Dioxin Emission Inventories: The Importance of Large Sources

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1. Introduction

The citizens of Columbus, Ohio were shocked to learn in 1993 that PCDD/PCDF measurements at the local 2000 ton/day trash incinerator extrapolated to an annual emission rate of nearly 1 kg/yr of TEQ. So were we. This quantity was greater than an estimate of atmospheric emissions for the whole of Germany, including all incinerators, traffic and industry.(1) It was also five times higher than what the USEPA had estimated with "high a degree of confidence" for all--approximately 160--US trash incinerators: 60-200 g/yr of TEQ.(2) This discrepancy probably increased the urgency of locating other such large sources. In February 1994, the USEPA began trying to identify other incinerators with high emissions, focusing on solid waste incinerators that, like Columbus, employed so-called "hot-sided" ESPs.(3)

Single large sources can have a dramatic impact on emissions inventories. Such estimates, discussed in this paper for US municipal solid waste incinerators, are typically constructed by averages or "typical values" estimated from non-random samples of the population of facilities. We argue for more careful attention to individual incinerators and for specification of the time period for which inventories are constructed. Large sources also have consequences for "mass balance" calculations that lead some to hypothesize significant missing sources.

2. Methods

We obtained stack measurements from a survey of incinerators that USEPA considered potentially large sources.(3) Measurements were in the form of PCDD/PCDF concentration per dscm corrected to 7% O_2. We calculated yearly emissions estimates assuming that stack measurements were representative of the emissions from that facility at that time. We averaged across similarly-equipped units within a facility; these values are assumed to apply to similarly-equipped, untested units. We present stack measurements prior to corrective measures (negotiated in many cases by USEPA).

Facility-specific information was available for the Columbus incinerator.(4,5) While preferred, such data are not generally available. In most cases we further assumed: flue gas flow rates of 3670 dscm at 7% O_2 per ton of unprocessed municipal solid waste; increased gas flow of 20% for refuse derived fuel facilities to adjust for higher heat content of the fuel; an average 43:1 conversion ratio from PCDD/PCDF to TEQ (calculated from USEPA(6)). We obtained average capacity utilization rates for many facilities from a 1991 report.(7) For the remainder, we assumed a 87.5% capacity utilization rate.(8)

Our flue gas flow rate for unprocessed municipal solid waste is based on that originally used by USEPA(8) and subsequently by Cohen et al.(9) But USEPA appears not to have corrected to 7% O_2. As a result, we increased flow rates by 50%. Our PCDD/F to TEQ conversion ratio, 43:1, is somewhat lower than the 60:1 used by USEPA(8), but higher than Jones' 30:1.(10)

We collected estimates of total PCDD and PCDF air emissions by U.S. municipal solid waste incinerators for the late 1980s and early 1990s.

3. Results and Discussion

Table 1 shows our estimates of emissions from ten U.S. incinerators. All measurements are from the time period 1992 to about mid-1994. Given the assumptions noted in the methods section, we believe that these are reasonable estimates for these facilities for this time period. Several other incinerators could reasonably be added to this list of high emitters. We omit test results from Montgomery Co.(Dayton) South as the temperature of the ESP was experimentally manipulated. No measurements were available from the Akron, Ohio facility from this period; it was however, a 1000 tpd RDF incinerator with a hot ESP. These could add substantially to the total.

The sum of the emissions from these ten incinerators is 3.6 kg/yr of TEQ. The sum for all U.S. incinerators should be at least this big for the same time period. 3.6 kg/yr may well be a minimum for a number of years before 1993 as well. We are currently doing more historical research on this question.

All facilities listed in Table 1 have since closed or retrofitted to reduce emissions, or are scheduled to do so.[3] Some facilities retested after retrofit had lower stack concentrations. Emissions estimates for times after USEPA intervention must carefully specify the period they cover.

Table 2 lists a number of estimates of total PCDD and PCDF air emissions (expressed as TEQ) from all U.S. municipal solid waste incinerators combined. We omit the study by Commoner et al.[17] for the year 1974 and the estimate by Travis and Hattemer-Frey[18]; the latter appears to only include 2,3,7,8-TCDD.

