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# Polychlorinated Dioxins, Furans, and Biphenyls, and Polybrominated Diphenyl Ethers in a U.S. Meat Market Basket and Estimates of Dietary Intake

JANICE K. HUWE* AND
GERALD L. LARSEN
*Biosciences Research Laboratory, USDA-ARS, P.O. Box 5674 University Station, Fargo, North Dakota 58105*

Persistent environmental contaminants including polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), non-ortho-polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) were analyzed in 65 meat samples collected from supermarkets across the U.S. in 2001. The samples included hamburger, sirloin steaks, pork chops, bacon, and whole chickens from nine different cities. The average PCDD/F/non-ortho-PCB toxic equivalency (TEQ) for all the samples was 0.55 pg/g of lipid (nd (nondetect) = DL (detection limit)/2) with a range from nondetectable to 3.21 pg/g of lipid. For PBDEs, eight congeners were routinely found in the meat samples with an average sum of 1.71 ng/g of lipid (nd = DL/2) and a range from nondetectable to 16.6 ng/g of lipid. While average TEQs were similar to recent values reported in Europe and Asia, the sums of PBDEs in chicken and pork were 3−20 times higher than levels reported in Spain and Japan for these foods. The presence of a few outliers raised the average PBDE sums and indicated that isolated sources of contamination may exist that, if identified, could be removed from the U.S. animal production chain. Using these TEQ and PBDE values and USDA food consumption data, the estimated dietary intake ranges from meat products were 5.3−16.0 pg TEQ and 14.9−44.7 ng of PBDEs/d or 0.1−0.3 pg TEQ and 0.3−0.5 (ng of PBDEs/kg of body mass)/d for an average individual, similar to intakes in other countries.

## Introduction

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) are ubiquitous, persistent, lipophilic pollutants. Polybrominated diphenyl ethers (PBDEs) are commonly used additive flame retardants that are also highly lipophilic and structurally similar to the PCDD/Fs and PCBs. While PCDD/F levels are decreasing in the environment and PCBs are no longer produced, PBDEs continue to be produced, and their levels appear to have increased in the environment and in humans over the past 10−20 years (*1*, *2*). The acute and chronic toxicities of dioxin-like compounds are well documented (*3*), and although PBDEs generally appear to have low acute toxicity, studies have shown developmental and neurological effects due to PBDE exposure (*4*). To better assess the risk from PBDEs, routes and magnitudes of exposure need to be investigated and characterized. Because human exposure to PCDD/Fs is almost entirely through the diet, it seems plausible that exposure to PBDEs may occur by this same route.

A number of studies have shown the presence of PBDEs in food samples and estimated that fish and animal products provide the largest amount of dietary exposure (*1*, *5*−*7*). Fish accounted for the majority of the European dietary intake of PBDEs (30−75%); however, the Canadian study showed meat products contributed 75% of the daily Canadian intake. A recent report on PBDE levels in U.S. retail foods showed a wide range of PBDE levels in fish and meat products; however, only one sample of each food type was analyzed in most cases (*8*). A more complete study on salmon ($N = 700$) has been reported that showed higher levels of PBDEs, PCDD/Fs, and PCBs correlated to certain areas of the world where the fish were raised and to the fish being farm-raised rather than wild (*9*, *10*).

In the U.S., the typical diet contains higher amounts of meat than fish (www.barc.usda.gov/bhnrc/foodsurvey). For PCDD/Fs, approximately 26−49% of human dietary exposure is attributed to meat and poultry, 11−15% to dairy products, and 4−9% to fish (*11*). For this reason, we have focused our investigation on the domestic meat and poultry supply (a meat market basket) to update data on the current levels of PCDD/Fs and PCBs in these retail foods and to begin to define the levels of PBDEs found in these products.

## Materials and Methods

**Samples.** To collect samples from across the U.S., 10 collaborators within the Agricultural Research Service (ARS) were contacted and asked to purchase specific meat and poultry items from large supermarkets in their local area. The cities where collections were made were Brooksville, FL, Richmond, VA, Storrs, CT, State College, PA, Fargo, ND, Miles City, MT, Corvallis, OR; Las Cruces, NM, and Tucson, AZ. All samples were collected in 2001 and shipped frozen to the USDA-ARS laboratory in Fargo, ND, for analysis. The samples were not meant to represent a statistical sampling of U.S. foods, but rather to include common types of meats such as bacon, whole chickens, sirloin steaks, 80% lean hamburger, and pork chops from each location.

**Sample Preparation.** The bacon and hamburger were analyzed whole; for the chickens, steaks, and chops, fat was trimmed and analyzed. PCDD/Fs and PCBs were analyzed together in one analysis, and PBDEs were analyzed using a separate sample portion. Ground bacon or fat trimmings (5 g) were spiked with the appropriate recovery surrogates: either 15 $^{13}$C-labeled PCDD/Fs and 3 $^{13}$C-labeled non-ortho-PCBs (nos. 71, 126, and 169) or 7 $^{13}$C-labeled BDEs (nos. 28, 47, 99, 153, 154, 183, and 209) (Wellington Laboratories, Guelph, ON, Canada, or Cambridge Isotope Laboratories, Andover, MA). The samples were homogenized in methylene chloride, dried with sodium sulfate, and then purified on a Power Prep instrument (Fluid Management Systems, Waltham, MA) for automated dioxin cleanup using jumbo triphasic silica, regular triphasic silica, basic alumina, and carbon cartridges. For PBDE purification, the carbon cartridge was omitted. The recovered product-containing fractions were concentrated into dodecane (10 or 20 $\mu$L) containing the internal standards ($^{13}$C-labeled 1,2,3,4,-TCDD, 1,2,3,7,8,9-HxCDD, and 3,3′,4,5′-TeCB or $^{13}$C-labeled BDEs-77 and -139) (Wellington, Laboratories).

