



Polychlorinated dibenzo-*p*-dioxins/dibenzofuran mass distribution in both start-up and normal condition in the whole municipal solid waste incinerator

Che-Kuan Chen^a, Chieh Lin^{a,*}, Yuan-Chung Lin^{b,c}, Lin-Chi Wang^{b,c}, Guo-Ping Chang-Chien^{b,c}

^a Department of Environmental Engineering and Science, National Pingtung University of Science and Technology, Nei Pu, Ping Tung 91207, Taiwan

^b Department of Chemical and Materials Engineering, Cheng Shiu University, Kaohsiung 833, Taiwan

^c Super Micro Mass Research & Technology Center, Cheng Shiu University, Kaohsiung County 833, Taiwan

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ABSTRACT

Although many researches focused on the polychlorinated dibenzo-*p*-dioxins/dibenzofuran (PCDD/F) emissions from stack, in the bottom ash and in the surrounding environment, researches focused on PCDD/F mass distributions in the whole incineration plant have seldom been addressed. This study determined PCDD/F emissions in the whole plant. A high-resolution gas chromatograph/high-resolution mass spectrometer was utilized for analyzing 17 PCDD/F species. Experimental results displayed that PCDD/Fs were formed during fly ash from super heater (SH), economizer (EC), semi-dryer absorber (SDA) and fabric filter (FF) was transferred to fly ash pit. Mass distribution ratios of PCDD/Fs in g I-TEQ (Toxicity Equivalency Quantity) per week from stack, SH, EC, SDA, FF, generation and bottom residue (BR) in start-up operations were 14.6%, 0.1%, 8.3%, 1.0%, 41.7%, 33.4% and 0.9%, respectively. Above results indicated that main PCDD/F source in the MSWI was from fly ash. However, the fly ash is easily controlled and PCDD/F emitted from stack flue gases will be difficult to be handled. Therefore, we should pay more attention on PCDD/F emission from flue gases especially from start-up procedure. Besides, fly ash should be controlled by sodium hypophosphite before being landfilled. MSWI did require further detoxification treatments for the solid residues and flue gases.

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1. Introduction

Waste-to-energy incineration was considered as a mainstream strategy for municipal solid waste management due to the lack of landfill spaces and the associated risks to water and soil. Besides, a landfill also gives risk to air because gas pollutants are formed by the reaction among wastes and then emit from the landfill. Approximately 85% of the waste volume and 60–75% of the mass of municipal solid waste incineration (MSWI) could be reduced [1,2]. In addition, MSWI could be located in the proximity of residential areas and generate electricity and/or heat from the energy content of municipal solid waste. Nevertheless, there are still negative impacts on the environment from MSWIs. Since polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) was first found in flue gases and in the fly ash of municipal solid waste incinerators (MSWIs) [3], extensive examinations on PCDD/Fs have been conducted due to their acute toxicity and the negative impacts on human health. Recent discussions of the possible mechanisms of PCDD/F formation have focused on two hypotheses: (1) Stieglitz et al. have suggested a de novo synthesis of PCDD/F from active

carbon particulates by gas–solid and solid–solid reactions with air, moisture, and inorganic chlorides, catalyzed by copper ions [4–6]. (2) Karasek and Dickson [7,8] have proposed that PCDD/F are formed from chloroaromatic precursors such as polychlorophenols and polychlorobenzenes by reactions which have been shown to occur by heterogeneous catalysis on the surface of fly ash particles at 250–400 °C.

There are many researches focused on the PCDD/F emissions from stack, in the bottom ash and in the surrounding environment [9–12]. However, there were only few researches focused on PCDD/F mass distributions in the whole plant have been reported. In 1992, Johnke concluded that the total PCDD/F emission factor was 211.6 µg I-TEQ ton-waste⁻¹ and the mass distribution of PCDD/Fs in the bottom ash, slag water, boiler ash, ESP (electrostatic precipitators) ash, scrubber water, filter cake and stack gas from the MSWI were 4.3%, 1.5%, 0.7%, 56.7%, 2.4%, 22.7% and 11.8%, respectively. Moreover, net 121.6 µg I-TEQ per waste was produced due to the incineration process [13]. Similar result was found. Giugliano et al. concluded that the total PCDD/F emission factor was 10.44 µg I-TEQ ton-waste⁻¹ and the mass distribution of PCDD/Fs in the slag, boiler ash, fabric filter ash, sludge and stack gas were 72.6%, 5.6%, 18.6%, 1.5% and 1.7%, respectively [14]. Recently, several studies have focused on the high PCDD/F emission during the start-up of incinerators [15–24]. In Taiwan, the elevated PCDD/F emissions

* Corresponding author. Tel.: +886 8 7703202x7087; fax: +886 8 7740393.

E-mail address: linchieh@mail.npust.edu.tw (C. Lin).

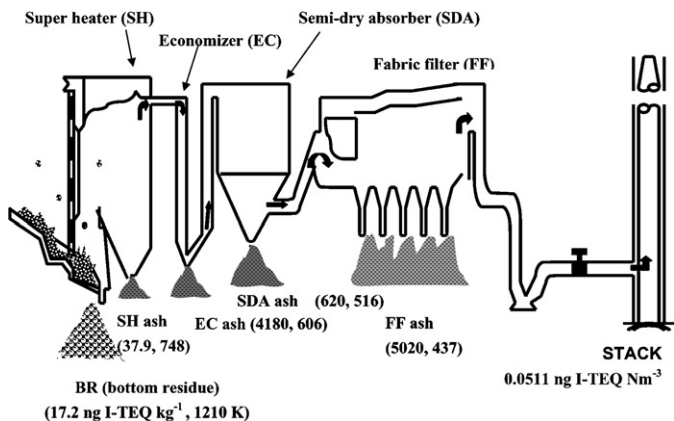


Fig. 1. The flow diagram of the KS MSWI.

of the MSWI during start-up could reach 96.9 ng I-TEQ Nm⁻³ and still maintained a high PCDD/F emission (40 times higher than the Taiwan emission limit) even 18 h after the injection of activated carbon, indicating the memory effect. Moreover, the PCDD/F emission quantity from stacks resulted from the start-up was 2.35 times larger than that of a whole year's normal operations [15]. Therefore, mass distribution of PCDD/Fs extended to the MSWI under the start-up operation condition should be investigated.

