

**DIOXINS, DIOXIN-LIKE COMPOUNDS, AND PBDES IN THE U.S. DOMESTIC MEAT SUPPLY: TRENDS AND LEVELS 2003 TO 2013**Lupton, SJ<sup>1\*</sup>, O'Keefe M<sup>2</sup>, Basu P<sup>2</sup>, Muñiz Ortiz J<sup>2</sup><sup>1</sup>USDA-ARS, Biosciences Research Laboratory, 1605 Albrecht Blvd, Fargo, ND, USA;<sup>2</sup>USDA-FSIS, Office of Public Health, 1400 Independence Ave SW, Washington D.C., USA**Introduction**

In the mid-1990s, 2002-2003, 2007-2008, and again in 2012-2013 the U.S. Department of Agriculture (USDA) conducted statistical surveys for dioxin and dioxin-like compound (PCDDs, PCDFs, and PCBs) residues in domestic meat and poultry.<sup>1-5</sup> The surveys were conducted to determine background levels of dioxins and dioxin-like compounds in domestic beef, pork, and poultry, as well as to track temporal trends, and uncover sources of dioxins to the food supply by investigating any unusual findings. Half way through the 2007-2008 survey, polybrominated diphenyl ethers (PBDEs) were analyzed in pooled samples because PBDEs are also found in animal food products, but most data on the levels in U.S. foods are from small market basket studies.<sup>6-8</sup> Again in the 2012-2013 survey, PBDEs were analyzed, however this time each individual sample was investigated. To provide a robust and statistically relevant dataset of PBDEs in domestic meat and poultry, we have now analyzed the 2002-2003, 2007-2008, and 2012-2013 USDA dioxin survey samples for PBDEs. These results not only provide the best assessment of PBDE levels in U.S. meat and poultry, but also, by comparing all three surveys, provide an opportunity to determine if levels have declined in these food groups since the production of PentaBDE and OctaBDE formulations in the U.S. ceased at the end of 2004. Additionally, these data sets being statistically based are able to be used to estimate human exposure to dioxins, dioxin-like compounds, and PBDEs based on consumption information of domestic meat and poultry.

**Materials and Methods**

The 2012-2013 dioxin survey was conducted using the same methodology from the previous surveys completed in 2002-2003 and 2007-2008.<sup>4-5</sup> Briefly, federal inspectors collected adipose tissue samples from U.S. slaughtering facilities on a monthly basis from October 2012 through September 2013. The statistically designed sampling framework represented the major categories of domestic meat and poultry production in the U.S. Operating facilities were sampled proportional to their production volumes and all facilities were eligible for sampling. The survey consisted of a total of 514 samples: 139 beef (heifers and steers), 137 pork (gilts and barrows), 153 young chickens, and 85 young turkeys, with each class weighted according to its production volume. Analyses were completed on 5 g of homogenized adipose tissue samples according to U.S. Environmental Protection Agency (U.S. EPA) Method 1613 (modified to include the *no*-PCBs). A method blank was analyzed with each set of 15 survey samples and used for blank subtraction as was done in the previous surveys. Limits of detection (LOD) were defined by the standard deviation of either method blanks or low level spikes (LOD = 2xSD). TEQs were calculated from the current and previous survey data using the WHO 2005 TEFs and reporting non-detects as zero or LOD/2. All values are expressed in pg/g lipid. Analysis for PBDEs (Congeners 28, 47, 99, 100, 153, 154, and 183) was also completed on each individual sample using U.S. EPA Method 1614 and values are expressed as ng/g lipid.<sup>8</sup>

**Results and Discussion:**

The USDA conducted surveys of PCDD/Fs and *no*-PCBs in beef, pork, and poultry in the mid-1990s with the assistance of the U.S. EPA<sup>3-5</sup> and again in 2002-2003 and 2007-2008 using in-house resources.<sup>2</sup> In the 2007-2008 survey, PBDE analysis was added midway through the survey on pooled samples for beef (N=8), pork (N=6), chicken (N=8), and Turkey (N=5). Also, samples from the 2002-2003 survey were pooled for PBDE analysis later on. The most recent survey conducted was in 2012-2013 for PCDD/Fs, *no*-PCBs, and PBDEs on all 514 individual samples. The mid-1990s and 2002-2003 surveys reported TEQs based on earlier TEF values, from 1993 and 1998, respectively.<sup>5</sup> Values for these surveys were converted to new TEQs using the 2005 TEFs for comparison purposes to the other surveys.<sup>5</sup>

The mean TEQ values (pg/g lipid) for PCDDs/Fs/PCBs and  $\Sigma$ PBDEs (ng/g lipids) are given from each slaughter class in Table 1. From the 2007-2008 survey to the current survey there is a slight increase in beef PCDD/F

