Summary of PCDD/PCDF Emissions in the United States: History and Relationship to Chlorine in Combusted Material

Valerie Thomas, Center for Energy and Environmental Studies;
Thomas Spiro, Department of Chemistry; Princeton University, Princeton, NJ 08544, USA

Based on emissions data from combustion and chemical sources of tetra- through octa-chlorinated dioxins and furans (PCDD/PCDFs), we have estimated total PCDD/PCDF air emissions in the United States. Emissions from the major sources are shown in Table 1. In order to compare the estimated emissions with PCDD/PCDF found in soils, PCDD/PCDF emissions which are broadly dispersed to soil, such as in pesticides, are also included. Total annual emissions of PCDD/PCDF in the US are estimated to be about 400 kilograms (7000 grams I-TEQ), as of 1989. In agreement with a number of previous studies, municipal waste incineration is estimated to be the largest source category for PCDD/PCDF emissions.[1),2),3),4)] The next largest source categories include hospital waste incineration, forest fires, and residential wood combustion. The full report also includes estimates of PCDD/PCDF emissions, from apartment incinerators, tire incineration, carbon regeneration, and other sources.[5)]

The emission factor for each source category is the average of PCDD/PCDF emissions data from individual sources. Emission factors within a category can vary significantly, and because the data is limited, the average emission factors given in Table 1 should be interpreted only on an order of magnitude basis. For most source categories, the measured emission factors from different source categories are indeed within the same order of magnitude. An important exception is municipal waste incineration. Because emission factors for municipal waste incineration can and do vary by more than an order of magnitude, depending on the combustion technology and operating conditions, emission factors were estimated for each major type of incinerator technology; the weighted average of these emission factors is given in Table 1.[6)] Table 1 also shows the number of sources tested in each category; those estimates based on measurements from only one or two sources (e.g. copper recycling) should be viewed as especially uncertain.

Figure 1 is a plot of the averages of the emission factor data for each combustion source category, versus the typical chlorine content of the combusted material. It shows that for poorly controlled combustion conditions, PCDD/PCDF emissions tend to increase with chlorine concentration. This trend indicates that the estimated emission factors given in Table 1 present a coherent picture of dioxin emissions.

Of particular interest are PCDD/PCDF emissions from forest fires and agricultural burning, for which there are no direct measurements. The estimated emission factor is based on data from poorly controlled biomass combustion.[7),8),9),10)]. In contrast, the

