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Survey of Polychlorinated Dibenzo-*p*-dioxins, Polychlorinated Dibenzofurans, and Non-*ortho*-polychlorinated Biphenyls in U.S. Meat and Poultry, 2007−2008: Effect of New Toxic Equivalency Factors on Toxic Equivalency Levels, Patterns, and Temporal Trends

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A statistically based survey of dioxins and dioxin-like compounds in domestic meat and poultry was conducted by the U.S. Department of Agriculture (USDA) from September 2007 to September 2008. Seventeen toxic polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and four non-*ortho*-polychlorinated biphenyls (no-PCBs) were measured in 510 beef (steer/heifer), market hog, young turkey, and young chicken samples. The results of the survey showed the sum of PCDD/F and no-PCB toxic equivalencies (sum-TEQs) ranging from not detected to 4.5 pg/g of lipid. Mean sum-TEQ levels for beef, turkey, chicken, and pork were 0.66, 0.61, 0.17, and 0.16 pg/g of lipid, respectively. To compare the new survey data with data from previous USDA surveys in the mid-1990s and 2002−2003, TEQs from all data sets were calculated using the most recent 2005 toxic equivalency factors (TEFs). The results of the recalculation on the older survey data was a small increase (4−13%) in mean TEQs for the mid-1990s data, which initially used pre-1994 TEFs, and a small decrease (2−4%) for the 2002−2003 data, which initially used 1998 TEFs. A comparison of the three surveys indicates declining TEQ trends in all slaughter classes over the 10 year period; however, the congener patterns remain relatively constant between 2002 and 2008, indicating similar animal exposures to dioxins and dioxin-like compounds during these time periods. Several samples from the 2008 survey with the highest TEQ values are undergoing follow-up investigations to determine possible sources that may be contributing to these levels.

KEYWORDS: Dioxins; PCBs; food survey; meat; poultry; TEFs; temporal trends

INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and polychlorinated biphenyls (PCBs) are ubiquitous and persistent environmental contaminants that bioaccumulate in the food chain and are considered to be toxic. The majority of human exposure to dioxins and dioxin-like compounds has been estimated to be from the dietary intake of animal and fish products (*1*, *2*). Thus, the U.S. Department of Agriculture (USDA) conducts periodic surveys to monitor these compounds in domestic meat and poultry to identify and contain possible contaminations, uncover and reduce input sources, provide statistically based information on the current level of dioxin-like compounds in slaughterhouse animals, and determine distribution profiles and temporal trends in the four slaughter classes that represent slightly >90% of the meat and poultry produced in the United States. Previous surveys of PCDD/Fs and PCBs in beef, hogs, chickens, and turkeys were conducted by the USDA in the mid-1990s with the assistance of the U.S. Environmental Protection Agency (*3*−*6*) and again in 2002−2003 using only USDA resources (*7*).

The value of surveillance for dioxin-like compounds in food and feed products has been proven numerous times in the past decades. Elevated dioxin residues in German dairy products led to the discovery and removal of dioxin-contaminated citrus pulp from the feeds (*8*). Other animal feed components including choline chloride contaminated by pentachlorophenol-treated wood (*9*) and bakery waste contaminated during the drying

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process (*10*) have also been discovered by European monitoring efforts with subsequent removal from the food chain. Recycled fat added to animal feeds was contaminated with PCB-laden transformer oil and resulted in widespread contamination of Belgian livestock and food products (*11*). This contamination episode lasted for an estimated 4 months before the issuance of recalls of the food products. Without routine surveillance, contaminations such as these may go undetected, leading to long-lasting increased exposures of the general population to dioxin-like compounds.

In the United States, surveys have also uncovered sources of contamination in animal feeds, thereby initiating the removal of these contaminated materials from the food chain. During a survey of domestic poultry in 1997, contaminated ball clay used as an anticaking agent in the feed was found to be responsible for elevated dioxin levels found in two chicken samples (*3*, *12*). The toxic equivalency (TEQ) levels in these two samples were elevated by >30 times the mean TEQ of the other young chickens in the survey. The contaminated ball clay had also been used in fish meals, and catfish raised on this feed showed significantly elevated levels of TCDD compared to other catfish (*13*). Following these discoveries, the U.S. Food and Drug Administration (FDA) banned the use of ball clay in animal feeds and issued advisories on the use of other anticaking agents (http://www.fda.gov, accessed June 4, 2009). In a 2002−2003 survey of U.S. meat and poultry, two hogs were found to have dioxin TEQs >10 times the average TEQ for market hogs (*7*). The source of contamination was traced to dioxin-contaminated mineral supplements added to the feed; the supplements were subsequently removed from the market (http://www.fda.gov, accessed June 4, 2009).

