

Emissions of Polychlorinated Dibenzo-*p*-dioxins and Polychlorinated Dibenzofurans from the Open Burning of Household Waste in Barrels

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Backyard burning of household waste in barrels is a common waste disposal practice for which pollutant emissions have not been well characterized. This study measured the emissions of several pollutants, including polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDDs/PCDFs), from burning mixtures designed to simulate waste generated by a "recycling" and a "nonrecycling" family in a 208-L (55-gal) burn barrel at the EPA's Open Burning Test Facility. This paper focuses on the PCDD/PCDF emissions and discusses the factors influencing PCDD/PCDF formation for different test burns. Four test burns were made in which the amount of waste placed in the barrel varied from 6.4 to 13.6 kg and the amount actually burned varied from 46.6% to 68.1%. Emissions of total PCDDs/PCDFs ranged between 0.0046 and 0.48 mg/kg of waste burned. Emissions are also presented in terms of 2,3,7,8-TCDD toxic equivalents. Emissions of PCDDs/PCDFs appear to correlate with both copper and hydrochloric acid emissions. The results of this study indicate that backyard burning emits more PCDDs/PCDFs on a mass of refuse burned basis than various types of municipal waste combustors (MWCs). Comparison of burn barrel emissions to emissions from a hypothetical modern MWC equipped with high-efficiency flue gas cleaning technology indicates that about 2–40 households burning their trash daily in barrels can produce average PCDD/PCDF emissions comparable to a 182 000 kg/day (200 ton/day) MWC facility. This study provides important data on a potentially significant source of emissions of PCDDs/PCDFs.

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Introduction

Open Burning of Household Waste. In many areas of the country, residential solid waste disposal practices consist of open burning using barrels or other similar devices instead of, or in addition to, disposal in municipal landfills or municipal waste combustors (MWCs). The motivations for households that open burn their garbage may include convenience, habit, or waste disposal cost avoidance (1). Modern refuse combustors have tall stacks, specially designed combustion chambers, and high-efficiency flue gas cleaning systems that serve to minimize the impact of emissions associated with waste combustion. In contrast, emissions from open burning of residential solid waste are released at ground level, possibly resulting in decreased dilution by dispersion. Additionally, the low combustion temperature and locally oxygen-starved conditions associated with backyard burning may result in incomplete combustion and increased pollutant emissions.

Only two previous studies characterized emissions associated with open burning of residential refuse in a backyard burner [e.g., a 208-L (55-gal) drum], and neither was published in the peer-reviewed literature (1, 2). Both study designs included a hood and stack constructed above the 208-L (55-gal) drum to capture the plume and facilitate pollutant emissions tests.

The Two Rivers Regional Council of Public Officials (Illinois) study estimated emissions per unit mass of waste initially present in the barrel for several different air toxics [e.g., total PCDDs/PCDFs, total volatile organic compounds (measured as methane), air toxics metals (antimony, arsenic, barium, beryllium, cadmium, chromium, lead, manganese, mercury, phosphorus, and titanium), particulate matter (PM), hydrogen chloride (HCl), carbon monoxide (CO), and oxides of sulfur and nitrogen] associated with open burning of residential solid waste. This study included a survey of 187 residents in rural counties of Illinois to determine the quantity and type of wastes burned, the management of the ash, and the motivation for burning. The Western Lake Superior Sanitary District (Minnesota) study evaluated emissions of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD); no other pollutants were measured. The results of these two studies indicated that the estimated emissions per unit mass of waste initially present in the barrel were higher than for controlled incinerators, with the Illinois study suggesting a factor of 17 higher than incinerators for total PCDDs/PCDFs and the Minnesota study suggesting a factor of 20 higher than incinerators for 2,3,7,8-TCDD.

In another study, emissions were quantified from the burning of municipal refuse on a burn table equipped with a cone to funnel the pollutants to a sampling port (3). Samples were analyzed for several combustion gases and hydrocarbons, including polycyclic aromatic hydrocarbons (PAHs), but not PCDDs/PCDFs. This study did not simulate the oxygen-starved conditions typically found in a burn barrel.

While these studies provide useful information, there are limitations associated with their results (e.g., the uncertainty in the dilution air and a limited number of target compounds). This paper presents a portion of the results of a study to qualitatively identify and quantitatively measure the emissions from open burning of residential solid waste in burn barrels using techniques that would minimize the limitations of previous studies. The full results of this study are contained in a U.S. Environmental Protection Agency (EPA) report (4).

Formation of PCDDs/PCDFs. Extensive research has been conducted to investigate the formation mechanisms of

PCDDs/PCDFs in the years since PCDDs/PCDFs were first discovered in the exhaust gases from MWCs in 1977 (5). Since then, multiple formation mechanisms have been proposed (6–12).

Field studies on MWCs have shown that the amount of fly ash (and its accompanying metallic catalysts) and organic precursors that pass through the temperature window between 250 and 700 °C as well as the amount of time spent in that optimal temperature window are the primary variables affecting PCDD/PCDF emissions (13). However, field studies have been unable to clearly demonstrate a correlation between chlorine input and PCDD/PCDF emissions in full-scale combustion systems, probably because other variables (PM carryover, PM control device temperature, and combustion efficiency) dominate (14). It is the authors' view that chlorine is present in excess relative to the other reactants in these systems.

