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PCBs in Food

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Abstract

We measured the concentrations of 205 polychlorinated biphenyl (PCB) congeners in 26 food items: beef steak, butter, canned tuna, catfish, cheese, eggs, french fries, fried chicken, ground beef, ground pork, hamburger, hot dog, ice cream, liver, luncheon meat, margarine, meat-free dinner, milk, pizza, poultry, salmon, sausage, shrimp, sliced ham, tilapia, and vegetable oil. Using Diet History Questionnaire II, we calculated the PCB dietary exposure in mothers and children participating in the AESOP Study in East Chicago, Indiana, and Columbus Junction, Iowa. Salmon had the highest concentration followed by canned tuna, but fish is a minor contributor to exposure. Other animal proteins are more important sources of PCB dietary exposure in this study population. Despite the inclusion of few congeners and food types in previous studies, we found evidence of a decline in PCB concentrations over the last 20 years. We also found strong associations of PCB congener distributions with Aroclors in most foods and found manufacturing by-product PCBs, including PCB11, in tilapia and catfish. The reduction in PCB levels in food indicates that dietary exposure is comparable to PCB inhalation exposures reported for the same study population.

INTRODUCTION

Food has long been considered the major source of polychlorinated biphenyls (PCBs) exposure.[1, 2] However, it is uncertain whether it is still true, particularly in the U.S. where commercial uses of PCBs have ceased for almost fifty years.[1, 3, 4] With the exception of seafood, there are few studies reporting PCB levels in foods sold in the U.S. While some of these studies reported a small number of PCB congeners in a wide range of foods, other

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Supplementary Material

Supplementary material related to this article (including food samples detail, GC/MS method detail, QA/QC results, individual concentrations of 205 PCB congeners in 26 foods, statistical analysis results, and the detail of the scoping review) can be found at https://, and the dataset can be found at https://doi.org/10.25820/data.001112.

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studies reported all 209 PCB levels in a small variety of foods.[1, 5–8] Some studies analyzed hundreds of samples across the country but quantified only 3 – 4 PCB congeners.[9–11]

The presence of any PCB congener in any food item has implications for human health because each PCB congener has specific physicochemical properties that affect bioaccumulation, bioavailability, and toxicities.[12–15] PCBs cause adverse human health effects by disrupting immune, reproductive, nervous, and endocrine systems.[16, 17] The International Agency for Research on Cancer (IARC) categorizes PCBs as group 1 human carcinogens.[18] In the U.S., PCBs were produced and sold as mixtures of 50–100 congeners called Aroclors. Although their commercial production was banned in the 1970s, hundreds of thousands of kilograms of these PCB mixtures are still contaminating the environment.[18–20] Moreover, some manufacturing processes, including the manufacture of paints and dyes, inadvertently produce some PCB congeners as by-products and contribute to environmental contamination.[21–25] These manufacturing by-product PCBs may be present in food. Analysis of the full set of PCB congeners is therefore important to examine the prevalence of manufacturing by-products PCBs and the full extent PCB contamination in the environment and in food.

Here we report one of the most comprehensive studies of PCBs in food since the 2000s. We measured 205 PCB congeners (represented as 171 chromatographic separations) in 26 food items purchased in a rural community in Iowa far from known significant PCB sources. We evaluated the trends of PCB levels in foods over the last twenty years and calculated the congener-specific PCB dietary exposure for mother-and-child cohorts in the U.S. Midwest.

MATERIALS AND METHODS

This research is part of the Airborne Exposures to Semi-volatile Organic Pollutants (AESOP) Study, a rural and urban community-based study assessing exposures to 209 PCBs in cohorts of mothers and children. The study has collected and reported exposure and health data from nearly 400 ethnically-diverse, urban/rural residential participants in East Chicago (EC), Indiana and Columbus Junction (CJ), Iowa since 2006. The details of the AESOP Study were described in Ampleman et al. (2015).[26] Starting in 2015, we administered to all participants the Diet History Questionnaire (DHQ) II developed by the U.S. National Cancer Institutes, National Institutes of Health (NIH).[27] DHQ II consists of 134 food items and their portion sizes. In our survey, additional location-specific dietary questions asked about fish consumption from local surface waters and locally-caught game. Of all 146 participants in the dietary survey, 93 participants from EC were 47 mothers, 27 girls, and 19 boys; and 53 participants from CJ were 27 mothers, 14 girls, and 12 boys. The participants were enrolled from middle and high schools in EC and CJ. The majority of the study population in both locations were Hispanic White (71% in EC and 68% in CJ) followed by African American (21% in EC) and non-Hispanic White (8% in EC and 32% in CJ).

Food Item Selection

The analysis of current PCB levels in foods that are commonly consumed is the key to precisely estimating contemporary PCB dietary exposures in our AESOP Study participants. We selected only food items that were most likely to contribute to dietary PCB exposures in

our participants for PCB analysis. First, we identified over forty food items commonly consumed by our study population from the diet histories. Next, we estimated the potential for each food item to contribute to dietary PCB exposures. We determined this using our participants' food intake and historical reports of PCB levels in those foods. For the latter, we referred to PCB levels in foods reported by the Bureau of Chemical Safety (BCS), Health Canada and selected the data for Toronto and Winnepeg in the mid-1990s (Table S1).[26, 28] This report was referred to because the geographical characteristics of a city by the Great Lakes of Toronto and a city in the Great Plain of Winnipeg are similar to EC and CJ, respectively. Using this approach, we prioritized and selected 26 food items for PCB analysis: beef steak, butter, canned tuna, catfish (farmed in China), cheese, eggs, french fries, fried chicken, ground beef, ground pork, hamburger, hot dog (including buns), ice cream, liver (beef and chicken), luncheon meat (ham and turkey breast), margarine, meat-free dinner (a mixture of frozen macaroni-and-cheese and tortillas), milk (2%), pizza, poultry, salmon (wild caught in Russia and processed in China), sausage, shrimp (farmed in Indonesia), sliced ham, tilapia (farmed in China), and vegetable oil (soybean oil). Locally caught fish was not selected because only one AESOP Study participant reported occasional consumption of locally caught fish. For each of the 26 food items, one or two specimen(s) of the most affordable items in the same stores and restaurants where our participants shop was purchased on April 21, 2018 or June 7, 2019 (See Table S2). Within four hours of purchase, we removed the samples from their packaging and homogenized each whole food in a food processor. The homogenate was transferred to residue-free amber glass jars (100 g jar^{-1}) with polytetrafluoroethylene (PTFE)-lined polypropylene caps (I-Chem Research, Hayward, CA, USA) and frozen at −20°C until extraction.

