ABSTRACT

Stack emissions measurements, resulting from burning actual wastes in commercial combustors with appropriate emissions control technology under usual operating conditions show significant reductions in particulate matter, acid gas, trace organic, and metal concentrations in exhaust gases from contemporary waste combustion systems. Current estimates of this industry’s contribution to the national dioxin burden, for instance, indicate that it is far less than 1 percent of that from known U.S. dioxin. However, the levels set in permits are much higher, high enough to allow for the variability of the waste and of the emissions control device performance, in order to avoid exceedances during periodic compliance tests which can occur due to statistical chance. Estimates of environmental impact and health risk which have been based on trial-burn data and permit levels are thus substantially higher than those which would result based on actual average emissions during normal operation. This paper focuses on dioxin/furan emissions from municipal, medical and hazardous waste combustion, showing their relationship to CO emissions, the emission control technology and operating temperature, as well as the variability of emissions over long term operation of toxic equivalent dioxins, PM and heavy metals.

INTRODUCTION

Emissions from combustion of municipal, medical and hazardous wastes are regulated under the Clean Air Act of 1990 (CAA) and the Resource Conservation and Recovery Act (RCRA). Many states have developed their own limitations, generally more stringent than the federal regulations prior to 1995, during permitting. As a result, new waste combustion systems have been developed to meet high standards. Older systems have often been retrofit or shut down.

The 1990 amendments to the CAA require federal regulation of emissions of particulate matter (PM), SO₂, HCl, opacity, NOₓ, lead, cadmium and mercury to be equivalent to Maximum Achievable Control Technology (MACT). At MACT, the emissions levels achieved by the best performing 12 percent of operating units are to be equaled by all other installations. The EPA has estimated that overall reductions in emissions of 98 percent will be achieved based on these new standards and guidelines, compared to those of old plants required to meet only the old 0.08 gr/dscf @ 12% CO₂ emissions standard, and lacking acid gas controls.

A great reduction in stack emissions from these various types of waste combustors had already taken place before the MACT standards become effective. A recent paper indicates that actual dioxin emissions from municipal waste combustors (MWCs) are 12 percent of EPA’s most recent estimate; hazardous waste incinerators (HWIs) are 19 percent; cement kilns (CKs) are 8 percent (only 2 percent, with newer information); and medical waste incinerators (MWIs) are 46 percent. In all, dioxin emissions from these sources, based on
current inventories and emissions tests, indicate they are no longer the predominant or even a major contributor to environmental dioxin, contrary to what the public has been led to believe.

**Comparison of dioxin emissions from waste combustion with all sources.**

It is difficult to compare the annual contribution of waste combustors in the U.S. with that from all sources. Estimates of dioxin and furan homologue (total PCDD/F) emissions from all *known* sources range from 4,000 to 6,000 g/y, and from *all* sources, perhaps from 20,000 to 40,000 g/y.² Early EPA emission estimates indicated that these sources contributed roughly 50 percent of the known emissions; hence, they were scheduled for MACT regulation. These estimates have been drastically reduced, however, as real data and updated inventories become available to replace permit values and to a limited extent, highly-biased trial-burn results.

For example an early EPA estimate for international toxic equivalent (TEQ) dioxin and furan annual emissions of MWIs was 670 g/y, but the EPA has now published a revised estimate, based on information volunteered by the industry. The estimate for 1990 is now 600 gTEQ/y; for 1995, 150 g/y; and for 2002, 14 to 23 g/y, depending upon various estimates of likely MACT rules.³

In the case of HWIs, the EPA’s estimate of 79 gTEQ/y was criticized. A more accurate estimate of 14.9 g/y for 1996 was derived taking into account the actual number, design and capacities of existing facilities, test data and operating hours.⁴ This estimate is still probably too high because it is based mainly on trial-burn, worst case emissions. In RCRA trial-burns, the feed is often spiked with artificially high feed rates of selected organic compounds and metals, in order to demonstrate 99.99 percent destruction and removal efficiency in the presence of background or even the use of non-detect levels. In general, tests of normal operation are either not performed or the data are not published. A series of continuous metals monitoring tests of the WTI hazardous waste combustion facility in East Liverpool, Ohio during normal operation showed metals emissions to be essentially at or below the detection limit of EPA stack test procedures.⁵ EPA has revised its estimated emissions from MWCs to 3,333 gTEQ/y in 1993, and to 733 g/y in 1995. When the 1995 rules are fully implemented, EPA expects the contribution from MWCs to be about 24 g/TEQ/y.⁶ As noted above, this is a very conservative estimate of actual environmental impact.