On the basis of current PCDD/PCDF formation theories, a "worst-case scenario" for the formation of PCDDs/PCDFs from combustion systems would combine the following features:

- (i) poor gas-phase mixing
- (ii) low combustion temperatures
- (iii) oxygen-starved conditions
- (iv) high PM loading
- (v) PM-bound copper
- (vi) presence of HCl and/or chlorine
- (vii) significant gas-phase residence time in the 250–700 °C temperature range

Open burning is a unique combustion source that appears to fit all of the requirements for such a worst-case scenario for the production of PCDDs/PCDFs. A burn barrel provides poor combustion conditions (providing a source of organic precursors). This paper will show that a burn barrel also provides significant gas-phase residence time in the optimal PCDD/PCDF formation temperature window in the presence of significant amounts of HCl and a plentiful supply of PM.

Focus of Paper. The main focus of this paper will be to examine a subset of the results from the EPA burn barrel tests: the emissions of PCDDs/PCDFs from open burning of household waste in barrels. These data will be presented in an effort to explain the magnitudes of the PCDD/PCDF emissions as well as the reasons for the differences between the PCDD/PCDF emissions measured under the different test conditions. Additionally, a comparison of burn barrel emissions to emissions from various types of MWCs will be presented.

Experimental Section

General. The objective of this project was to qualitatively identify and quantitatively measure the emissions of numerous pollutants from the open burning of household waste in barrels. Analytical work was divided between ARCADIS Geraghty & Miller (ARCADIS) and the New York State (NYS) Department of Health, Wadsworth Center for Laboratories and Research (WCL&R).

Due to the highly variable nature of household waste generation, a reasonable representation of a waste stream for disposal in a burn barrel was prepared based on the typical percentages of various materials characterized and quantified for NYS residents. The characterization was performed by the NYS Department of Environmental Conservation, Division of Solid Waste. On the basis of this detailed characterization, waste-stream scenarios for an "avid recycling" and a "nonrecycling" family of four were simulated for this project. For each scenario, the waste composition was simplified by combining the percentages for similar materials [e.g., the percentages for high-density polyethylene (HDPE), low-density polyethylene (LDPE), and polypropylene (PP) were combined]. For each waste category, items characterized

TABLE 1. Composition of Waste

	nonrecycler (%)	avid recycler (%)
paper		
newspaper, books, and office paper	32.7	3.3
magazines and junk mail	11.1	
corrugated cardboard and kraft paper	7.6	
paperboard, milk cartons, and drink boxes	10.3	61.9
plastic ^a		
PET no. 1	0.6	
HDPE no. 2, LDPE no. 4, and PP no. 5	6.6	10.4
PVC no. 3	0.2	4.5
PS no. 6	0.1	0.3
mixed no. 7	0.1	0.3
food waste	5.7	
textile/leather	3.7	
wood (treated/untreated)	1.1	3.7
glass/ceramics		
bottles/jars	9.7	
ceramics (broken plates and cups)	0.4	6.9
metal, ferrous		
iron, cans	7.3	4.0
non-ferrous		
aluminum (cans, foil)	1.7	1.0
other non-iron (wire, copper pipe, batteries)	1.1	3.7
% total	100	100

^a PET, poly(ethylene terephthalate); HDPE, high-density polyethylene; LDPE, low-density polyethylene; PP, polypropylene; PVC, poly(vinyl chloride); and PS, polystyrene.

as "miscellaneous" were included by adding that percentage to the item(s) within each category that possessed the largest percentage(s). For the plastic category, the miscellaneous percent was relatively large and was divided between the two largest percentages within that category [for the avid recycler it was divided between poly(vinyl chloride) (PVC) and LDPE and for the nonrecycler it was divided between HDPE and LDPE]. Although this simplification was dealt with in a consistent manner, the percentage of PVC for the avid recycler rose from 0.7% to 4.5%, which was greater than expected. Table 1 presents the composition of the simulated waste streams for the two scenarios. The simulated household waste was prepared primarily from raw materials diverted from the household wastes of ARCADIS staff members. Moisture content of the waste was not measured or altered. Household hazardous wastes (e.g., household chemicals, paint, grease, oils, tires, and other vehicle parts) were not included in the waste burned.