In addition to discovering episodic contaminations, food survey data are valuable in risk assessments to estimate human dietary intakes (see, for example, ref *2*), to determine temporal trends of dioxins in the food supply, and to validate the quality of domestic foods with regard to dioxin residues. For intake assessments and residue determinations, individual dioxin-like compounds are generally summed to yield a single toxic equivalency value or TEQ. A TEQ is the summation of the products of individual congener concentrations and their toxic equivalency factors (TEFs). TEFs are values based on the toxicity of an individual congener relative to the most toxic congener, 2,3,7,8-TCDD (TEF = 1), and assigned values of ≤1. Because TEQs are based on TEFs, a relative toxicity that is re-evaluated as new toxicity studies become available, the TEQ is a dynamic value. The first set of international TEFs (I-TEFs) was established in 1988 for PCDD/Fs (*14*) and in 1994 for PCBs (*15*). In 1998 the World Health Organization (WHO) published a revised set of TEFs for PCDD/Fs and PCBs (WHO 1998 TEFs) (*16*); the major changes were an increase in 1,2,3,7,8-PeCDD TEF from 0.5 to 1.0 and a decrease in OCDD/F TEFs from 0.001 to 0.0001. Again in 2005 a WHO panel of experts re-evaluated TEFs for dioxins and dioxin-like compounds incorporating a recently revised and updated database of relative effect potencies (WHO 2005 TEFs) (*17*). The outcome of this latest re-evaluation was a change in several TEFs from the previous WHO 1998 values: 1,2,3,7,8-PeCDF decreased from 0.05 to 0.03; 2,3,4,7,8-PeCDF decreased from 0.5 to 0.3; OCDD, OCDF, and PCB-81 increased from 0.0001 to 0.0003; PCB-169 increased from 0.01 to 0.03; and mono-*ortho*-PCBs were all set at 0.00003, a decrease of 70−99% for most of these congeners. As a result, previously calculated TEQs based on earlier TEFs no longer reflect the most recent toxicological data.

In preparation for a new survey on PCDDs, PCDFs, and PCBs in domestic meat and poultry by the USDA, we recalculated the TEQs of the raw data from previous USDA surveys in the mid-1990s and 2002−2003 using the WHO 2005 TEFs. The resulting data can now be directly compared to the data collected in the 2008 USDA dioxin survey. This approach will provide the most current and meaningful toxicologically based values for risk assessments and allow TEQ comparisons between the old surveys and the new survey to investigate temporal trends.

MATERIALS AND METHODS

The 2008 dioxin survey was conducted using the same methodology and instrumentation as a previous survey completed in 2003 (*7*). Because the methods are based on well-established and published method, details are given in the Supporting Information. The sample design was intended to obtain 510 samples over the course of 1 year, beginning in September 2007. The samples were allocated across product classes as follows: 136 market hogs, 139 steers and heifers, 151 young chickens, and 84 young turkeys. All establishments that slaughtered each product class were eligible for sampling, and slaughter totals from the previous year, as recorded for each establishment in the FSIS' electronic Animal Disposition Reporting System (eADRS), were used to generate a sampling frame on a quarterly basis. Specific establishments were chosen for sample collection using a probability-proportional-to-size design, where slaughter totals were used as the size variable. Under this design, establishments were scheduled to collect approximately the same percentage of samples in a product class as the percentage of national slaughter they performed. The four product classes surveyed represent slightly >90% of the meat and poultry produced in the United States, and the majority (90%) of it is consumed domestically; therefore, production can be used as a surrogate for consumption (for more details on the sampling frame see ref *7* and the Supporting Information).

Sample collection requests and supplies were distributed on a weekly basis to FSIS inspectors at the specified establishments. Inspectors collected approximately 250 g of back fat from cattle, 250 g of belly fat from hogs, or 250 g of abdominal fat from young chickens and turkeys. The poultry samples were composites from three birds in the same flock. The samples were placed in clean glass jars, frozen overnight, and shipped in sealed boxes to the USDA ARS Biosciences Research Laboratory, Fargo, ND, for analysis.

Individual samples were homogenized, and a subsample (5 g) was analyzed for 17 PCDD/Fs and four non-*ortho*-PCBs (no-PCBs) (no. 77, 81, 126, and 169) according to a method based on EPA Method 1613B (*18*) modified to include the no-PCBs (*19*). A method blank was analyzed with each set of nine survey samples and used for blank subtraction. A known spiked sample was analyzed at least twice each month to provide ongoing quality assurance. Detection limits (DL) were calculated according to the method of Glaser et al. (*20*) as two standard deviations of either method blanks or low level spikes in a clean fat matrix (DL = 2 × SD) and ranged from 0.03 to 0.15 pg/g for all congeners except OCDD (DL = 0.87 pg/g) and PCB-77 (DL = 6.65 pg/g). The elevated DLs for OCDD and PCB-77 were due to the high and variable amounts measured in our method blanks. TEQs were calculated using the WHO 2005 TEFs and reporting nondetects as zero or DL/2. In most cases, the middle-bound data (nd = DL/2) will be used for discussion. All values are expressed in picograms per gram of lipid after gravimetric determination of the lipid content.

