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Statistical Survey of Persistent Organic Pollutants: Risk Estimations to Humans and Wildlife through Consumption of Fish from U.S. Rivers

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Abstract

U.S. EPA conducted a national statistical survey of fish tissue contamination at 540 river sites (representing 82 954 river km) in 2008–2009, and analyzed samples for 50 persistent organic pollutants (POPs), including 21 PCB congeners, 8 PBDE congeners, and 21 organochlorine pesticides. The survey results were used to provide national estimates of contamination for these POPs. PCBs were the most abundant, being measured in 93.5% of samples. Summed concentrations of the 21 PCB congeners had a national weighted mean of 32.7 μg/kg and a maximum concentration of 857 μg/kg, and exceeded the human health cancer screening value of 12 μg/kg in 48% of the national sampled population of river km, and in 70% of the urban sampled population. PBDEs (92.0%), chlordane (88.5%) and DDT (98.7%) were also detected frequently, although at lower concentrations. Results were examined by subpopulations of rivers, including urban or nonurban and three defined ecoregions. PCBs, PBDEs, and DDT occur at significantly higher concentrations in fish from urban rivers versus nonurban; however, the distribution varied more among the ecoregions. Wildlife screening values previously published for bird and mammalian species were converted from whole fish to fillet screening values, and used to estimate risk for wildlife through fish consumption.

Graphical Abstract

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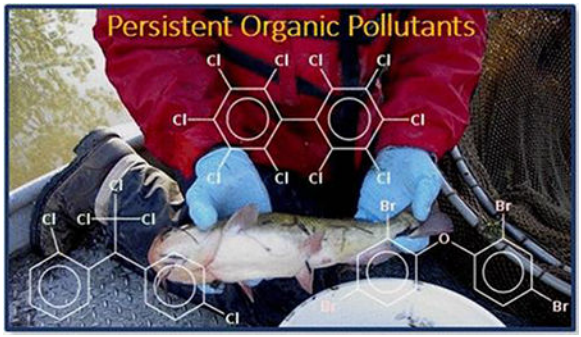
Supporting Information

The authors declare no competing financial interest.



Introduction

Polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and many organochlorine pesticides, such as dichlorodiphenyltrichloroethane (DDT), dieldrin, and chlordane, are well-known persistent organic pollutants (POPs) that have been shown to be ubiquitous in the environment.(1, 2) PCBs were widely used in industry and manufacturing as dielectric and coolant fluids, but because of their environmental toxicity and persistence, were banned for use by the U.S. Congress in 1979.(3) PBDEs share a similar chemical structure to PCBs. They are used as flame retardants in numerous consumer products, such as building materials, electronics, furniture, and textiles.(2) The production of octaBDE and pentaBDE commercial mixtures in the United States ended in 2004, leaving decaBDE as the only congener still in production in the U.S., which is also scheduled to be phased out of use.(4) DDT is one of the most recognizable POPs. After concerns were raised that its widespread agricultural use as a pesticide was having serious impacts on the health of the environment,(5) it was banned for use in the United States in 1972.(6)Chlordane and dieldrin were used as pesticides mainly to control termites and other insects in agricultural areas, and both substances having been banned for all agricultural uses in the U.S. by the U.S. EPA since the late 1980s. After its agricultural use was ended, chlordane could continue to be used to control termites around foundations.

The list of possible health effects in humans and wildlife from exposure to these POPs is extensive, and includes cancer, reduced reproduction rates, changes in immune and endocrine end points, and developmental neurotoxic effects.(2, 3, 7–13) Effects for some POPs, such as PCBs, can also be transgenerational, as the compounds have been found to cross over into the placenta and result in poor attention and behavioral problems in exposed infants and children.(14, 15) Most POPs are lipophilic, and have been measured in human serum, blood, breast milk, and other human tissue,(16–21) and in the tissue of a variety of wildlife.(22–29) Not only do they accumulate in tissues, but also they tend to biomagnify in the food chain,(30) and a link between dietary consumption of fish and marine mammals and human blood or serum levels has been observed for several POPs.(31, 32) DDT is still detected in food supplies, and food-borne DDT, especially from seafood, remains a significant source of human exposure.(20, 33) Although most of these POPs have been banned for use in the U.S. for decades, their persistence and toxicity still make it important to study their potential for exposure today.