Experimental Conditions. Initial charge mass for each test burn was determined by loading the barrel to the same level for each test. Thus, approximately the same volume (uncompacted) of waste was used for each test burn. Since the nonrecycler's waste was less dense than the avid recycler's waste, the test burns for the nonrecycler had a smaller initial waste mass. There were two test burns for each scenario, and these were performed in EPA's Open Burning Test Facility (4). The mass burned ranged from 6.4 to 13.6 kg (14–30 lb) per test. The burn device was a 208-L (55-gal) steel drum, modified for ventilation by placing 24 1.27-cm (0.5-in.) diameter holes evenly spaced around the bottom of the barrel (as informal observations suggest is typically done by homeowners). The barrel was sandblasted prior to use to remove paint in an effort to simulate the use of a weathered, used burn barrel that would represent the most common residential burn device as well as to remove any potential contaminants that would bias emission measurements. The barrel was placed on an electronic scale platform to allow the mass consumed by combustion to be monitored. A

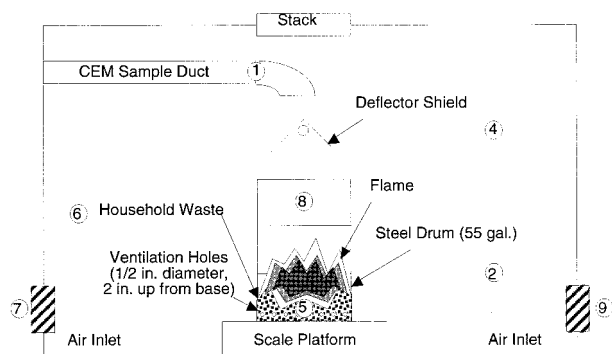


FIGURE 1. Burn hut diagram (numbers represent thermocouple locations).

schematic diagram of the burn hut portion of the Open Burning Test Facility is shown in Figure 1. The burn hut is a small outbuilding 335 cm long (132 in.), 272 cm wide (107 in.), with a peak height of 221 cm (87 in.). High-volume air handlers provided metered dilution air into the burn hut yielding approximately 2.46 air exchanges per minute. Additional fans were set up inside the burn hut to enhance mixing within the hut. Samples for the continuous emission monitors (CEMs) were collected from the sampling duct. Other sampling devices were set up and operated like ambient air samplers inside the burn hut.

Before each test, the material to be combusted was placed in the barrel; air flow through the facility was initiated; temperature measurements with thermocouples were begun; and 15 min of background data was obtained on CEMs for oxygen (O₂), carbon dioxide (CO₂), CO, and total hydrocarbons (THCs). The material to be combusted was then ignited for a short period (<3 min) using a propane torch. Sampling was initiated at least 2 min after the removal of the propane flame. Propane flames generally produce only water, CO₂, and small quantities of low molecular weight products of incomplete combustion. Since these products were expected to have largely dissipated prior to initiation of sampling, this procedure should not have biased the results. This assumption was verified by a hut blank experiment during which the propane torches were lit, but household waste was not burned. Additionally, the hut blank experiment provided information for the assessment of background contaminant concentrations in the ambient air that was pumped through the facility. A single integrated sample was collected over the course of each active burn, and sampling was terminated when the burn mass did not change over an extended period.

Sampling and Analysis. Metals were sampled in accordance with Method 101A (15) modified to be nonisokinetic since sampling was not done from a duct. At WCL&R, the metals were extracted from the particulates, and the extracts were analyzed using inductively coupled plasma mass spectrometry, atomic absorption (AA) spectrophotometry, electrothermal AA spectrophotometry (graphite furnace), and cold vapor AA using EPA standard methods.

HCl was sampled and analyzed in general accordance with EPA Method 26 (16), except that isokinetic sampling procedures were not utilized.

PM with an aerodynamic diameter <2.5 μm (PM_{2.5}) was measured using a dichotomous sampler placed inside the burn hut (17).

PCDDs/PCDFs were sampled using a Graseby PS-1 sampler operated within the burn hut. This train, designed to comply with the EPA's ambient sampling method TO-9 (18), consists of an open-faced filter holder followed by a polyurethane foam (PUF)-sandwiched XAD-2 bed vapor trap. Because this sampler does not have a particulate size separation device, fairly low flow rates can be used. Given

TABLE 2. Run Duration and Burn Mass

test no.	test	start mass (kg)	final mass (kg)	mass burned (kg)	amt burned (%)	duration (min)
1	avid recycler	12.5	4.4	8.1	65.3	77
2	avid recycler	13.6	4.4	9.2	68.1	83
3	hut blank	0.0	0.0	0.0		92
4	nonrecycler	6.4	3.1	3.3	51.6	62
5	nonrecycler	8.8	4.7	4.1	46.6	91

the expected high concentrations of analytes in these tests, this sampler was operated at approximately 28.3 L/min (1 ft³/min) for approximately 1.5 h. The temperature of air entering the train and within the PUF cartridge was assessed during preliminary tests in order to decide if further precautions were necessary to cool the system. Since further precautions were required, a copper cooling coil was fabricated to enclose the exterior of the PUF module. The method of operation of this sampling train differs from method TO-9 in other respects:

(i) Due to constraints of facility size, the sampler location criteria in TO-9 were modified (i.e., the sampler was located inside the burn hut).

(ii) The flow through the sampler was measured by a separate dry gas meter rather than a Venturi and Magnehelic gauge as discussed in TO-9.

(iii) Analysis was performed using high-resolution gas chromatography and low-resolution mass spectrometry (HRGC/LRMS) using EPA Methods 23 and 8280 (19, 20).

(iv) The filter and vapor-phase module were extracted and analyzed together.

These samples were spiked, extracted, and concentrated by ARCADIS. The extracts were shipped on ice to WCL&R for analysis by EPA Method 8280.

