration
TEQ	Toxic equivalence quantity
PCDD/Fs	Polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans
I-TEQ	International toxic equivalence quantity
EPA	Environmental protection agency
HRGC	High resolution gas chromatograph
HRMS	High resloution mass spectrum
PeCDF	Pentachlorinated dibenzofurans
TCDD	Tetrachlorinated dibenzo-p-dioxins
HpCDF	Heptachlorinated dibenzofurans
OCDD	Octachlorinated dibenzo-p-dioxins
ICPOES	Inductively coupled plasma-optical emission spectrometer
BJH	Barret-Joyner-Halenda

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