Comparisons with our estimate are most apt for the year 1993 and several years before. Comparisons with studies for 1994 and after are less straightforward due to the impact of USEPA's intervention. **We estimate that the ten incinerators emitted 3.6 kg/yr of TEQ circa 1993. This value exceeds the central estimates of total emissions from all 170-180 U.S. trash incinerators as computed by every study but one.** The one exception is USEPA 's 1987 report.[11]

All of the studies appear to have used the same basic method. Divide the population of incinerators into categories. Based on measurements or engineering judgment, decide on a "typical" stack concentration for each subgroup. Multiply the stack concentration by the total annual throughput, making various assumptions similar to those outlined in our methods. Several studies do not present sufficient information regarding methodology to allow much analysis: Schaum et al.[2,14]; Rigo[13,16]; IWSA [15]. Table 3 aggregates results from the other five studies into comparable categories. USEPA[8], Cohen et al.[9] and Thomas and Spiro[12] are reasonably similar on this level of aggregation. Jones[10] has a much lower total that is skewed towards modular incinerators instead of RDF and mass burn. For some unexplained reason, Jones appears to have omitted RDF and mass burn facilities equipped only with ESPs, large sources in the other analyses.

The two most detailed inventories are the USEPA's dioxin reassessment study[8] and Cohen et al.[9] Cohen et al. were primarily interested in the geographic distribution of sources and long-distance transport of pollutants. They used USEPA's emission factors, but employed a more detailed incinerator categorization scheme, somewhat different throughput values, and a slightly lower PCDD/PCDF:TEQ ratio. As the results are qualitatively similar, we will focus on USEPA.[8]

Why is our minimum for ten incinerators higher than USEPA's best estimate for 171? First, as noted earlier, USEPA employed a low value for the amount of flue gas per ton of waste burned: our value increases emissions by 50% for mass burn facilities, 80% for RDF. Second, our PCDD/PCDF to TEQ conversion factor further increases emissions.

Third, USEPA used a very small number of emissions tests to derive emission factors. Small samples can distort results. They estimated that 96% of total incinerator emissions came from refractory mass burn and RDF incinerators equipped with ESPs (Table 3). USEPA's estimate for ESP-equipped refractory facilities, 0.7 ug-TEQ/kg, was based on tests from one plant. Their estimate for ESP-equipped RDF burners, 0.15 ug-TEQ/kg was based on tests from two plants. Sensibly, these two categories were among those highly represented in the Agency's search for high emitters. USEPA's emission factor is much lower than two of the RDF plants listed in Table 1. On the other hand, their emission factor is somewhat bigger than four of five refractory incinerators listed in Table 1.

Fourth, the essence of the emission factor idea is that all facilities within a category have the about same level of emissions, allowing for random variation. Investigators typically categorize by air pollution control device and type of incinerator (e.g., mass burn vs. RDF). USEPA now realizes that ESP temperature is also important. What is the correct level of categorization? We argue that emissions from individual facilities should be directly used in source inventories. Average or "typical" emission factors should only be used for plants that have not been tested. Data on long term-emissions from particular incinerators are virtually non-existent. There is little evidence that stack concentrations deemed "typical" of a category of incinerators are really a better estimate of the emissions from a given facility than its actual measurements. Furthermore, many pieces of information are used to compute an emissions estimate: stack concentrations of PCDD/F or TEQ, flue gas flow rate, amount of material burned per day, capacity utilization. A number of these may covary, leading to error when averages are used. Individuals plants have their own quirks. For instance, the Columbus incinerator has a high flue gas flow rate even for an RDF plant.

There is a further argument for the facility-based approach. Investigators constructing emission factors may have a tendency to omit "outliers," especially high test results not deemed "typical." One danger of this is that the alleged outlier is real, not a fluke. Hot-sided ESPs are an illustration. Large emitters can have dramatic impacts on inventories. Table 1 suggests that three RDF plants have a yearly output of TEQ (2700 g) greater than that estimated by USEPA[8] for all RDF facilities (720 g). Pinellas, a mass burn water wall facility in Table 1, appears to have an annual emission of TEQ (290 g) greater than that calculated by USEPA for all such plants (74 g). **The sum should not be less than the parts**. It is possible that some of our assumptions are incorrect. One might, for instance, argue that the test results we used are not "representative" for a facility. Such debates only argue for closer attention to individual incinerators.