In this study, the PCDD/F contents in the output residues and PCDD/F concentration in the stack gas from the MSWI were measured. Secondly, the PCDD/F congener profiles were compared and discussed. Finally, emission factor and mass distribution ratios of PCDD/Fs in the MSWI under the start-up operation condition were assessed.

2. Experimental methods

2.1. Basic information about the selected MSWI

The stack samples and ash samples were collected from KS MSWI, located in southern Taiwan. There are four incinerators, each of which includes own heat recovery systems (350 °C), selective non-catalytic reduction, dry scrubber (250–230 °C), activated carbon injection, fabric filter (180–160 °C) and stack. The treatment processes are the most common ones in Taiwan, which are recognized as the most effective technique for PCDD/F emission control. Operation of the KS MSWI began in 2000 and its total capacity is 1800 ton/day with lower heating value of 2500 kcal/kg-waste. The flow diagram of the KS MSWI was shown in Fig. 1.

2.2. Sample collection

The PCDD/F samples were collected isokinetically from the stack flue gas of the selected incinerators 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. Prior to sampling, XAD-2 resin was spiked with PCDD/F surrogate standards pre-labeled with isotopes, including ³⁷C₁₄-2,3,7,8-TCDD (tetrachlorodibenzo-*p*-dioxin), ¹³C₁₂-1,2,3,4,7,8-HxCDD (hexachlorinated dibenzo-*p*-dioxin), ¹³C₁₂-2,3,4,7,8-PeCDF (pentachlorinated dibenzofuran), ¹³C₁₂-1,2,3,4,7,8-HxCDF (hexachlorinated dibenzofuran) and ¹³C₁₂-1,2,3,4,7,8,9-HpCDF (heptachlorinated dibenzofuran). The recoveries of PCDD/F surrogate standards were 95–117%, and met the criteria within 70–130%. To ensure the free contamination of the collected samples, one trip blank and one field blank were also taken during the field sampling was conducted. Details are similar to that given in our previous work [19].

There are four incinerators at the KT MSWI. Therefore, the sampled incinerator was selected by random sampling for stack samples and ash samples. To obtain good representative ash samples, sampling was done according to regulation specified in ordinance NIEA R118.02B and also referred to Minnesota regulation 7035.2910 in US. In the sampling period, ash was collected directly from super heater (SH), economizer (EC), semi-dry absorber (SDA), fabric filter (FF) and bottom residue (BR). Above ash was weighed in order to calculate ash amounts. Then ash from SH, EC, SDA and FF was put back to transfer process and sent to fly ash pit. Ash from SH, EC, SDA and FF was mixed well in fly ash pit and the mixed ash was named FAP (fly ash pit) ash. Every 12 h for 3 days, about 200 g of samples were collected from the SH, EC, SDA, FF, FAP and BR for PCDD/Fs analysis. As a result, approximately 1.2 kg of samples was collected from the incinerator. The collected samples should be stored in properly sealed containers to eliminate the effects from air circulation and water. Appropriate labels should be used for the delivery of samples. A sampling report should also be supplied for the samples. Except for samples that were in solid state, all samples must be stored in the refrigerator with temperature maintained at 4 ± 1. The ferrous and non-ferrous metals, glass and stone were removed and after well-mixing and diagonal sectioning, 600 g of different ash was retained. The samples were then spread out on a clean aluminum foil and naturally dried. The samples were then mixed thoroughly for the determination of PCDD/F analysis.

2.3. Analyses of PCDD/Fs

Analyses of stack flue gas followed the US EPA modified Method 23. Analyses of PCDD/Fs in the ash samples followed the US EPA modified method 1613A. All chemical analyses were carried out by the Super Micro Mass Research and Technology Center at Cheng Shiu University—the accredited laboratory in Taiwan for PCDD/F analyses. Prior to analysis, each collected sample was spiked with a known amount of the ¹³C₁₂-labeled internal standard to the extraction thimble. Add toluene to fill the reservoir approximately 2/3 full. Adjust the heat source to cause the extractor to cycle 3 times/h. After being extracted for 24 h, the extract was concentrated, treated with concentrated sulfuric acid, and then followed by a series of sample cleanup and fractionation procedures, including multilayer silica gel column, alumina column and activated carbon chromatography. 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. Immediately prior to analysis, the standard solution for recovery checking was added to the sample. The recoveries of PCDD/F internal standards for the tetra-through hexachlorinated homologues were between 71% and 95%, and met the criteria within 40–130%, while that for the hepta and octachlorinated homologues were between 57% and 106%, and met the criteria within 25–130%. A high-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) was used in the PCDD/F 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. Helium was used as the carrier gas. 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. The electron energy and source temperature were at 35 eV and 250 °C, respectively. Details of the analysis could be found in our previous work [10,20]. The toxic equivalent quantity of PCDD/Fs is given by I-TEQ = ΣXi × li, where I-TEQ denotes the international toxic equivalent quantity, Xi represents the concentration of PCDD/F congeners, and li is the

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