Mono-*ortho*-PCBs (mo-PCBs) (no. 105, 114, 118, 123, 156, 157, 167, and 189) were analyzed at the U.S. EPA laboratory, John C. Stennis Space Center, MS, according to a previously published method (*21*). Preliminary results of the mo-PCB analyses will be discussed in this paper.

The TEQ data from each animal class were not normally distributed; therefore, for paired comparisons across slaughter classes and surveys, nonparametric statistical tests (Kolmogorov−Smirnov and Wilcoxon rank-sum) were applied to the unweighted raw data. Significance was set at a p value of <0.05. For comparison

Table 1. Mean PCDD/F, Non-*ortho*-PCB, and Mono-*ortho*-PCB TEQs from Previous USDA Surveys of Meat and Poultry Calculated Using Pre-1994, 1998, or 2005 TEF Values[a]

		mid-1990s survey				2002−2003 survey			
		N	pre-1994 TEFs	2005 TEFs	% change	*N*	1998 TEFs	2005 TEFs	% change
beef	PCDD/F	51	0.867	0.941	8.5	139	0.744	0.713	−4.2
	no-PCB		0.354	0.366	3.5		0.127	0.133	4.7
	mo-PCB		0.091	0.018	−80.2				
pork	PCDD/F	56	1.314	1.380	5.1	136	0.207	0.193	−6.8
	no-PCB		0.038	0.042	11.1		0.024	0.030	25.0
	mo-PCB		0.027	0.005	−81.5				
chicken	PCDD/F	41	1.790	2.044	14.2	151	0.220	0.207	−5.9
	no-PCB		0.187	0.188	0.5		0.072	0.080	11.1
	mo-PCB		0.091	0.021	−76.9				
turkey	PCDD/F	15	0.925	0.974	5.3	84	0.411	0.377	−8.3
	no-PCB		0.446	0.456	2.3		0.177	0.193	9.0
	mo-PCB		0.210	0.046	−78.1				

[a] The percent change in TEQ due to the change in TEF values is also given. All values are expressed as pg/g of lipid with nd = DL/2. *N* = number of samples in each slaughter class.

purposes, the results discussed in this paper do not include PCB-81 in the TEQ sum because it was a minimal contributor and was not reported in the earlier surveys. Additionally, results from the mid-1990s surveys include only data from steers/heifers, market hogs, and chickens/turkeys, which accounted for 81, 72, and 70% of the beef, pork, and poultry sampled in the mid-1990s, respectively. Chickens and turkeys were further separated for comparison.

RESULTS

In two earlier USDA surveys, TEQs were reported on the basis of TEF values existing at the time, the mid-1990s surveys using TEFs established prior to 1995 (*14*, *15*), and the 2002−2003 survey using WHO 1998 TEFs (*16*). The effect of converting average TEQs from these surveys to TEQs based on the recent 2005 TEFs is shown in **Table 1**. When the 2002−2003 survey data were converted from the 1998 to 2005 TEFs, mean PCDD/F TEQs decreased 4−8% in each slaughter class, and the no-PCB TEQs increased 5−25%. If the sum of PCDD/F and no-PCB TEQs (sum-TEQ) is considered, levels decreased 2−4%. The mid-1990s survey data in **Table 1** show the effects of the conversion of TEQs from earlier TEFs (I-TEFs for PCDD/Fs and WHO 1994 TEFs for PCBs) to the 2005 TEFs. The change results in a 5−14% increase in mean PCDD/F TEQs, a slight increase in no-PCB TEQ, a large drop (>75%) in mo-PCB TEQ, and total TEQ changes from −6.6% for turkeys to +8.5% for chicken.

Because the mid-1990s survey data showed mo-PCBs contributed minimally (≤3%) to the total TEQ after conversion to the WHO 2005 TEFs, the mo-PCBs were measured in only those meat and poultry samples for which no-PCB TEQ exceeded a set limit, that is, the 90th percentile of the 2002−2003 survey. These no-PCB TEQ limits were 0.13, 0.29, 0.37, and 0.50 pg/g of lipid for pork, chicken, beef, and turkey, respectively. In preliminary results, mo-PCBs were measured in 15 samples and contributed only an additional 0.1−16.5% (average = 2.4 ± 4.1%) to the total TEQ. Most of these 15 samples were related to a localized PCB contamination that is under investigation, and the results will be discussed in more detail in a future paper.