Sample Extraction

Each food item was analyzed in triplicate. About 15–20 g of foods were weighed, mixed with 20 g of combusted diatomaceous earth (DE, Thermo Fisher Scientific), and spiked with 1 ng each of 3,5-dichlorobiphenyl (PCB14; AccuStandard, New Haven, CT, USA), 2,3,5,6-tetrachloro(2',3',4',5',6'−2H5)biphenyl (d-PCB65; Cambridge Isotope Laboratories, Andover, MA, USA), and 2,3,4,4',5,6-hexachlorobiphenyl (PCB166; AccuStandard) as surrogate standards (SS) to assess laboratory efficiency .The lipophilic components were extracted using pressurized and heated solvent (ASE350, Thermo Fisher Scientific) first with hexane:acetone (1:1 v/v) and then with hexane (pesticide grade; Fisher Chemical, Fair Lawn, NJ, USA). The combined solution was concentrated under a stream of nitrogen and water was removed with brine and/or Na_2SO_4. Then, PCBs in sample extracts were separated from lipid matrix by gel permeation chromatography (GPC). The samples were eluted through 60 g of Bio-Beads S-X3 (40–80 μm styrene-divinylbenzene beads with 3% cross-linkage; Bio-Rad Laboratories, Los Angeles, CA, USA) packed in 2.54 cm interior diameter glass column with dichloromethane:hexane (1:1 v/v; pesticide grade; Fisher Chemical). The first fraction of 175 mL was discarded, and the following 75 mL was collected for PCB analysis. The remaining interferences were removed from sample extracts by washing with concentrated sulfuric acid (Fisher Chemical), passing through sulfuric acid:silica gel (1:2 w/w) columns (Flash Chromatography Grade; 70–230 Mesh; Fisher Chemical), and eluting with hexane. Finally, the samples were concentrated to 1 mL and spiked with 1 ng each of internal standards (IS): 2,4,6-trichloro(2',3',4',5',6'−2H5)biphenyl

(d-PCB30; Cambridge Isotope Laboratories) and 2,2',3,4,4',5,6,6'-octachlorobiphenyl (PCB204; AccuStandard). For butter, cheese, and margarine, 5 g of starting material was used instead of 20 g. For vegetable oil, 5 g was spiked with the SS without pressurized extraction but was passed though the GPC column twice. 20 g of DE was used as method blank. In the analysis of National Institute of Standards and Technology (NIST) Standard Reference Material (SRM 1994; New York/New Jersey waterway sediment), 0.10 g was analyzed in quintuplicate.

Instrument Analysis, Quantification, and Statistical Methods

Gas chromatography (GC) with mass spectrometry (MS) was employed for identification and quantification (Agilent 7890B GC equipped with Supelco SPB-Octyl capillary column, 30 m, 0.25 mm i.d., 0.25 μm film thickness; coupled with Agilent 7000D Triple Quadrupole (QqQ) MS). The samples were analyzed in positive electron impact (EI) mode in multiple reaction monitoring (MRM) mode (Table S3–S5). PCB masses in samples were quantified for 205 PCB congeners (171 chromatographic peaks) by the internal standard method by comparing with those in the calibration standard solution (1 ng mL^{-1}) containing 209 PCB congeners and isotope-labeled surrogate and internal standards. PCB congener names are in accordance with the U.S. Environmental Protection Agency (US EPA).[29] PCB dietary exposures were calculated using total PCB concentrations in 26 foods and individual consumption rates from the dietary survey. Statistical analyses were computed in the R statistical computing environment (version 4.0.0).[30] A significance level of 0.05 is used throughout this report. Dietary exposure to PCBs was calculated as the product of AESOP participants' food consumption rates and total PCB concentrations in foods.

Quality Assurance and Quality Control

The quantification of PCB levels in foods is challenging because the matrices differ. Our method efficiently extracted a small quantity of PCBs from matrices that contain a high and variable lipid content. We assessed the capability of our method through the recovery of SS, the limits of quantification (LOQ), the analysis of National Institute of Standards and Technology (NIST) Standard Reference Material (SRM 1994; New York/New Jersey waterway sediment), and the reproducibility of individual congener concentrations using median ($\tilde{x}$) and arithmetic mean ± standard deviation ($\bar{x} \pm SD$). The extraction efficiency was represented by the recoveries of SS injected in every sample (Figure 1 and Table S6–S7). The SS recoveries ranged from 48.5% to 123.1% ($\tilde{x}$ = 93.2%; $\bar{x} \pm SD$ = 91.5% ± 12.9%). We did not observe any pattern for SS recoveries by food type. We used the SS recoveries to correct the PCB masses in samples and method blanks. The LOQs were obtained from the analysis of 10 method blanks and calculated using the upper end of the 95% confidence interval ($\overline{X} + t_{0.95} \times SD$) (Figure 1 and Table S8). The LOQs ranged from 2.3 to 578.6 ($\tilde{x}$ = 22.9; $\bar{x} \pm SD$ = 57.4 ± 84.5) pg $sample^{-1}$. We assumed a value of zero for congeners whose masses in all triplicates were below LOQs. For congeners that had one or two of the triplicates above LOQs, we substituted the replicate(s) below LOQs with LOQs over the square root of 2 ($LOQ/\sqrt{2}$).[31] PCB masses were then converted to concentrations. The reported PCB concentrations are the mean of the triplicates. In analysis of SRM, our method yielded PCB recoveries ranging from 61.9% to 152.0% ($\tilde{x}$ = 106.6%; $\bar{x} \pm SD$ = 101.2% ±

