EVIDENCE FOR A DECLINE IN BACKGROUND EXPOSURE OF AMERICANS TO DIOXINS FROM THE 1990'S TO THE 2000'S

Matthew Lorber[1], Donald G. Patterson, Jr.[2], Janice Huwe[3], and Larry L. Needham[2]

[1]National Center for Environmental Assessment, Office of Research and Development, United States Environmental Protection Agency, 1200 Pennsylvania Ave, Washington, DC; [2]National Center for Environmental Health, Centers for Disease Control and Prevention, Atlanta, GA; [3]Biosciences Research Laboratory, Agricultural Research Service, U.S. Department of Agriculture, Fargo, ND.

Introduction

The United States Environmental Protection Agency (EPA) characterized exposure of Americans to dioxin-like compounds based on data originating in the mid to latter 1990s[1]. This characterization included an average background intake dose of 60.4 pg TEQ/day (WHO 2005 TEFs; recalculated from the originally published 66 pg TEQ/day, which was developed using WHO 1998 TEFs) and an average background body burden of 22.9 pg TEQ/g lipid (ppt lwt; WHO 2005 TEFs). Both of these estimates were based on mean values from different surveys. Intake estimates combined mean population food intakes (and other exposure contact rates, such as inhalation, but food is primary vector of dioxin exposure) with mean food concentrations from statistically designed national surveys. The body burden estimate was developed as the mean from 6 different surveys covering 316 individuals. By duplicating this characterization in the most analogous way possible using newer data, from surveys conducted between 2000 and 2004, the intake dose was calculated to be 41.0 pg TEQ/day. The mean body burden from newer surveys was found to be between 17.2 and 21.7 ppt TEQ lwt, although a more precise characterization of average background body burden is not possible due to high detection limits and a resulting high frequency of non-detects in the NHANES 2001/2002 data set of individual results. These revised estimates used mean values from essentially analogous surveys, redone in the early 2000s, as noted. The food surveys were redone by the same organization that did the statistically-based surveys of the 1990s. They found declines in TEQ concentrations of all food animals surveyed. They also found that, based on practices of raising food animals in the US (i.e., in confined settings), that terrestrial food animal concentrations tend to cluster near, and mostly less than, 1.0 ppt TEQ lwt with little variability. The variability in overall TEQ intakes is based primarily on food intake variability. Food intake surveys suggest that the 95% intake of animal fats in the US diet is twice the mean and that the 99% percentile is three times the mean. Hence, dioxin intakes characteristic of the early 2000s can range as high as three times the mean intakes, or possibly above 100 pg TEQ/day for a segment of the population. Regarding body burdens, the mean background was determined from over 1000 individuals sampled as part of the statistically based 2001/2 NHANES survey. The 25% extrapolation from this population is about two-thirds of this mean and the 95% is about twice this mean. Prior work has shown that higher body burdens in the typical population (i.e., not occupationally or unusually exposed) are found in older individuals due to past high exposures. The primary uncertainty in these background exposure estimates is in the treatment of non-detects. The ability to state with confidence that exposures have declined, and by how much, will be examined further with appropriate statistical tests and presented at the conference. The evidence for a decline is consistent from food concentrations to body burdens, however, as described below.