The hamburger (5 g) was spiked with recovery surrogates, mixed with Celite, and extracted in an automated solvent

* Corresponding author phone: (701) 239-1288; fax: (701) 239-1430; e-mail: huwej@fargo.ars.usda.gov.



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**TABLE 1. Concentrations of PCDD/Fs and Non-Ortho-PCBs in Hamburger, Bacon, and Meat Trimmings on a pg/g Lipid Basis[a]**

| | hamburger, $n = 11$ | | bacon, $n = 11$ | | chicken fat, $n = 22$ | | pork fat, $n = 11$ | | beef fat, $n = 10$ | |
|---|---|---|---|---|---|---|---|---|---|---|
| congener | av | range | av | range | av | range | av | range | average | range |
| 2378TCDF | 0.1 (0) | nd | 0.1 (0) | nd | 0.1 (0.1) | nd−0.2 | 0.04 (0) | nd | 0.03 (0) | nd |
| 12378PeCDF | 0.2 (0) | nd | 0.1 (0) | nd | 0.1 (0.02) | nd−0.3 | 0.1 (0) | nd | 0.1 (0) | nd |
| 23478PeCDF | 0.3 (0.2) | nd−1.6 | 0.1 (0.1) | nd−0.4 | 0.1 (0.1) | nd−0.4 | 0.04 (0.01) | nd−0.1 | 0.1 (0.1) | nd−0.2 |
| 123478HxCDF | 0.4 (0.3) | nd−1.6 | 0.2 (0.1) | nd−1.2 | 0.2 (0.2) | nd−2.8 | 0.1 (0.04) | nd−0.3 | 0.3 (0.3) | nd−0.9 |
| 123678HxCDF | 0.4 (0.1) | nd−1.5 | 0.2 (0.1) | nd− 0.6 | 0.2 (0.1) | nd−0.8 | 0.1 (0.04) | nd−0.4 | 0.2 (0.2) | nd−0.7 |
| 234678HxCDF | 0.4 (0.3) | nd−2.1 | 0.1 (0.1) | nd−0.5 | 0.1 (0.04) | nd−0.9 | 0.1 (0) | nd | 0.1 (0.1) | nd−0.2 |
| 123789HxCDF | 0.1 (0.03) | nd−0.3 | 0.1 (0) | nd | 0.04 (0.01) | nd−0.2 | 0.04 (0) | nd | 0.03 (0) | nd |
| 1234678HpCDF | 1.4 (1.1) | nd−6.3 | 0.5 (0.3) | nd−3.5 | 0.7 (0.6) | nd−8.0 | 0.6 (0.5) | nd−2.5 | 0.9 (0.9) | nd−2.0 |
| 1234789HpCDF | 0.2 (0.04) | nd−0.5 | 0.1 (0.02) | nd−0.3 | 0.1 (0) | nd−0.1 | 0.1 (0) | nd−0.1 | 0.1 (0.03) | nd−0.2 |
| OCDF | 0.4 (0.3) | nd−1.6 | 0.1 (0.1) | nd−0.2 | 0.2 (0.2) | nd−2.9 | 0.3 (0.2) | nd−1.9 | 0.3 (0.3) | nd−0.8 |
| 2378TCDD | 0.2 (0.1) | nd−0.6 | 0.1 (0) | nd | 0.1 (0) | nd | 0.1 (0) | nd | 0.1 (0) | nd |
| 12378PeCDD | 0.3 (0.2) | nd−0.8 | 0.04 (0) | nd | 0.04 (0.02) | nd−0.3 | 0.03 (0) | nd | 0.2 (0.2) | 0.1−0.3 |
| 123478HxC DD | 0.5 (0.5) | nd−1.3 | 0.1 (0.1) | nd−0.2 | 0.04 (0.02) | nd−0.3 | 0.1 (0.03) | nd−0.2 | 0.2 (0.2) | 0.1−0.4 |
| 123678HxC DD | 2.5 (2.5) | 0.47−5.6 | 0.1 (0.1) | nd−0.3 | 0.4 (0.4) | nd−6.2 | 0.2 (0.2) | nd−0.6 | 1.4 (1.4) | 0.3−2.8 |
| 123789HxC DD | 0.5 (0.5) | nd−1.4 | 0.1 (0) | nd | 0.1 (0.02) | nd−0.5 | 0.1 (0) | nd | 0.2 (0.20 | nd−0.4 |
| 1234678HpCDD | 7.5 (7.5) | 0.86−21.5 | 0.6 (0.5) | nd−1.1 | 2.6 (2.5) | nd−47.4 | 1.7 (1.7) | nd−12.1 | 6.0 (6.04) | 0.5−21.0 |
| OCDD | 9.1 (5.6) | nd−32.4 | 2.6 (0.5) | nd−5.5 | 9.0 (8.0) | nd−139.6 | 11.4 (10.5) | nd−87. 5 | 12.2 (11.7) | nd−69.3 |
| PeCB-126 | 1.4 (1.5) | nd−3.8 | 0.1 (0) | nd | 0.9 (0.9) | 0.2−1.76 | 0.1 (0.03) | nd−0.2 | 1.2 (1.1) | 0.5−2.7 |
| HxCB-169 | 0.4 (0.3) | nd−1.3 | 0.4 (0.3) | nd−1.2 | 0.4 (0.3) | nd−1.1 | 0.2 (0.1) | nd−0.4 | 0.3 (0.3) | 0.1−0.5 |
| PCDD/F TEQ | 1.3 (1.0) | 0.07−3.1 | 0.3 (0.1) | nd−0.7 | 0.3 (0.2) | nd−2.3 | 0.2 (0.1) | nd−0.4 | 0.6 (0.5) | 0.2−0.9 |
| PCB TEQ | 0.2 (0.2) | nd−0.3 | 0.01 (0) | nd−0.02 | 0.1 (0.2) | 0.02−0.2 | 0.01 (0) | nd−0.02 | 0.1 (0.1) | 0.1−0.8 |
| total TEQ | 1.4 (1.1) | 0.2−3.2 | 0.3 (0.1) | nd−0.7 | 0.4 (0.3) | 0.02−2.4 | 0.2 (0.1) | nd−0.4 | 0.7 (0.6) | 0.2−1.0 |
| % lipid | 18.8 | 11.7−22.0 | 39.1 | 30.7−46.3 | 65.7 | 56.0−71.6 | 51.0 | 37.2−71.6 | 62.6 | 45.2−73.4 |

