The Inventory of Sources of Dioxin In the United States

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I. Introduction

Since 1992, the United States Environmental Protection Agency (USEPA) has been conducting a study of human exposures and health effects associated with polychlorinated dibenzodioxins and polychlorinated dibenzofurans (i.e., dioxin). To better understand the sources and environmental releases of dioxin, many countries throughout the world have undertaken assessments or inventories of existing source activity, to include the United Kingdom, Canada, Germany, Sweden, Denmark, New Zealand, and The Netherlands. This paper presents and describes the draft inventory of sources of dioxin in the United States.

II. Methods

In the development of a national inventory, the EPA has generally classified sources of dioxin as: combustion sources; metals smelting and refining and processing; chemical manufacturing; biological and photochemical processes; and reservoir sources. Combustion sources include waste incineration (municipal solid waste, medical waste, sewage sludge, and hazardous waste), the combustion of various fuels (coal, wood, and petroleum products), and various other high-temperature combustion sources (i.e., cement kilns) as well as poorly controlled combustion sources (such as building fires). Metal smelting includes various types of both primary and secondary metals operations (iron ore sintering, steel production, and scrap metal recovery). Chemical manufacturing sources include the chlorine bleached wood pulp, chlorinated phenols, polychlorinated biphenyls, phenoxy herbicides and chlorinated aliphatic compounds. In biological and photochemical process, it is possible to form dioxin under certain environmental conditions (e.g., from the action of microorganisms in compost, and from the microbial degradation of phenolic compounds; from the photolytic decomposition of highly chlorinated phenols. Reservoir sources are materials or places which contain previously formed dioxin and have the potential for redistribution and circulation of dioxin back into the open environment. Potential reservoirs include: soils, sediments, vegetation and PCP-treated wood. EPA has made annual release estimates of dioxin to the air, land, and water for all of these sources,

ORGANOHALOGEN COMPOUNDS Vol. 36 (1998) with the exception of the reservoir sources, biological an photochemical processes.

The sources were evaluated for environmental releases from the perspective of timedependency. Two reference years were selected: 1987 and 1995. 1987 was selected because few potential sources had been characterized in the U.S. for dioxin emissions prior to this time, and this also represented sources that were largely uncontrolled with respect to dioxin. 1995 is representative of the latest time period that could practically be addressed with existing information, but also represents a time when many identified sources had controlled or reduced dioxin releases. The use of reference years allows for time-trend analysis.

In developing an inventory, imphases was given to the use of primary information on the measurement of dioxin releases from the sources. Primary information includes original engineering test reports utilizing adequate quality control and quality assurance procedures for both the sampling and laboratory protocols. Over several thousand potential sources of dioxin exist in the U.S., dioxin releases have been characterized from less than 10% of the sources. Given this circumstance, and because testing all suspected sources is impractical and prohibitively expensive, it was necessary to extrapolate dioxin releases from the few tested facilities to the many non-tested sources. Such extrapolation involves deriving emission factors that are considered representative of the industrial class involved. An emission factor is mathematically derived from the tested facilities, and relates the mass of dioxin released per unit of activity level. Activity level considers the type of materials or waste processed or produced such as kg waste combusted, liters industrial effluent discharged into surface water; kg of material produced by an industry, ctc. As examples, the emission factors for combustion, manufacturing, and transportation sources may be expressed as g dioxin emitted per kg waste combusted; g dioxin released per L of wastewater discharged to surface water; and g dioxin emitted per km driven, respectively. When emissions and releases from tested facilities are presented in these terms, annual releases from a single source or a class of sources can be estimated by multiplying the emission factor by the annual activity level, as in the equations below.

$$E_{total} = \sum E_{tested_i} + \sum E_{untested_i}$$

$$E_{total} = \sum E_{tested_i} + \sum (EF_i \times A)_{untested_i}$$

Where:

Ettotal is the annual dioxin emissions from all sources; g I-TEQ/yr

 $E_{tested,i}$ is the annual dioxin emissions from all untested facilities in source class i; g TEQ/yr

 $E_{untested,i}$ is the annual emissions from all untested facilities in source class i, g TEQ/yr EFi is the mean emission factor for the tested facilities in source class i, g TEQ/kg A_i is the measure of activity level for untested facilities in source class i, kg/yr

The quantitative estimates of emissions and environmental releases for each source category are contained in both an EPA report¹ and supporting electronic database².

III. Results

Table 1 summarizes the annual TEQ emissions from sources in the United States for

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both reference years. The ranges in the release estimates are intended to reflect overall confidence in both the emission factor estimate and the measure of activity of the source class. Despite relative uncertainty in these numbers, the following conclusions can be made from the National Source Inventory.

 The environmental releases of dioxin-like compounds in the United States occur from a wide variety of sources, but are dominated by releases to the air from combustion sources.
 Insufficient measurement data are available to comprehensively estimate source releases of dioxin to water and to land.

3. Central estimates of releases of dioxin-like compounds to all environmental media were approximately 12 kg TEQ in 1987 and 3 kg TEQ in 1995. As a reflection of uncertainties in the estimates for individual sources, dioxin releases ranged from 5 kg to about 30 kg TEQ and 1 kg to about 8 kg TEQ for 1987 and 1995, respectively.

4. Comparison of the central estimates suggests that there was approximately a 75% decrease in dioxin releases to the U.S. environment between 1987 and 1995. This reduction was primarily caused by reductions in air emissions from municipal and medical waste incinerators.