Major reductions in emissions which have already been achieved. The minor contribution modern waste combustion systems are currently making to the environment may seem incredible to the public. After all, they have been told repeatedly that waste combustors are major contributors.

**Toxic equivalent versus total PCDD/F emissions and concentrations.**

It is essential to establish the relationship between total PCDD/F and TEQ. A system for converting the overall mixture of 2,3,7,8 substituted PCDD/F congeners to an equivalent amount of 2,3,7,8 tetrachlorodibenzo-p-dioxin has been developed. EPA has chosen to suggest that a factor of 60 to be used to divide into total PCDD/F concentration to get TEQ. Actually, this factor is not constant. Figure 1 shows that for MWCs, biomedical waste combustors (BMWCs) and HWIs, at old facilities with ESPs, as well as newer ones with spray-dry scrubbers and ESPs (SD/ESP), or fabric filters (SD/FF) with or without carbon injection, the ratio of PCDD/F to TEQ ratio is between 30:1 to 100:1. The spread between the lines depends upon a number of factors, such as the waste itself, and the congener distribution between dioxins and furans. For MWCs the average seems to be around 50:1,⁷ but for HWIs the ratios have been found to be as high as 100:1.

**Historical measures performed to reduce dioxin emissions.**

In the early 1980s, total PCDD/F emissions from some municipal waste incinerators and waste-to-energy facilities were found to be as high as 10,000 ng/dsm³ corrected to 7% O₂. Concern over such high concentrations led to world-wide research, from laboratory to full-scale testing, to determine the cause and universality of these emissions. Stack gas emissions controls for waste combustors have evolved as follows:

- no control;
- wet scrubbers or low efficiency ESPs;
- improved ESPs with spray-dryer absorber and dry reagent injection scrubbers;
- dry lime injection plus a fabric filter;
- spray-dryer absorber with a fabric filter; and
- Powdered Activated Carbon (PAC) injection.

The reductions in PCDD/F emissions achieved during this evolution have to be presented on logarithmic coordinates since they span six orders of magnitude (a 1,000,000-time reduction from top to bottom). Figure 2 shows total furans plotted versus dioxins indicating that the data falls in a clear but wide band; wherein the furans range from equal to dioxins to about 10 times dioxins. Most important is the great progress made in a series of steps, reducing dioxins and furans by improvements in the combustion of municipal solid waste (MSW) improvement, by acid gas controls and fabric filters (SD/FF), and finally by the use of activated carbon injection, from over 10,000 down to less than 2 ng/Nm³. Data from hazardous waste and medical waste incinerators also falls in the same band.

- A landmark test was performed at Pittsfield, MA, under ASME sponsorship with New York State Energy Research and Development Authority (NYSERDA) financing, to obtain data over a wide range of operating conditions, burning industrial waste as well as municipal waste, including tests spiked with high PVC levels. These tests found that furnace temperature had an affect on PCDD/F leaving the boiler. As seen in Figure 3, tests at various furnace temperatures show a relationship with carbon monoxide (CO) emissions, (and, not shown, corresponding changes in excess oxygen). The jump at low CO levels indicates that increasing furnace temperature ultimately is counterproductive due to the corresponding reduction in oxygen levels which result in a decrease in mixing effectiveness. The data from Hampton show the improvements in combustion from 1986 to 1994 resulting from improved overfire air mixing and control, among other factors such as better control of waste feeding.