[a] Sample data are blank-subtracted and corrected for recovery. Values below the detection limit were considered nondetects (nd's) and set to either DL/2 or zero (in parentheses) before averages were calculated.

extractor (Dionex, Sunnyvale, CA) using 2-propanol/hexane/methylene chloride (35:30:35) at 125 °C and 1500 psi. The extract was purified on the Power Prep system as described above. The percent lipid in each sample was determined gravimetrically before application to the Power Prep system.

**Analysis.** A 2 $\mu$L aliquot of the final extract was analyzed for 17 PCDD/Fs and PCBs-77, -126, and -169 according to EPA method 1613A (tetra- through octachlorinated dioxins and furans by isotope dilution HRGC/HRMS, 1994) modified to include the non-ortho-PCBs. PBDEs were analyzed by a modification of a previously described method (*12*). This method was modified to include seven $^{13}$C-PBDE recovery standards, two $^{13}$C-PBDE injection standards (in place of the $^{13}$C-octa-CDE used previously), and quantification of 42 native PBDEs by isotope-dilution methods (at least one from each homologue group). PBDEs were analyzed on an Autospec mass spectrometer (Micromass, Beverly, MA), and PCDD/Fs and PCBs were analyzed on an Autospec Ultima instrument (Micromass). Both isotope dilution methods corrected native analytes for recoveries of the $^{13}$C-labeled standards.

A method blank or a method spike containing the native analytes was run every fifth sample. Because method blanks contained detectable levels of several analytes, all data were blank-subtracted. Limits of detection were calculated from the standard deviations of blanks or low-level spikes in a clean pork fat matrix as described by Glaser et al. (*13*) and as used in a previous survey of PCDD/Fs in meat and poultry (*14*). All values below the detection limit (DL) were treated as nondetects (nd's) and were set equal to either zero or 1/2 the detection limit. Toxic equivalencies (TEQs) were calculated from the 1998 World Health Organization toxicity factors (*15*) as either medium-bound (nd's = DL/2) or lower-bound (nd's = 0) values to reflect the quality of the data.

**QA/QC.** The recoveries of all $^{13}$C-surrogates were between 35% and 150%, except that of BDE-209, which occasionally fell below 20%. All reported analytes showed accuracy and precision better than 20% in the method spikes. At the limit of quantitation (roughly 3.5 times the DL), replicate spiked fat samples demonstrated acceptable accuracy and precision (<25%) for each analyte, and the ratios of each isotope pair were within 15% of the theoretically calculated values.

Our laboratory has successfully participated in interlaboratory comparison studies coordinated by the Norwegian Institute of Public Health for the analysis of PCDD/Fs, PCBs, and PBDEs in food samples using these methods (www.fhi.no).

## Results and Discussion

Tables 1 and 2 show the results from the analysis of 65 meat samples for PCDD/Fs, non-ortho-PCBs, and PBDEs on a lipid-mass basis. Fat trimmings were analyzed for chicken, pork, and beef to expedite the cleanup and because these lipophilic contaminants concentrate into the fats. Dioxins and furans have been shown to equally distribute into inter- and intramuscular fat stores (*16*, *17*), making fat trimmings a practical matrix from which to estimate dioxin levels in retail meat cuts. The high lipophilicity of the PCBs and PBDEs suggests they may behave in a similar manner.