Estimated Emissions. Estimated emissions of PCDDs/PCDFs per unit mass consumed by combustion were calculated by assuming thorough mixing of air inside the burn hut and using

$$E = (C_{\text{sample}} Q_{\text{hut}} t_{\text{run}}) / (m_{\text{burned}}) \quad (1)$$

where E is the estimated emissions in milligrams per kilogram (mg/kg) of waste burned, C_{sample} is the concentration of the pollutant in the sample in milligrams per cubic meter of air (mg/m³), Q_{hut} is the flow rate of dilution air into the burn hut in cubic meters per minute (m³/min), t_{run} is the run time in minutes, and m_{burned} is the mass of waste consumed by combustion over the duration of the test burn in kilograms.

Results

CEM and Thermocouple Data. Table 2 lists the burn masses and duration for the four run conditions and the hut blank. Table 3 lists the CEM and thermocouple data from the four runs that burned waste. Thermocouple locations are labeled in Figure 1. Average data are reported as the mean across the entire run duration from ignition until sampling ceased. Note that O₂ and CO₂ data are not reported in Table 3 because they were very nearly at ambient concentrations over the entire run duration. Figure 2 shows the time/temperature history of thermocouple 8, which was mounted near the top opening of the barrel. Note that the temperatures are in the 250–700 °C temperature window for a significant portion of the run time.

Statistical analyses were performed using the JMP software to examine whether the average temperature over the run or the maximum temperature over the run gave a correlation with PCDD/PCDF emissions. No statistically significant correlations were found. It is likely that temperature has an

TABLE 3. CEM and Thermocouple Data Averaged over Run Duration

	run 1, avid recycler	run 2, avid recycler	run 4, nonrecycler	run 5, nonrecycler
THC (ppm)	8.1	6.5	10.4	11.1
CO (ppm)	10.0	7.3	14.1	15.4
temp 1 (°C)	101.1	114.2	78.6	74.4
temp 2 (°C)	58.0	56.6	51.8	50.0
temp 3 (°C)	na ^a	na	74.7	71.0
temp 4 (°C)	69.5	110.8	na	na
temp 5 (°C)	276.1	161.7	317.2	592.2
temp 6 (°C)	64.3	54.7	59.9	52.7
temp 7 (°C)	53.9	53.4	48.4	50.0
temp 8 (°C)	117.7	192.5	107.7	109.7
temp 9 (°C)	44.0	44.5	38.1	40.5

^a na, not available (thermocouple failure).

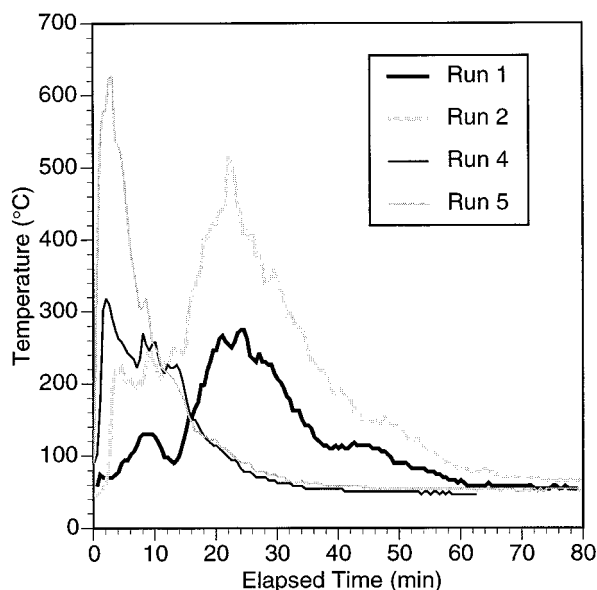


FIGURE 2. Temperature vs. time.

influence on emissions of PCDDs/PCDFs; however, given the transient nature of the experiments and the limited number of data points, no effect was discernible from these data.

PCDD/PCDF Data. Table 4 lists the estimated emissions for PCDDs/PCDFs for each test burn. Where no peak was detected above the detection limit, estimated emissions were calculated based on the method detection limits and reported as “less than” values. For the most part, emissions of PCDDs (but not PCDFs) were below detection limits for the non-recycler, except for OCDD, which was higher than that of the avid recycler. The hut blank sample showed levels of OCDD comparable to the combustion runs, and the nonrecycler runs also exhibited high levels of OCDD while other PCDDs/PCDFs were low. Subsequent informal laboratory audits have suggested the presence of native OCDD in the supposedly isotopically labeled spiking mixture that was used at the APPCD analytical laboratory during that time. For this reason, OCDD data should be treated as suspect; therefore, all discussions are based on the PCDD/PCDF data with the OCDD data excluded. It must be noted that some internal standard recoveries were outside of the generally accepted range between 40 and 120%, so quantitation for some congeners may be questionable, although qualitatively the data are sound. PCDD/PCDF results are corrected for recovery of internal standards, with recoveries ranging