40.9%) when compared with concentrations reported by NIST (Table S9). The overall concentrations are comparable indicating our method is accurate and representative. We made no correction for SRM recoveries. The reproducibility was represented by the coefficient of variation (CV) of PCB-congener concentrations from the triplicates (Figure 1 and Table S10). The CV are between 1.0% and 158.4% ($\tilde{x}$ = 42.9%; $\bar{x} \pm$ SD = 47.0% ± 31.2%). As expected, the variation of CV increased with the decrease of concentrations in food items. Our analytical method shows efficient extraction, small interferences, and reproducible results; thereby giving reliable picogram-scale PCB concentrations in foods representative of the diet of our study participants. 127 PCB congeners (as 103 peaks) out of 205 PCB congeners (as 171 peaks) were detected in the foods in this study. The individual PCB congener concentrations can be found in the Supporting Information, Table S10, and the full dataset including associated metadata can be found in Saktrakulkla et al. (2020).[32] Our study was designed to reduce the uncertainty in the analytical measurements but was not designed to assess PCBs in all foods or PCB variability in different items.

Scoping Review

No comprehensive review of PCBs in food has been published, despite the common assumption that dietary sources are important sources of human exposure to PCBs. To explore the overall picture of PCB levels in foods, we conducted a scoping review using the six-stage methodological framework proposed by Arksey and O'Malley (2005) and refined by Levac et al. (2014) and the Joanna Briggs Institute (2017).[33–35] In summary, we focused our attention on the PCB levels in foods relevant to 26 food items in this study that has been reported in the U.S. since 2000. We started by developing a set of search strings to identify all English language peer-reviewed journal articles of PCB levels in foods since 2000 but excluded other types of PCB studies (e.g. toxicity, environmental matrix, or wild animal). The literature search was conducted on September 30, 2019 through two databases PubMed (n = 1,135) and Web of Science (n = 1,831). We also included 6 government reports: 3 reports from the U.S. Department of Agriculture (USDA) and 3 years from the Canadian BCS (2000–2002).[10, 11, 28] The reports from BCS were included because they were the most complete dataset of PCB levels in food in North America, and because the similarity between the origins of foods in U.S. and in Canada. After excluding duplicate publications (n = 2,023), we then reviewed every abstract to screen relevant studies.[36] The result was 192 reports worldwide relevant to foods in this study. We then conducted full-text review of 8 research articles of foods in the U.S., 6 government reports, and 11 research articles of fish in multiple regions. The studies of fish collected outside the U.S. were included because the canned and frozen fish products consumed by our study population can be imported. Three reports of PCBs in salmon that used the same dataset were excluded. Finally, 22 reports were extracted for the total PCB concentrations and the number of PCB congeners studied. The Arksey and O'Malley's methodological framework and the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) flow diagram of the scoping review of PCB levels in foods are in Table S16 and Figure S4, respectively. The search string and the list of articles can be found in Saktrakulkla et al. (2020).[32]

RESULTS AND DISCUSSION

The Studies of PCB Levels in Foods in The United States

According to our scoping review, there are 192 worldwide reports of PCB levels in relevant foods published since 2000; however, there are only 11 reports of PCB levels in foods in the U.S. (Figure S5). Most studies focused on fish (marine or fresh water and farmed or wild caught) followed by meats (beef, pork, and poultry), dairy, and chicken eggs (Figure S6). Two-thirds of the 192 reports studied foods in Europe and another quarter in Asia (Figure S5). Only 8 research articles reported PCB levels of foods in the U.S.[1, 5, 7, 37–41] and another 8 multi-regional studies of fish provided relevant results.[6, 8, 42–47] Although another 3 reports by BCS and 3 reports by USDA are included,[9–11, 28] totaling 22 reports, the number of studies of PCB levels in foods relevant to the U.S. is surprisingly small.

The number of PCB congeners studied is inconsistent among reports. We conducted a full-text review of the 22 reports (Table 1). Of 22 reports, there are 3 studies that quantified a full set of PCB congeners: in salmon by Hites et al. (2004), in shrimp by Fillos et al. (2012), and in yellowfin tuna by Nicklisch et al. (2017).[6–8] Half of the 22 studies reported only 5 to 20 PCB congeners. BCS reported an 11-consecutive-year study of total PCB concentration of 40 congeners in 50 food items.[28] This was the most complete dataset of PCB levels in food in North America although the study was conducted in 1992 – 2002. Schecter et al. (2010) reported the levels of PCBs in 31 food items from Texas.[1] Although considering only 7 PCB congeners, this study is the most recent study in the U.S. that covers a wide variety of foods. McKelvey et al. (2010) reported the total concentrations of 101 PCB congeners in 19 different fish species sampled from Chinese neighborhoods in New York City.[39] Hoffman et al. (2006), Huwe et al. (2009), and Lupton et al. (2017) analyzed hundreds of beef, pork, chicken, and turkey across the country as a part of a quinquennial survey of the USDA. However, only 3 – 4 congeners were reported.[9–11] Chen et al. (2017) reported 19 congeners in 8 brands of milk from California.[41] Of 22 studies, 8 studies report PCB concentrations as PCB mass per gram of lipid, and six of which did not provide the %lipid content for conversion back to PCB mass per gram of wet weight.[7, 9–11, 37, 42, 44, 47] One article reported PCB levels only as their toxicological equivalent (TEQ) which requires approximation to convert back total PCB levels.[43, 48]

PCB Levels and Congener Distribution in Foods

We found the total PCB concentrations in all 26 foods in this study to be below 400 pg g^{-1} WW (Figure 2). Salmon (wild caught) had the highest concentration (380 pg g^{-1} WW). However, this level is about one-tenth of that in wild salmon reported in a large-scale salmon study by Hites et al. (2004) and that in salmon by Schecter et al. (2010) but is comparable to those reported by BCS (2000 – 2002).[1, 6, 28] This finding suggests a decrease of total PCB concentration in wild salmon with time. The second highest total PCB concentration was found in canned tuna (330 pg g^{-1} WW). The level was in the same order of magnitude as that reported in a recent large-scale yellowfin tuna study by Nicklisch et al. (2017) but was lower than most previous reports.[1, 5, 8, 28] Marine fish is clearly contaminated with PCBs at higher levels than the other foods. Among the livestock-products, beef steak had the highest concentration (290 pg g^{-1} WW) followed by butter (270 pg g^{-1} WW) and fried chicken (210

pg g^{-1} WW). While the concentration in beef steak remains at similar levels when compared with other previous reports, butter and fried chicken are lower.[1, 5, 28, 42] Catfish (180 pg g^{-1} WW) and ground beef (120 pg g^{-1} WW) were the other two foods that had total PCB concentrations above 100 pg g^{-1} WW. The remaining 19 food items had total PCB concentrations below 100 pg g^{-1} WW. None of the vegetable-source foods (vegetable oil, french fries, and margarine) had a concentration above 25 pg g^{-1} WW.

