CONTROLLING PCDD/PCDF EMISSIONS FROM
INCINERATORS BY FLUE GAS CLEANING*

by

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ABSTRACT

New source performance standards for municipal waste combustors (MWCs) and guidelines for existing incinerators in the U.S., proposed on December 20, 1989, are to be promulgated in December 1990. The proposed national regulations require more stringent control of particulate matter and include pre-combustion, combustion, and post-combustion controls, the last two depending on size and age of the facility.

Dry scrubbing processes have been used exclusively on recent MWCs and are planned for future MWCs in the U.S. for flue gas cleaning (post-combustion control). They inherently include particulate matter control along with their primary function of acid gas removal. These technologies are also generally effective in controlling polychlorinated dibenzo-p-dioxins (PCDD), dibenzofurans (PCDF), and trace heavy metals. Test results quantifying air pollutant emissions, especially PCDD/PCDF, and their control will be presented and compared with the proposed regulations.

INTRODUCTION

The combustion of municipal solid waste (MSW) in the U.S. has almost doubled in the past 3 years, with waste-to-energy conversion complementing the reduction of waste volume for landfill disposal. About 15% of the MSW generated is incinerated, equivalent to 68,000 tonnes/day. Since 95% of the MSW burned is in waste-to-energy facilities, the test results reported here will be for municipal waste combustors (MWCs) with energy recovery.

Over the same 3 years, the U.S. Environmental Protection Agency (EPA) has been developing stricter rules to control pollutant emissions from MWC facilities. On December 20, 1989, these rules were proposed for new facilities [new source performance standards (NSPS)] and existing facilities (emission guidelines).[1] They require all facilities to use three complementary methods to control air polluting emissions: material separation (pre-combustion fuel cleaning), combustion control [good combustion practice (GCP)], and post-combustion control or flue gas cleaning (FGC). The final rules, to be promulgated in December 1990, are now being prepared following the public comment period and the consideration of additional data.

While the present rule for MWCs requires only particulate matter (PM) control, the proposed rules also limit "MWC emissions," nitrogen oxides (NO_x), and carbon monoxide (CO). "MWC emissions" include "MWC metals" as measured by total PM, "MWC organics" as measured by total polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDD), and "MWC acid gas" as measured by hydrogen chloride (HCl) and sulfur dioxide (SO_2). Table I summarizes the proposed emission limits for these air pollutants, the limits being a function of the facility's size (capacity) and age (new or existing unit).[1] While all facilities must comply with PM, PCDD/PCDF, and CO limits, the acid gas emission limits apply to all but small existing sources ($\leq$225 tonnes/day). Only large new sources (>225 tonnes/day) must achieve the NO_x emission limits. While only the proposed MWC emission limits (except CO) are given in Table I, the proposed rules also cover personnel training and operating and monitoring requirements for each facility to ensure compliance with emission limits.

* This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.

TABLE I. PROPOSED MUNICIPAL WASTE COMBUSTION EMISSION STANDARDS[a,1]

	New Source Performance Standards (NSPS)		Emission Guidelines		
CAPACITY, tonnes/day (tons/day)	≤225 (≤250)	>225 (>250)	≤225 (≤250)	>225,≤2000 (>250,≤2200)	>2000 (>2200)
METAL EMISSIONS					
•Particulate Matter, mg/dscm (gr/dscf)	34 (0.015)	34 (0.015)	69 (0.030)	69 (0.030)	34 (0.015)
•Opacity, %	10	10	10	10	10
ORGANIC EMISSIONS					
•Chlorinated Dibenzo-p-dioxins & Dibenzofurans (PCDD/PCDF), ng/dscm	75 250[c]	5-30[b]	500 1000[c]	125 250[c]	5-30[b]
ACID GAS CONTROL, %, OR EMISSIONS (ppmv)					
•HCl	80 (25)	95 (25)	None	50 (25)	95 (25)
•SO_2	50 (30)	85 (30)	None	50 (30)	85 (30)
•NO_x	None	(120-200)[b]	None	None	None

[a]All emission limits are referenced to dry gas with 7% O_2 concentration (20°C, 101.3 kPa).

[b]Single value, probably in this range, will be supplied at promulgation of rules.

[c]Value for refuse-derived fuel (RDF) combustors in the capacity category shown.

Dry acid gas scrubbing processes (or dry scrubbers) for FGC are used on the more recent MWC facilities in the U.S., and this trend is expected to continue for units now being planned. The dry acid gas FGC processes convert the acid gases to a solid form which can be collected as PM. Similarly, organics and metals as solids prior to or after scrubbing are also collected as PM.