This work presents the results of the 2008–2009 National Rivers and Streams Assessment (NRSA) fish tissue indicator, where 50 POPs (Table 1) were measured in 540 composite fish fillet samples collected across the conterminous states (Figure 1). The NRSA is one of several U.S. EPA national probabilistic surveys designed to evaluate the overall condition and health of the nation's waters, and a wide variety of indicators are evaluated. NRSA study results for pharmaceuticals measured in urban surface water, and fish tissue contaminant results for both mercury and perfluorinated compounds have been reported previously.(34–36) Mercury was detected in every one of the 540 samples included in this study, and fillet tissue concentrations in an estimated 25% of the sampled population exceeded the U.S. EPA 300 μg/kg fish-tissue based water quality criteria for mercury.(34) The probabilistic design for this survey provides national estimates of the distribution of these 50 POPs in fifth order and greater U.S. rivers for the assessment of human health impacts of fish consumption. A series of cancer(37) and noncancer(37, 38) human health screening values (SVs) were applied to the fillet tissue results. They provide estimates of the percentage of all U.S. river km represented in this study (i.e., the sampled population of rivers) that would be expected to contain POPs that exceed these SVs. Fish fillet tissue contaminant concentrations were also compared to wildlife screening values that were converted from whole fish values to fillet screening values in order to estimate exposure risks to mammals and birds. Geographical distributions of these contaminants were compared, first between urban and nonurban river segments, and then among three aggregated National Aquatic Resource Assessment ecoregions: Eastern Highlands (EHIGH), Plains and Lowlands (PLNLOW), and the West and Mountains (WMTS) (Figure 1).

Materials and Methods

2.1 NRSA Design and Site Selection

The NRSA included 1924 sites within the conterminous United States that were sampled for a range of indicators.(39) Fish samples were analyzed from 540 sites on rivers fifth order and greater to determine concentrations of chemicals in fish, including pesticides, PCBs, PBDEs, selenium, and mercury. The 50 POPs listed in Table 1 include 21 out of a possible 209 PCB congeners, 8 PBDE congeners, and 21 organochlorine pesticides and metabolites. The number of congeners of PCBs and PBDEs was limited to allow for the simultaneous analysis of multiple classes of 50 POPs with a single analysis, and the specific congeners were chosen as they have been reported in previous U.S. EPA studies.(22) The mercury findings and fish tissue sample collection design were described previously.(34) Fish tissue samples were collected at sites that had a permanent fish population. Rivers were designated as fifth order or greater based on Strahler stream order.(40) The sampling framework was derived from the National Hydrography Data set (NHD) and included Strahler stream order attributes.(41) The fish sampling sites were classified as two subpopulations: urban and nonurban river segments. NHD-Plus and the U.S. Census Bureau national urban boundary GIS coverage layers were used to identify urban sampling areas, which were defined as densely settled census block groups with a minimum population density of 50 000 people. Sampling sites were selected using a probability-based approach,(42, 43)generally applying the spatial methodology used for lakes in U.S. EPA's National Lake Fish Tissue Study(44) to major U.S. rivers. Fish samples were collected from 164 randomly selected urban river

sites and 378 nonurban river sites. This included sampling locations in 46 states with the following distribution of river sites: fifth order, 154 sites; sixth order, 161 sites; seventh order, 99 sites; and eighth order and above, 126 sites.