Our findings suggest that the PCB levels in food are decreasing. We examined the change of PCB level in food by comparing the total PCB concentrations in foods in this study with those in 22 studies found from our scoping review (Table S13). Using Wilcoxon signed-rank test for statistical analysis, we found that the PCB levels in food in our study are significantly lower than those in 9 previous reports.[1, 5, 9–11, 28, 37] The decrease is not significant in 2 previous reports because only 3 – 4 foods were reported, so the powers were likely to be too low to detect significant differences.[43, 46] Statistical comparisons could not be made with the remaining 11 studies because only one food was reported. Furthermore, using Spearman rank correlation test, we found that the ranks of total PCB concentrations in foods we measured were similar to those reported by BCS (2000 – 2002)[28] and Schecter et al. (2001 and 2010)[1, 5] (Table S14). In the remaining 17 studies, too few food items were reported to make any conclusion about relative rankings of PCB concentration by food type. The limited variety of foods analyzed by other studies, different analytical techniques used, variable number of congeners analyzed, and different sampling locations precluded additional statistical comparisons of PCB concentrations in food over time.

We found that the lower-chlorinated PCBs were less than 20% of the total PCBs measured in most foods (Figure 2). This is interesting because approximately half of the total PCBs sold as Aroclor mixtures were mono-, di-, and tri-chlorinated PCBs.[14, 15, 18] The low levels of lower-chlorinated PCBs in foods may be due to their preferential metabolism in animals, weak bioaccumulation in plants and animal feed, and/or removal from food processing equipment and facilities in the decades since Aroclor production was halted.[14, 15, 18, 49, 50] The lower-chlorinated PCBs can also be lost to volatilization during heating and cooking.[51, 52] Indeed, processed foods such as canned tuna, butter, and fried chicken contain a lower fraction of the lower-chlorinated PCBs than the other foods.

We found strong associations with PCB congener distributions of Aroclors in most foods despite the low levels of lower-chlorinated PCBs. We used Pearson correlation coefficient (R) to evaluate the association between PCB congener distributions in foods and Aroclor (Table S11) and the association among foods (Table S12). Most foods showed significant association with the Aroclors that contained a larger portion of higher-chlorinated PCBs (e.g. Aroclors 1254 and 1260).[14, 15, 18] Three foods with the highest total PCB concentrations (salmon, canned tuna, and beef steak) had the congener distributions most similar to Aroclor 1254 ($R > 0.70$). We also found some similarities of PCB congener distributions among food items: between salmon and canned tuna ($R = 0.94$); across butter, ground beef, and hamburger ($R > 0.89$); and between liver and sausage ($R = 0.97$). The PCB concentrations and the number of measured PCB congeners in the other foods were too small to provide reliable comparisons.

PCB congeners produced as by-products are present in catfish, tilapia, and french fries (Table S10). PCB11, PCB51, and PCB68 are markers of manufacturing by-product PCBs because they are absent (less than 0.5%) in most Aroclors[12, 13] but commonly observed in the environment as by-products of some chemical manufacturing processes (e.g. paint pigments and polymer resin).[21, 22, 24, 25, 53, 54] Recent studies showed that one PCB11 metabolite, 3,3'-dichlorobiphenyl-4-ol (4-OH-PCB11), inhibited cell proliferation, increased oxidative stress, and disturbed mitochondrial respiration and fatty acid metabolism.[55, 56] Other toxicities of manufacturing by-product PCBs and their mechanisms are yet to be studied. We found that the concentration of PCB11 in tilapia (farmed, frozen; 63.3 pg g^{-1} WW) was the highest among manufacturing by-product PCB congeners in this study. PCB11 can be rapidly metabolized and has a very short half-life (about 2 hours in rat organs).[57, 58] An accumulation of PCB11 in catfish, tilapia, and potato suggested 2 possible contamination sources: (1) continuous exposure during farming, or (2) exposure during the manufacturing process. While tilapia and french fries contained one manufacturing by-product PCB congener, catfish (farmed, frozen) contained PCB51 and PCB68 in addition to PCB11. Manufacturing by-product PCBs have been speculated to be the major source of PCBs in the environment in the near future,[59] and our findings confirm that that they contribute to human exposure through the diet.

Salmon had the highest toxicological equivalent (TEQ) and neurotoxicity equivalents (NEQ) among foods (Figure 3). Congener-specific concentrations of 205 congeners in foods enabled us to assess 2 known PCB toxicities. First is the toxicity mediated through the aryl hydrocarbon receptor (AhR) of 12 dioxin-like PCBs. The other is neurotoxicity of 86 PCB congeners including 2 manufacturing by-product PCB congeners (PCB11 and PCB51). We calculated TEQ and NEQ values in foods using the 2005 WHO toxicological equivalent factor[60] and the neurotoxic equivalent factor (NEF) recently updated by Pradeep et al.[61], respectively. Salmon showed a much higher TEQ value (48 pg g^{-1} fat) than any other food. The NEQ value was also highest in salmon (120 pg g^{-1} WW) followed by canned tuna (100 pg g^{-1} WW) and beef steak (96 pg g^{-1} WW). The ranks of NEQ values in foods were in concordance with total PCB concentrations (Figure 3) because a larger number of PCB congeners were used in the calculation. If only the 12 dioxin-like PCBs were considered, only salmon would be a food of concern. However, congener-specific concentrations of 86 PCBs showed that other foods were also potential risks of neurotoxicity.