Only the 2,3,7,8-substituted PCDD/Fs were found in the meat samples. Because of a measurable laboratory background for PCB-77 (4.6 ± 2.2 pg), this congener is not reported. Aside from the nine reported PBDE congeners, no other PBDEs were consistently found in the meat samples. Although mono- and disubstituted diphenyl ethers were not recovered from the cleanup procedure, spiked samples showed that 32 other tri- through decasubstituted congeners could be adequately recovered and quantitated by the method. The presence of BDE-209 is uncertain in many cases due to the large amounts found in the blanks (459 ± 493 pg), so it is not included in the PBDE results.

Of the four types of meat included in the study, pork (including bacon) had the lowest average TEQ levels followed by chicken and beef. This is similar to the results of a recent USDA dioxin survey in meat and poultry which showed that beef animals had the highest TEQ levels (0.87 ppt, nd = DL/2) compared to chickens (0.29 ppt, nd = DL/2) and hogs (0.23 ppt, nd = DL/2) (ref *14* and Table 3). The average TEQs found in the retail products were nearly identical to the average TEQs found in the survey, which had analyzed adipose tissues collected directly from slaughtering facilities in 2002−2003. Except for one chicken sample (TEQ = 2.4 ppt), all of the market basket samples fell within the TEQ

**TABLE 2. Concentrations of Major PBDEs in Hamburger, Bacon, and Meat Trimmings on a pg/g Lipid Basis[a]**

| | hamburger, $n = 11$ | | bacon, $n = 11$ | | chicken fat, $n = 22$ | | pork fat, $n = 11$ | | beef fat, $n = 10$ | |
|---|---|---|---|---|---|---|---|---|---|---|
| PBDE no. | av | range | av | range | av | range | av | range | av | range |
| tri-28/33 | 4.3 (2.4) | nd−18 | 3.7 (1.7) | nd−11 | 2.9 (2.1) | nd−12 | 6.5 (5.1) | nd−48 | 2.3 (0.9) | nd−9.1 |
| tetra-47 | 180 (170) | nd−590 | 230 (220) | nd−1380 | 810 (806) | 64−4890 | 1070 (1070) | 72−8130 | 70 (54) | nd−250 |
| penta-85 | 22 (0) | nd−27 | 23 (0) | nd | 47 (36) | nd−310 | 34 (19) | nd−140 | 14 (0) | nd |
| penta-99 | 260 (260) | nd−1030 | 300 (290) | nd−1890 | 1380 (1380) | 86−7870 | 1040 (1040) | 87−6200 | 110 (100) | nd−400 |
| penta-100 | 42 (42) | nd−130 | 41 (40) | nd−260 | 280 (280) | 14−1610 | 180 (180) | 19−1170 | 20 (16) | nd−69 |
| hexa-153 | 100 (98) | nd−560 | 78 (73) | nd−420 | 220 (220) | 24−1010 | 120 (120) | nd−350 | 19 (16) | nd−60 |
| hexa-154 | 29 (28) | nd−96 | 42 (41) | nd−260 | 88 (88) | nd−620 | 87 (87) | 10−290 | 11 (9.9) | nd−34 |
| hepta-183 | 29 (17) | nd−61 | 120 (110) | nd−380 | 130 (130) | nd−680 | 84 (77) | nd−270 | 12 (0) | nd |
| ∑(tri−he pta) | | | | | | | | | | |
| mean | 670 (610) | nd−2470 | 840 (790) | 100−4620 | 2960 (2940) | 190−16620 | 2620 (2600) | 190−16330 | 250 (200) | nd−840 |
| median | 380 (330) | | 420 (370) | | 1500 (1490) | | 1000 (980) | | 170 (110) | |
| % lipid | 20.2 | 16.1−24.6 | 39.1 | 30.7−46.3 | 65.7 | 56.0−71.6 | 51.0 | 37.2−71.6 | 62.6 | 45.2−73.4 |

[a] Sample data are blank-subtracted and corrected for recovery. Values below the detection limit were considered nondetects (nd's) and set to either DL/2 or zero (in parentheses) before averages or medians were calculated.

**TABLE 3. Comparison of PCDD/F and Non-Ortho-PCB TEQs from This Market Basket Study and Other Studies (pg/g of lipid)[a]**