Emission inventories should specify the time period to which they apply. A July 1994 study by Rigo and Rigo[13] for the Columbus Health Department estimated total TEQ emissions from US trash incinerators at 850 g/yr. The report discusses neither the results of the 1992 tests for the Columbus incinerator nor the subsequent March 1994 tests. As our Table 1 indicates, the 1992 tests of this one incinerator extrapolate to a total larger than that estimated by Rigo for all incinerators combined. The 1994 test results are lower, extrapolating to between 100 and 300 g/yr (We do not have the test results and cannot verify the number. These tests are controversial due to some evidence that trash was specially selected to try to lower dioxin emissions[19]). While Rigo[13] states that the 850 g/yr estimate describes "today" (i.e., presumably mid 1994), there is insufficient methodological detail in the report to determine if or how the specific test results for Columbus were handled.

Many of the studies in Table 2 compare estimated air emissions in the U.S. with deposition. This has led some authors to hypothesize large unknown sources. But some mass balance calculations rely on a very small number of deposition measurements, principally data from two Indiana locations measured prior to 1992[20] Given the paucity of these data, care is need in nationwide mass balances, a point noted by USEPA[8] and others.[21] We would like to add to these cautions. First, we believe that timing should also be considered. It is probably not appropriate to compare deposition in 1991--presumably reflecting emissions in 1991 or before--with inventories for 1994 and after when a number of large sources were shut down or modified. Such comparisons will tend to exaggerate the "unknown" source fraction. Second, few large sources are needed to alter emissions estimates. Third, stack emissions are often collected under ideal circumstances. Actual emissions can be larger for a number of reasons including seasonal variation, upset conditions, start-up, shut-down and periods of soot blow off. An increasing number of trash incinerators facilities are now also burning medical, pharmaceutical and industrial wastes. The use of ranges of emission estimates, as USEPA and others have done, is imperative to deal with such uncertainties.

4. Conclusion

We estimate that ten U.S. trash incinerators emitted 3.6 kg of TEQ per year circa 1993. This is greater than the central estimates for all 170-180 U.S. trash incinerators computed by several investigators for this time period. We recommend the establishment of a publicly accessible repository of test results to allow better emissions inventories. It's about time. After all, we have known about PCDD/PCDF emissions from trash incinerators since the late 1970s.

Table 1. Emission Estimates for Incinerators Equipped with "Hot-Sided" ESPs

facility	type	tpd	average capacity	PCDD/PCDF (ng/dscm)	TEQ (ug/kg)	TEQ (g/yr)
Norfolk	RDF	2000	.70	32000	3.6	1700
Columbus	RDF	2000	.70	13000	2.1	980**
Pinellas	MB-WW	3000	.90	3400	.32	290
Pulaski	MB-REF	1500	.875	5530	.52	230
McKay Bay	MB-REF	1000	.844	3860	.36	100
Waipahu	MB-REF	600	.875	5690	.53	90
Detroit	RDF	1100	.725	2850	.32	85
Dayton-North	MB-RC-REF	300*	.85	8100	.76	60
Clinton	MB-REF	600*	.875	3250	.31	50
Harrisonburg	MB-MOD	100	.65	8460	.79	20
Total						3600

* only one of three units is included

** TEQ value estimated from 1992 test results[4,5]

Key: MB = mass burn; RDF=refuse-derived fuel; REF=refractory; MOD=modular; WW=Water wall; RC=rotary combustor

Note: USEPA[3] lists Harrisonburg and McKay Bay as MB-WW. We follow Berenyi & Gould[7]

Table 2. Estimates of PCDD/PCDF Air Emissions by all U.S. MSW Incinerators Combined (expressed as TEQ)

Year for estimate	TEQ (g/yr)	Reference
1987?	24000	USEPA[11,22]
1989	3000	Thomas & Spiro[12]
1990	379.8	Jones[10]
1993?	60-200	Schaum et al.[2]
1993?	3000 (1300-6700)	USEPA 1994[8]
1993	1900 (1700-14000)	Cohen et al.[9]
1994	850	Rigo[13]
1994?	1800-9000	Schaum et al.[14]
1994	2000	IWSA[15]
1995	1030	Rigo[16]

Table 3. Comparison of Emissions Inventories by Incinerator Type (kg TEQ/yr)

type	USEPA(11)	Thomas** & Spiro(12)	Jones(10)	USEPA(8)	Cohen(9)
Mass burn	16.7	2.3	.02	2.3 [2.2]*	1.3 [1.2]*
RDF	7.1	1.1	.001	.72 [.72]*	.52 [.51]*
Modular	.1	.07	.36	.03	.04

* Estimate for facilities equipped with only ESPs.

** Conversion from PCDD/PCDF to TEQ using their ratio of 60:1.(12,21)

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