UPDATED ANALYSIS OF U.S. SOURCES OF DIOXIN-LIKE COMPOUNDS AND BACKGROUND EXPOSURE LEVELS

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The United States Environmental Protection Agency (EPA) is currently reassessing the exposure to and human health/ecological impacts of dioxin and related compounds. This effort began in 1991 and is expected to be completed in 1995. At Dioxin 93, the authors presented a paper describing the progress and tentative findings from this effort on sources which release these compounds to the environment and background exposure levels[1]. The purpose of this paper is to present an updated analysis of these issues. This analysis should still be regarded as tentative, pending completion of the final Reassessment document.

The updated results of a nationwide inventory of emission estimates for the U.S. are summarized in Table 1. This table lists emission estimates for the major known or suspected sources which could have releases of CDD/F (chlorinated dibenzodioxins and dibenzofurans) to the environment. The emissions are expressed in terms of 2,3,7,8-TCDD toxic equivalents (TEQ). In order to make each source emission estimate, information was required concerning both the "emission factor" term for the source (e.g., mass of contaminant released per mass of feed material processed) and the "production" term for the source (e.g., mass of feed material processed annually in the U.S.). Because the quantity and quality of the available information for both terms for each emission source varies considerably, a confidence rating scheme was developed. The scheme assigns ratings of "high", "medium", or "low" to both terms. In addition, the uncertainty in these national release estimates is reflected by presenting a possible range from a lower to upper estimate. In general, the emission estimates are quite uncertain since they were derived by extrapolating tests at only a few facilities to a nationwide basis.

New data has resulted in major changes in emission estimates for some source categories and relatively minor changes have occurred to others due to changes in computational procedures. Key source categories and major changes are discussed below:

- Hospital Waste Incinerators - A reanalysis of these data have resulted in somewhat higher emission estimates for this category. This still appears to be the largest known source in the US. This is due to the facts that these incinerators typically do not have extensive pollution controls, number about 6000 and burn high chlorine content waste. New standards will be proposed soon which should lead to substantial reductions in emissions.
- Municipal Waste Incinerators - The emission estimate for this category has increased significantly since that presented in 1993. The earlier estimate was developed before the

reassessment effort had assembled individual facility test data. In the absence of such data, the earlier paper used the emission estimate associated with implementation of the standard promulgated by EPA in 1991. The revised estimate reflects a review of test data received within the last year. The Agency is still in the process of collecting test data on these facilities and it is likely that further adjustments will be needed before a final estimate of current emissions can be made. Many municipal waste incinerators are currently taking actions to reduce their emissions and the Agency will soon be proposing stricter standards than the 1991 rules.