The average congener concentrations for each slaughter class in the 2008 survey are presented in **Table 2** along with the mean, median, and range of TEQs. The contribution of no-PCBs to the mean sum-TEQ ranged from 13% in market hogs to >40% in turkeys. Nonparametric tests (Kolmogorov−Smirnov and Wilcoxon rank-sum) showed significant differences in the distribution of TEQs across all slaughter classes ($p < 0.0001$). In general, turkeys

Table 2. Mean Concentrations of 17 PCDD/Fs and 4 Non-*ortho*-PCBs, and Mean, Median, and Range of TEQs in 4 Domestic Slaughter Classes from the 2008 Survey[a]

congener	WHO 2005 TEF	beef *N* = 139	pork *N* = 136	chicken *N* = 151	turkey *N* = 84
2378-TCDD	1	0.04 (0.02)	0.02 (0.00)	0.02 (0.00)	0.05 (0.04)
12378-PeCDD	1	0.19 (0.18)	0.05 (0.00)	0.04 (0.00)	0.17 (0.17)
123478-HxCDD	0.1	0.22 (0.22)	0.04 (0.03)	0.03 (0.02)	0.09 (0.08)
123678-HxCDD	0.1	1.34 (1.34)	0.14 (0.12)	0.09 (0.07)	0.41 (0.41)
123789-HxCDD	0.1	0.24 (0.23)	0.06 (0.03)	0.04 (0.01)	0.05 (0.04)
1234678-HpCDD	0.01	3.46 (3.46)	0.93 (0.87)	0.45 (0.44)	0.33 (0.32)
OCDD	0.0003	4.19 (4.00)	4.91 (4.78)	4.18 (4.09)	0.88 (0.50)
2378-TCDF	0.1	0.04 (0.00)	0.05 (0.00)	0.06 (0.03)	0.16 (0.15)
12378-PeCDF	0.03	0.03 (0.01)	0.03 (0.01)	0.03 (0.02)	0.07 (0.06)
23478-PeCDF	0.3	0.11 (0.10)	0.05 (0.01)	0.05 (0.02)	0.14 (0.14)
123478-HxCDF	0.1	0.29 (0.29)	0.07 (0.04)	0.05 (0.02)	0.09 (0.08)
123678-HxCDF	0.1	0.16 (0.16)	0.04 (0.02)	0.04 (0.01)	0.05 (0.04)
234678-HxCDF	0.1	0.15 (0.14)	0.04 (0.01)	0.03 (0.01)	0.04 (0.02)
123789-HxCDF	0.1	0.05 (0.00)	0.05 (0.00)	0.05 (0.00)	0.04 (0.00)
1234678-HpCDF	0.01	0.66 (0.62)	0.26 (0.18)	0.11 (0.03)	0.09 (0.01)
1234789-HpCDF	0.01	0.10 (0.02)	0.10 (0.01)	0.09 (0.00)	0.08 (0.00)
OCDF	0.0003	0.20 (0.14)	0.16 (0.10)	0.11 (0.06)	0.09 (0.03)
PCB-77	0.0001	4.29 (0.33)	4.45 (0.27)	4.52 (0.82)	4.36 (0.91)
PCB-81	0.0005	0.20 (0.06)	0.18 (0.03)	0.22 (0.09)	0.25 (0.13)
PCB-126	0.1	0.96 (0.96)	0.12 (0.05)	0.44 (0.42)	2.18 (2.18)
PCB-169	0.03	0.35 (0.35)	0.15 (0.12)	0.21 (0.17)	0.95 (0.95)
mean PCDD/F TEQ		0.55 (0.51)	0.14 (0.04)	0.12 (0.04)	0.36 (0.34)
mean no-PCB TEQ		0.11 (0.11)	0.02 (0.01)	0.05 (0.05)	0.25 (0.25)
median PCDD/F TEQ		0.31 (0.27)	0.12 (0.02)	0.11 (0.02)	0.31 (0.28)
median no-PCB TEQ		0.09 (0.09)	0.01 (0.00)	0.03 (0.03)	0.14 (0.14)
sum-TEQ range		0.15−4.86 (0.05−4.85)	0.10−1.37 (0.00−1.27)	0.10−1.28 (0.00−1.21)	0.14−4.60 (0.05−4.53)

[a] Middle bound concentrations (nd = DL/2) are reported in pg/g of lipid with lower bound concentrations (nd = 0) in parentheses. TEQs are calculated with WHO 2005 TEFs.

