Using a State-of-the-art Air Pollution Control System to Lower Pollutant Emissions

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ABSTRACT

This study focused on the pollutant characteristics of a renovated laboratory waste treatment plant that adopted a state-of-the-art design for its air pollution control system. The output materials, including slag, primary quenching tower ash, scrubbing wastewater, sludge, fabric filter ash, and flue gas, were sampled and analyzed. Incorporating an organic liquid waste injector into the primary combustion chamber enabled the local temperature to be increased to approximately 1300–1400°C, thereby reducing the PCDD/F concentration in the slag to 0.019 ng I-TEQ g⁻¹ and the PCDD/F emission factor to 699 µg I-TEQ ton-waste⁻¹. In addition, the concentration of the flue gas was much lower than the regulated standard, and the low level of emissions contributed to the performance of the air pollution control devices. The fly ash contained crystalline-phase NaCl or other sodium salts that resulted from the neutralization of NaOH by acidic pollutants. However, high concentrations of Cl and PCDD/Fs were measured in the scrubbing wastewater and sludge, respectively. The scrubbing unit of the renovated air pollution control system was placed in front of the fabric filter to remove HCl, thus providing excellent protection for the filter.

Keywords: Air pollution control devices; Fly ash; Incinerator; Laboratory waste.

INTRODUCTION

To treat the laboratory waste (LW) generated during research and education in schools, in 2005, the Ministry of Education subsidized the Sustainable Environment Research Center (SERC) at National Cheng Kung University to build a treatment plant located in Tainan City to dispose of LW (Wu et al., 2014a). This LW treatment plant has three major systems: a physicochemical treatment system, an incineration system, and a plasma melting system. Among these systems, the incineration system was assigned to treat combustible LW and plastic containers. The plastic containers were composed of polyethylene, polypropylene, or plastic materials. The combustible LW included medical or biochemical experiments, which usually had high levels of Cl (Coutinho et al., 2006). Previous studies have shown that a large amount of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) or polycyclic aromatic hydrocarbons (PAHs) are generated when waste with high levels of Cl is incinerated (Wang and Chang-Chien, 2007; Wang et al., 2010; Liao et al., 2014). It has been reported that the PCDD/Fs and PAHs are harmful to public health, and thus the flue gas and ashes needed further treatment to avoid secondary pollution (Hung et al., 2018; Liu et al., 2019a). However, even with appropriate treatment using air pollution control devices (APCDs), the PCDD/F content in fly ashes is still extremely high and requires final disposal to avoid threatening public health (Hsieh et al., 2018). Plasma melting systems are set up to dispose of hazardous residues, including sludge and toxic ashes or other wastes from incineration and physicochemical
In addition to the formation of high level of PCDD/Fs, the Cl in input materials caused another problem: It is transformed into HCl during the incineration process. This acid gas seriously corrodes the components of the system. After a decade of operation, the incineration system will be worn out and require renovation. Therefore, the APCD process was newly designed following a state-of-the-art concept to eliminate the corrosion due to the presence of HCl.

This study focused on the emission characteristics and the distribution of pollutants over the entire incineration process. The PCDD/F and HCl concentrations in output materials were analyzed to determine their mass distributions among the output materials. The crystalline characteristics and surface structure of solid specimens were investigated. In addition, the operating characteristics of the old and renovated incineration systems were compared and discussed.