- Wood Burning - Reductions have occurred in the emission estimates for residential wood burners. The earlier estimate was made by combining total particulate generation rates with dioxin levels found in chimney soot. This approach is likely to lead to over estimates since dioxin levels in soot can be much higher than what is actually emitted on particulates due to accumulation in chimneys. New data are now available based on direct measurements in smoke from wood stoves under various conditions[2,3]. These new data obviously provide a much better basis for making emission estimates. The estimates for industrial wood burning have not changed significantly.
- Forest Fires - The estimate for emissions from forest fires have been reduced substantially. The earlier estimate was made in the same fashion as described above for wood stoves, ie. combining total particulate generation rates with levels found in chimney soot. New data has been published on dioxin emissions from burning natural wood in stoves under conditions of uncontrolled draft[2,3]. Although these conditions do not precisely match those of forest fires, it is believed that they offer a more credible basis for the calculation than the method used earlier. The theory that much of today's body burden could be due to natural sources (such as forest fires) has been largely discounted by testing of ancient tissues which show levels much lower than those found today[4]. Only one test has been conducted that directly measured CDD/F in smoke of forest fires[5]. Low levels were detected, but the authors caution that all or a portion of these emissions could represent resuspended material from aerial deposits rather than originally formed material.
- Cement Kilns - New data has allowed addition of this category and suggests that this is a potentially important source. About 15% of the U.S. kilns burn hazardous waste. It appears that dioxin levels in emissions from kilns burning hazardous waste are higher than those not burning hazardous waste.
- Diesel Vehicles - The data on dioxin emissions from diesel vehicles remains quite limited and somewhat contradictory. A relatively high emission rate is suggested by a study conducted in Norway which measured the increase in dioxin concentration in air between two points in a tunnel[6]. Although the air samples were collected in a manner representing hundreds of vehicles, the indirect method of analysis introduces uncertainty. Much lower emission rates are suggested by a Swedish study involving direct tailpipe measurements.[4] This study reported no emissions at a detection limit of 100 pg/l or approximately 0.01 to 0.05 ng/km - a factor of 100 to 500 lower than the emission rate reported in the tunnel study. Because this study's results are based on only one vehicle using Swedish fuel, this emission factor is also quite uncertain. These two studies yield a very wide range of emission estimates and clearly suggests that further testing is needed.
- Metal Smelters - New data on emissions from secondary lead smelters suggest that this is a relatively small source in the U.S. No significant changes have been made to the emission estimates for secondary copper smelters. New data from Europe[8] suggests that dioxin emissions occur during iron sintering operations, but estimates could not be made for U.S. operations. Sufficient data are still lacking to estimate emissions from other

types of metal smelters in the U.S.

- Pulp and Paper Mills - These facilities can have dioxin releases to water, land and paper products. The paper industry has recently made process changes which they estimate have reduced dioxin emissions by about 80%. The upper and lower estimates for water discharges, 150 and 74 g TEQ/yr respectively, are based on an analysis of 1993 estimates separately determined by EPA and the paper industry.

The other combustor categories evaluated in this report are estimated to have much lower emissions and thus appear to be relatively minor sources on a national scale (although their local impacts could be important to evaluate). These include sewage sludge incinerators, hazardous waste incinerators, Kraft liquor boilers, drum and barrel reclaimers, tire combustors, carbon reactivation furnaces and scrap electric wire recovery facilities. The importance of coal-fired utilities remains unknown, although testing is now underway.

Dioxin-like compounds can also be formed during the manufacture of certain compounds such as chlorinated phenols, chlorinated benzenes and others. The releases associated with chemical manufacturing could not be quantified due to the lack of test data. Potentially such releases could occur via the product itself or as emissions to the air, land or water. Such problems have lead to the termination of production of PCBs and some phenoxy herbicides. Recently, some claims have been made that significant dioxin emissions may occur during the production of vinyl chloride monomer and associated products. However, insufficient emission data are currently available to evaluate these reports.

The 1993 paper presented a preliminary "mass balance" analysis on the estimates of national dioxin releases to the environment. Basically, this procedure involves comparing estimates of the emissions to estimates of aerial deposition. Such studies in Sweden[9] and Great Britain[10] have suggested that the deposition exceeds the emissions by about 10 fold. These studies are acknowledged to be quite speculative due to the strong potential for inaccuracies in emission and deposition estimates. In addition, the apparent discrepancies could be explained by long range transport from outside the country, resuspension and deposition of reservoir sources or unidentified sources. Bearing these limitations in mind, this procedure was used in the earlier paper and updated here to compare the estimated emissions and deposition in the U.S.

Deposition measurements have been made at several locations in Sweden[11] and two places in the U.S.[12] These limited data suggest that a deposition rate of 1 ng TEQ/m^2-yr is typical of remote areas and that 2 to 6 ng TEQ/m^2-yr is more typical of populated areas. Applying these values, the total U.S. deposition can be estimated as 20,000 to 50,000 g TEQ/yr. This range can be compared to the range of emissions for the US (3,300 to 26,000 g TEQ/yr). The current range of deposition estimates was broadened over the earlier point estimate to better reflect the uncertainty.