- Tests at the Montgomery County facility in Dayton, OH brought to light the production of PCDD/F in the electrostatic precipitator (ESP). As seen in Figure 4, toxic equivalent TCDD concentrations were found to be a clear function of ESP temperature. TEQ emissions from the facilities with ESP’s showed the same trend with temperature, but distinctly different absolute values due to differences in combustion efficiency, that is, the ability to reduce precursors to dioxins. Even the data from a Danish facility having a spray-dry scrubber with fabric filter (SD/FF) shows a clear trend with baghouse temperature, with a major dip below 320°F.

- Slip stream tests performed by Environment Canada at Quebec City, and later confirmed in full-scale facilities, showed that dry lime or spray-dry lime scrubbers followed by fabric filters could drastically reduce PCDD/F (as well as acid gases and metals) emissions.

- The use of acid gas scrubbers, with either spray-dry or dry injection of lime, followed by an ESP or fabric filter was found to provide a 90% or greater reduction in TEQ, as can be seen in Figure 5. Facilities with ESPs, without acid gas controls, showed no reduction, or even an increased while passing through the ESP.

- When acid gas controls are used, the reduction in dioxins was found to be about the same, regardless of the levels of CO measured in the stack. Figure 5 shows that the high levels of CO at the inlet of the scrubber/baghouses in Quebec (marked Q) and in Hartford (marked H), TEQ levels were reduced to less that 0.1 ng/dscm and even as low as 0.01 ng/dscm.

- Joint tests by Environment Canada/EPA/DOE at the Mid-Connecticut refuse derived fuel fired (RDF) plant in Hartford, CT showed that the carbon produced by less-than-perfect combustion of MSW removed both mercury and PCDD/F. Other tests found that dry powdered activated carbon (PAC) injection effectively removed mercury and PCDD/F.

- Recent tests sponsored by ASME and supported by the DOE/NREL plus a consortium of facilities and trade organizations have shown that existing waste-to-energy facilities with ESPs could be upgraded to reduce PCDD/F, mercury and acid gases (SO₂ and HCl) by dry injection of calcium or
sodium based reagents and reduction of the ESP operating temperature to levels meeting the USEPA’s 1995 guidelines for small facilities.\textsuperscript{14}

**Carbon monoxide as an indicator of good combustion.**

Figure 3 shows the relationship between TEQ emissions from MSW combustors (prior to the emission controls) versus carbon monoxide (CO). The upper line shows a good fit to dioxin data taken at the Hampton, Hamilton, and Pulaski facilities, MWCs. An enormous reduction in TEQ was obtained by more effective mixing of oxygen and organics, and reduction in ESP operating temperature in the process of upgrading the old-style refractory incinerator at the Pulaski facility.\textsuperscript{15} Similar improvements were obtained at Hampton.\textsuperscript{16} These two sets of data fall close to the same line, even though Pulaski has a refractory furnace followed by a water quench without boiler and Hampton has a refractory furnace followed by a waste heat boiler. The fundamental mechanisms are the same.

The Pittsfield data, however, plotted on the same graph as a series of points representing tests at various furnace temperatures, showed that both CO and PCDD/F correlate with furnace temperature until low CO levels are achieved. At low CO, PCDD/F and CO are no longer proportional.\textsuperscript{17} These results, expressed as TEQ, are shown in Figure 3. Evidently the Pittsfield combustion system resulted in lower TEQ than Hampton and Pulaski, at furnace temperatures lower than 1800°F.

**Formation of PCDD/F**

Extensive research has shown that one formation mechanism for PCDD/F is particulate and metal surface reactions, especially at the temperatures experienced between the boiler outlet and ESP exhaust. Higher PCDD/F concentrations were found when the ESPs were operated at the historic 450-500°F temperature to avoid condensation and corrosion than at the 285-325°F operating range which is now commonly used.

**Temperature of emissions control device**

Figure 4 shows the effect of the temperature at which the ESP emissions control system is operated on TEQ emissions. Here we see that TEQ data falls on lines with similar slopes, but at different levels, indicating that other variables are involved. The differences in level may be attributed to the quality of combustion, hence the quantity of dioxin precursors available for conversion on flyash, boiler and ESP surfaces.