Methods

EPA provided adult dose estimates for inhalation, soil ingestion, soil dermal contact, water ingestion, and for 10 food ingestion categories including beef, pork, poultry, milk, dairy, fish, and other products[1]. Each pathway estimate was derived as a point estimate using a mean adult per capita contact rate coupled with a concentration in the exposure media derived as a mean concentration from one or more surveys. The dose estimates were dominated by ingestion of animal food products: ingestion of beef, pork, poultry, dairy, eggs, milk, and fish comprised 93% of total exposures. For this reason, and for the sake of expediency, other pathways (water/soil/vegetable oil ingestion, inhalation, soil dermal contact) are not updated here. Further, contact rates are also not adjusted as evidence suggests a small change in food consumption patterns between the 1990s and 2000s. EPA and the United States Department of Agriculture (USDA) conducted national surveys of dioxin-like compounds in beef, pork, and poultry based on a statistical sampling from national slaughterhouses in the mid-1990s[2-4]. Newer surveys of dioxin-like compounds in beef, pork, and poultry were conducted by the USDA in 2002/3 using the same statistical framework[5]. Also, an updated milk survey by the EPA was used in this exercise to characterize milk/dairy exposures [6]. To describe background body burden levels, EPA derived an average lipid-based blood concentration profile using data from 6 different surveys of general background population exposures conducted in the mid to latter 1990s totaling 316 individuals. Substantially more valid characterizations of national background adult body burdens are now available from over 1000 individuals in the 2001/2 National Health and Nutrition Examination Survey (NHANES)[7,8], and these are used here. Table 1 shows the results of the revised background intake calculations. Congener profiles from the averages of the mid-1990s food surveys and the redone 2002/3 USDA surveys, and the mid-1990s body burden surveys compared against the 2001/2 NHANES body burdens, are shown in Table 2.

Discussion

As seen in Table 1, this exercise suggests a reduction in dose from 60.4 pg TEQ/day to 41.0 pg TEQ/day, or about a one-third reduction from the mid 1990s to early 2000s. However, this needs to be carefully considered. At trace levels in food, analytical capability, and in particular the impact of the limit of detection (or limit of quantification) and the substitution method used to characterize non-detects, has to be considered. For example, a reduction in pork concentrations from 1.41 ppt TEQ lwt in the mid-1990s to 0.22 ppt TEQ lwt in 2002/3 seems like a large reduction – an 84% reduction, but it should be recognized that these concentrations were both calculated assuming ND = ½ DL. At ND = 0, the earlier pork average concentration was 0.40 ppt TEQ lwt, while the later average drops only to 0.15 ppt TEQ lwt at ND = 0. Hoffman et al.[5] recognized the importance of the substitution method for non-detects when they reported on these updated national food surveys, and in comparing their new data with the earlier mid-1990s surveys, they presented results at both ND = 0 and ½ DL. At ND = ½ DL, they found pork and poultry concentrations decreased by 65% and 84%, respectively, and beef decreased by 37%. At ND = 0, they found that the pork and poultry concentrations still dropped 63% and 62%, respectively, but in fact the beef concentrations increased by 22%. This increase could reflect a relatively stable concentration in beef or possibly a small rise in concentrations. Another way to compare the data from the two sets of surveys is to focus on congeners quantified most often. The hepta dioxin congener, 1234678-HpCDD, and the octa congener, OCDD, are almost always found at frequencies greater than 50%, and the average concentrations at ND = 0 and ND = ½ DL are nearly identical. From Table 2, it is seen that there are reductions in average concentrations of the hepta and octa dioxin congeners in all of the food profiles, including that for beef and particularly for pork: 1234678-HpCDD drops from 10 to 1 ppt, and OCDD from 53 to 9 ppt.

The analysis of congener-specific body burden data is also influenced by the detection limit and the treatment of non-detects. The TEQ concentration of the profile provided in Table 2 from the mid-1990s was 22.9 ppt TEQ lwt. EPA[1] stated that the average TEQ concentration dropped by only 1 ppt TEQ lwt when recalculating at ND = 0. This suggests acceptably low detection limits. In contrast, samples from NHANES 2001/2 had very low amounts of serum available for measurements of dioxins, furans, and PCBs; this resulted in high detection limits and thus a large number of non-detects for certain key congeners. For example, 2378-TCDD was only detected 13% of the time. As seen in Table 2, there is a meaningful disparity with 2378-TCDD averages calculated at ND = 0 and ND = detection limit divided by square root of two (DL/sr(2)); it dropped from 2.5 to 0.7 ppt lwt. Five of ten furan congeners were never quantified in the NHANES 2001/2 individual data set. As a result, there is a large difference in average TEQ concentrations derived with the two substitution methods, 17.2 and 21.7 ppt TEQ lwt. So, with only these data, it is appropriate to observe that TEQ concentrations may have declined, but the amount of decline is not known with certainty. Like the food samples, however, information can be gleaned from congeners found most often. The hepta and octa dioxin congeners were found in 99% and 82% of the individual samples, so the means calculated with both substitution methods are similar. It can be seen that there were meaningful declines in these congeners: for 1234678-HpCDD, the concentration declined from 79 to 54 ppt lwt, and for OCDD, the concentration declined from 664 to 452 ppt lwt. Interestingly, PCB 126, which has the highest TEF of the dioxin-like PCBs at 0.1 and was also detected very frequently in the individual samples (89%), appeared to about double in the time frame studied, from about 18 to 35 ppt lwt.

