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HUMAN EXPOSURE TO DIOXIN FROM COMBUSTION SOURCES

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Holly A. Hattemer-Frey

Curtis C. Travis

Office of Risk Analysis
Health and Safety Research Division
Oak Ridge National Laboratory *
P.O. Box 2008, Bldg. 4500S
Oak Ridge, Tennessee 37831-6109

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INTRODUCTION

Because of their extreme toxicity, much concern and debate has arisen about the nature and extent of human exposure to dioxin. Since municipal solid waste (MSW) incinerators are known to emit polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) [1], many people who live near MSW incinerators fear that they will be exposed to high levels of dioxin and subsequently develop cancer. What is often overlooked in this debate, however, is the fact that the general population is continuously being exposed to trace amounts of dioxin as exemplified by the fact that virtually all human adipose tissue samples contain dioxin at levels of 3 parts per trillion (ppt) or greater [2,3]. This paper provides a perspective on MSW incineration as a source of human exposure to dioxin by comparing this exposure source with exposure to background environmental contamination and evaluates some of the potential key sources of PCDD/PCDF input into the environment.

BACKGROUND EXPOSURE

Background levels of 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) have been measured in air, soil, sediment, suspended sediment, fish, cow milk, human adipose tissue, and human breast milk samples [2,4-13]. Travis and Hattemer-Frey [14] used measured environmental concentrations to quantify the amount of TCDD entering the food chain and the extent of human exposure. Potential pathways of human exposure include: (1) inhalation; (2) accumulation in vegetation via direct

deposition and root uptake and ingestion of contaminated fruit, vegetables, and grains; (3) ingestion of contaminated soil and forage by terrestrial organisms, bioaccumulation of TCDD in their tissues, and human consumption of contaminated beef and milk products; (4) deposition onto water surfaces, accumulation in aquatic biota, and transferral to humans through the aquatic food chain; and (5) ingestion of contaminated drinking water.

Because TCDD is highly lipophilic, it partitions mainly into soil (85%) and sediment (14%) and readily bioaccumulates in living organisms [14]. Less than 1% of the TCDD released into the environment partitions into air, water, suspended sediment, and biota [14]. Table 1 shows that the food chain, especially meat and dairy products, accounts for more than 98% of human exposure to TCDD and that the long-term, average daily intake by the general population of the U.S. is 47 picograms (pg)/day [14]. Consumption of contaminated vegetation and fish were minor pathways of human exposure, and inhalation was not a major pathway of human exposure [14].

This environmental pathway estimate was verified using a one compartment pharmacokinetic model [15], which was used to estimate daily intake from human body burdens (i.e., adipose tissue levels). Using a half-life of 2,120 days for TCDD in the human body [16] and an average human adipose tissue concentration of 6 ng/kg [2], the long-term, average daily intake of TCDD was estimated to be 28 pg/day. Both the environmental pathway and the body burden estimates agree well with an estimate of 25 pg/day reported by Beck et al. [17].

Beck et al. [17] and Ono et al. [18] analyzed randomly collected food samples of the human diet and estimated the average, background daily intake of all PCDDs and PCDFs as a whole [expressed in Toxic Equivalents Dioxins and Furans (TEDFs)] resulting from ingestion of contaminated food items to be about 90 and 60 pg TEDFs/day, respectively. [When PCDDs and PCDFs concentrations are expressed in TEDFs, the toxicity of all isomers is weighted relative to the known toxicity of 2,3,7,8-TCDD. Thus, ingesting one gram of TEDFs is equivalent to ingesting one gram of TCDD.] The Japanese intake estimate, however, reflects the dietary consumption of an individual weighing 50 kg. If the Japanese estimate is adjusted for an individual weighing 70 kg, then the average daily intake of PCDDs and PCDFs is 84 pg TEDFs/day.

EXPOSURE TO MSW INCINERATOR EMISSIONS

It is widely believed that MSW incineration is the major source of human exposure to dioxin. To assess the extent of human exposure to facility-emitted PCDDs and PCDFs, Travis and Hattemer-Frey [19] evaluated 11 risk assessment documents prepared for proposed MSW incinerators designed to use modern, efficient pollution control equipment. The geometric mean of the data from those 11 documents was used to represent exposure to a typical, modern MSW incinerator. (Hence, the following results may not apply to older incinerators that do not use efficient pollution control equipment.) Results suggest that MSW incineration is not a major source of human exposure to dioxins and furans for the following reasons.

First, exposure to background levels of PCDDs and PCDFs accounts for more than 99% of the total daily intake by the maximally exposed individual living near a typical, modern MSW incinerator. Table 2 shows that the predicted daily intake of PCDDs/PCDFs by the maximally exposed individual is 80 times less than exposure to background levels. These data indicate that human exposure to PCDDs and PCDFs emitted from a typical, state-of-the-art MSW incinerator is not excessive relative to exposure to background levels, since the individual lifetime cancer risk associated with exposure to facility-emitted PCDDs and PCDFs is one in a million ($1x10^{-6}$), while the cancer risk associated with exposure to background environmental contamination is two in ten thousand ($2x10^{-4}$).