PCB Dietary Exposure in AESOP Study Subjects

While PCB dietary exposures varied among groups of children, those of mothers are comparable between our two study communities (Figure 4 and Table S15). Using total PCB concentrations in 26 foods that we measured and using individual consumption rates from the survey with DHQ II, we calculated PCB dietary exposures in 6 groups of AESOP Study participants (EC mothers, girls, and boys; and CJ mothers, girls, and boys) together with median ($\tilde{x}$) and arithmetic mean ± standard deviation ($\bar{x} \pm SD$). EC boys showed the highest PCB dietary exposure ($\tilde{x}$ = 20; $\bar{x} \pm SD$ = 26 ± 18 μg $year^{-1}$) among the 6 groups of participants. The next highest were mothers: EC mothers ($\tilde{x}$ = 14; $\bar{x} \pm SD$ = 19 ± 19 μg $year^{-1}$) and CJ mothers ($\tilde{x}$ = 15; $\bar{x} \pm SD$ = 20 ± 19 μg $year^{-1}$). Children in other groups showed lower PCB dietary exposure than mother groups: EC girls ($\tilde{x}$ = 11; $\bar{x} \pm SD$ = 24 ± 31 μg year

$^{-1}$), CJ girls ($\tilde{x}$ = 14; $\bar{x} \pm SD$ = 16 ± 11 μg year^{-1}), and CJ boys ($\tilde{x}$ = 10; $\bar{x} \pm SD$ = 18 ± 19 μg year^{-1}). The differences were due to the participants' reports of their food choices and consumption rates, and PCB content in those foods. EC boys reported consuming higher amounts of meat dietary sources with relatively higher PCB concentrations than the other groups.

Meat was the most important source of PCB dietary exposures in AESOP Study participants (Figure 4 and Table S15). We categorized dietary sources into 4 groups: meat, dairy, fish, and other. We found that meat dietary sources contributed about 50% of PCB dietary exposures followed by dairy (about 25%), fish (about 15%), and other (about 10%) dietary sources. The distributions of PCB dietary sources were similar among the 6 groups of AESOP Study participants. This finding confirmed our previous hypotheses[26] that fish plays a less important role than meat and dairy dietary sources in PCB dietary exposures in this U.S. Midwest study population.

Dietary and inhalation exposure to PCBs are similar in magnitude in AESOP Study participants (Figure 4 and Table S15). We have confirmed this important finding through our partnership with two Midwestern communities who shared their dietary patterns and allowed us to place air samplers in their homes, use of highly comparable data sets of congener-specific measurements of food and air, and use of consistent methods for calculation of both dietary and inhalation routes.[26, 62] This coordinated approach enabled us to directly compare PCB dietary exposure and inhalation exposure. We found that PCB dietary exposures (ranging from 1.2 to 120 μg year^{-1}) are comparable with PCB inhalation exposures (ranging from 0.2 to 160 μg year^{-1})[26] and with PCB inhalation exposures from school air (ranging from 0.7 to 116 μg year^{-1})[62]. Although diet remains a significant route of PCB exposure, its importance when compared with inhalation is likely to decline with the decrease of PCB levels in food.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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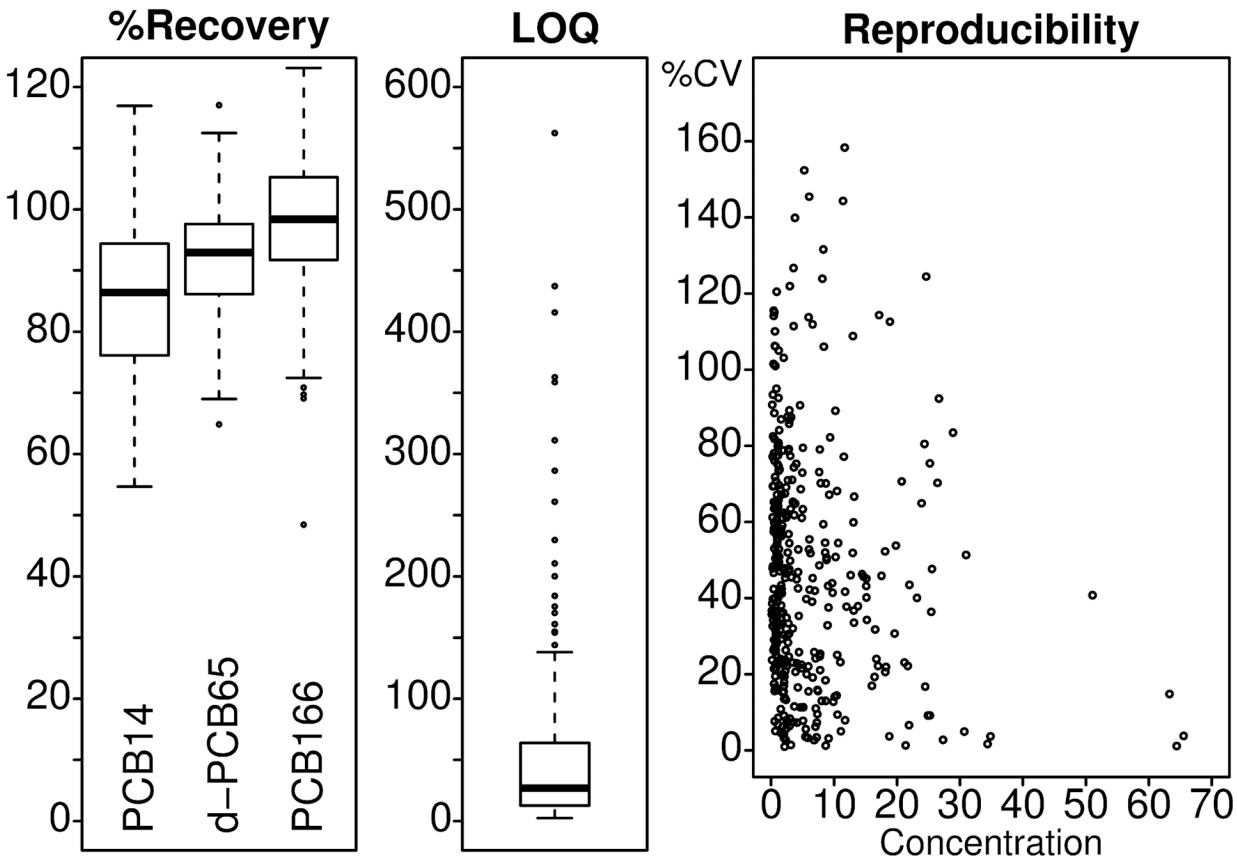

Figure 1.