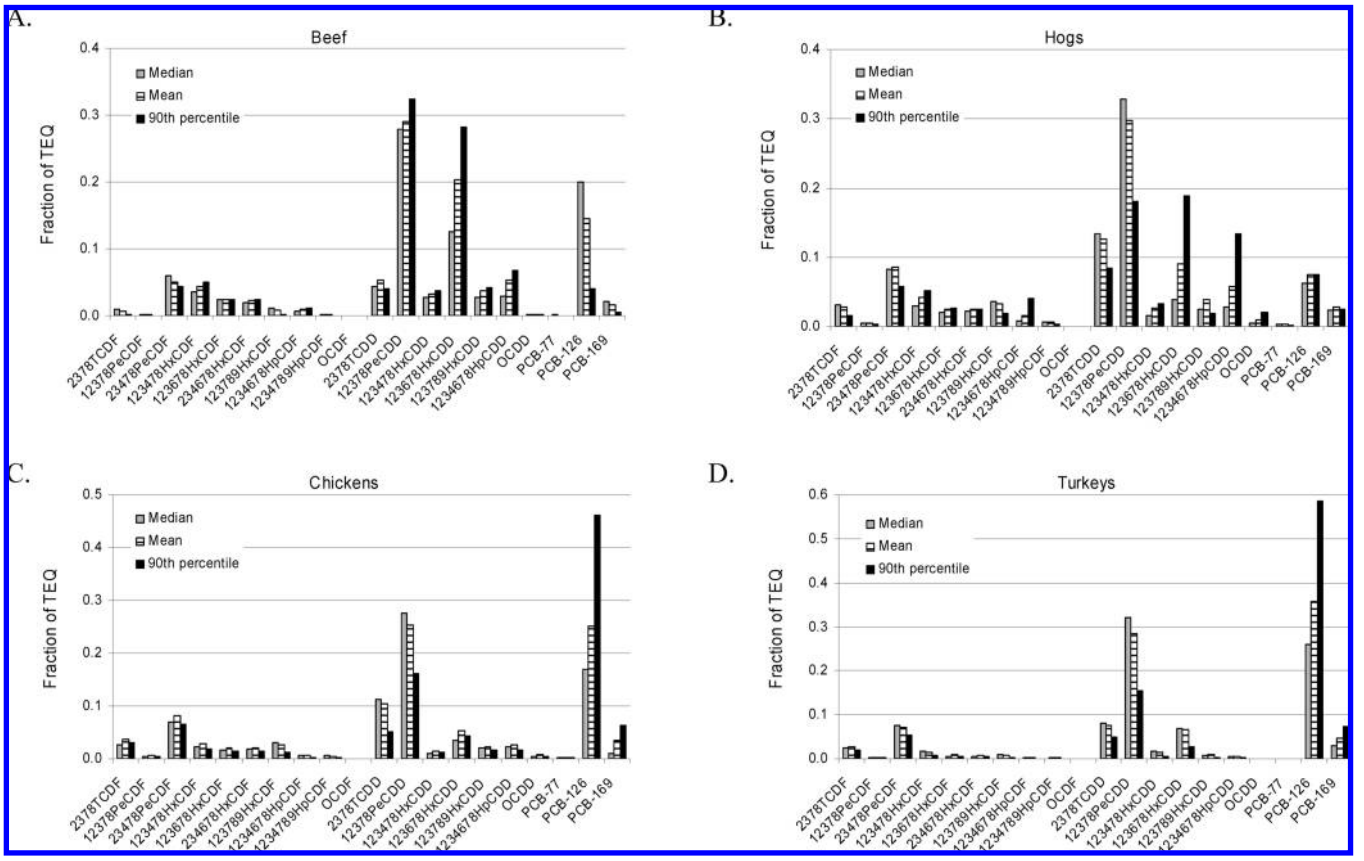

Figure 1. Fractional contribution of each individual congener to sum-TEQ of the median, mean, and 90th percentiles in each slaughter class for the 2008 survey: (**A**) steers and heifers; (**B**) market hogs; (**C**) young chickens; (**D**) young turkeys. Data are calculated with nd = DL/2.

had the highest levels of PCBs in the survey, and beef had the highest PCDD/F levels. Both turkey and beef had a rather wide range of TEQs, with one sample in each class exceeding 4.0 pg/g of lipid. Excluding PCB-77, the concentration of which was driven by its high detection limit, the dominant congeners were OCDD and HpCDD in beef and pork, OCDD, HpCDD, and PCB-126 in chicken, and OCDD, PCB-126, and PCB-169 in turkeys. In addition, 1,2,3,6,7,8-HxCDD was the third most dominant congener in beef. It was detected in 99% of the beef samples, in 96% of the turkey samples, and in > 40% of the pork and chicken samples, making it one of the most prevalent congeners found.

On a toxicity basis, 1,2,3,7,8-PeCDD and PCB-126 contributed most to the overall TEQ averages, with contributions ranging from 25 to 30% and from 8 to 36%, respectively (**Figure 1**). Although TCDD contributed 5−13% to the average TEQs, it was detected in one-third or fewer of the samples in three of the slaughter classes (detections rates: 4% in pork and chicken, 34% in beef, and 62% in turkey). 1,2,3,6,7,8-HxCDD contributed 5−20% to TEQ averages and was a frequently detected congener, as explained above. **Figure 1** also shows individual congener contributions to the median and 90th percentile TEQs. The median patterns are quite similar to the means in most cases. The patterns for the 90th percentile show TEQ redistributions compared to the median and mean patterns. For example, the TEQ contributions from 1,2,3,6,7,8-HxCDD and HpCDD double from the median to the 90th percentile in beef and quadruple in pork, whereas contributions of other congeners correspondingly decrease. This pattern change is mainly driven by the increasing concentrations of 1,2,3,6,7,8-HxCDD and HpCDD, a > 12-fold increase from the median to the 90th percentile. Similarly in poultry, the TEQ contributions of PCB-126 and