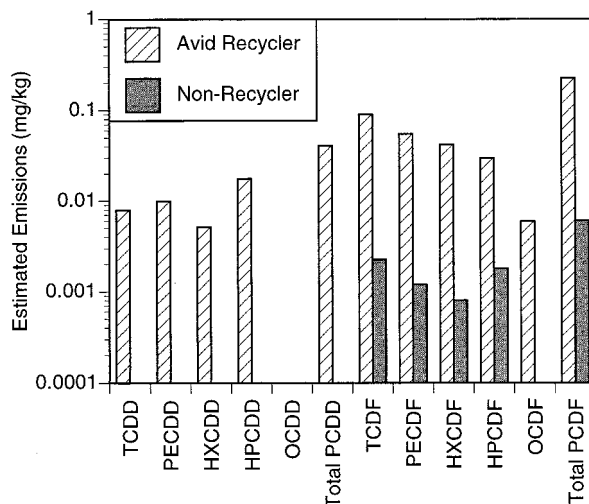


FIGURE 3. Congener distribution of PCDDs/PCDFs. Results are averaged across duplicate run conditions with nondetects set to zero. Note that OCDD data are suspect due to the presence in hut blank samples and are not shown.

between 40 and 120% being desirable. The calculations correcting for standard recovery can lead to quantitation errors when recoveries are outside this range. The complete recovery data can be found in the full report (4).

Figure 3 illustrates the distribution of congener emissions across all runs. PCDFs were higher than PCDDs, which is consistent with results seen from MWCs, hazardous waste incinerators (HWIs), and other combustion sources (14). In general, PCDDs/PCDFs emissions were higher for the avid recycler than for the nonrecycler. Chlorobenzene emissions also exhibited this same general trend (4), although a strong correlation between chlorobenzenes and PCDDs/PCDFs was not seen, although chlorobenzenes are believed to be one of the primary organic precursors leading to formation of PCDDs/PCDFs. The high PCDD/PCDF emissions of run 1 strongly influenced the average value for the avid recycler.

Discussion

Differences between Runs. Household waste is an extremely heterogeneous material; its composition varies greatly among households and also week by week for individual households. These tests were performed on a simulated waste stream intended to represent two types of households, with two samples of each household’s waste. This prevented statistical examination of relationships between waste composition, combustion conditions, and pollutants that were emitted. However, it is still possible to gain an understanding of potentially important factors that contribute to the emissions of PCDDs/PCDFs from open burning of garbage.

There are several possible explanations for the observation that the avid recycler’s emissions of PCDDs/PCDFs were higher than those of the nonrecycler. As pointed out above, the procedure for preparing waste samples yielded a PVC content of 4.5% for the avid recycler’s waste versus 0.2% for the nonrecycler. The higher proportion of PVC plastic in the avid recycler’s waste stream could potentially increase formation of chlorinated organic compounds. Combustion conditions such as time/temperature history, mixing patterns, and oxygen availability as well as the particular mixture of carbon molecules and chlorine in the presence of a metal catalyst are all important factors in the formation of PCDDs/PCDFs. All of these variables potentially changed between the avid recycler and the nonrecycler test cases.

Another possible explanation for the differences in emissions between run conditions and between runs at the

TABLE 4. Estimated Emissions of PCDDs and PCDFs (mg/kg)

isomer	compound	avid recycler		nonrecycler	
		run 1	run 2	run 4	run 5
2378	TCDD	<0.0009	<0.0005	<0.0003	<0.0003
12378	PECDD	0.0013	<0.0005	<0.0003	<0.0003
123478	HXCDD	0.0002	<0.0001	<0.0004	<0.0003
123678	HXCDD	0.0014	<0.0006	<0.0006	<0.0005
123789	HXCDD	0.0008	<0.0006	<0.0005	<0.0004
1234678	HPCDD	0.0153	0.0008	<0.0006	<0.0005
12346789	OCDD ^a	0.0115	0.0005	0.0448	0.0317
2378	TCDF	0.0022	0.0002	0.0001	<0.0003
12378	PECDF	0.0035	<0.0004	0.0001	<0.0002
23478	PECDF	0.0013	0.0002	0.0002	<0.0003
123478	HXCDF	0.0012	0.0001	0.0001	0.0001
123678	HXCDF	0.0067	0.0006	0.0002	<0.0003
234678	HXCDF	0.0094	0.0009	0.0001	<0.0003
123789	HXCDF	0.0024	0.0003	<0.0004	<0.0003
1234678	HPCDF	0.0439	0.0015	0.0002	0.0034
1234789	HPCDF	0.0004	<0.0003	<0.0005	<0.0003
12346789	OCDF	0.0114	0.0005	<0.0007	<0.0006
total	TCDD	0.0141	0.0018	<0.0003	<0.0003
total	PECDD	0.0191	0.0008	<0.0003	<0.0003
total	HXCDD	0.0099	0.0004	<0.0006	<0.0005
total	HPCDD	0.0338	0.0015	<0.0006	<0.0005
total	OCDD ^a	0.0115	0.0005	0.0448	0.0317
total	TCDF	0.158	0.0224	0.0038	0.0007
total	PECDF	0.0995	0.0106	0.0024	<0.0002
total	HXCDF	0.0781	0.0056	0.0011	0.0005
total	HPCDF	0.0576	0.0021	0.0002	0.0034
total	OCDF	0.0114	0.0005	<0.0007	<0.0006
total	PCDD	0.0884	0.005	0.0448	0.0317
total	PCDF	0.4046	0.0412	0.0075	0.0046
total	PCDD/PCDF ^b	0.4815	0.0457	0.0075	0.0046
total	TEQ ^c	0.0054	0.00123	0.000759	0.000903

^a OCDD results are suspect due to presence of elevated levels in the blanks. ^b Without OCDD results included. ^c TEQ, toxic equivalency (25). TEQ contributions from non-detects were calculated at the detection limit and OCDDs were neglected.