| | TEQ | U.S., 2001 | U.S. survey, 2002−3 | MS, U.S., 1994 | EU, 1997−2003 | Greece, 2002 | Spain, 2000 | Belgium, 2000−1 | Taiwan, 2003 | Korea, 2001 |
|---|---|---|---|---|---|---|---|---|---|---|
| beef | PCDD/F | 0.64 | 0.78 | | 0.46[c] | 0.55 | 0.45 | 1.56 | 0.97 | |
| | PCB | 0.11 | 0.13 | | 0.80[c,d] | 0.41 | | 3.34 | 0.36[d] | 0.38 |
| hamburger | PCDD/F | 1.54 | | 0.86 | | | | | | |
| | PCB | 0.15 | | | | | | | | |
| pork | PCDD/F | 0.37 | 0.29 | 0.99[b] | 0.21 | 0.39 | 0.31 | 0.17 | 0.31 | |
| | PCB | 0.01 | 0.02 | | 0.23[d] | 0.69 | | 0.02 | 0.14[d] | 0.20 |
| chicken | PCDD/F | 0.37 | 0.29 | 0.78 | 0.65[e] | 0.30[e] | 1.32 | 0.35 | 0.73 | |
| | PCB | 0.10 | 0.07 | | 1.22[d,e] | 0.06[e] | | 0.43 | 0.77[d] | 0.25 |
| *N* | | 11 or 22 | 136−151 | 3 | 12−25 | 3 or 4 | 4 composites (10 each) | 25−48 | 12 or 18 | 30−66 |
| nd's = | | DL | DL | DL | DL | DL | DL/2 | 0 | DL | DL |
| ref | | this work | *14* | *18* | *19* | *20* | *21* | *22* | *23* | *24* |

[a] TEQ values are calculated using 1998 WHO toxic equivalency factors. Mono-ortho-PCBs are not included in PCB TEQs unless noted. The number of samples analyzed in each study is given (*N*), and the method used to treat nondetected congeners is also reported. [b] Pork sausage. [c] Ruminants, may include ruminants other than beef animals. [d] Includes all 12 dioxin-like PCBs. [e] Poultry, may include poultry other than chicken.

ranges of the previous survey. This indicates that processes related to retail food marketing do not substantially contribute dioxins, furans, or non-ortho-PCBs to the food supply.

Table 3 also compares this market basket data with selected data from other countries and an earlier market basket study in the U.S. Because of the low number of samples from the previous U.S. market basket study (*18*), it is difficult to draw any firm conclusions about temporal trends in U.S. retail meats. PCDD/F levels may have declined by 50% or more in retail pork and chicken from 1994 to 2001, but PCDD/Fs did not appear to decline in hamburger. This trend was also observed in the USDA survey of beef, pork, and poultry (*14*). Compared to recent European and Asian data (*19*−*24*), TEQs are not notably different given that various methods were used to calculate detection limits in these studies. Non-ortho-PCBs contributed less than 20% to the TEQ in each of the U.S. meats, but more than 40% in several European meats. Some of the European PCB levels may reflect residual contamination from a PCB-contamination incident that occurred in Belgium in 1999 (*25*), especially the Belgian beef samples. It may also be indicative of higher environmental levels of PCBs in certain parts of Europe and Asia.

The major PCDD/F congeners contributing to the TEQ in the U.S. meats were 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF. Each of these congeners contributed approximately 10−30% of the total TEQ in each meat type. In the European meats, these two congeners contributed somewhat more to the TEQ, 20−50%. 1,2,3,6,7,8-HxCDD was another significant contributor in U.S. beef (20%). 2,3,4,7,8-TCDD was not detected in 95% of the samples in this market basket and only contributed to the TEQ when nondetects were not considered as zero. A similar congener pattern was also observed in the USDA survey of meat and poultry (*14*).

Unlike the PCDD/Fs and PCBs, beef appeared to have the lowest amounts of PBDEs, while chicken and pork had the highest (Table 2). Two explanations for this difference are (1) that the source of PBDE exposure for livestock is different from the source of dioxin exposure, which is generally thought to be from environmental fallout onto forages and feed or (2) that ruminants, such as cattle, absorb less or metabolize and excrete more PBDEs than other livestock species, resulting in lower body burdens. Little or no data on PBDE levels have been collected from slaughter house meat samples or from animal feeds and forages, and no ruminant metabolism work has been conducted, making it impossible to speculate as to the source(s) of the PBDEs found in the meats. Hites et al. (*9*) have shown that fish feed can be contaminated with PBDEs and may be related to elevated concentrations in farm-raised fish.

Inspection of the individual samples and the mean and the median values for the PBDE data show that a few high samples were driving the average upward. One pork and two chicken samples were found to have PBDE sums >15 ng/g of lipid. The two chickens were collected at the same time and location and were the same commercial brand. The presence of these outliers suggests that if isolated sources of PBDEs exist and can be tracked down, PBDEs can be removed from the food production chain.

**TABLE 4. Comparison of Average Sums of PBDEs from This Market Basket Study and Other Studies (ng/g of lipid)[a]**

| | PBDE ∑ | U.S., 2001 | TX, U.S., 2003 | U.S., 1997 | Sweden, 1999 | Spain, 2000 | Japan, 2001−2 |
|---|---|---|---|---|---|---|---|
| steak/hamburger | five major[b] | 0.42 | | | | | 0.32[f] |
| | tetra−octa | 0.46 | 0.58[c] | | | 0.29 | |
| pork/bacon | five major[b] | 1.59 | | | | | 1.26[f] |
| | tetra−octa | 1.73 | 1.64[c] | | | 0.60 | |
| chicken | five major[b] | 2.78 | | 12.9 | | | 0.13[f] |
| | tetra−octa | 2.95 | 4.62[d] | 13.7 | | 0.25 | |
| *N* | | 21 or 22 | 1 or 4 | 13 | | 2 composites (10 each) | 2 |
| all meats | five major[b] | 1.60 | | | 0.36 | | |
| | tetra−octa | 1.71 | 2.35[d] | | | 0.57[e] | |
| *N* | | 65 | 9 | | NR | 15 composites (10 each) | |
| nd's = | | DL/2 | 0 | DL/2 | DL/2 | DL/2 | NR |
| ref | | this work | *8* | *12* | *1* | *7* | *5* |

[a] The number of samples analyzed in each study is given (*N*), and the method used to treat nondetected congeners is also reported. [b] Includes BDEs-47, -99, -100, -153, and -154. [c] Includes two tri and deca congeners. [d] Summation without tri or deca congeners was estimated from the graphs. [e] Estimated 15% lipid for all meats from paper. [f] Estimated 5% lipid for each meat type. NR = not reported.