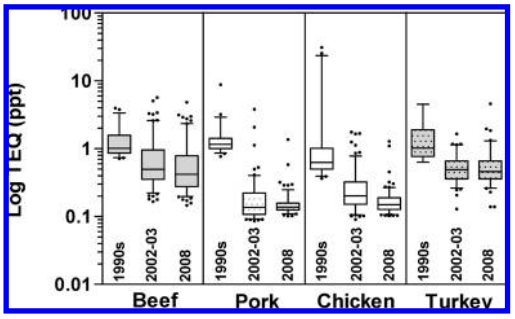

Figure 2. Comparison of data (pg of sum-TEQ/g of lipid, nd = DL/2, WHO 2005 TEFs) from surveys in the mid-1990s, 2002−2003, and 2008 across four slaughter classes. Horizontal lines represent the 5th, 25th, 50th (median), 75th, and 95th percentiles. Dots represent individual values outside the 5th and 95th percentiles.

PCB-169 more than double from the median to the 90th percentile due to a > 8-fold increase in concentrations, and contributions from other congeners (especially TCDD and PeCDD) correspondingly decrease.

Figure 2 indicates a decreasing trend in TEQs from the mid-1990s to 2008. Higher detection limits in the mid-1990s surveys make it difficult to directly compare these data with the later surveys. Hoffman et al. (*7*) concluded that although TEQ levels in poultry and pork were most likely declining from the mid-1990s to 2003, no trend could be established for beef due to the detection limit issue. A statistical comparison of the 2002−2003 and 2008 data was possible because the methodology and detection limits of the two surveys were virtually identical. Because the TEQ data from both surveys and in each animal class were not normally

Table 3. Median TEQs from the 2002−2003 and 2008 Surveys and Probability (*p* Value) from Wilcoxon Rank-Sum Test of the Significance of Each Slaughter Class Sharing Different Underlying Distributions[a]

		PCDD/F TEQ (pg/g of lipid)	no-PCB TEQ (pg/g of lipid)	sum-TEQ (pg/g of lipid)
beef	2002−2003 TEQ	0.36 (0.31)	0.11 (0.11)	0.49 (0.45)
	2008 TEQ	0.31 (0.27)	0.09 (0.09)	0.42 (0.38)
	distribution *p* value	0.02 (0.04)	0.005 (0.005)	0.01 (0.01)
chicken	2002−2003 TEQ	0.13 (0.06)	0.05 (0.05)	0.20 (0.14)
	2008 TEQ	0.11 (0.02)	0.03 (0.03)	0.15 (0.06)
	distribution *p* value	0.03 (<0.001)	<0.0001 (<0.0001)	<0.0001 (<0.0001)
turkey	2002−2003 TEQ	0.29 (0.25)	0.15 (0.15)	0.49 (0.45)
	2008 TEQ	0.31 (0.28)	0.14 (0.14)	0.46 (0.44)
	distribution *p* value	0.34 (0.34)	0.12 (0.12)	0.37 (0.41)
hogs	2002−2003 TEQ	0.12 (0.03)	0.02 (0.02)	0.14 (0.05)
	2008 TEQ	0.12 (0.02)	0.01 (0.004)	0.14 (0.02)
	distribution *p* value	0.06 (0.02)	0.35 (<0.0001)	0.37 (<0.0001)

[a] $p < 0.05$ indicates paired data have different distributions. TEQs are calculated with nd = DL/2 (nd = 0 in parentheses).

distributed, nonparametric tests [i.e., Kolmogorov−Smirnov (K−S) test and Wilcoxon rank-sum test] were used to test whether the 2002−2003 and 2008 samples had the same distribution or were significantly different. Both tests gave similar results. A paired comparison for 2002−2003 and 2008 TEQ data within each slaughter class (**Table 3**) found that the beef and chicken data were independent (not similarly distributed across surveys, $p < 0.05$), and median TEQs had declined 14 and 25% for beef and chicken, respectively, from 2002 to 2008 with nearly equal declines in both PCDD/F and PCB TEQs. The turkey data were similarly distributed ($p > 0.1$), and median TEQs were relatively constant between the two survey time periods (6% decrease in sum-TEQ). The distribution similarity between 2002−2003 and 2008 TEQ data for market hogs depended on the approach used for treating nondetects. Using middle-bound values (nd = DL/2) for the hog TEQ data leads to the conclusion of similar distribution between 2002 and 2003 and 2008 ($p > 0.05$) and median TEQs showing no change.