Use of acid gas controls produced a startling reduction in PCDD/F emissions. Figure 5 compares data from tests of facilities having just ESPs with those using scrubber/fabric filters in more detail, specifically showing TEQ levels entering and leaving the emissions control system. It is apparent that TEQ levels in the stack emissions of SD/FF facilities are essentially independent of CO, as well as of the inlet TEQ concentrations. Closer investigation of the ESP inlet/outlet points shows that TEQ has been produced rather than reduced in the ESP.

The early pilot tests, marked Quebec (Q), show inlet levels at about 100 ngTEQ/dsm\textsuperscript{3}, with an outlet level of about 0.1 ng/dsm\textsuperscript{3}, a reduction of 99.9 percent.\textsuperscript{10} It was doubted that these pilot test results could be repeated: however, the tests at the Hartford, CT facility (H) show similarly high TEQ levels entering and similar low exit levels.\textsuperscript{11} During these tests, the Hartford facility was burning RDF under relatively erratic conditions resulting in high uncontrolled TEQ and CO levels. Better operating conditions and lower CO readings (80 ppm) showed similar inlet and outlet TEQ levels. It appears that the carbon associated with the relatively poor combustion conditions was serving as an effective Total PCDD/F removal agent. The carbon also captured most of the mercury!

Conventional waterwall MWCs, which have CO emissions in the 30 ppm range, show stack concentrations similar to those at the Hartford, CT facility. Marion County, OR (M) and Commerce, CA (C) were two of the first with SD/FF to show this performance.\textsuperscript{7}

The next step in reducing PCDD/F was the addition of PAC prior to the baghouse, either separately in the duct, or along with dry injected lime. This produced further (90 percent or greater, in some case) reduction in dioxin emissions.
In summary, the levels of TEQ emissions from contemporary MWCs are well below the guideline for existing small MWCs of 2.1 ng/dsm³, and large ones with ESPs of 1.0 ng/dsm³, and the regulations for new facilities of 0.2 ng/dsm³ established by the USEPA.

**Emissions Variability**

Stack emissions of PM, dioxins and metals from fifteen compliance tests of a single facility over a four year period, each point being the average of three compliance tests, are shown in Figure 6. From the statistics of the data shown in Table 1, these observations may be made:

- Each data set represents 1/15 of the tests, so 93% of the tests are at or less than the maximum.
- The standard deviation ranges from 62% to 2.06% of the average.
- The average plus three SD is close to the maximum value in the test series.
- The coefficient of variation, SD/Avg, is close to 1.0 for PM and TEQ, but varies up to 2.0 in the case of lead, which is more dependent upon the waste burned.
- The average plus three SD approaches the maximum value.
- The ratio of the maximum reading to the average ranges from 2.2 to 8.3.

These data from a WTE facility show that even if permit conditions are set to allow for the maximum value which occurred once in 15 tests, the actual average of emissions would be far less, in fact 2 to 8 times less, depending upon the specific pollutant in question. This relationship is found in much of the available municipal, medical and hazardous waste emissions data.

The variability exhibited at four HWIs having complex, state-of-the-art air pollution control systems (APCS) is illustrated in Figure 7. The TEQ concentrations differed by more than two orders of magnitude between plants, but are comparatively uniform within a facility. Many factors contribute to this behavior, including the waste burned, incinerator design and operation, and the specific APCS employed. The difficulty in establishing a single permit level for this group of incinerators is evident.

**Chlorine in waste versus dioxins in stack**

Some have raised the concern that chlorine in waste burned in incinerators and waste-to-energy facilities is the principal causative element of dioxin emissions. Anecdotes are cited that indicate there is a correlation between chlorine in the waste and uncontrolled dioxin emissions. An ASME study which analyzed all the available data with HCI concentrations entering the APCS (an indication of chlorine in the waste feed) and PCDD/F concluded that there is no real correlation in the entire data set. The principal factors influencing emitted PCDD/F concentrations are the type of emissions control device employed and its operating temperature.