Literature on occurrence of these compounds in environmental and exposure media were compiled and summarized. The average levels found in the media have not changed significantly since the 1993 paper and are presented in Table 2. These media concentrations were computed assuming that nondetects equal half the detection limit. However, estimates were also computed assuming that nondetects equal zero. For some food groups such as eggs, much lower TEQ estimates resulted indicating a high percent of nondetects among individual congeners. The upper mean TEQ estimates are generally comparable to the TEQ estimates derived from studies conducted in Germany[13] and Canada[14]. In summary, the limited number of U.S. food samples and the high incidence of nondetects in some groups make an uncertain basis for estimating national background levels. However, the general agreement with food level estimates reported for Canada

and Germany provides some reassurance that these U.S. values are reasonable. It is clear though that a large survey is needed to confirm residue levels of CDD/F in the U.S. food supply.

For the purposes of calculating background exposures to CDD/Fs via dietary intake the upper-range background TEQs (i.e., those calculated using one-half the detection limit for the non-detects) were used. The ingestion rates have been adjusted slightly since the 1993 paper, resulting in a slightly higher estimate for background exposure (120 pg/d, see Table 2). This estimate compares favorably with analogous estimates of 85 - 158 pg TEQ/day exposures via food consumption in Germany[13], 121-126 pg TEQ/day exposure via numerous routes in the Netherlands[15], and 140-290 pg TEQ/day for the typical Canadian exposed mainly through food ingestion[14]. These exposure estimates are further supported by similar estimates derived from the application of pharmacokinetic models to levels of these compounds found in human tissue.

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Table 1. U.S. Annual Dioxin Emission Rates for Various Sources[1]

Source	Annual Emission Rate (g TEQ/yr)	Confidence Rating[2]	Ratio of Facilities Tested to Total
Medical Waste Incinerators	1600 - 16000	M/L	6/6700
Municipal Waste Incinerators	1800 - 9000	H/M	30/171
Cement Kilns	110 - 1100	H/L	17/212
Industrial Wood Burning	100 - 1000	H/L	2/?
Secondary Copper Smelting	74 - 740	H/L	1/24
Forest Fires	27 - 270	M/L	[1 study]
Diesel Fueled Vehicles	27 - 270	H/L	[See Note 3]
Hazardous Waste Incinerators	11 - 110	M/L	6/190
Residential Wood Burning	13 - 63	H/L	[2 studies]
Sewage Sludge Incinerators	10 - 52	H/M	3/199
Drum and Barrel Reclaimers	0.5 - 5.0	L/L	1/?
Kraft Black Liquor Boilers	0.9 - 4.3	H/M	3/104
Secondary Lead Smelters	0.7 - 3.5	M/M	3/23

1. These emission estimates are preliminary and have not yet been peer reviewed.
2. First letter represents confidence rating for production estimate and second letter is confidence rating for emission estimate, L = Low, M = Medium and H = High.
3. Lower emission estimate is based on test of one Swedish truck and upper estimate is based on a tunnel study in Norway which represented 100's of vehicles.

Table 2. Estimated Background Exposures in the U.S.

Media	North America			
	Conc. TEQ	Contact rate	Daily intake pg/day	% of total
Soil ingestion	8.0 ppt	100 mg/day	0.8	0.7
Fish ingestion	1.2 ppt	6.5 g/day	7.8	6.6
Inhalation	0.095 pg/m^3	23 m^3/day	2.2	1.8
Water ingestion	0.0056 ppq	1.4 L/day	0.008	0.01
Milk ingestion	0.07 ppt	250 g/day	18	15
Dairy ingestion	0.36 ppt	67 g/day	24	20
Eggs ingestion	0.14 ppt	29 g/day	4.1	3.4
Beef ingestion	0.48 ppt	77 g/day	37	31
Pork ingestion	0.26 ppt	47 g/day	12	10
Poultry ingestion	0.19 ppt	68 g/day	13	11
	Total		120	100