IV References

1. *The Inventory of Sources of Dioxin in the United States*. The Office of Research and Development, National Center for Environmental Assessment, Washington DC. External Review Draft. EPA/900/P-98/002Aa, April 1998.

2. Database of Sources of Environmental Releases of Dioxin-Like Compounds in the United States. The Office of Research and Development, National Center for Environmental Assessment, Washington DC. External Review Draft. On Compact Disk. EPA/600/P-98/002Ab, April 1998.

Source Type/environmental Media	1995 Releases (g TEQ/yr)	Range Reflecting Uncertainty (g TEQ/yr)	# Tested out of Total	1987 Releases (g TEQ/уг)	Range Reflecting Uncertainty (g TEQ/yr)	# Tested out of Total	Change in Dioxin Releases
Municipal Solid Waste Incinerators, to air	1,100	500 - 2,500	38/138	7900	3,500 - 18,000	19/115	-86%
Secondary Copper Smelting, to air	541	170 - 1,700	1/24	304	100 - 900	1/24	78%
Medical Waste Incinerators, to air	477	150- 1,500	19/2400	2470	800 - 8,000	6/5000	-80%
Forest Fires, to air	208	65 - 650	(A)	170	50 - 500	(A)	22%
Cement Kilns (hazardous waste), to air	153	50 - 500	12/34	117	40 - 400	12/180	30%
Sewage Sludge, land applied	207	120 - 375	174 /16,000	207	120 - 375	174 /16,000	0%
Coal Fired Utilities, to air	73	30 - 160	11/1000	63	30 - 140	11/1000	16%
Residential Wood Burning, to air	63	20 - 200	(B)	90	30 - 300	(B)	-30%
Diesel Trucks, to air	34	10 - 100	(C)	26	8 - 80	(C)	30%
Industrial Wood Burning, to air	30	13 - 65	4/80	27	12 - 60	4/80	10%
Bleached Pulp and Paper Mills, to water	20	10 - 30	104/104	360	250 - 500	104/104	-94%
Cement Kilns (nonhazardous waste), to air	18	6 - 60	11/178	14	4 - 40	11/178	30%

Table 1. Inventory of Sources and Environmental Releases of Dioxin in the United States: 1995 and 1987

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Source Type/environmental Media	1995 Releases (g TEQ/yr)	Range Reflecting Uncertainty (g TEQ/yr)	# Tested out of Total	1987 Releases (g TEQ/yt)	Range Reflecting Uncertainty (g TEQ/yr)	# Tested out of Total	Change in Dioxin Releases
Secondary Aluminum Smelters, to air	17	5 - 50	4/76	10	3 - 30	4/67	70%
Oil Fired Utilities, to air	9	3 - 30	(D)	16	5 - 50	(D)	-44%
Leaded Gasoline Vehicles, to air	<1	<1	(E)	32	10 - 100	(E)	-99%
Unleaded Gasoline Vehicles, to air	6	2 - 20	(F)	4	1 - 12	(F)	50%
Sewage Sludge Incineration, to air	6	3 - 13	11/199	6	3 - 13	11/1 99	0%
Hazardous Waste Incineration, to air	6	3 - 13	17/162	5	2 - 10	17/227	20%
Kraft Black Liquor Boilers, to air	2	1 - 5	6/124	2	1 - 5	6/124	0%
Secondary Lead Smelting, to air	2	1 - 4	3/23	1	0.5 - 3	3/24	100%
Boilers/Industrial Furnaces, to air	0.4	0.1 - 1	2/136	1	0.2 - 20	1/136	-60%
Paper Mill Sludge, to land	1	1 - 2	13/104	14	10 - 20	6/104	-93%
Cigarette Smoke, to air	0.8	0.3 - 3	(G)	1	0.3 - 3	(G)	-20%
Total number facilities tested			430			390	
TOTAL grams TEQ	3,000	1,200 - 8,000		12,000	5,000 - 30,000		
% REDUCTION 1987 TO 1995:	75%						

Footnotes:

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A. Dioxin have been measured in forest fire, but could not be used to derive emission factors. Estimates were based on 9 wood stoves experiments of the combustion of a variety of wood types, and the results were extrapolated to the amount of biomass consumed by fire for 1995 and 1987.

B. Estimates were based on 9 wood stoves experiments of the combustion of a variety of wood types. About 3.5 million and 5 million households burned wood in wood stoves or fireplaces in 1995 and 1987, respectively.

C. The emission factor was developed from an API study of dioxin in air within the Baltimore Harbor Tunnel predominantly used by heavy duty trucks. More than 50,000 heavy duty diesel trucks passed through the tunnel during the sampling of tunnel air.

D. Emission factor developed from the stack test at 2 oil-fired electric utility boilers. Total dioxin emissions estimated on basis of total oil consumed by all utilities. Actual number of utilities not known

E. No leaded gasoline powered vehicles tested in the U.S. Data derived from German and Swedish studies of 4 cars operated on leaded gasoline.

F. No tests for dioxin emissions of unleaded-gasoline powered vehicles has been conducted in the U.S. Data derived form Swedish and German EPA-sponsored studies. Three cars tested varied in age, and were equipped with catalytic converters. The unleaded fuel in Europe is comparable to unleaded gasoline in the U.S. G. Data derived from a Japanese study of 20 brands of cigarettes (7 of which were U.S. brands). Number of cigarettes smoked: 487 billion in 1995, and 575 billion in 1987.

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