Since the main focus here is on controlling PCDD/PCDF, FGC process conditions affecting their control as PM will be discussed, and test results mainly from compliance tests will be compared with the proposed U.S. rules. The potential effects of material separation and NO_x control on PCDD/PCDF and PM emissions will not be discussed. Data are not available on these effects, but properly controlled ammonia flows (minimum slip) in non-selective and selective catalytic reduction are believed to have little or no effect on PCDD/PCDF and PM emissions from dry scrubbers. The treatment or disposal of ash or solid residue will not be discussed as this waste is subject to solid waste rules in the U.S.

DRY FLUE GAS CLEANING

Dry acid gas control processes on MWCs produce a dry powdery waste, and flue gases leaving these processes are not saturated with water. In contrast, wet processes discharge slurries and flue gases saturated with water. Two dry processes are applied on MWCs: dry sorbent injection (DSI) into flue gas followed by PM collection, usually by a fabric filter (FF), and spray dryer absorption with either a FF or an electrostatic precipitator (ESP) for PM collection. While designed primarily to remove HCl and SO_2, both processes achieve multi-pollutant control, including PM, trace organics, and trace heavy metals. In waste-to-energy MWCs with dry scrubbers, the flue gas leaves the boiler at 200°C or less and enters the PM collector at 150°C or less. Therefore, the net production of PCDD/PCDF by de novo synthesis in the PM collector would not be a problem.

Figure 1 shows the DSI process with two options. The dirty flue gas may be humidified and cooled via water injection prior to its contacting powdered sorbent [usually hydrated lime, $Ca(OH)_2$] in a reactor, or it may be cooled indirectly using a heat exchanger before sorbent injection. Both options reduce the gas temperature to about 150°C and lower the approach to saturation temperature (difference between temperature of the gas and its dewpoint) to enhance the acid-gas/sorbent reaction.

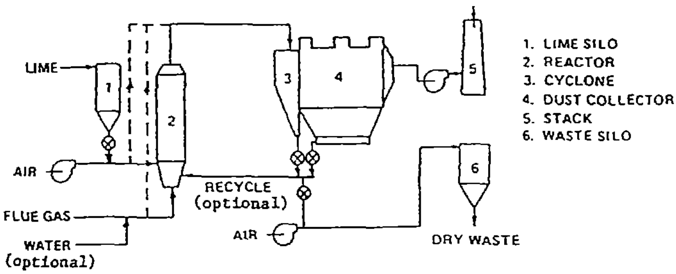

Figure 1. Dry sorbent injection into fluid bed reactor or flue gas duct (dashed line).

Several plants, generally with combustors rated at 180 tonnes/day or less, use the DSI/FF system. $Ca(OH)_2$ is the sorbent for each plant.

The lime spray dryer absorber (SDA) in combination with PM collection is shown in Figure 2. Flue gas at about 200°C enters the SDA and is cooled to around 140°C as the lime slurry droplets are dried in contact with the hotter gas. The gas/lime slurry reaction leads to neutralization of the acid gases and production of solids. Since the gas into the SDA is sprayed with a wet sorbent slurry or solution and, while the collected solids in the process are dry, this process is sometimes referred to as a semi-dry rather than a dry process.

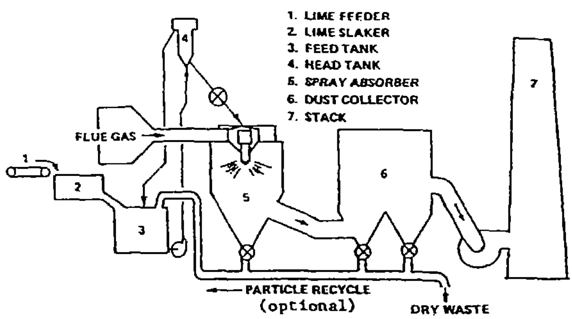

Figure 2. Spray dryer absorption (semi-dry) process.

Most recent MWCs having over 180 tonnes/day capacity and those now being planned for the U.S. use the SDA/FF or SDA/ESP system for acid gas control. As with the DSI/FF system, the approach to saturation temperature and reagent (or stoichiometric) ratio (moles of sorbent per mole of acid gases entering the scrubber) are the major parameters affecting acid gas removal.

PCDD/PCDF removal in dry scrubbers is enhanced by lower gas temperature which leads to condensation of vapor and improved adsorption onto particles, especially fine particles. The presence of lime-based sorbents may also enhance the removal of PCDD/PCDF, by providing additional particle surface area from the reaction products for adsorption, chemical reaction, or both. The limited vapor pressure data on chlorinated dioxins suggest that condensation of these compounds is not their sole mechanism of removal.[2] The carbon (C) content of the flyash particles appears to affect PCDD/PCDF capture. A recent patent proposes the addition of activated C to flue gas to improve removal of PCDD/PCDF [as well as NO_x and mercury (Hg)].[3]