METHODS

Processes in the Old and Renovated Incineration Systems

The process flow diagram of the old incineration system is shown in Fig. 1. The LW was fed with 375 kg h⁻¹ through a feeding unit into the primary combustion chamber, and the incineration temperature was maintained above 900°C. The bottom ash was quenched using water and collected in the ash pit. The flue gas was heated to 1150°C by a diesel combustor in the secondary combustion chamber to destroy any unburned residues, CO, and persistent organics, and then the flue gas passed through a series of APCDs. The primary and secondary quenching towers quenched the flue gas rapidly to 400°C and 160°C, respectively, in order to reduce the formation of PCDD/Fs. The activated carbon was injected into the flue gas to adsorb the PCDD/Fs and was trapped with residual particulate by the fabric filter. After passing through the fabric filter, a wet scrubber was used to remove residual HCl to protect the induced-draft fan. Without this unit, the induced-draft fan would be broken within two weeks. The flue gas was heated to 110°C before being emitted into the ambient air to avoid the formation of white mist. The pollutant emission characteristics of this incineration system have been reported in previous studies (Chang et al., 2012; Wu et al., 2014b).

Fig. 2 shows the process flow diagram of the renovated system. An organic liquid waste injector was integrated with the primary combustion chamber to generate a local high temperature (ranging from approximately 1300–1400°C) to melt the burned materials and bottom ash. The melted material was then quenched to form inert slag and was collected in the pit. The extra heat also elevated the temperature of the flue gas and reduced the concentration of particulates and PCDD/Fs (Tsai et al., 2018). Similar to the old incineration system, the flue gas was heated to 1150°C and then quenched to 400°C. After the primary quenching tower, the flue gas was directly quenched using a Venturi scrubber and wet scrubber to 60°C to remove the particulates.
and HCl. The wastewater was treated using a wastewater treatment system, and the pollutants were removed from the wastewater as sludge. The treated wastewater was stored in a tank and recirculated into the two scrubbers to save water. The flue gas then passed through a dewatering unit to remove moisture to avoid clogging the fabric filter. The activated carbon was injected into the flue gas and filtrated using the fabric filter, and the flue gas was emitted into the ambient air. The primary quenching tower ash, sludge, and fabric filter ash were further treated using the plasma melting system to form inner slag.

**Determination of Pollutants from Stack**

The incineration tests were conducted in triplicate. The feeding materials were mainly LW, and the Cl level was estimated to be as high as 10%. The slag, ashes, scrubbing wastewater, sludge, and flue gas were all sampled and analyzed to evaluate the performance of the renovated incineration system. The pollutant concentrations from the stack, including SO₂, NOₓ, O₂, CO, HCl, particulate, PCDD/Fs, and heavy metals, were determined in this study. The sampling and analysis procedure all followed the standard method established by the Taiwan Environmental Protection Administration (TEPA). A gas analyzer (PG-250A; Horiba) was used to measure the NOₓ concentration via a chemiluminescence detection method (TEPA, 2006a), the concentrations of SO₂, CO₂, and CO were obtained via non-dispersive infrared absorption (TEPA, 2006b, c, 2016), and the O₂ concentration was obtained via the galvanic cell method (TEPA, 2006d). The HCl concentration of the flue gas was determined using a mercury(II) thiocyanate colorimetric method (TEPA, 2008). The detailed procedure for measuring these gases is given in the corresponding methods (TEPA, 2006a, b, c, d, 2008, 2016).

For the analysis of the particulate and heavy metals (Cd, Hg, and Pb), the flue gas sampler, equipped with a filter holder, a cooling device, a pump, and a flow meter, was used to isokinetically sample at an average flow rate of 3 L min⁻¹. The sampling procedure followed the TEPA standard method (TEPA, 2005, 2006e) and were given in our previous work (Kuo et al., 2004). For the PCDD/Fs, the sampling train for the flue gas met the TEPA A807.75C requirements (TEPA, 2010). Before sampling, the probe was washed with acetone, dichloromethane, and toluene, in that order.

A cartridge was filled with 20–40 g adsorbent (Amberlite XD-2®) and was spiked with five PCDD/F surrogate standards pre-labeled with isotopes, ³⁷Cl-2,3,7,8-TCDD, ¹³C₁₂-2,3,4,7,8-HxCDD, ¹³C₁₂-2,3,4,7,8,PeCDD, ¹³C₁₂-2,3,4,7,8-HxCDF, and ¹³C₁₂-2,3,4,7,8,9-HpCDF. The flue gas was sampled isokinetically, and the samples were preserved and transported to the laboratory at 10°C for further analysis.