Quality control metrics. The performance of our analytical method is assessed by the extraction efficiency for 3 surrogate compounds (%Recovery, left); congener-specific limits of quantification in 10 method blanks (LOQ in pg sample^{-1}, middle); and the reproducibility of triplicate analysis (Reproducibility, right) as the coefficient of variation (%CV) versus concentration (pg g^{-1} WW).

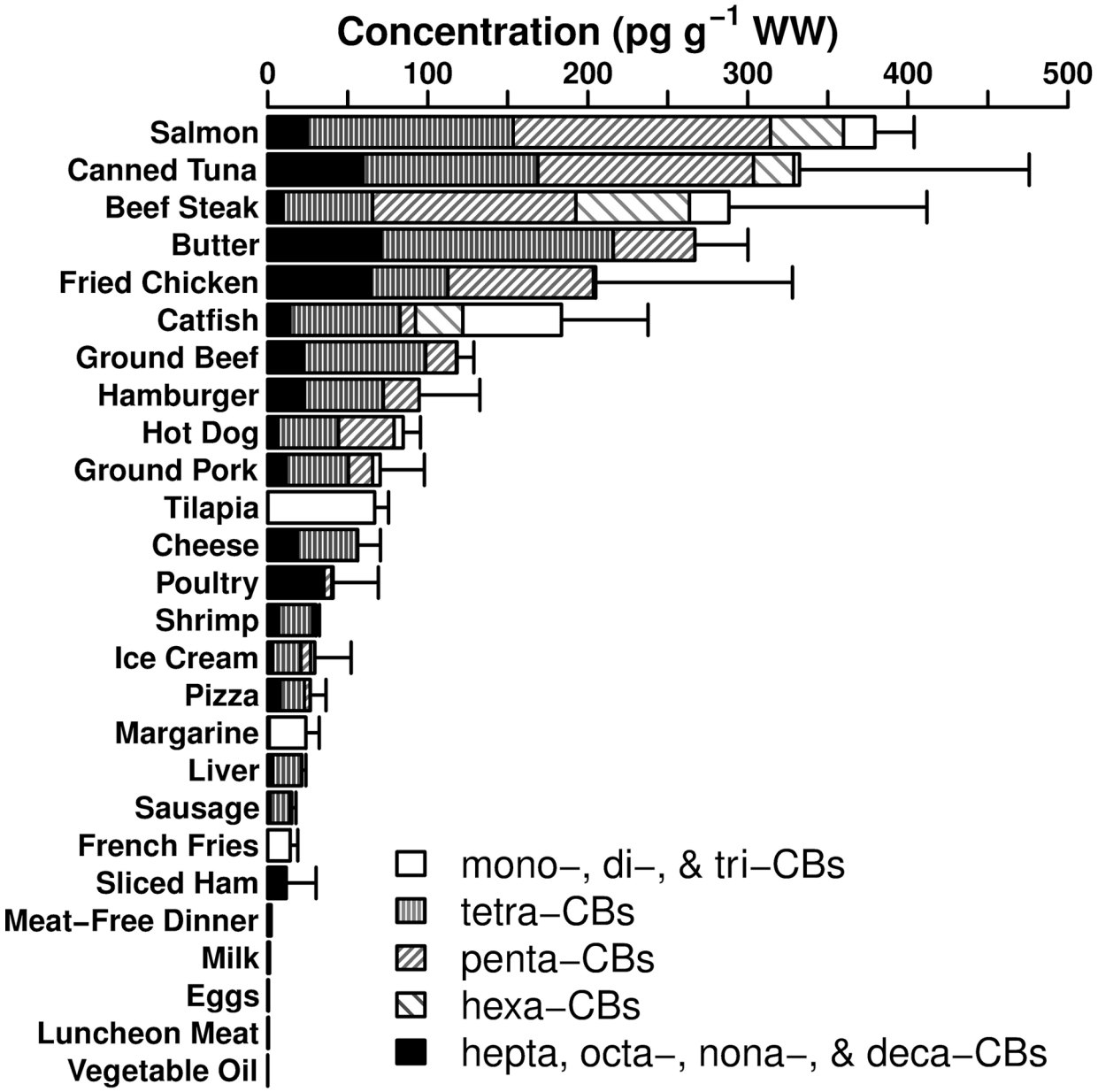

Figure 2.

PCB concentrations in foods (pg g^{-1} WW), calculated as the sum of the mean of the congeners measured in triplicate in each food item. The error bars indicate the standard deviation. Salmon was wild caught frozen; catfish and tilapia were farmed frozen; liver was beef and chicken; meat-free dinner was a mixture of frozen macaroni-and-cheese and tortillas; milk was 2%; and luncheon meat was ham and turkey breast.