TEST RESULTS AND COMPARISON WITH PROPOSED RULES

Test results from facilities with dry scrubbers are shown in Tables II and III. These results indicate that only SEMASS Unit 2 had a greater average PCDD/PCDF emission (311 ng/dscm) than the proposed limit (125 ng/dscm, see Table I). While this average is based on three tests, with emissions of 18.0, 6.6, and 907 ng/dscm, respectively, no explanation was given for the highest value reported or any mitigating factors. Limited test data indicate that the lime SDA/FF system is more effective in controlling PCDD/PCFF emissions than the lime SDA/ESP system. This may be a result of better fine particle collection in the FF and/or secondary absorption/adsorption in the filter cake. The available PM control data show that, except for the SDA/ESP system, good PM control (>99%) parallels high PCDD/PCDF control (>95%). However, all PCDD/PCDF data in Table III meet the proposed emission guidelines, since all facilities listed have capacities below 2000 tonnes per day.

TABLE II. CONTROL OF PARTICULATE MATTER (PM), POLYCHLORINATED DIBENZO-P-DIOXINS (PCDD) AND DIBENZOFURANS (PCDF), AND SELECTED HEAVY METALS WITH DRY LIME INJECTION/FABRIC FILTER SYSTEMS[4,5]

	Average PM Concentration[a] mg/dscm @ 12% CO_2		Average Total PCDD/PCDF Concentration ng/dscm @ 7% O_2
Location and Test Date	inlet	outlet	outlet
•Claremont, NH			
Unit 1, 5/87	NA[b]	25	NA
7/87	NA	NA	37.6
Unit 2, 5/87	NA	9.8	NA
7/87	NA	NA	32.3
•St. Croix, WI			
6/88	NA	34	7.73
10/88	NA	27	NA
•Springfield, MA[c]			
7/88	2059	3.7	0.15[d]
•Dutchess County, NY			
Unit 1, 2/89	NA	22	4.83
Unit 2, 2/89	NA	80	17.9
3/89	NA	25	NA
5/89	NA	18	NA

[a]dscm = dry standard cubic meter (20°C, 101.3 kPa).

[b]Not available or not measured.

[c]All concentrations are referenced to dry gas with 12% CO_2.

[d]Dioxin at outlet reported as 2,3,7,8 tetrachlorinated dibenzodioxin equivalent (EPA method).

TABLE III. CONTROL OF PARTICULATE MATTER (PM), POLYCHLORINATED DIBENZO-P-DIOXINS (PCDD) AND DIBENZOFURANS (PCDF), AND SELECTED HEAVY METALS WITH LIME SPRAY DRYER ABSORBER (SDA)/FABRIC FILTER (FF) OR SDA/ELECTROSTATIC PRECIPITATOR (ESP) SYSTEMS[4,6,7,8]

Location, Control System, and Test Date	Average PM Concentration[a] mg/dscm @ 12% CO_2		Average Total PCDD/PCDF Concentration ng/dscm @ 7% O_2	
	inlet	outlet	inlet	outlet
•Marion County, OR Unit 1, SDA/FF 9/86	2016	5.3	43.0	1.26
•Biddeford, ME Unit A, SDA/FF 12/87	7322	32	903	4.38
•Mid-Conn.[b] Unit 11, SDA/FF 7/88	5514	9.2	996	0.646
2/89[c]	4073	4.1	747	0.368
•Millbury, MA Unit 1, SDA/ESP 2/88	NA[d]	4.1	NA	NA
Unit 2, SDA/ESP 2/88	NA	19	170	59.2
•SEMASS Unit 1, SDA/ESP 3/89	9793	18	NA	9.3
Unit 2, SDA/ESP 4/89	8832	27	NA	311[e]

[a] dscm = dry standard cubic meter (20°C, 101.3 kPa).
[b] All concentrations are for dry gas with 12% CO_2.
[c] Values are averages for normal SDA/FF temperatures (performance tests 6, 8, 12, 13, and 14).
[d] NA = not available or not measured.
[e] Average value of 18.0, 6.6, and 907.

Tables IV and V compare PCDD/PCDF emissions systems on a toxic equivalency basis. Table IV shows that the EPA method gave values below those from the International (1989) method. It also shows that the ratio of the total outlet PCDD/PCDF to toxic equivalency varies greatly, so that it is not possible to simply and easily convert from total PCDD/PCDF to toxic equivalencies, given one or the other. Table IV also shows that the total PCDF exceeded the total PCDD, except for the toxic equivalents for the mass burn combustor with the SDA/FF system. Table V indicates that the reported units with SDA/FF systems met the 0.1 ng/Nm^3 toxic equivalent (I-89) standard used in some European countries, while the units equipped with other FGC systems did not.

TABLE IV. COMPARISON OF TOTAL PCDD/PCDF AND 2,3,7,8 TCDD TOXIC EQUIVALENTS (All concentrations are in ng/dscm referenced to dry gas at 20°C with 12% CO_2.)