There is little published data which directly studies the relationship between HCl and PCDD/F, since, for one thing, few data sets exist for the same facility with a wide range of uncontrolled HCl. However, the USEPA tests of the Morristown, NJ facility, and tests of the Bronx-Lebanon, NY facility do provide a limited set of such data, shown in Figures 8, 9, and 10. Both of these data sets also show the effect of powdered activated carbon (PAC) injection.

Morristown, NJ has a rotary kiln combustor and heat recovery boiler followed by a SD/FF. Without carbon injection, the data show a range of TEQ emissions between 0.8 and 7 ng/dsm³. During this set of tests, there was little variation in HCl, hence no meaningful correlation could be obtained. However, the two points obtained with PAC injection shows a wider range of HCl from 600 to 1700 ppmv which reflects the typical range of medical waste when general waste is mixed with the types containing more PVC. The PAC reduced the TEQ level to 0.05 ng/dsm³.

The Bronx-Lebanon facility has a two-stage reciprocating hearth refractory combustor followed by a heat recovery boiler, air-to-gas-cooler, dry lime injection, lime control tower, and fabric filter. The data points at about 200 ppmv HCl were obtained during testing with trash, not medical waste, whereas the points in the 1,000
ppmv range were medical waste. Here, TEQ was reduced from 0.8 ng/dsm³ to about 0.1 ng/dsm³ with PAC injection. The Morristown data for Total PCDD/F show a strong reduction in PCDD/F as HCl levels increased. What is most striking about the Bronx data is that there was essentially no difference in TEQ emissions in spite of the range from 600 to 1700 ppmv HCl without carbon, and a range from 200 to 1,000 ppmv HCl with PAC. These data indicate that combustors equipped with acid gas controls reduce PCDD/F to low levels, quite independent of the amount of HCl entering the system. It is also especially interesting to note that PCDD/F actually decreases with increased HCl, and that PAC has a relatively greater effect at higher HCl levels. These, of course, may be artifacts due to the limited size of these data sets rather than a real phenomena.

**Summary**

Since emissions from waste combustion have come under regulation, enormous reductions in emissions of PCDD/F have occurred. Large reductions have resulted from improvements in combustion and the use of acid gas scrubbers. Carbon injection has further reduced dioxin emissions to levels under the practical quantitation limit (0.2-0.5 ng/dsm³ @ 7% O₂). These reductions support the EPA estimate that the overall reduction will be about 98 percent compared to uncontrolled emissions, not the actual state of affairs in 1995 when the MWC rule was promulgated, and even show that the EPA estimate is very conservative: the emissions are and will be far lower. The contribution of PCDD/F to the environment from regulated waste combustors is already an extremely small fraction of the total environmental burden. With anticipated further reductions, this contribution will approach insignificance.

The EPA, in an effort to establish MACT standards, is challenged by the variability of emissions from individual facilities, as well as from different facilities employing essentially the same emissions control technology. The stack emissions from a given facility vary under normal operating conditions, approaching a log-normal rather than normal distribution. The level which probably will not be exceeded in 95 percent of the tests is likely to be may range from two or three to as much as eight times the data mean; hence permit conditions must allow for this possibility. On the other hand, environmental impacts depend on the average or median emissions since short-term peaks are not, by definition, sustained.

Waste burning facilities having essentially the same emissions control technology, such as scrubber/baghouse technology, display a wide, but low, range of uncontrolled PCDD/F concentrations due to differences in design, operating conditions and waste composition (but not chlorine content at usual levels). Controlled concentrations depend on the temperature of the particulate APCS and retention time. Outlet PCDD/F is relatively independent of inlet concentrations.

While CO is a reasonably good indication of good combustion, PCDD/F stack concentrations are independent of CO when acid gas controls are used, depending mainly on the temperature at which the controls are operated, and the design and operation of these controls.