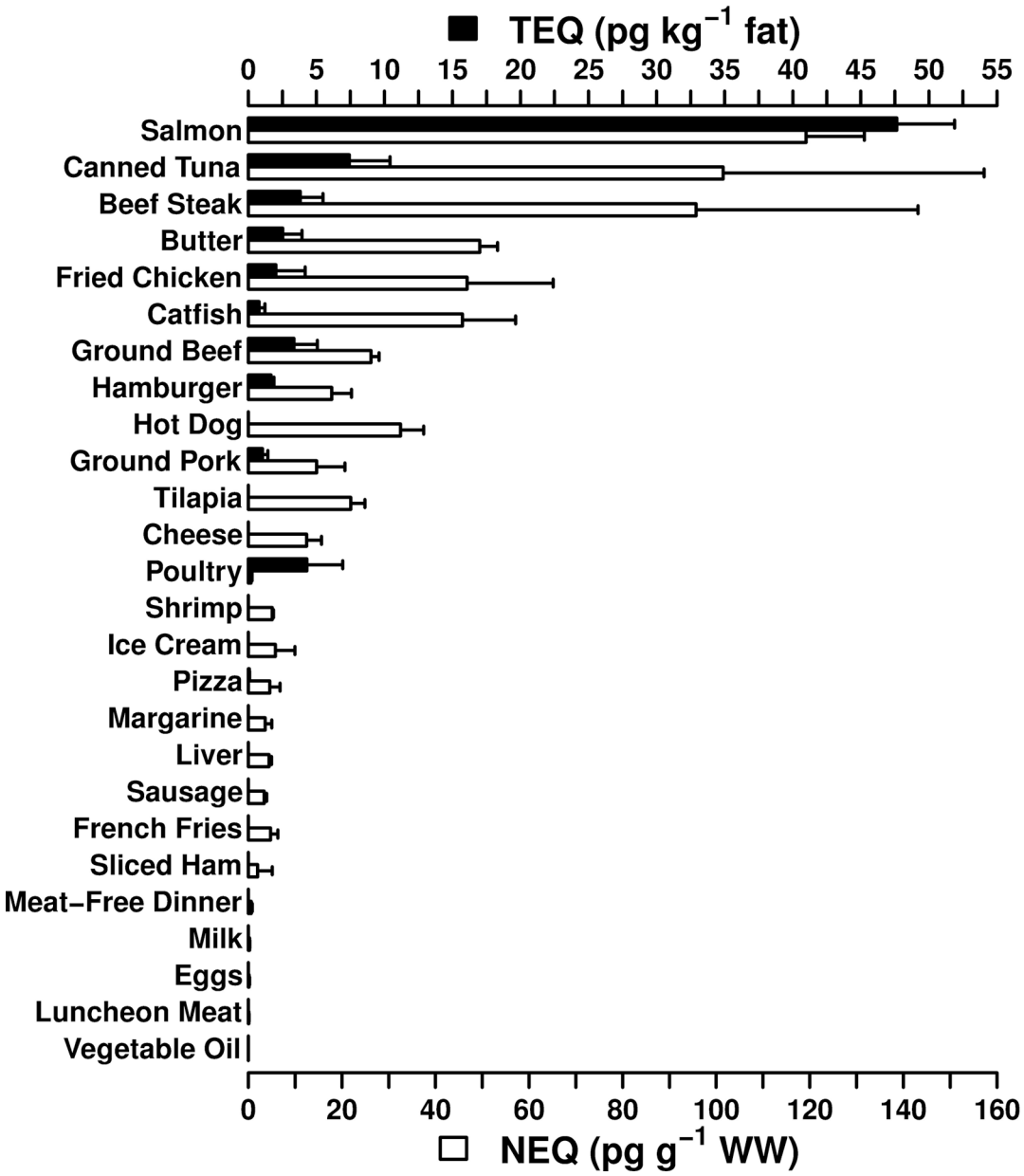

Figure 3.

Toxic equivalency (TEQ in pg kg^{-1}fat; filled bars and top axis) and neurotoxic equivalent (NEQ in pg g^{-1} WW; open bars and bottom axis) values in foods. The error bars indicate the standard deviation. Salmon was wild caught frozen; catfish and tilapia were farmed frozen; liver was beef and chicken; meat-free dinner was a mixture of frozen macaroni-and-cheese and tortillas; milk was 2%; and luncheon meat was ham and turkey breast.

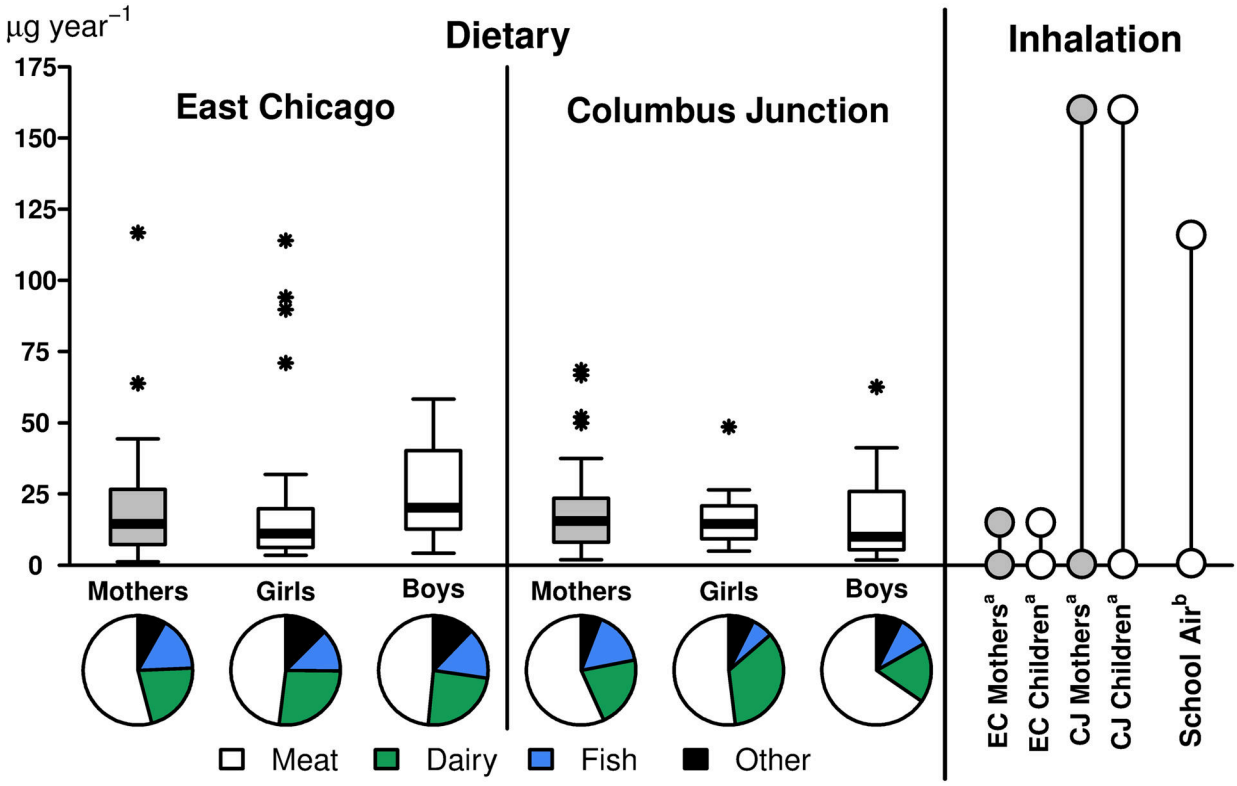

Figure 4.