Because of the low sample volumes resulting in high detection limits, serum pools from the NHANES 2001/2002 were developed by the Centers for Disease Control and Prevention (CDC) and analyzed for dioxin-like compounds [8]. Geometric means of congeners were provided for these serum pools for this study. These geometric means suggest a TEQ concentration of about 14 ppt lwt. This may be a better descriptor of a central tendency to describe background body burdens for the U.S. population, but it is distinctly different from the arithmetic average; it is substantially lower. Since the intake dose was developed using arithmetic averages of food survey intakes and average food intakes, it may be appropriate to use average population concentrations in this exercise. However, there may be other reasons to characterize the central tendency of the US population by use of a geometric mean concentration, so this has been provided in Table 2 as a comparison to individual mean concentrations.

The presentation of results as averages neglects variability in exposures. EPA[1] found that intakes of dioxin TEQs ranged up to three times the mean intake, based on an evaluation of food intakes: the 95% intake of animal fats is about twice the mean and 99% intake is three times the mean. The body burden data support this general variability in the population: the 95% concentration from NHANES individual samples equals 59.5 ppt TEQ, just about 3 times the mean at 21.7 ppt TEQ. The 25% concentration is about two-thirds the mean concentration[7]. However, it is noted that higher concentrations of dioxins in the general population are mostly found in older people as a result of past high exposures rather than current exposures[1]. Hoffman et al[5] note small variability in poultry and pork data, with nearly all samples less than 1.0 ppt TEQ, which they attribute to the fact that these food animals are raised to slaughter in confined settings limiting exposures to dioxins. More variability was seen in beef data, however, because they are raised in variable settings, including outdoor grazing, and feedlot conditions. In any case, these data suggest that variability in background exposures in the US is currently based on dietary patterns, along with historical trends in exposure.

Table 1. Updated estimates of background dose of dioxin-like compounds compared to the EPA estimates pertinent to mid-1990s[1] (note: EPA[1] quantities adjusted to WHO 2005 TEFs).

Description	Contact Rate	CDD/F conc, pg TEQ/g wwt	CDD Dose, pg TEQ/d	PCB conc, pg TEQ/g wwt	PCB Dose, pg TEQ/d
Milk	175 g/day	0.01	2.1	0.005	0.9
Dairy	55 g/day	0.08	4.4	0.035	1.9
Eggs	16.8 g/day	0.06	1.0	0.10	1.5
Beef	49.7 g/day	0.12	6.0	0.02	1.1
Pork	15.4 g/day	0.04	0.6	0.006	0.1
Poultry	35.0 g/day	0.02	0.7	0.007	0.3
Other meat	.24.5 g/day	0.06	1.4	0.011	0.3
Fish	15.5 g/day	0.33	5.1	0.61	9.5
TOTAL, meat/dairy/fish/egg updated, pg TEQ/day			(a) 21.2		(a) 16.0
TOTAL, meat/dairy/fish/egg, EPA mid-1990s[1], pg TEQ/day			(b) 36.8		(b) 19.8
TOTAL, other pathways (air, water, soil, veg oil), pg TEQ/day from mid-1990s			(c) 2.9		(c) 0.9
OVERALL TOTAL, updated, (a) + (c)			24.1		16.9
OVERALL TOTAL, mid-1990s, (b) + (c)			39.7		20.7