The samples were initially spiked with the ¹³C₁₂-labeled internal standard. A series of steps, including a Soxhlet extraction for 24 h, measurement of the nitrogen blowing concentration, the cleanup and fractionation procedure, and measurement of the nitrogen blowing re-concentration, were conducted to pretreat the samples. The final concentrated eluate (about 1 mL) was analyzed using a high-resolution
gas chromatograph (HRGC; 6970 Series gas; Hewlett-Packard, CA, USA) and a high-resolution mass spectrometer (HRMS; Micromass AutoSpec Ultima; Waters, Manchester, UK) to determine the concentration of the PCDD/Fs. The PCDD/F analysis followed the standard methods announced in TEPA NIEA A808.75B. The methods and details of the procedure followed a previous study (TEPA, 2013; Chen et al., 2018).

**Sampling and Analysis of Wastewater and Solid Specimens**

Output materials, including the slag, primary quenching tower ash, scrubbing wastewater, sludge, and fabric filter ash, were collected from the bottom ash pit and air pollution control devices of the incineration systems. The solid specimens were dried, pulverized, and ground to a particle size that could pass through a mesh with a pore size of 149 μm to ensure homogeneity. The ground powder was pretreated and analyzed following the above-mentioned method for the PCDD/F analysis. For the analysis of metals, the digestion procedure, which was modified from the standard method provided in NIEA R317.11C (TEPA, 2004), was conducted to pretreat the solid specimens. Ground samples (~0.1 g) were placed in Teflon tubes to which 2 mL HCl (32 wt%) and 4 mL HNO3 (67 wt%) were added. The Teflon tubes were sealed and heated using a microwave digestor (MARSXpress; CEM) with the following program: heated tubes were sealed and heated using a microwave digester oxygen-rich conditions, and the LW could be effectively decomposed (Aurell and Marklund, 2009; Kuo et al., 2019). The NOx concentration was 76.5 ppm, which was slightly higher than that in the old system and was nearly half the regulated concentration in the emission standard. This was probably due to the thermal NOx caused by the local high temperature (~1300°C) in the primary combustion chamber. In addition, the main objective of the renovated APCD system was not to remove NOx. If confronted with a stringent NOx regulation, a unit of activated carbon adsorption or selective catalytic reduction could be integrated into the proposed system to reduce the NOx concentration (Fang et al., 2019; Liu et al., 2019b).

The Cl level of the input materials was estimated to be 10% or even higher, where, according to previous reports, the incineration process generates a huge amount of HCl and PCDD/Fs (Hatanaka et al., 2005). However, the concentrations of SO2, HCl, and PCDD/Fs were only 6.5 ppm, 0.775 ppm, and 0.109 µg I-TEQ Nm–3, respectively.

**RESULTS AND DISCUSSION**

**Pollution Characteristics of the Output Material**

Table 1 shows the characteristics of the pollutant emissions from the flue gas. The O2, CO2, and CO concentrations in the flue gas were 15.7%, 4.2%, and 14 ppm, respectively. Based on the CO2 and CO concentrations, the combustion efficiency (= [CO2]/([CO2] + [CO]) × 100%) was 99.97%. The results show that the combustion behavior in the primary and secondary combustion chamber occurred under oxygen-rich conditions, and the LW could be effectively decomposed (Aurell and Marklund, 2009; Kuo et al., 2019). The NOx concentration was 76.5 ppm, which was slightly higher than that in the old system and was nearly half the regulated concentration in the emission standard. This was probably due to the thermal NOx caused by the local high temperature (~1300°C) in the primary combustion chamber. In addition, the main objective of the renovated APCD system was not to remove NOx. If confronted with a stringent NOx regulation, a unit of activated carbon adsorption or selective catalytic reduction could be integrated into the proposed system to reduce the NOx concentration (Fang et al., 2019; Liu et al., 2019b).