TABLE 5. Estimated Emissions of PCDDs/PCDFs, Copper, Hydrogen Chloride, Chlorobenzenes (CBs), Chlorophenols (CPs), and PM_{2.5} (mg/kg)

run	PCDD ^a	PCDF	total PCDD + PCDF	Cu	HCl	CBs	CPs	PM _{2.5}
run 1, avid recycler	0.0769	0.4046	0.4815	15.01	3280	0.287	0.576	6930
run 2, avid recycler	0.0045	0.0412	0.0457	6.18	1510	1.73	1.79	3580
run 4, nonrecycler	nd ^b	0.0075	0.0075	2.16	481	0.416	0.413	20070
run 5, nonrecycler	nd	0.0046	0.0046	0.573	86.4	0.432	1.5	14800

^a OCDD measurements are not included in total of PCDDs. ^b nd, none detected.

same condition may be a cause due to the heterogeneous nature of the household waste itself. Despite the efforts to create an artificial waste stream that simulated “real” household waste, duplicated for each run condition, there undoubtedly were slight differences between the batches used for the runs, such as bleaching agents, coatings, inks, dyes, or foreign matter, or how the material was stacked in the barrel. One possible explanation can be inferred from examining the metals and HCl emissions.

Effect of Exhaust Gas Constituents. On the basis of current PCDD/PCDF formation theory, it would be expected that exhaust gas constituents such as PM, copper, or HCl might affect PCDD/PCDF emissions. Table 5 lists the total PCDD + PCDF emissions (excluding the suspect OCDD data), copper emissions, HCl emissions, and emissions of PM_{2.5}. Although PM with an aerodynamic diameter less than 15 μm was also measured, greater than 85% of the PM was less than 2.5 μm in diameter. PM_{2.5} emissions did not correlate with PCDD/PCDF emissions. It may be that there was an excess of PM available in all run conditions, with the availability of solid-phase surface area not being a limiting factor. Other measured metallic species as well as chlorobenzenes and

chlorophenols were also compared to emissions of PCDDs/PCDFs, and no correlations were found.

Note that run 1 exhibited higher copper and HCl emissions than the other runs. Specifically, the emissions of copper, HCl, and PCDD/PCDF for run 1 were higher than those for run 2 by factors of approximately 2, 2, and 10, respectively. These observations provide a possible explanation for the higher PCDD/PCDF emissions observed during run 1 and are consistent with current theories of heterogeneous dioxin formation.

Figure 4 shows emissions of PCDDs/PCDFs as a function of copper and HCl emissions. These data suggest that increases in emissions of either copper or HCl may be associated with increases in PCDD/PCDF emissions. The chlorine- and copper-containing fractions of the waste may have been distributed in the waste mixture in a manner that introduced these elements into the combustion zone of the burn barrel, thereby making copper and HCl available as reactants for PCDD/PCDF formation. Such observations are consistent with currently accepted theories of heterogeneous PCDD/PCDF formation from combustion of municipal waste. More conclusive statements about the relationship between

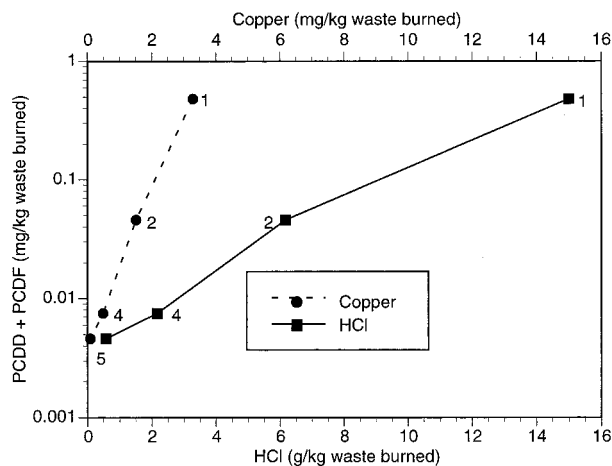


FIGURE 4. PCDD + PCDF vs airborne copper and HCl emissions. Numbers next to data points correspond to run numbers.

PCDD/PCDF emissions and copper or HCl emissions are not possible since the limited number of data points precluded rigorous statistical analyses.

Past studies on MWCs and HWIs have not been able to prove a causal relationship between chlorine input (and subsequently HCl emissions) and PCDD/PCDF emissions (14), because other more important parameters (e.g., flue gas cleaning temperature, particulate matter carryover, effectiveness of combustion) completely masked any contribution from chlorine input rates. To heterogeneously form PCDDs/PCDFs, a chlorine source, a carbon source, and a solid-phase metal catalyst must be present. It may be that, for MWC or HWI facilities, chlorine is present in excess even at very low concentrations, while in an open-burning scenario, the known precursor organics and/or metal catalysts may be present in excess.