**CONCLUSIONS**

The ratio of total PCDD/F to the TEQ ranges from 30 to 100 creating inconsistent control requirements in view of EPA’s vacillation in choosing between PCDD/F or TEQ in regulatory standards. This effect is most pronounced at low concentrations where zero is used for non-detects in calculating TEQs. The variability of emissions is similar for MWCs, HWIs, and MWIs. The standard deviation ranges from less than one to over two times the mean.

Estimates of annual emissions of organics and metals must be based on the average of readings taken periodically throughout a year or several years. The maximum measurement reported over the course of time may range from two to eight times the average, depending upon the pollutant. This variation should be allowed for in the permit, but using such maximum values to evaluate environmental impact results in serious over-estimation.
While CO has been found to be a good surrogate for good combustion, it is not a surrogate for dioxin stack emissions at low CO levels. It has been found that regardless of the PCDD/F levels entering scrubber/baghouse emissions controls, outlet levels are consistently low, and strongly dependent on the temperature at which the baghouse is operated.

The “control efficiency” for organics (such as PCDD/F) is not a constant factor, but rather is highly dependent on the temperature of the gases in the emission control system, relatively independent of inlet pollutant concentrations. The same may be the case for many heavy metals.

**Table 1 - Compliance Test Stack Emissions Measured over Four+ Years**

WTE Facility with Dry lime-Injection and Fabric Filter

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<tr>
<th></th>
<th>TSP gr/dscf</th>
<th>Cadmium µg/dscm</th>
<th>Lead µ/dscm</th>
<th>Mercury µg/dscm</th>
<th>Chromium µg/dscm</th>
<th>Nickel µg/dscm</th>
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<td>2.95</td>
</tr>
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</table>

**REFERENCES**

6 USEPA, “National Dioxin Emissions from Medical Waste Incinerators,” (also Municipal Waste Combustors); USEPA, Emissions Standards Division, TRP, NC, June 1996.


Figure 1  Total Dioxins versus Toxic Equivalent (TEQ): Stack Emissions from Waste-to-Energy Facilities Equipped with Electrostatic Precipitators (ESPs) [triangle] or Spray-dry Scrubbers with Fabric Filters (SD/FF).[square].

Figure 2 - Total dioxins versus furans, showing reductions due to improved combustion, scrubber/baghouse control, and carbon injection.
Figure 3  Toxic Equivalent versus Carbon Monoxide. Hampton, Hamilton and Pulaski Points Fall on the Same Line. Pittsfield Data from Diagnostic Tests Fall on the Same Line Except at Temperatures above 1550°F.
Figure 4  ESP Gas Temperatures versus TEQ Emissions from WTE Facilities with ESPs, Compared with Danish Facility with Spray-Dry Baghouse, Tested Over a Range of Temperatures. Sets of Two or Three Test Runs Show Trend with Gas Temperature. Dayton (Montgomery) Data Fall on Same Line (open box) Except Point With Furnace Lime Injection.
Figure 5 - TEQ versus Carbon Monoxide Showing Inlet and Outlet Conditions of ESPs and baghouse-equipped WTE Facilities. Down triangles are inlet, up triangles are outlets. P and MD have ESPs, the others have scrubber/baghouses. M is Marion County, C is Commerce, H is the RDF facility in Hartford, CT.
Figure 6  Distribution of Test Data from a Single WTE Facility with Dry Lime-injection Baghouse. Metals, Particulate (PM) and Dioxins (TEQ) Show a Similar Range of Variation.
Figure 7 - Distribution of TEQ Emissions from Individual Hazardous Waste Combustors.
Figure 8 TEQ versus Hydrogen Chloride Measured at Inlet to Scrubber/Baghouse of Two Medical Waste Combustion Systems, With and Without Carbon Injection, Comparing Morristown and Bronx Data.
Figure 9  TEQ versus Hydrogen Chloride, Without Carbon, Measured at the Inlet of the Dry Lime-injection Baghouse and in the Stack.

Figure 10  TEQ versus Hydrogen Chloride Showing Measurements at Inlet of Dry Line-injection Scrubber/baghouse and in the Stack, With Carbon Injection.