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![Table 1: Emission characteristics of the flue gas.](image)
and 0.109 µg I-TEQ Nm⁻³, respectively, which were all much lower than the regulated standard (TEPA, 2003). In addition, the concentrations of particulate and heavy metals were near the detection limits. The results showed that the APCDs effectively reduced the pollutant emissions.

Table 2 shows the metal content, PCDD/Fs, and Cl in the output materials. In the primary combustion chamber, the LW was incinerated and melted into slag. The levels of PCDD/Fs and Cl were 0.019 ng I-TEQ g⁻¹ and 1080 mg kg⁻¹, respectively, which were much lower than those of the old incineration system (1.87 ng I-TEQ g⁻¹ and 105,000 mg kg⁻¹; Wu et al., 2014b). This could be explained by the fact that the local high temperature destroyed the PCDD/Fs and caused more Cl to vaporize into the flue gas. The slag also had Fe (6390 mg kg⁻¹), Cu (2610 mg kg⁻¹), and Mn (2570 mg kg⁻¹), and other metals were in trace amounts.

The primary quenching tower ash was mainly composed of Na (202,000 mg kg⁻¹) and Cl (312,000 mg kg⁻¹), which came from the reaction (as shown in Eq. (1)) of the NaOH solution injected with the HCl in the flue gas. Except for K (32,600 mg kg⁻¹), there were very small amounts of other metals in the primary quenching tower ash.

\[
\text{NaOH} + \text{HCl} \rightarrow \text{NaCl} + \text{H}_2\text{O} \quad (1)
\]

The PCDD/F level in the primary quenching tower ash was 0.242 ng I-TEQ g⁻¹, which was much lower than that in the municipal solid waste incinerator (Lin et al., 2008). The solution of 40% NaOH was used as the scrubbing water, and, thus, the wastewater had high levels of Na⁺ (174,000 mg L⁻¹). In addition, due to the vaporization of water and neutralization of acid gas, it was necessary to refill the NaOH solution to maintain the alkalinity of the recirculated wastewater and the water level. A previous study reported that scrubbers could effectively remove PCDD/Fs (Lee et al., 2004). Therefore, similar to HCl, the PCDD/Fs also accumulated in the recirculation units, and thus, the PCDD/F concentration in the wastewater was as high as 10,900 pg L⁻¹. On the other hand, the suspended solids in the scrubbing wastewater were removed by the wastewater treatment system and collected as sludge.

Due to low vapor pressure and solubility, the PCDD/Fs in the flue gas mainly existed in the particulate phase. The particulate phase was scrubbed out from the flue gas, and the PCDD/Fs were highly concentrated in the suspended solids. Therefore, the PCDD/F levels were as high as 83.5 µg I-TEQ g⁻¹. In addition, the sludge had Mn (29,500 mg kg⁻¹), Fe (21,300 mg kg⁻¹), Na (9700 mg kg⁻¹), and Cl (7460 mg kg⁻¹). After passing through the scrubbing unit, the activated carbon and residual particulates were trapped as fabric filter ash. The fabric filter ash had Cl (38,000 mg kg⁻¹), Na (24,100 mg kg⁻¹), Fe (5340 mg kg⁻¹), and K (3240 mg kg⁻¹), and the PCDD/F level was 1.14 µg I-TEQ g⁻¹.

Fig. 3 shows the PCDD/F patterns of the output materials. The major three PCDD/F congeners in the ingot were 2,3,4,7,8-PeCDF (47.9%), OCDF (25.6%), and 2,3,7,8-TeCDF (16.5%). The PCDD/F flue gas profile was similar to those of the other output materials. The PCDD/F mass of these output materials was mainly distributed in 2,3,7,8-TeCDF (~20%). Next to 2,3,7,8-TeCDF, the other congeners, including 1,2,3,7,8-TeCDF (9.94%), 1,2,3,4,7,8-HxCDD (12.2%), 1,2,3,6,7,8-HxCDD (10.2%), 1,2,3,7,8,9-HxCDD (9.5%), and OCDF (9.3%), all accounted for about 10%.