Comparison to Other Burn Barrel Study Results. In general, the PCDD/PCDF estimated emissions presented in this paper are higher than the estimated emissions presented in the Minnesota and Illinois studies but not unreasonably higher given the variance between test/fuel conditions observed in duplicate test burns in this study. The Minnesota

burn barrel study reported 4.0×10^{-6} mg of 2,3,7,8-TCDD/kg of garbage fed into the barrel. The 2,3,7,8-TCDD emission estimate presented in this paper was based on the method detection limit and was estimated to be $<5 \times 10^{-4}$ mg/kg of trash consumed by combustion. The Illinois study reports 6.2×10^{-4} mg of total PCDD/PCDF/kg of refuse fed into the barrel (average of three test burns). These results can be directly compared to results presented in this paper by adjusting for the percent of mass reduced during each test burn. The adjusted average emissions are 1.6×10^{-3} mg of total PCDDs/PCDFs/kg of refuse consumed by combustion (average of three test burns). These Illinois data compare well with the PCDD/PCDF emissions factors of 7.6×10^{-4} – 4.6×10^{-3} mg of total PCDD/PCDF (excluding OCDD)/kg of refuse consumed by combustion presented in this paper for the nonrecycler scenario.

Comparison with Emissions from Incineration. To place burn barrel emissions in perspective, estimated emissions for PCDDs/PCDFs from burn barrels were compared to average emission factors for three types of incinerators [mass burn, modular, and refuse-derived fuel (RDF) combustors] (21) in Figure 5. Although, most of these selected MWCs possess various types of air pollution control devices [electrostatic precipitators (ESP), spray dryers, and fabric filters], an emissions factor for source test data measured at the inlet to the air pollution control device, essentially uncontrolled, also is presented (21).

The MWC emission factors used for this comparison were developed from a compilation of data published on 107 separate test reports (21). These data are averages and are not representative of any particular facility (21) and are based on the amount of mass fed into the incinerator. A quality rating was assigned to each emission factor presented in the EPA report (21). Specific information on the bases for each rating for each MWC is in the EPA report (21). In general, the MWC emission factor data presented in Figure 5 were assigned high to average ratings (21). The emission factor rating for the RDF combustor possessing a spray dryer and fabric filter was assigned a poor rating (21).

Figure 5 shows that burn barrel PCDD/PCDF emissions are generally much higher than for MWCs that possess air pollution control equipment. Additionally, this figure shows

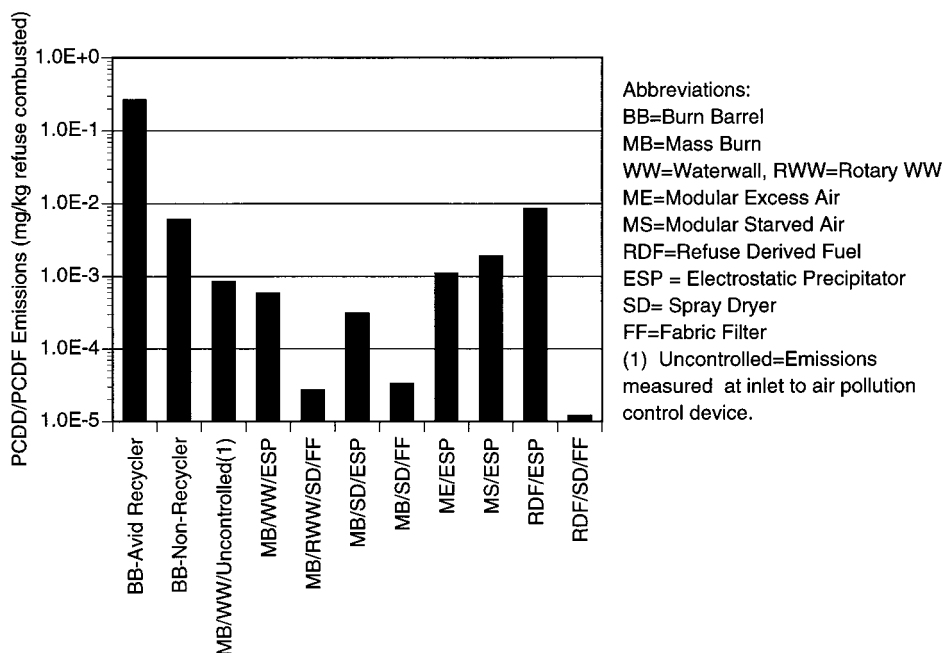


FIGURE 5. Comparison of total dioxin and furan emissions for burn barrels and municipal waste combustors (source of MWC data: *Locating and Estimating Air Emissions from Sources of Dioxins and Furans*; U.S. EPA: Washington, DC, May 1997; EPA-454/R-97-003).