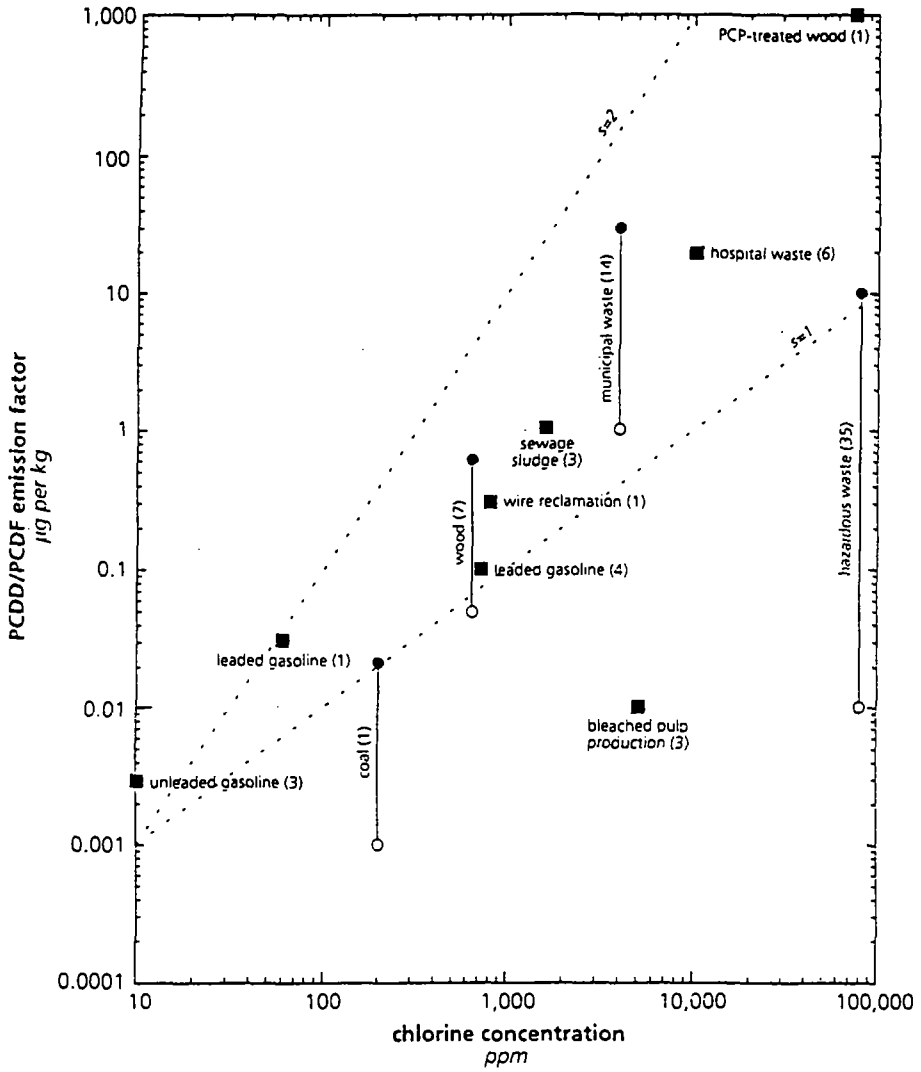

Figure 1: Average PCDD/PCDF emission factors for combustion processes, versus the average chlorine concentration of the combusted material (1989). Where information is available, poorly controlled combustion is indicated by a black dot and well-controlled combustion, connected by a line, is indicated by a white dot. In all other cases, the black square indicates the average emission factor. The lines s=1 and s=2 have slopes of 1 and 2, respectively.

Table 1: Order of Magnitude Emissions of PCDD/PCDF in the US (1989)
(Asterisks indicate estimates based on emissions from similar sources.)

Source Category (Numbers indicate number of facilities tested)	Emission Factor µg/kg feed	Material Consumed 10^9 kg /yr	PCDD/F Emissions kg/yr	PCDD/F Emissions TEQ g/yr
Consumer Waste Combustion				
Municipal Waste Incineration (14)	10	20	200	3000
Hospital Waste Incineration (6)	20	2	40	700
Sewage Sludge Incinerators (3)	1	3	4	70
Industrial Waste Combustion				
Hazardous Waste Incineration (35)	3	4	~ 10	200
Copper Recycling (2)	20	0.7	~ 10	200
Steel Recycling (2)	0.1	30	3	50
Bleached Pulp Production (3)	0.01	30	0.4	7
Biomass Combustion, etc.				
Forest and Agricultural Burning*	0.4	80	30	500
Residential Wood Burning *	0.4	50	20	300
Industrial Wood Combustion (8)	0.06	60	4	70
Structural Fires*	0.4	5	2	30
PCP-Treated Wood Combustion (1)	1000	10^{-3}	1	20
PCB Fires*	1000	7×10^{-4}	0.7	10
Fossil Fuel Combustion				
Oil Combustion (except gasoline)*	0.003	500	1	20
Leaded Gasoline (5)	0.03	30	1	20
Unleaded Gasoline (3)	0.003	300	0.8	10
Coal Combustion (1)	0.001	600	0.6	10
Dioxin-Contaminated Chemicals				
PCP Wood Preservative (to air only) (1)	2,000,000	—	20	200
2,4-D Herbicide (1)	200	2×10^{-2}	4	70
Tetra-chloroethylene (4)	10	0.3	3	50
Total	0.2	2000	400	7000

emission factor for industrial wood combustion is based on data from biomass combustion with pollution control and/or from industrial settings.[7),11),12),13)].