**Mass Distribution of PCDD/Fs and HCl among Output Materials**

Table 3 shows the mass distribution of PCDD/Fs and HCl among the output materials in the two incineration systems. Due to the local high temperature in the primary combustion chamber, the mass fractions of PCDD/Fs and Cl in the slag were only 0.04% and 0.01%, respectively, which were much lower than those in the bottom ash in the old incineration system (31.6% and 24.9%). For the renovated incineration system, the PCDD/F mass (97.02%) mainly existed in the scrubbing units and the loading in the fabric filter for PCDD/F removal was much lighter than that in the other systems (Wang and Chang-Chien, 2007; Wang et al., 2010). The mass fraction of PCDD/Fs emitted from the flue gas was only 0.23%, indicating good performance of this APCD in terms of the removal of PCDD/Fs.

For the renovated incineration system, the Cl mass fraction removed by the primary quenching tower was only 10.3%,
Fig. 3. PCDD/F patterns of output materials: (a) slag, (b) primary quenching tower ash, (c) scrubbing wastewater, (d) sludge, (e) fabric filter ash, (f) flue gas.

Table 3. Mass percentage of PCDD/Fs and Cl in the output materials in the renovated and old incineration systems.

| Item            | Slag | Primary quenching tower ash | Sludge | Scrubbing wastewater | Fabric filter ash | Flue gas |
|-----------------|------|-----------------------------|--------|----------------------|------------------|----------|
| Renovated system | PCDD/Fs | 0.04 | 1.66 | 92.97 | 4.05 | 1.06 | 0.23 |
|                  | Cl    | 0.01 | 10.30 | 0.05 | 89.38 | 0.17 | 0.08 |
| Old system       | PCDD/Fs | 31.6 | 28.7 | 1.75 | 32.6 | 1.8 | 3.52 |
|                  | Cl    | 24.9 | 20.1 | 4.99 | 19.9 | 31.0 | 0.17 |

and, thus, the Cl mass existing in the flue gas was still as much as 89.7%. The Venturi and wet scrubbers removed 89.4% of the Cl mass, and the Cl mass fraction reaching the fabric filter was greatly reduced to 0.25%. This prevented the fabric filter from being corroded by HCl. When it came to the old incineration system, the mass fraction of HCl removed by two quenching towers was only 25.09%, and thus, the fabric filter still had to process an HCl mass of 51.07%. Therefore, the welding bead and the plate edge of the components in the fabric filter were seriously corroded.
and had to be repaired at least once a year. After the fabric filter, the wet scrubber removed 31.0% of the Cl mass, which explained why the induced-draft fan broke in two weeks without the wet scrubber.

**Surface Microstructure and Crystalline Phase of the Solid Specimens**

Figs. 4 and 5 show the SEM images and XRD patterns of the solid specimens. Due to being melted, the structure of the slag was bulky and rigid and had flat, sharp-edged surfaces, which was completely different from the other ashes and sludge. The major crystalline phase in the slag was Ca₅(SiO₄)₂(SO₄) (ternesite) and Ca₁₀(SiO₄)₃(SO₄)₃(OH, F, Cl)₂ (hydroxylellestadite), which were mainly composed of igneous rock and neutralized salts. These crystalline phases are often found in high-temperature environments, which was consistent with the conditions in the primary combustion chamber (melting and then quenching).