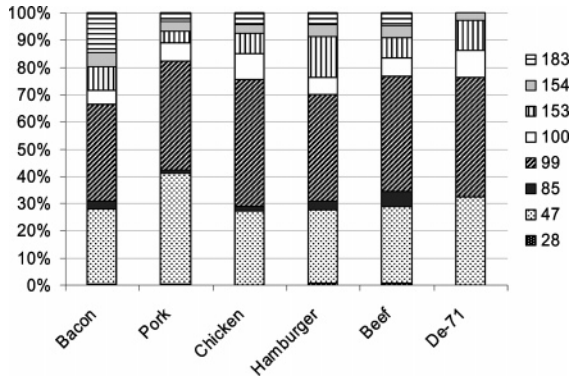


**FIGURE 1. Contribution of individual PBDEs to the sum in various meats and a penta-BDE commercial formulation (DE-71) analyzed in our laboratory. Values are for lipid-adjusted concentrations with nondetects set to DL/2.**

Table 4 compares the average sum of selected PBDEs found in this market basket with data from Europe and other U.S. studies and with estimated data from Japan. For all beef samples, the sums on a lipid-mass basis were similar to those of meat products analyzed in Sweden (*1*) and in a previous U.S. study (*8*) and slightly higher than levels found in Spain (*7*) and estimated from Japanese data (*5*).

The average lipid-mass total in pork and bacon was comparable to the value found previously in the U.S. and estimated in Japan but almost 3 times higher than that of pork products from Spain. Total PBDEs in U.S. chickens averaged 10−20 times higher than those in chickens from Spain or Japan. The PBDE content of chickens in this study was lower than that of chicken(s) analyzed in two previous U.S. studies (*8*, *12*). While PBDE levels may have decreased between 1994 and 2001, the differences may merely reflect the small number of samples in each study and the large variability that can occur between samples.

The average PBDE pattern for each meat type is shown in Figure 1, along with a typical pattern from a penta-BDE commercial formulation (DE-71) analyzed in our laboratory. For meat samples, BDEs-47 and -99 were the largest contributors to the PBDE sum, and the ratio of BDE-47 to BDE-99 was generally less than unity (average 0.78, range 0.3−3.0). This pattern strongly resembles the penta-BDE formulation shown in Figure 1, which has a BDE-47/BDE-99 ratio of 0.4. A similar congener pattern has been seen in other animal products from Canada (*6*), Japan (*5*), and the U.S. (*8*, *12*). In contrast, the congener patterns in fish and seafood (*5*, *8*, *9*) and human samples (*5*, *6*, *26*) generally show a ratio of BDE-47 to BDE-99 greater than 2. For populations who are not largely fish eaters, this implies that sources in addition to diet may be contributing to their PBDE body

**TABLE 5. Estimated Dietary Intakes from This Meat Market Basket Using PCDD/F TEQs, Non-Ortho-PCB TEQs, and Tri- to Hepta-PBDE Sums with nd = DL/2, the USDA Continuing Survey of Food Intakes by Individuals (1994−96), and Low or High Fat Content Meat Types (5% or 15% Lipid)**

| | | lean meats, 5% lipid | | | higher fat meats, 15% lipid | | |
|---|---|---|---|---|---|---|---|
| | consumption[a] (g/d) | PCDD/F TEQ (pg/d) | PCB TEQ (pg/d) | PBDE sum (ng/d) | PCDD/F TEQ (pg/d) | PCB TEQ (pg/d) | PBDE sum (ng/d) |
| beef | 24 | 0.68 | 0.13 | 0.3 | 2.04 | 0.39 | 0.9 |
| pork | 10 | 0.11 | 0.01 | 1.3 | 0.33 | 0.02 | 3.9 |
| chicken | 21 | 0.29 | 0.11 | 3.1 | 0.87 | 0.32 | 9.3 |
| sausage, wieners, etc. | 21 | 0.50[b] | 0.08[b] | 1.8[b] | 1.50[b] | 0.25[b] | 5.4[b] |
| mixed meat products | 99 | 2.38[b] | 0.40[b] | 8.4[b] | 7.14[b] | 1.19[b] | 25.2[b] |
| sum | | 4.6 | 0.73 | 14.9 | 13.8 | 2.17 | 44.7 |

| | lean meats, 5% lipid | | | higher fat meats, 15% lipid | | |
|---|---|---|---|---|---|---|
| | PCDD/F TEQ (pg/kg of bm) | PCB TEQ (pg/kg of bm) | PBDE sum (ng/kg of bm) | PCDD/F TEQ (pg/kg of bm) | PCB TEQ (pg/kg of bm) | PBDE sum (ng/kg of bm) |
| daily intake[c] | 0.09 | 0.01 | 0.28 | 0.26 | 0.04 | 0.84 |
| monthly intake[c] | 2.6 | 0.41 | 8.4 | 7.8 | 1.23 | 25.2 |

[a] Based on data for all individuals. [b] Calculated using the average levels of all meat groups. [c] Based on an average 53 kg of body mass (bm) for all individuals.

burdens or that PBDE congener pattern changes occur in humans via selective uptake from foods, selective elimination, or differential metabolism.