DISCUSSION

Updating past survey data to the latest TEF system should provide the most current and relevant data for exposure assessments but can lead to confusion due to the fact that many regulations and health recommendations are based on earlier sets of TEFs (*22*). In applying the WHO 2005 TEFs to the data from the USDA surveys conducted in the mid-1990s and 2002−2003, we found PCDD/F TEQs increased from the I-TEF-based data (mid-1990s) and decreased to some extent from the WHO 1998-based data (2002−2003) driven mainly by the TEF changes made for two key congeners, 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF. The 1,2,3,7,8-PeCDD TEF increased from 0.5 to 1.0 in 1998, and the 2,3,4,7,8-PeCDF TEF decreased from 0.5 to 0.3 in 2005. Non-*ortho*-PCB TEQs increased after conversion for both data sets due to the increased toxicity of PCB-169 from 0.01 to 0.03. In the mid-1990s survey data, mono-*ortho*-PCB TEQs showed a dramatic decrease (approximately 80%) due to TEF decreases of nearly an order of magnitude for seven of the eight mo-PCBs. Overall total dioxin-like PCB TEQ decreased 14−28%, and no-PCBs accounted for 90−96% of the newly calculated PCB TEQ. Because PCDD/Fs are the dominant TEQ contributors in U.S. meat and poultry (generally > 70% of TEQ), the sum-TEQ followed the trend of the PCDD/F TEQ, showing small decreases.

Wittsiepe et al. (*23*) showed similar changes due to updating TEF data for human milk and serum samples from Germany. Converting from WHO 1998 to WHO 2005 TEFs decreased the PCDD/F TEQ by approximately 15%, increased the no-PCB TEQ by 8−15%, and decreased mo-PCB TEQ by 88%. Converting from I-TEQs had little effect on the PCDD/F TEQs in the data set. Bhavsar et al. (*24*) also found similar TEQ changes in fish (decreases of 7.5% in PCDD/F TEQ and 25% in PCB TEQ from WHO 1998 TEFs, but little change from I-TEFs and WHO 1994 TEFs) and proposed regression models to predict TEQ changes for the conversion between TEF systems. Given the similar changes observed for these three data sets (U.S. meat and poultry, German blood and milk, and Canadian fish), the proposed regression models may be applicable to a large number of data sets. However, because the total TEQ value is based on individual congener concentrations, the congener pattern of any sample set will strongly influence the change produced by a TEF update. This emphasizes the importance of reporting full congener data when possible (as in **Table 2**). For instance, U.S. meat and poultry TEQs tracked the changes to PCDD/F TEQ, whereas the Canadian fish TEQs tracked the PCB TEQ changes because dioxin-like PCBs accounted for > 60% of the TEQ in those samples.

Even within the 2008 U.S. meat and poultry data set, different congeners contributed differently to the sum-TEQs of each slaughter class (**Figure 1**). Poultry had relatively higher contributions from PCB-126 to the mean than beef and pork. This may reflect different dietary regimens or different metabolizing capabilities between these animal classes. The fact that PCB-126 strongly dominates the TEQ of the top 10% of the sampled poultry (45−60% of the sum-TEQ) and the contribution of PCB-169 steadily increases from the median to the 90th percentile implies an exposure to PCBs in a small portion of the poultry that drives the overall PCB contribution. In fact, a small cluster of turkeys and chickens from this survey are being investigated by the FDA to determine the possibility of a localized PCB contamination. This investigation is ongoing, and results will be published at a later date.

The beef and hog median and mean TEQs are dominated by 1,2,3,7,8-PeCDD, a congener with one of the highest TEFs (TEF = 1). However, the contribution of three other congeners consistently increases from the median through the mean to the 90th percentile. These congeners are 1,2,3,6,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDD, and OCDD. Together these three congeners have been shown to be the major congeners accumulated in cattle (perirenal fat or milkfat) following exposure to pentachlorophenol-treated wood (*25*, *26*). Given their prevalence in the top 10% of beef and hog samples, exposure to pentachlorophenol-treated wood in some animal facilities may be contributing to dioxin levels in these animal classes. Evidence of a connection between pentachlorophenol-treated wood and the PCDD/F patterns observed in survey animals from the mid-1990s has also been proposed (*27*, *28*). Several follow-up investigations resulting from the 2008 survey are exploring this hypothesis.

One important congener that has diminished in its contribution to TEQ is 2,3,4,7,8-PeCDF. Under the I-TEF and WHO 1998 TEF schemes, 2,3,4,7,8-PeCDF had a TEF of 0.5 and contributed 15−20 and 9−14% to the TEQs in the mid-1990s and 2002−2003 surveys, respectively (*3*−*7*). After conversion to the WHO 2005 TEFs, where 2,3,4,7,8 PeCDF has a TEF of 0.3, 2,3,4,7,8-PeCDF contributes 8−12, 5−10, and 5−9% to the sum-TEQs in each of

the three U.S. surveys in the mid-1990s, 2002−2003, and 2008, respectively. Other data sets will be similarly or more greatly influenced by the new TEF value. For example, in a dietary intake study in Finland (*29*), 2,3,4,7,8-PeCDF contributed > 60% to the PCDD/F TEQ intake. Under the 2005 TEF system, both the absolute and relative PCDD/F TEQ contributions of 2,3,4,7,8-PeCDF would decline, resulting in a > 24% decrease in PCDD/F TEQ.