Using historical combustion data, it is also possible to roughly estimate past PCDD/PCDF emissions. Table 2 shows estimates of US PCDD/PCDF emissions in 1940, 1970, and 1989. The emission factor for municipal waste incineration in 1970 is based on emissions estimates from 1970s-vintage municipal waste incinerators (30 µg/kg), and on the highest measured emissions from municipal waste incineration (60 µg/kg). The emission factor for municipal waste incineration in 1940 is bounded below by the emission factor for poorly controlled biomass combustion (0.4 µg/kg) and bounded above by the highest emission factor for municipal waste combustion (60 µg/kg). The total PCDD/PCDF dispersed to soil is dominated by pentachlorophenol (PCP), which in 1970 was being used as a pesticide; the pesticide 2,4,5-T also contributed significantly.

These estimates can be tested against measured PCDD/PCDF concentrations in air, soil, and sediments. For air concentrations of most common pollutants, including SO_2, NOx, benzene, lead, and total suspended particulates, the ratio of total annual US

emissions to average US urban air concentrations is on the order 10^{17} m^3/yr, with a range of (0.5 - 11) x 10^{17} m^3/yr.[14),15)] On this basis, for total PCDD/PCDF emissions of 400 kg/yr, typical urban air concentrations of PCDD/PCDF can be expected to be on the order of 0.4 - 8 pg/m^3, which is in good agreement with the available data.[16)]

Table 2: Estimated PCDD/PCDF Emissions to Air and Soil, 1940 to 1989. [17)]

Source	Emission Factors Total PCDD/F, µg/kg			Activity Rate 10^{10} kg/yr			Total PCDD/F Emissions kg/yr		
	1940	1970	1989	'40	'70	'89	1940	1970	1989
MSW Combustion	0.4-60	30-60	10	1.5	2	2	6-900	600-1200	200
Agricultural Fires	0.4	0.4	0.4	2	2	2	8	8	8
Forest Fires	0.4	0.4	0.4	20	6	6	80	20	20
Wood Combustion	0.4	0.4	0.2	9	9	10	40	40	20
Coal Combustion	0.02	0.02	0.001	40	40	60	8	8	0.6
Chemicals to Air							0	200	20
Other							0	100	100
Total to Air							140 - 1000	1000 - 1600	400
Pesticides to Soil							0	1000	0
Total to Air & Soil							140 - 1000	2000 - 2600	400

The expected concentration of PCDD/F in soil, C, can be calculated from the estimated annual emissions, En, using the following expression:

$$C = \sum \frac{E_n}{V} \exp(- n \ln 2/\tau)$$

where τ is the half-life, V is the effective soil volume (given by the product of the mixing depth and the area of the US), and the sum is over the number of years, n, since deposition of the PCDD/F. The effective half-life of PCDD/PCDF in soil is not well known, but here will be taken to be roughly 20 years.[18),19)] For a 2.5 cm mixing depth (corresponding to the typical soil sampling depth), the estimates given in Table 2 indicate average US soil concentrations of PCDD/PCDFs of about 100 pg/cm^3. Average soil concentrations of PCDD/PCDFs in the US are not known, but the most representative survey, of rural soil PCDD/PCDF concentrations in Ontario and the US mid-west, reported an average of 100 pg/cm^3.[20)]

Historical air emissions of PCDD/PCDF are recorded in lake sediments. Sediment cores confirm that peak emissions were in the 1965 - 1975 time frame. PCDD/PCDF content in lake sediments from before 1940 are roughly an order of magnitude less than in 1970, and PCDD/PCDF concentrations have generally decreased since the 1970s.[21),22)] The sediment record is in rough agreement with the estimates given in Table 2, and indicates that emissions in 1940 were closer to the lower end of the estimated range.

Thus while there is considerable uncertainty in both the estimated emissions and in the interpretation of the environmental data, the environmental concentrations of PCDD/PCDFs are roughly consistent with those expected from the emissions estimates.

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