To estimate dietary intakes from meats, food consumption data from the USDA Continuing Survey of Food Intakes by Individuals, 1994−96 (www.barc.usda.gov/bhnrc/foodsurvey) were paired with the TEQ and PBDE data collected in this study. Because fat trimmings were analyzed in this market basket, data on the typical fat content of various meats were obtained from the USDA (www.nal.usda.gov/fnic/foodcomp). Table 5 shows the results of two scenarios: one from a diet of low-fat meat products (e.g., skinless chicken and trimmed lean beef and pork, 5% lipid) and one from higher fat meats (e.g., skin-on chicken, 80% lean hamburger or sausage, and untrimmed beef and pork, 15% lipid). The PCDD/F TEQ intakes from the higher fat meat and poultry were similar to intakes found in a recent U.S. total diet survey for meat and poultry products (6.7 (pg TEQ/kg of body mass)/month) (*11*) and to values reported for adults in other countries for these meat products (12.1 and 10.4 pg TEQ/d) (*21*, *22*). The estimated non-ortho-PCB TEQ intakes were lower from meats in this study than reported for Belgian adults (16.2 pg TEQ) (*22*). The PBDE input estimates from this meat market basket ranged from 15 to 45 ng/d or from 8.4 to 25.2 (ng/kg of body mass)/month for an average individual (53 kg of body mass, all age categories). This intake will vary on the basis of different consumption patterns related to age and gender. The daily adult intakes from meat and meat products in Canada (33 ng/d) (*6*) and Spain (20.2 ng/d) (*7*) are in the range of the U.S. intake calculated here.

In conclusion, although dioxin TEQs are similar in many meat types around the world, higher levels of PBDEs are present in certain U.S. meats compared to European and Japanese meats. This may reflect the larger amount of penta-BDE being used in North America (www.bsef.com). How PBDE contaminants enter the food chain is unknown, but may include animal feeds (e.g. fish meal), environmental inputs (housing or litter), or even food-packaging materials. Results of this study combined with U.S. food consumption data estimated an average PCDD/F TEQ intake from meat and poultry in agreement with that from a recently published total diet study in the U.S. and provided a first estimate of PBDE dietary inputs from U.S. meats and poultry.

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## Supporting Information Available

Two tables providing data on PCDD/F, non-ortho-PCB, and major PBDE concentrations for all individual meat samples. This material is available free of charge via the Internet at http:/pubs.acs.org.

## Literature Cited

(1) Darnerud, P. O.; Eriksen, G. S.; Jóhannesson, T.; Larsen, P. B.; Viluksela, M. Polybrominated diphenyl ethers: occurrence, dietary exposure, and toxicology. *Environ. Health Perspect.* **2001**, *109* (1), 49−68.

(2) de Wit, C. An overview of brominated flame retardants in the environment. *Chemosphere* **2002**, *46*, 583−624.

(3) Schecter, A., Ed. *Dioxins and Health*; Plenum: New York, 1994.

(4) Darnerud, P. O. Toxic effect of brominated flame retardants in man and in wildlife. *Environ. Int.* **2003**, *29*, 841−853.

(5) Ohta, S.; Ishizuka, D.; Nishimura, H.; Nakao, T.; Aozasa, O.; Shimidzu, Y.; Ochiai, F.; Kida, T.; Nishi, M.; Miyata, H. Comparison of polybrominated diphenyl ethers in fish, vegetables, and meats and levels in human milk of nursing women in Japan. *Chemosphere* **2002**, *46*, 689−696.

(6) Ryan, J. J.; Patry, B. Body burdens and food exposure in Canada for polybrominated diphenyl ethers (BDEs). *Organohalogen Compd.* **2001**, *51*, 226−229.

(7) Bocio, A.; Llobet, J. M.; Domingo, J. L.; Corbella, J.; Teixidó, A.; Casas, C. Polybrominated diphenyl ethers (PBDEs) in foodstuffs: human exposure through the diet. *J. Agric. Food Chem.* **2003**, *51* (10), 3191−3195.

(8) Schecter, A.; Päpke, O.; Tung, K.; Staskal, D.; Birnbaum, L. Polybrominated diphenyl ethers contamination of United States food. *Environ. Sci. Technol.* **2004**, *38* (20), 5306−5311.

(9) Hites, R. A.; Foran, J. A.; Schwager, B. A.; Knuth, M. C.; Carpenter, D. O. Global assessment of polybrominated diphenyl ethers in farmed and wild salmon. *Environ. Sci. Technol.* **2004**, *38* (19), 4945−4949.