TEQs (+1.8%), however all other slaughter classes decreased, market hogs are -28%, young chickens are -8.3% and young turkeys are down -28%. The range for PCDDs/Fs in the 2012-2013 beefs samples was 0.060 to 6.314 pg/g lipid and for the 2007-2008 survey the range was 0.108 to 4.640 pg/g lipid, but the 2012-2013 survey had one high sample at 6.314. If the sample that resulted in 6.134 pg/g lipid is removed, the mean decreases to 0.51 pg/g lipid which is a decrease from the previous survey of -7.2% with a range of 0.060-3.602 pg/g lipid. Beef had the widest range of total TEQs found in the 2012-2013 survey. Given that cattle in the U.S. graze on a wide variety of pastureland across the country, whereas, poultry and pork production is typically confined and vertically-integrated, a larger range of TEQ values is not a surprising observation.

Mean TEQs for the *no*-PCBS have also decreased from 2007-2008 to 2012-2013 for all classes except the market hogs. The increase for *no*-PCBs in market hogs is due to one high PCB sample. The range of *no*-PCBs in the current survey is 0.001 to 1.949 pg/g lipid (mean = 0.03) compared to the 2007-2008 survey which was 0.009 to 0.190 pg/g lipid (mean=0.02), however if the high value 1.949 is removed from the 2012-2013 mean then the new mean would decrease to 0.01 and that corresponds to a 50% decrease in the *no*-PCB mean with a range of 0.001 to 0.048 pg/g lipid. In the 2012-2013 survey, the contribution of *no*-PCBs to the total mean TEQ were 15% in beef and chicken, 23% in pork, and 24% in turkeys, however with the removal of the high pork sample the *no*-PCB contribution in pork becomes 9%. When the contribution of *no*-PCBs to the total TEQ from the 2012-2013 survey is compared to the 2007-2008 survey, all slaughter classes displayed a decrease in the *no*-PCB contribution.

The differences of the mean  $\Sigma$ PBDEs between the 2007-2008 and 2012-2013 surveys were +27% for beef, -31% for market hogs, -77% for young chickens, and -57% for young turkeys. From the whisker plot in Figure 1b we see a downward trend of the median values. A true comparison of the previous surveys and the current one is difficult, due to the previous survey using pooled samples. In figure 2, the wider range of PBDE levels in the current survey can be compared to the range of pooled samples from the 2007-2008 survey and it should be noted that the medians for each class overall have decreased from the 2007-2008 survey.

Using these data, consumer exposure can be estimated for these compounds and compared to the EPA reference dose of 0.7 pg/kg bw-day.<sup>9</sup> Table 2 shows the consumption rates and lipid content of lean and non-lean meat for all production classes. Mean consumption rates can be calculated for PCDD/F/PCB and PBDEs using the lipid content for lean and non-lean meat for all production classes. Mean consumer exposure estimates from meat classes for PCDD/F/PCBs (Table 2) are below the daily reference dose. A comparison of the survey data (Table 1, Figure 1) shows that for all slaughter classes TEQs appear to have declined over the past two decades, but this decline may be slowing down or leveling off in at least one slaughter class. During each survey, outlier values have been identified that do not reflect the general population. In all outlier cases, on-farm investigations were completed by the U.S. Food and Drug Administration (U.S. FDA) to determine contamination sources.<sup>4,5</sup> Most investigations revealed sources (old fence posts and utility poles) that could easily be corrected.

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**Table 1.** Mean values of TEQs (pg/g lipid) for PCDD/Fs and *no*-PCBs, and  $\Sigma$ PBDEs (ng/g lipid) from the 2012-2013 survey of domestic meat and poultry with % change from the 2007-2008 survey. Values assume non-detects = LOD/2 and non-detects = 0 in parentheses.

Slaughter Class	Survey Years	PCDD/F TEQ pg/g lipid	<i>no</i> -PCB TEQ pg/g lipid	$\Sigma$ PBDE ng/g lipid <sup>1,2</sup>
Steers/Heifers N=139	2007-2008	0.55 (0.51)	0.11 (0.11)	0.33 (0.30)
	2012-2013	0.56 (0.53)	0.10 (0.10)	0.42 (0.42)
	% change	+1.8% (+3.9%)	-9.1% (-9.1%)	+27% (+40%)
Market Hogs N=137	2007-2008	0.14 (0.04)	0.02 (0.01)	0.52 (0.51)
	2012-2013	0.10 (0.04)	0.03 (0.03)	0.36 (0.36)
	% change	-28% (0%)	+50% (+200%)	-31% (-29%)
Young Chickens N=153	2007-2008	0.12 (0.04)	0.05 (0.05)	0.78 (0.76)
	2012-2013	0.11 (0.06)	0.02 (0.02)	0.18 (0.18)
	% change	-8.3% (+50%)	-60% (-60%)	-77% (-76%)
Young Turkeys N=85	2007-2008	0.36 (0.34)	0.25 (0.25)	1.76 (1.76)
	2012-2013	0.26 (0.24)	0.085 (0.08)	0.76 (0.76)
	% change	-28% (-29%)	-66% (-68%)	-57% (-57%)

<sup>1</sup> Individual samples for the 2007-2008 survey were analyzed for PBDEs in pools. Number of sampling pools were beef (N=8), pork (N=6), chicken (N=8) and turkey (N=5).

