Removal Efficiency and Homologue Patterns of PCDD/Fs at Different Sampling Points in MSWIs

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Introduction

Incineration has been widely used in treating wastes because of its reduction of volume and weight, and high destruction efficiency. However, since incinerators have been pointed out the largest contributor of polychlorinated dibenzo-p-dioxin/dibenzofurans (PCDD/Fs) [1] numerous investigations have been conducted on the best ways of controlling and regulating such emission.

Different mechanisms of PCDD/Fs formation in incinerator have been described. One of them is that PCDD/Fs is formed from chlorinated aromatic precursors. And, these gas-phase precursors react with the fly ash surface to form PCDD/Fs. Another mechanism is the *de novo* synthesis of PCDD/Fs by catalytic reaction of HCl on the fly ash surface (containing Cu^{2+}) resulting in the formation of Cl radicals which react further with organic carbon.[4].

The method of achieving PCDD/Fs removal in flue gas is to use good combustion practice in combination with appropriate flue gas cleaning techniques. There are many processes to reduce PCDD/Fs emission. The most well known processes of removing PCDD/Fs are adsorption and catalytic destruction. Fabric bag filter house with activated carbon injection is popular for the removal of PCDD/Fs. The fly ash cake on bag filter surface was effective to adsorb PCDD/Fs in flue gas and the injection of activated carbon increased the removal efficiency of PCDD/Fs from the flue gas. The activated carbon also provides adsorption sites of PCDD/Fs in flue gas. Another process is selective catalytic reactor(SCR), which is known to destroy PCDD/Fs as well as NOx in flue gas. The catalysts for incinerators are TiO_2, WO_3, and V_2O_5, while V_2O_5 is excellent to destruct PCDD/Fs. The destruction efficiency are depends on the area velocity and operating temperature.

In Korea, eleven municipal solid waste incinerators (MSWIs) are being in operation for over 10 years and the first survey of PCDD/Fs emissions from incinerators was conducted in 1997. Among them, seven incinerators were reported to emitted PCDD/FS over the emission limit (0.5 ng-TEQ/Nm^3). After the measurement results were published in 1997, many efforts to reduce the emission of PCDD/Fs amount were made such as activated carbon injection and changing operating condition.[4]

Therefore, we conducted this study to evaluate PCDD/Fs removal efficiency of each flue gas treatment system. And we also examined the peculiar patterns of PCDD/Fs homologue in each system.

Experiments

Sampling ; Samples of two municipal solid waste incinerators (MSWIs) are collected following the Korean Standard Method which is a modified US EPA method 23.

The sampling point is shown in Figure 1. The flue gases are sampled at the boiler outlet(a), bag filter(B.F) inlet(b), Selective Catalytic Reactor (SCR) inlet(c), and outlet(d) in MSWI A.

Figure 1. Schematic flow and sampling points of MSWI A

Another stack gas sampling is done to evaluate the PCDD/Fs removal efficiency of activated carbon. The stack gas samples are collected with/without injection of activated carbon in the MSWI B. The activated carbon was injected at the inlet of spray dryer adsorber (SDA).

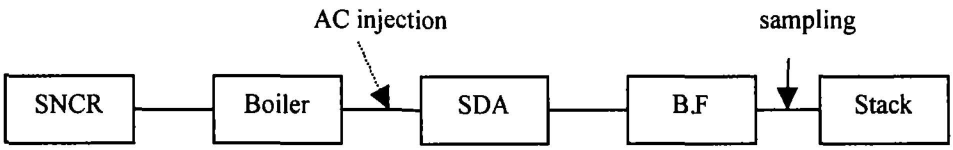

Figure 2. Schematic flow and sampling points of MSWI B

The waste composition, the flue gas composition and the operating conditions of incinerators are investigated continuously during the stack gas sampling. The operating conditions of each incinerator are summarized in Table 1.

<Table 1> Operating conditions of incinerators

Incinerator	Operating condition
MSWI A	Combustor outlet : 840°C Boiler outlet : 193 °C Bag Filter outlet : 140 °C SCR inlet : 205°C SCR outlet : 194°C
MSWI B	Furnace outlet : 1032°C SDA inlet : 194 °C Bag Filter inlet : 161°C Bag Filter outlet : 155°C

Analysis ; Sample preparation was done according to the US EPA method 23. PCDD/Fs were analyzed by high-resolution gas chromatography / high-resolution mass spectrometry (Hewlett-Packard Model 5890 serious _/ Micromass Autospec Ultima). The MS was operated at 10,000 resolution under positive EI conditions (35 eV electron energy), and data were acquired in the single ion recording (SIR) mode.