Secondly, the maximum air concentration of PCDDs and PCDFs predicted to occur at the point of maximum individual exposure is slightly less than measured background levels of PCDDs and PCDFs in urban air (Table 3). Background concentrations of PCDDs and PCDFs have been measured in urban air in the U.S. ($1.2x10^{-5}$ ng TEDFs/m^3), West Germany ($3.3x10^{-5}$ ng TEDFs/m^3), and Sweden ($9.3x10^{-6}$ ng TEDFs/m^3), respectively [7,9,11], with a geometric mean of $1.6x10^{-5}$ ng TEDFs/m^3. Thus, atmospheric concentrations of PCDDs and PCDFs emitted from modern MSW incinerators should be indistinguishable from measured background levels, which suggests that, on the local level, MSW incinerators are not substantially increasing background atmospheric levels of dioxin.

This conclusion is supported by actual measurements of PCDDs and PCDFs in ambient air near operating MSW incinerators. Rappe et al. [9] reported that the concentration of PCDDs and PCDFs measured at 13 sites

1500 meters downwind from a MSW incinerator near Hamburg, West Germany, was 5.8×10^{-5} ng TEDFs/m^3. Similarly, Olie et al. [20] found that concentration of PCDDs and PCDFs in the air two kilometers from a MSW incinerator near Amsterdam was 4.1×10^{-5} ng TEDFs/m^3. Thus, measured air concentrations around these two operating MSW incinerators are about three to four times higher than the mean background urban air concentration of PCDDs and PCDFs (1.6×10^{-5} ng TEDFs/m^3). These data indicate that air concentrations of PCDDs and PCDFs near operating MSW incinerators are slightly elevated relative to background urban air levels. The fact that measured air concentrations of PCDDs and PCDFs near operating MSW incinerators are at or near background levels, however, supports modeling conclusions that MSW incinerators are not substantially elevating background levels of PCDDs and PCDFs (model predictions of PCDD/PCDF concentrations around modern MSW incinerators plus the geometric mean background air concentration equals 2.8×10^{-5} ng TEDFs/m^3 versus 4.1×10^{-5} to 5.8×10^{-5} ng TEDFs/m^3 actually measured around operating MSW incinerators).

One might wonder why, if concentrations of PCDDs and PCDFs around operating MSW incinerators are at or near measured background levels, is the contribution of facility-emitted PCDDs and PCDFs to total daily intake by the maximally exposed individual (Table 2) so small? One reason is that risk assessments for proposed MSW incinerators generally assume that only a small percentage of food consumed by the maximally exposed individual is actually produced in the local area (and therefore affected by incinerator emissions). Since the food chain is the primary pathway of human exposure to a large class of organic

compounds, including TCDD, DDT, and most pesticides [14,21], this assumption can substantially reduce intake estimates. In areas of high agricultural production, for example, the 0.6 pg TEDFs per day daily intake estimate for incinerators could be low.

SOURCES OF PCDD/PCDF INPUT

If MSW incineration is not the major source of human exposure to PCDDs and PCDFs, what is? Although the magnitude of PCDD/PCDF input into the environment remains unknown, principal sources of PCDDs and PCDFs are suspected to be: (1) high temperature industrial processing facilities, such as metal processing/treatment plants and copper smelting plants [1,22]; (2) motor vehicles [9,23-25]; (3) chemical manufacturing processes [26]; (4) chemical/industrial waste incineration [26,27]; (5) MSW incinerators [1,9,22,26]; and (6) pulp and paper mills [9,28]. The magnitude of emissions from sources for which empirical data are available are discussed below.

Industrial Sources

Rappe et al. [1] and Marklund et al. [22] reported that emissions of PCDDs and PCDFs from high-temperature industrial processing plants seem to be of the same magnitude as emissions from MSW incinerators. The Swedish EPA [29] estimated that emissions of PCDDs and PCDFs from these types of plants operating in Sweden range from 50 to 150 grams TEDFs per year, while normal-sized, modern MSW incinerators (50 to 200,000 tons of waste per year capacity) emit about 1 to 50 grams TEDFs per year [1,19].

Rappe et al. [9] found that levels of PCDDs and PCDFs in air collected from a West German industrial area located 600 meters from a copper smelting plant ($3.6x10^{-2}$ ng/m^3) were about 12 times higher than measured background levels in suburban air collected about 13 km from Hamburg, West Germany ($3.0x10^{-3}$ ng/m^3) and about three times higher than levels measured around a MSW incinerator operating near Hamburg ($1.3x10^{-2}$ ng/m^3).

Rappe et al. [1], moreover, concluded that because there are many more industrial sources, their contribution to total PCDD/PCDF emissions could be much greater than the contribution from MSW incinerators. Marklund et al. [22] observed that "total emissions from industrial incinerators could be of the same magnitude or even higher than emissions from MSW incinerators," while Nakano et al. [30] reported that "PCDDs and PCDFs in the urban air are surmised to be derived from domestic and industrial waste incinerators." Thus, various researchers agree that industrial sources may contribute equal or even larger amounts of PCDDs and PCDFs into the environment than MSW incinerators.