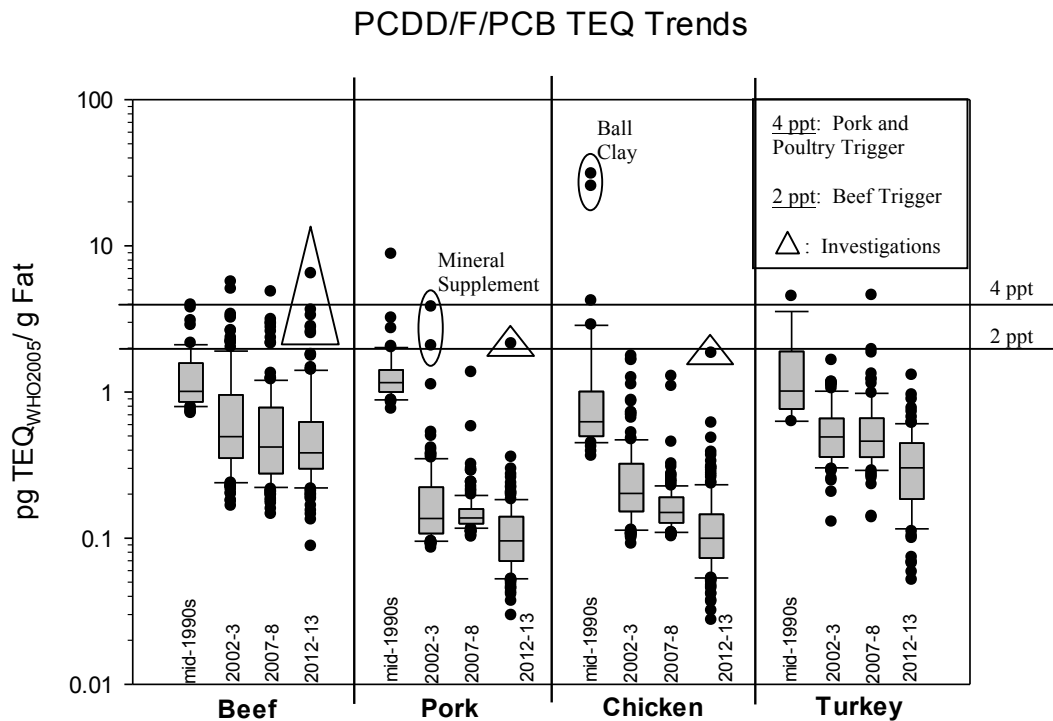
<sup>2</sup>  $\Sigma$ PBDE is the sum of BDEs-28, 47, 99, 100, 153, 154, and 183.

**Table 2.** Exposure assessments based on consumption data and 2012-2013 survey mean concentrations for dioxins/furans/*no*-PCBs and  $\Sigma$ PBDEs.

	Unit	Beef	Pork	Chicken	Turkey
Consumption Rate	g of meat/kg bw-day	0.77	0.39	0.63	0.14
Mean Concentration	D/F/PCBs (pg/g lipid)	0.66	0.13	0.13	0.35
	$\Sigma$ PBDEs (ng/g lipid)	0.42	0.36	0.18	0.76
Non-lean Meat	Non-lean % Lipid	19.24	14.95	15.06	8.02
Non-lean Exposure	D/F/PCBs (pg/kg bw-day)	0.098	0.008	0.012	0.004
	$\Sigma$ PBDEs (ng/ kg bw-day)	0.062	0.021	0.017	0.009
Lean Meat	Lean % Lipid	6.16	5.88	3.08	2.86
Lean Exposure	D/F/PCBs (pg/kg bw-day)	0.031	0.003	0.003	0.001
	$\Sigma$ PBDEs (ng/ kg bw-day)	0.020	0.008	0.003	0.003

Figure 1. Comparison of concentration levels from 4 different surveys for PCDD/F/PCB (A) and 3 different surveys for PBDEs (B). All values are background subtracted, lipid adjusted, and based on  $nd=LOD/2$ .

A)



B)