Comparisons of the 2008 data with the previous U.S. survey data show several other commonalities. The average congener patterns show that the most prevalent PCDD/Fs in each survey and slaughter class were OCDD and 1,2,3,4,6,7,8-HpCDD, followed, in most cases, by 1,2,3,6,7,8-HxCDD. The levels of OCDD and HpCDD are not surprising because they are the most dominant congeners found in the U.S. environment (*30*, *31*). The increased prominence of 1,2,3,6,7,8-HxCDD along with OCDD and 1,2,3,4,6,7,8-HpCDD may indicate a possible link to pentachlorophenol that has continued over the 10 year span. Comparison of TEQs (**Figure 2**) shows that in each survey outliers were found that were subsequently traced to inadvertent dioxin exposures through feeds. These include two chicken samples in 1997 contaminated by dioxin-containing ball clay (*12*) and two pork samples in 2002−2003 contaminated by dioxin-containing mineral supplements (*7*). Elevated samples from the 2008 survey are undergoing follow-up investigations led by the FDA for the identification of the potential sources of contamination.

Excluding these outliers, steers and heifers generally had the widest range of TEQs, which is not surprising given that cattle in the United States graze on a wide variety of pastureland across the country, whereas poultry and pork production is typically confined and integrated with large feed suppliers (vertically integrated). In the 2002−2003 survey, heifers had an average TEQ 28% higher than that of steers; however, in the 2008 survey average steer and heifer levels (0.63 and 0.72 pg/g of lipid, respectively) differed by only 13%, suggesting any discrepancy seen in the 2002−2003 data was likely an artifact of the small sample size and not gender differences. Gilts and barrows again showed similar mean TEQs (0.16 and 0.17 pg/g of lipid, respectively).

Comparison of these survey data to recent beef, pork, and chicken data from Europe and Asia required conversion to the WHO 1998 TEF system, under which most of the previous studies have been published (*32*−*37*). Overall, the mean PCDD/F TEQ of all the U.S. samples using WHO 1998 TEFs was 0.30 ± 0.23 pg/g of lipid and the no-PCB TEQ was 0.08 ± 0.08 pg/g of lipid. This PCDD/F TEQ level is similar to the levels found in Europe and Asia, which ranged from 0.02 pg/g of lipid in South Korean chicken (*35*) to 1.56 pg/g of lipid in Belgian beef (*34*) (mean from five countries = 0.35 ± 0.41 pg/g of lipid). The U.S. PCB level was in general lower than that of most other countries, where no-PCB TEQs ranged from 0.01 pg/g of lipid in Dutch chicken (*32*) to 3.34 pg/g of lipid in Belgian beef (*34*) (mean from five countries = 0.45 ± 0.82 pg/g of lipid). It is not known whether the relatively high PCDD/F and PCB levels in the Belgian beef are residual contaminations from the Belgian PCB incident that occurred in 1999 (*11*) or are typical of Belgian beef (the Belgian samples were collected in 2000 and 2001).

Despite the difficulties of comparing data from different laboratories and time periods, data from three USDA surveys show a decreasing trend in TEQ over the past decade for domestic meat and poultry. A more rigorous comparison of the data from the two most recent surveys shows that, in general, both PCDD/Fs and no-PCBs are declining. One exception was increasing PCB levels in turkeys, which was likely due to a localized contamination that is being further investigated, as mentioned above. Similar downward trends of TEQs in foods have recently been reported in The Netherlands (*32*) and Spain (*38*). Likewise, downward trends are suggested by PCDD/F measurements in serum levels of the U.S. population from 1999 to 2004 (*39*). Taken together, it may be that emission regulations and food and feed monitoring programs, which at times have identified and removed adulterated feed components, have resulted in continuing declines of dioxin-like compounds in the food supply. In the United States, the TEQ trends and the individual congener patterns imply that although the same environmental sources may be entering the food chain (i.e., the congener patterns remain similar), the levels are declining over time.

ABBREVIATIONS USED

PCDD, polychlorinated dibenzo-*p*-dioxin; PCDF, polychlorinated dibenzofurans; PCB, polychlorinated biphenyl; TEF, toxic equivalency factor; TEQ, toxic equivalency.

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Supporting Information Available: Tables showing the sampling design by slaughter class and the levels of PCDD/Fs and PCBs in the method blanks and limits of detection for each congener and a more detailed description of the sampling design and analytical method. This material is available free of charge via the Internet at http://pubs.acs.org.

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