Motor Vehicles

Studies have shown that motor vehicle emissions may be a larger source of PCDD/PCDF input than emissions from MSW incinerators. Ballschmiter et al. [23] argued that a more prevalent source of PCDD/PCDF input than MSW incineration must exist to account for the widespread, background contamination of PCDDs and PCDFs and suggested that the ubiquitous, non-point character of motor vehicle emissions

"strongly recommends this source for consideration as a major environmental input."

Marklund et al. [25] found that total emissions of PCDDs and PCDFs from cars in Sweden using unleaded gasoline were 10 to 100 grams TEDFs per year, which is equivalent to the amount of PCDDs and PCDFs emitted from two t 20 MSW incinerators of normal size and technology. Thus, in Sweden, motor vehicles and MSW incinerators emit about the same amount of PCDDs and PCDFs. Jones [31] reached the same conclusion for the United States and contends that due to the widespread source of motor vehicle emissions, roadside exposures are equal to or greater than exposures from the elevated stacks of MSW incinerators.

Rappe et al. [9] also measured PCDD/PCDF levels in ambient air within a traffic tunnel. PCDD/PCDF levels in the tunnel ($2.8x10^{-2}$ ng/m^3) were two times higher than levels downwind from the MSW incinerator ($1.3x10^{-2}$ ng/m^3) and nine times higher than the suburban air concentration ($3.0x10^{-3}$ ng/m^3) [9]. Rappe et al. [9] concluded that "measurements made in the traffic tunnel clearly indicate that motor vehicles are a source of PCDDs/PCDFs in the ambient air." Hence, several researchers confirm that motor vehicle emissions may also be a significant source of PCDD/PCDF in the environment.

Pulp and Paper Mills

The US EPA [13] first reported that pulp and paper mills using a chlorine bleaching processes may be another source of PCDD/PCDF input into the environment. Swanson et al. [32] analyzed the effluents from one mill producing bleached pulp in Sweden and found that PCDD/PCDF emissions ranged from 2 to 5.8 grams TEDFs per year. Since the mill

they sampled used a less efficient processing method than most operating mills in Sweden, they estimated that Swedish pulp and paper mills as a whole discharge about 5 to 15 grams of TEDFs per year [32]. These preliminary findings suggest that the amount of PCDDs and PCDFs formed in pulp and paper mills in Sweden is small relative to other sources.

Thus, these data indicate that environmental concentrations cannot be linked to any one combustion source. Combustion processes in general (not just MSW incinerators) are the dominant source of PCDDs and PCDFs in the environment. It is premature to conclude that MSW incineration is the major source. The magnitude of PCDD/PCDF emissions from and PCDD/PCDF concentrations in the ambient air and other environmental media (e.g., soil and cow's milk) around other operating combustion sources known to emit dioxins and furans (i.e., copper smelting plants, steels mills, motor vehicles, and pulp and paper mills) are needed before definitive statements about the major source(s) of PCDD/PCDF input can be made.

CONCLUSIONS

This paper in not intended resolve the incineration debate or to criticize or recommend MSW incineration as a waste management tool. It does, however, reevaluate some widely held beliefs concerning human exposure to PCDDs and PCDFs emitted from MSW incinerators. MSW incinerators are one source of PCDD/PCDF input into the environment. Empirical evidence demonstrates, however, that well-operated, modern MSW incinerators may not be the major source of human to PCDDs and

PCDFs, since exposure to background levels overwhelms exposure to facility-emitted contaminants. The relatively small contribution by incinerators to total daily intake suggests that some, as of yet unidentified, source(s) (possibly automobiles or industrial sources) are substantially contributing to background levels of dioxins and furans. We recommend that future research efforts focus on characterizing the source(s) of background levels of PCDDs and PCDFs, because they may pose far greater threats to human health than PCDD/PCDF emissions from modern MSW incinerators.

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Table 1. Predicted average background daily intake of TCDD by the general population of the U.S.

Source	Daily intake (pg/day)	Percentage of the total daily intake
1. Air	1	2%
2. Water	0.007	<.01%
3. Food (total)	46	98%
a. Produce	5	11%
b. Milk	13	27%
c. Meat	23	50%
d. Fish	5	10%
Total Intake	47	100%

Table 2. Total daily intake of PCDDs/PCDFs by maximally exposed individual living near a typical, modern MSW incinerator

Source	Daily Intake (pg TEDFs/day)	Percentage of the Total Daily Intake	Reference
Background*	87.0	99.3%	17,18
Incinerator	0.6	0.7%	19
Total	87.6	100%	

* Geometric mean of estimates reported by Beck et al. [17] and Ono et al. [18] for a 70 kg individual.

Table 3. Predicted concentration of PCDDs and PCDFs around a typical, modern MSW incinerator versus background urban air concentrations

Source	Concentration (ng TEDFs/m^3)	Reference
Predicted	1.2×10^{-2}	19
Measured Background[a]	1.6×10^{-2}	7,9,11

ng = nanogram = 10^{-9} grams

[a] Represents the geometric mean of reported measurements for the U.S., Germany, and Sweden [7,9,11].