Combustor Type	Flue Gas Cleaning System	Total PCDD/PCDF		Toxic Equivalent Method[a]		Ratio[b]	
		A. inlet	B. outlet	C. EPA	D. I-89	B/C	B/D
Mass Burn	SDA/FF	70.8 (26.5/44.2)[c]	2.20 (1.01/1.19)	0.063 (0.060/0.002)	0.079 (0.072/0.006)	35	28
Mass Burn	SDA/ESP	279 (50.6/228)	76.0 (15.6/60.4)	0.485 (0.247/0.278)	1.159 (0.247/0.911)	157	66
Refuse Derived Fuel	SDA/FF	996 (328/668)	0.646 (0.271/0.375)	0.0017 (0.000/0.0017)	0.0050 (0.0008/0.0042)	380	129
Processed Fuel	SDA/ESP	NR[d] (NR)	8.65 (NR)	0.105 (NR)	0.142 (NR)	82	61

[a] EPA = U.S. Environmental Protection Agency, I-89 = International Method, 1989.
[b] Ratio is the total PCDD/PCDF value in column B to toxic equivalent value in column C or D.
[c] Values in parentheses are total PCDD and PCDF, respectively.
[d] NR = Not reported.

TABLE V. 2,3,7,8 TCDD TOXIC EQUIVALENTS
(All concentrations are in ng/Nm^3 referenced to dry gas at 0°C with 11% O_2[a].)

Combustor Type	Flue Gas Cleaning System	2,3,7,8 TCDD Toxic Equivalent[b]	
		EPA	I-89
Mass Burn	DSI/FF	0.116	NA
Mass Burn	SDA/FF	0.049	0.061
Mass Burn	SDA/ESP	0.377	0.900
Refuse Derived Fuel	SDA/FF	0.0013	0.0037
Processed Fuel	SDA/ESP	0.0815	0.110

[a]Conversion from 12% CO_2 to 11% O_2 assumes % CO_2 + % O_2 = 21.
[b]EPA = U.S.Environmental Protection Agency, I-89 = International Method,1989.

SUMMARY

Dry scrubbing processes are widely used on modern MWCs in the U.S. The lime SDA in combination with a FF or ESP is the predominant system, with the hydrated lime DSI/FF system being applied on several small units (<180 tonnes/day). Generally, existing MWCs with dry acid gas control are achieving the proposed PCDD/PCDF limits, with the SDA/FF system achieving lower PCDD/PCDF emissions and greater PCDD/PCDF removal than the SDA/ESP system. For the FGC systems compared, only the SDA/FF systems complied with the 0.1 ng/Nm^3 toxic equivalency (I-89) standard of some European countries.

REFERENCES

1. Air Pollution Standards of Performance for New Stationary Sources; Rule and Proposed Rules (40 CFR Parts 60, 51, and 52), Federal Register, Vol. 54, No. 243, Wednesday, December 20, 1989, pp. 52188-52304.

2. Damle, Ashok S., David S. Ensor, and Norman Plaks, "Condensible Emissions from Municipal Waste Incinerators," The Eighth Symposium on the Transfer and Utilization of Particulate Control Technology, San Diego, CA, March 1990.

3. Moller, Jens T., Niels Jacobsen, Kirsten K. Nielsen, and Stig Rasmussen, "Process for Removal for Mercury Vapor and/or Vapor of Noxious Organic Compounds and/or Nitrogen Oxides from Flue Gas from an Incinerator Plant," U.S. Patent No. 4,889,698, December 26, 1989.

4. Municipal Waste Combustors-Background Information for Proposed Standards: Post Combustion Technology Performance, EPA-450/3-89-027c (NTIS PB90-154865), U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC, August 1989.

5. McClanahan, D., A. Licata, and J. Buschmann, "Operating Experience with Three APC Designs on Municipal Incinerators," International Conference on Municipal Waste Combustion, Hollywood, FL, April 1989.

6. Brna, T.G., M.G. Johnston, C.E. Riley, and C.C. Masser, "Performance of Emissions Control Systems on Municipal Waste Combustors," Paper No. 89-109.2, Air and Waste Management Association Annual Meeting, Anaheim, CA, June 1989.

7. Brna, T.G. and J.D. Kilgroe, "Control of PCDD/PCDF Emissions from Municipal Waste Combustion Systems," Ninth International Symposium on Chlorinated Dioxins and Related Compounds (Dioxin 89), Toronto, Ontario, Canada, September 1989.

8. SEMASS Waste-to-Energy Resource Recovery Facility: Compliance Test Report. Prepared by Eastmount Engineering, Inc., Walpole, MA, for SEMASS Partnership, Rochester, MA, August 1989.