(10) Hites, R. A.; Foran, J. A.; Carpenter, D. O.; Hamilton, M. C.; Knuth, B. A.; Schwager, S. J. Global assessment of organic contaminants in farmed salmon. *Science* **2004**, *303*, 226−229.

(11) South, P.; Egan, S. K.; Troxell, T.; Bolger, P. M. Dietary PCDD/PCDF exposure estimates for the U.S. population. *Organohalogen Compd.* **2004**, *66*, 2695−2701.

(12) Huwe, J. K.; Lorentzsen, M.; Thuresson, K.; Bergman, X. Analysis of mono- to deca-brominated diphenyl ethers in chickens at the part per billion level. *Chemosphere* **2002**, *46*, 635−640.

(13) Glaser, J. A.; Foerst, D. L.; McKee, G. D.; Quave, S. A.; Budde, W. L. Trace analyses for wastewaters. *Environ. Sci. Technol.* **1981**, *15* (12), 1426−1435.

(14) Huwe, J. K.; Hoffman, M.; Deyrup, C.; Hulebak, K.; Larsen, G.; Zaylskie, R.; Lorentzsen, M.; Clinch, N. A survey of PCDD/Fs and co-planar PCBs in the US meat and poultry supply in 2002−2003. *Organohalogen Compd.* **2004**, *66*, 1942−1946.

(15) Van den Berg, M.; Birnbaum, L. S.; Bosveld, A. T. C.; Feeley, M.; Giesy, J. P.; Hanberg, A.; Hasegawa, R.; Kennedy, S. W.; Kubiak, T.; Larsen, J. C.; van Leeuwen, F. X. R.; Liem, A. K. D.; Nolt, C.; Peterson, R. E.; Poellinger, L.; Safe, S.; Schrenk, D.; Tillitt, D.; Tysklind, M.; Younes, M.; Wærn, F.; Zacharewski, T. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. *Environ. Health Perspect.* **1998**, *106* (12), 775−792.

(16) Ferrario, J.; Bryne, C. The concentration and distribution of 2,3,7,8-dibenzo-*p*-dioxins/furans in chickens. *Chemosphere* **2000**, *40*, 221−224.

(17) Feil, V. J.; Huwe, J. K.; Zaylskie, R. G.; Davison, K. L.; Anderson, V. L.; Marchello, M.; Tiernan, T. O. Chlorinated dibenzo-*p*-dioxins and dibenzofurans concentrations in beef animals from a feeding study. *J. Agric. Food Chem.* **2000**, *48* (12), 6163−6173.

(18) Fiedler, H.; Cooper, K. R.; Bergek, S.; Hjelt, M.; Rappe, C. Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF) in food samples collected in Southern Mississippi, USA. *Chemosphere* **1997**, *34*, 1411−1419.

(19) Gallani, B.; Verstraete, F.; Boix, A.; Von Holst, C.; Anklam, E. Levels of dioxins and dioxin-like PCBs in food and feed in Europe. *Organohalogen Compd.* **2004**, *66*, 1893−1900.

(20) Papadopoulos, A.; Vassiliadou, I.; Costopoulou, D.; Papanicolaou, C.; Leondiadis, L. Levels of dioxin and dioxin-like PCBs in food samples on the Greek market. *Chemosphere* **2004**, *57*, 413−419.

(21) Llobet, J. M.; Domingo, J. L.; Bocio, A.; Casas, C.; Teixidó, A.; Müller, L. Human exposure to dioxins through the diet in Catalonia, Spain: carcinogenic and non-carcinogenic risk. *Chemosphere* **2003**, *50*, 1193−1200.

(22) Focant, J. F.; Eppe, G.; Pirard, C.; Massart, A. C.; De Pauw, A. E. Levels and congener distributions of PCDDs, PCDFs, and non-*ortho* PCBs in Belgian foodstuffs. Assessment of dietary intake. *Chemosphere* **2002**, *48*, 167−179.

(23) Hsu, M. S.; Wang, S. M.; Chou, U.; Chen, S. Y.; Huang, N. C.; Liao, G. Y.; Yu T. P.; Ling Y. C. Levels of dioxin-like compounds in Taiwan foodstuffs. *Organohalogen Compd.* **2004**, *66*, 2111−2116.

(24) Kim, M.; Kim, S.; Yun, S.; Lee, M.; Cho, B.; Park, J.; Son, S.; Kim, O. Comparison of seven indicator PCBs and three coplanar PCBs in beef, pork, and chicken fat. *Chemosphere* **2004**, *54*, 1533−1538.
(25) van Larebeke, N.; Hens, L.; Schepens, P.; Covaci, A.; Baeyens, J.; Everaert, K.; Bernheim, J. L.; Vlietinck, R.; De Poorter, G. The Belgian PCB and dioxin incident of January-June 1999: exposure data and potential impact on health. *Environ. Health Perspect.* **2001**, *109* (3), 265−273.
(26) Hites, R. A. Polybrominated diphenyl ethers in the environment and in people: a meta-analysis of concentrations. *Environ. Sci. Technol.* **2004**, *38* (4), 945−956.

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