PCB dietary exposures of mothers (gray) and middle and high school children (white) participating in AESOP Study in East Chicago, Indiana (EC; top-left boxplot) and in Columbus Junction, Iowa (CJ; top-middle boxplot) together with their proportions of PCB dietary exposure sources (bottom pies); and PCB inhalation exposures in the same study populations (right lines) in µg year^{-1}.

[a]Inhalation exposures in AESOP Study populations were calculated as time-integrated products of airborne PCB concentrations and inhalation rates.[26]

[b]Inhalation exposures through school air in AESOP Study children were calculated as the sum of seasonal exposure during spring, autumn, and winter.[62]

Table 1.

The total PCB concentration (pg/w wet weight) in foods relevant to 26 food items in this study in the United States that has been reported since 2000.

Collection Year	Beef Steak	Butter	Canned Tuna	Catfish	Cheese	Eggs	French Fries	Fried Chicken	Ground Beef	Ground Pork	Hamburger	Hot Dog	Ice Cream	#Cor Stud
1995	181.4	3223.5	1433.2	6972.5	588.1	212.8			181.4	181.4		3329.7	1.6	
1998–1999		2070.0												
1997–2001			555.0											
2000	206.0	1648.8	12428.6	1002.0	357.3	393.5			447.4	310.2	159.4	150.0	346.3	
2001	244.5	1513.6	16201.5	1307.9	384.8	165.5	96.8		582.4	419.4	280.2	148.6	241.3	
2002	139.2	842.9	7209.8	4207.9	337.6	363.4	107.5		296.0	96.0	134.2	126.7	131.5	
2001								170.0			330.0			
2001														
2001	0.3								0.3	0.1	0.2			
2002														2
2002–2003	1.0								1.0	1.2				
2003–2004														
2004–2005			94500.0											
2006–2007			34000.0											
2007														1
2007–2008				47.0										
2007–2008	1.2								1.2	1.5				
2009														2
2009		1130.0	700.0	1200.0	1060.0	480.0	700.0				2400.0		1580.0	
2012–2013	1.5								1.5	2.9				
2014														
2014–2015[k]			2470.0											2
2018–2019	288.2	267.0	332.2	183.6	56.3	0.4	14.1	205.1	118.3	70.4	94.7	84.7	29.4	2

Collection Year	Liver	Luncheon Meat	Margarine	Meat-Free Dinner	Milk	Pizza	Poultry	Salmon	Sausage	Shrimp	Sliced Ham	Tilapia	Vegetable Oil	#Cor Stud
1995				159.5	0.5		470.1	1433.2				6972.5		
1998–1999														
1997–2001														

2000	292.8	359.8	5.0	342.0	37.5	266.1	116.8	539.0	393.8		1002.0	4.7	
2001	660.8	328.8	26.5	158.6	40.4	305.0	168.9	299.0	421.2		1307.9	10.0	
2002	145.4	219.0	303.3	13.2	45.3	161.5	93.2	387.9	277.1		4207.9	0.0	
2001						300.0							
2001								1850.0					
2001							0.2						
2002								41500.0					2
2002–2003							0.7						
2003–2004								18350.0					
2004–2005													
2006–2007													
2007											13000.0		1
2007–2008								10860.0		117.0	47.0		
2007–2008							0.7						
2009										206.0			2
2009			1130.0		540.0		580.0	3800.0	1340.0		540.0	890.0	1170.0
2012–2013							0.9						
2014					208.8								
2014–2015[k]													2
2018–2019	21.3	0.4	23.8	1.6	1.0	26.6	40.8	379.3	15.0	30.2	11.7	66.9	0.0	2

Catfish and tilapia were farmed frozen; liver was beef and chicken; luncheon meat was ham and turkey breast; meat-free dinner was a mixture of frozen macaroni-and-cheese and tortilla; milk was 2%; and salmon was wild caught frozen.

Concentrations in fresh weight units are reproduced with the significant figures of the original reports. Concentrations reported as TEQ, lipid-adjusted, or other units were estimated as fresh weight and reported here in fewer significant figures.

[a] Lipid content of 92% is applied.[63]

[b] Lipid contents as reported are applied.

[c] Lipid contents of 17%, 10%, and 7% are applied to fried chicken, hamburger, and pizza, respectively.[63] The geometric mean of World Health Organization (WHO) toxic equivalency factors (TEFs, 1998) at 0.000322 is used for the conversion from pg WHO TEQ/g to pg g^{-1} WW.[48]

[d] Lipid contents of 10%, 15%, 30%, and 10% are applied to hamburger, chicken, pork, and beef, respectively.[63]

[e] Lipid contents of 20%, 30%, 15%, and 7.5% are applied to steers-heifers, market hogs, young chickens, and young turkeys, respectively[63]

[f] Lipid content of 4% is applied.[63]

[g] Lipid content of 20%, 30%, 15%, and 7.5% are applied to beef, pork, chickens, and turkeys, respectively.[63]

[h] Lipid of 0.44% as reported is applied.

[i] Lipid contents of 20%, 30%, 15%, and 7.5% are applied to steers-heifers, market hogs, young chickens, and young turkeys, respectively.[63]

[j] Density of 1.03 g/mL is applied.[64]

[k] The collection year was not stated. The collection year is then assumed based on article submission date.