The crystalline phases in the primary quenching tower ash mainly comprised NaCl with some Na₄(SO₄)₂·NaCl·NaF (sulfohalite), which were all salts due to the neutralization of NaOH with acid pollutants. The primary quenching tower ash had a powdery, porous structure, which mainly came from the aggregation of NaCl and the particulates in the flue gas. The XRD analysis and SEM images of the primary quenching tower ash matched the metal composition (Na = 202,000 mg kg⁻¹ and Cl = 312,000 mg kg⁻¹).

Similar to the primary quenching tower ash, the structure of the sludge was also powdery and porous, and the crystalline phases were also NaCl, with some Na₄(SO₄)₂·NaCl·NaF. However, the sludge had some flat, bulky structures that might have been due to using the frame filter for dewatering compression. Except for the crystalline phases of NaCl and Na₄(SO₄)₂·NaCl·NaF, the fabric filter ash still had another crystalline phase, Na₂₉(SO₄)₁₀·ZnCl₂·NaCl (D’Ansite [Zn]), which also came from the neutralization of NaOH with acid pollutants. The fabric filter ash was powdery and porous, and its appearance matched the profile of crystalline phases.

**Comparison of the Old and Renovated Incineration Systems**

Table 4 shows a comparison of the renovated incineration system and the old one. Due to the injection of waste solvent to generate the local high-temperature environment, the PCDD/F emission factor in this system was slightly reduced from 888 to 699 µg I-TEQ ton-waste⁻¹. In addition, the PCDD/F concentration in the flue gas was greatly reduced from 0.491 to 0.109 ng I-TEQ Nm⁻³. This contributed to the good performance of the renovated APCDs. In the old system, the PCDD/F masses (all were approximately 30%) were roughly equally distributed in the bottom ash, primary quenching tower ash, and fabric filter ash. In the renovated incineration system, the PCDD/F mass fraction in the primary quenching tower ash was only 1.66%, but that in the scrubbing units (sludge and scrubbing wastewater) was as high as 97.02%. The PCDD/Fs mainly existed as the particulate phase, so the sludge accounted for 92.97% of the PCDD/F mass.

The emitted concentration of HCl in both incineration systems was lower than 1 ppm, and both systems relied on scrubbers to wash HCl out of the flue gas. However, in the old incineration system, the wet scrubber was set up after the fabric filter and could not prevent the fabric filter from being corroded. By contrast, the new design positioned the scrubbing units in front of the fabric filter, thus providing excellent protection for the fabric filter.

**CONCLUSIONS**

This study investigated the performance of a state-of-the-art air pollution control system. The integration of waste solvent into the primary combustion chamber improved the combustion efficiency to 99.97% and also reduced the formation of PCDD/Fs. The APCDs effectively reduced the pollutive flue
Fig. 5. XRD patterns of the output materials: (a) slag, (b) primary quenching tower ash, (c) sludge, (d) fabric filter ash.

Table 4. Comparison of the performance of the old and renovated incineration systems.

| Item                                                | Old system        | Renovated system   |
|-----------------------------------------------------|-------------------|--------------------|
| PCDD/F emission factor (µg ton-waste⁻¹)             | 888               | 699                |
| PCDD/F concentration of flue gas (ng I-TEQ Nm⁻³)   | 0.491             | 0.109              |
| Output materials where PCDD/Fs were mostly concentrated | Secondary quenching tower ash | Recirculation water tank sludge |
| HCl removal efficiency of APCDs                     | Excellent         | Excellent          |
| Main unit to remove HCl                             | Wet scrubber      | Venturi/scrubbing tower |
| Protection of fabric filter                         | Poor              | Excellent          |
gas emissions, resulting in concentrations significantly below the regulated standard. The primary component of the fly ash was NaCl or other sodium salts, which were formed when acidic pollutants neutralized the NaOH. The HCl and PCDD/Fs were distributed mainly in the scrubbing wastewater and the sludge, respectively. The scrubbing unit provided excellent protection for the fabric filter by removing most of the HCl before the sample passed through the filter.

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