TABLE 6. Comparison between Open Burning of Household Waste and Controlled Combustion of Municipal Waste in a Municipal Waste Combustor (22)^a

	avid recycler	non recycler	MWC
PCDD ($\mu\text{g}/\text{kg}$ of waste burned)	40.7	nd ^b	0.0016
PCDF ($\mu\text{g}/\text{kg}$ of waste burned)	222.9	6.05	0.0019
PCDD + PCDF ($\mu\text{g}/\text{kg}$ of waste burned)	263.6	6.05	0.0035

^a Note that OCDD data were not included in the open-burning results. ^b nd, none detected.

TABLE 7. Number of Open-Burning Households to Equal the Total PCDD/PCDF Emissions (mg/day) from a Full-Scale MWC Facility^{a,b}

	avid recycler	nonrecycler
PCDD + PCDF ^c	2.5	37

^a Using refuse generation rate supplied by NYSDEC; MWC burns 182 000 kg/day (200 ton/day). ^b Emissions data for a modern, clean-operating MWC with good combustion and flue gas cleaning technology. ^c OCDD measurements not included in total of PCDDs.

that burn barrel PCDD/PCDF emissions are higher than for the mass burn MWC with uncontrolled emissions (i.e., emissions measured prior to the flue gas cleaning system).

The emissions for burn barrels were also compared to a modern, clean-operating MWC with good combustion and flue gas cleaning technology (22). On the basis of data from a field test at a RDF MWC (22) and averaging the "Normal Good" performance tests (PT-08, PT-09, and PT-11) conditions, using the samples taken at the pollution control device (a spray dryer and fabric filter) outlet, the comparison in Table 6 was generated. Note that the PCDD/PCDF emission factor for this particular RDF facility is approximately 3.5 times lower than the average emission factor for this type of facility presented in Figure 5. Additionally, this emission factor was based on the mass of refuse fed into the incinerator; an adjustment to account for the amount of mass burned for each performance test was not possible. Such an adjustment would raise the emission factor by an undetermined amount.

This particular MWC study (22) was used for comparison in the EPA's burn barrel study report because it contained a large set of target compounds over a wide range of operating conditions. Additionally, target compounds from this full-scale MWC study included not only PCDDs/PCDFs but also many of the other compounds (e.g., PCBs, PAHs, etc.) that were measured in the EPA burn barrel study.

From these comparisons, it is readily apparent that even the differences between the avid recycler and nonrecycler emissions are minor in comparison to the difference between open burning of household waste and the controlled combustion of municipal waste at most kinds of dedicated MWC facilities. The emissions from open burning are several orders of magnitude higher than for controlled combustion in a modern, clean-operating MWC.

As an additional comparison of a burn barrel and a well-controlled MWC, an estimate of the number of open-burning households that emit PCDDs/PCDFs in amounts comparable to a hypothetical RDF incinerator was derived. Table 7 was generated by calculating the total mass of PCDDs/PCDFs emitted per day using the estimated emissions per unit mass burned from Table 6, the waste generation rates derived from the NYS analysis (4.9 and 1.5 kg/day for a nonrecycling and an avid recycling family of four, respectively), assuming a household burns their waste daily, and comparing those values to a hypothetical 182 000 kg/day (200 ton/day)

modern, well-controlled RDF incinerator emitting PCDDs/PCDFs at the rate described in Table 6. To perform this comparison and to be consistent with the MWC emission rate, the burn barrel estimated emissions were adjusted to account for the refuse fed into the barrel. It should be noted that this size MWC facility processes the equivalent waste from 37 000 nonrecycling and 121 000 avid recycling households (based on the above waste generation rates). By dividing the daily estimated emissions from a MWC by the daily estimated emissions from open burning, it is possible to estimate how many open-burning households it would take to equal the amount of PCDDs/PCDFs emitted by this moderately sized MWC facility. The number is surprisingly low, with estimates ranging between 2.5 and 37 households burning their trash in barrels producing PCDDs/PCDFs comparable to a well-operated full-scale RDF MWC facility serving thousands of households.

Significance of Study and Future Work. This particular source could potentially be significant in the overall national PCDD/PCDF budget (23). The EPA 1994 Draft Dioxin Reassessment document (24) attempted to conduct a mass balance for dioxin emissions in the United States and identified a significant gap between current deposition estimates and emission estimates. The deposition estimates were considerably higher than the emissions estimates. The EPA speculated that this indicated that there were unknown dioxin emission sources. Given the prevalence of open burning in barrels (the Illinois study included a survey that indicated that 40–50% of rural Illinois residents burn at least a portion of their household waste in barrels), it is very possible that the PCDD/PCDF emissions from burn barrels may be an important missing link to help close the gap between measured deposition rates and the emissions inventories.

Based on the complex set of variables that affect emissions of pollutants from burn barrels, further study would be warranted. The study reported in this paper consisted of a limited number of tests measuring a wide range of pollutants. Further studies should concentrate on examining the emissions of PCDDs/PCDFs. The effect of waste composition, chlorine content, chlorine type (organic vs inorganic), heating value, bulk density, and their subsequent effect on resulting combustion conditions (e.g., temperature) and emissions of pollutants would be primary variables to investigate. Additionally, the types of wastes that are typically disposed of via open burning in a burn barrel and the frequency of burning should be investigated.

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