Table 2. Average concentrations (pg/g lipid) of individual congeners and TEQs in major terrestrial food product groups and human blood from the mid-1990s compared to comparable data collected in the 2000 – 2003 time frame (* = no data available; NH = NHANES 2001/2; ND = non-detect; GM = geometric mean)

Congener	Beef		Pork		Poultry		Human Blood, ng/g lipid			
	Mid-1990s (n=63)	2002/3 (n=139)	Mid-1990s (n=80)	2002/3 (n=136)	Mid-1990s (n=80)	2002/3 (n=151)	Mid-1990s (n=316)	NH Individual samples (n=1081)		NH Pooled GM (n=51 pooled)
								ND = DL/sr(2)	ND = 0	
2378-D	0.05	0.06	0.10	0.04	0.16	0.04	2.1	2.5	0.7	1.1
12378-D	0.35	0.23	0.45	0.03	0.24	0.06	5.2	4.6	3.7	4.4
123478-D	0.64	0.30	0.52	0.08	0.18	0.05	6.2	5.1	2.9	3.0
123678-D	1.42	1.63	1.10	0.18	0.39	0.26	73.1	47.1	46.9	30.8
123789-D	0.53	0.32	0.47	0.03	0.39	0.06	7.1	6.0	4.0	3.8
1234678-D	4.48	3.97	10.15	1.20	1.53	1.23	79.2	53.8	53.7	35.0
OCDD	4.78	3.92	52.77	9.14	5.31	4.97	664.0	452.1	419.2	315.9
2378-F	0.03	0.03	0.09	0.02	0.28	0.07	0.7	1.8	ND	0.4
12378-F	0.31	0.05	0.45	0.05	0.21	0.07	0.8	1.9	ND	0.4
23478-F	0.36	0.16	0.56	0.08	0.25	0.06	6.2	6.5	5.8	4.9
123478-F	0.55	0.41	0.98	0.17	0.23	0.09	6.5	6.4	6.0	3.4
123678-F	0.40	0.25	0.58	0.13	0.20	0.08	5.3	5.4	4.8	3.1
123789-F	0.31	0.03	0.45	0.03	0.15	0.03	0.7	2.0	ND	0.4
234678-F	0.39	0.21	0.57	0.09	0.21	0.06	2.2	2.2	0.4	0.6
1234678-F	1.00	0.81	3.56	0.68	0.27	0.21	13.2	11.6	11.4	5.2
1234789-F	0.31	0.05	0.57	0.05	0.17	0.02	1.2	2.4	ND	0.4
OCDF	1.88	0.15	2.30	0.44	0.34	0.15	2.1	7.4	ND	2.0
PCB 77	1.0	3.6	1.6	3.6	9.3	5.19	31.1	*	*	*
PCB 81	*	*	*	*	*	*	3.2	9.2	ND	2.3
PCB 118	448.6	*	95.5	*	522.0	*	*	14760	13830	10706
PCB 105	92.7	*	33.4	*	132.0	*	*	4420	1630	1286
PCB 126	4.1	1.2	0.3	0.2	1.8	0.68	18.1	35.4	34.9	18.8
PCB 156	60.7	*	21.6	*	41.0	*	*	7660	6060	4847
PCB 157	13.8	*	5.1	*	10.5	*	*	3530	370	1096
PCB 169	0.7	0.3	0.3	0.3	0.2	0.38	19.4	23.7	23.2	14.0
PCB 189	*	*	*	*	*	*	*	10	3350	461
CDD/F TEQ	**1.00**	**0.71**	**1.37**	**0.19**	**0.71**	**0.21**	**20.5**	**17.5**	**12.9**	**12.0**
cPCB TEQ	**0.43**	**0.13**	**0.04**	**0.03**	**0.19**	**0.08**	**2.4**	**4.3**	**4.3**	**2.3**
TOTAL TEQ	**1.43**	**0.84**	**1.41**	**0.22**	**0.89**	**0.29**	**22.9**	**21.7**	**17.2**	**14.3**

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