Results and Discussion

The PCDD/Fs removal efficiency of pollution control devices ; Table 2 and 3 show the results of PCDD/Fs measurement. Removal efficiency of each pollution control devices is calculated based on TEQ value. The PCDD/Fs removal efficiency of each device in MWSI A is as follows ; 87% in SDA, 77. 6% in bag filter and 77% in SCR. In MSWI B, the removal efficieny of PCDD/Fs is about 99 % on activated carbon injection. Many studies reported good efficiency of activated carbon to reduce PCDD/Fs emission regardless of the type of activated carbon.[2-3] Shinoda reported the high PCDD/Fs removal rate (99.9%) by means of using the activated carbon and Reimerink also showed 90-99% removal efficiency of PCDD/Fs in flue gas. [8,9] Compared to that in this study, the injection of activated carbon is the most powerful method to remove PCDD/Fs emission in incinerator.

<Table 2> PCDD/Fs measurement results in MSWI A

Sampling point	A	B	C	D
Ng-TEQ/Nm^3	2.599	0.339	0.076	0.022
Total ng/Nm^3	158.1	21.2	7.2	3.5
Removal efficiency	SDA : 87 %	Bag Filter : 77.6 %	SCR : 77.1 %	

<Table 3> PCDD/Fs measurement results in MSWI B

	w/o injection of AC		With injection of AC		
Times	1	2	1	2	3
Ng-TEQ/Nm^3	1.157	0.916	0.01	0.013	0.008
Removal efficiency	AC : 99 %				

Homologue patterns of PCDD/Fs in MSWI A ; Figure 3 shows the pattern of PCDD/Fs homologe in MSWI A and 10 other MSWIs in Korea and all data are normalized to the total sum of [PCDDs]+[PCDFs]=1 Unlike that of 10 other MSWIS, the emission of PCDDs is mch higher than that of PCDFs in MSWI A. At this we further investigate into the reason of this behavior and check the emission patterns of each sampling point. As shown in Figure 5, while more PCDFs are formed in boiler, more PCDFs are also removed in SCR, and so, final emission levels of PCDDs becom higher than that of PCDFs, which make the emission patterns of MSWI A distinctive. Another comparison is made between MSWI A and C which also has SCR in its pollution control devices. The results are shown in Figure 4. Hiraoka reported that the PCDD/Fs detruction ratio depended on the operating temperature, space velocity, and catalyst geometric properties in SCR.[7] Ok reported that the destruction efficiency of PCDFs was fairy high even at low temperature compared to that of PCDDs in pilot test.[6] The operating temperature of SCR is 205°C in MSWI A and 320°C in MSWI C. Therefore, destruction efficiency of PCDFs might be higher than that of PCDDs in MSWI A. In summary, this discrepancy of destruction efficiency between PCDFs and PCDDs might be due to the operating temperature of SCR. From these results, the emission patterns of PCDD/Fs in incinerator should be varied with operating temperature.

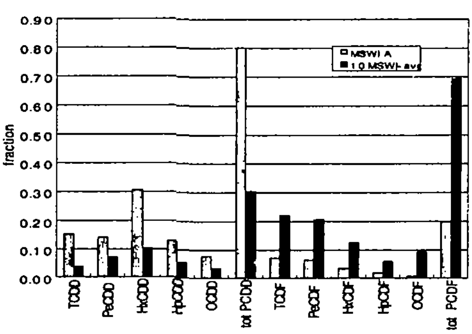

Figure 3. The homologue patterns of MSWI A and the average of 10 MSWIs

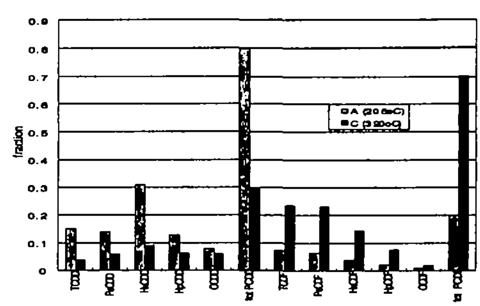

Figure 4. The comparison of homologue patterns with operating temperature in SCR

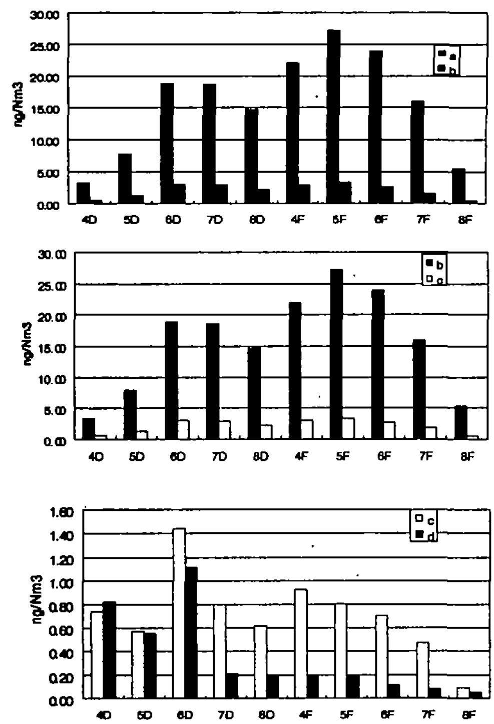

Figure 5. Homologue patterns of PCDD/Fs at different positions of MSWI A

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