2.2 Sample Collection

One composite sample of a single fish species was collected from each site (Figure 1). A routine composite sample consisted of five fish, but composites containing fewer or greater than five fish were accepted in an effort to retain a sample from each target river segment (51.3% and 1.1% of the composites, respectively). Species were selected to be ubiquitous, abundant, and easily identified. Individual specimens of the species were selected to be adults of similar size (the length of the smallest individual in a composite could not be less than 75% of the total length of the largest individual) and sufficiently large to provide adequate tissue.(45) A total of 15 species were identified in a targeted list, including members of the sunfish (with largemouth and smallmouth bass preferred), trout/salmon, pike, temperate bass, perch, and catfish families. Field teams used active methods, primarily electrofishing, to collect fish samples from each site during the May through September field sampling period in 2008 and 2009. Whole fish were shipped on dry ice to the designated sample preparation laboratory for storage until subsequent fillet tissue sample preparation and analysis. Other aspects of fish collection and handling methods are further described elsewhere.(45)

2.3 Sample Preparation and Analysis

Fish were filleted in the laboratory. Scales were removed, then lateral muscle fillets from both sides of each fish were prepared with skin on and the belly flap (ventral muscle and skin) attached. Fillets from individual specimens that comprised the sample were homogenized together, regardless of the proportional weight of individual fish. Composites were homogenized using a tissue grinder and an 8 g aliquot of homogenate was used for analysis. Wet tissue samples were extracted using pressurized fluid extraction (PFE), followed by sample cleanup with gel permeation chromatography (GPC) and alumina, and clean extracts were analyzed by gas chromatography with electron capture detection (GC-ECD). GC-ECD was chosen as the analysis method since the ECD detection method was found to be more sensitive for the target analytes than some mass spectrometry methods, and provided a very cost-effective analysis for the range of compounds in this study. Full details of the extraction and GC-ECD analysis methods are included in Supporting Information (SI) Document 1. Briefly, 8.0 g of tissue were added to 20 g of anhydrous sodium sulfate and allowed to dry for 1 h. Samples were spiked with a surrogate standard solution and transferred to 33 mL PFE cells fitted with a cellulose filter, and remaining cell volume was filled with drying material. PFE cells were extracted with methylene chloride (50%) and hexane (50%) at 100 °C and 1000 psi for three 7 min cycles. Extracts were collected into a 60 mL glass vial, and the volume was reduced to approximately 3 mL using a stream of nitrogen in a 50 °C water bath. Concentrated sample extracts were dried using a 1-in. diameter glass chromatography column packed with 20 g of sodium sulfate and glass wool, and rinsed three times with hexane. Further sample extract cleanup was performed using GPC with a Waters HPLC, followed by solid-phase extraction using columns packed with deactivated alumina-N. Final sample extracts were adjusted to a volume of 1 mL after the

addition of 25 μL of an internal standard solution consisting of pentachloronitrobenzene, PCB 96, and PCB 166. These extracts were analyzed using Agilent 6890 gas chromatographs equipped with pressure-pulsed splitless injection, narrow-bore columns and micro ECD detectors. Since ECD detection is not as selective as some mass spectrometry methods available, two separate GC-ECD analyses were performed on instruments equipped with different GC columns, first a primary analysis (Agilent HP-5 capillary column: 30 m length, 0.25 mm diameter and 0.25 μm film thickness), followed by a confirmatory analysis (J&W DB - XLB column, 30 m length, 0.25 mm diameter and 0.25 μm film thickness), with the two different oven programs for both methods being described in SI Document 1. Concentrations of detected analytes were calculated for both the primary and confirmatory analyses. Reported concentrations are a result of the average of the primary and confirmatory analyses, unless the relative percent difference (RPD) in concentrations between the two columns was greater than 30%, in which case, the lesser concentration was reported. If an analyte was not detected in both analyses, it was reported as not-detected.

The method detection limit (MDL, Table 1), which is defined as the minimum concentration of an analyte that can be identified and detected with 99% confidence that the analyte concentration is greater than zero, was determined for each analyte using the procedure described at 40 CFR Part 136 Appendix B.(46) The quantitation limit (QL) was defined as 3 times the MDL, and both the MDL and QL were calculated on the primary and confirmatory columns, and were updated on a yearly basis. Concentrations were reported to the MDL that applied at the time of analysis, however, any concentrations greater than the MDL but less than the QL were flagged as estimated. The maximum MDL and QL observed during the course of the study for each analyte is listed in Table 1. Four quality control samples were analyzed with each extraction batch, including a laboratory reagent blank (LRB), a laboratory fortified blank (LFB), and two laboratory fortified matrix (LFM) samples. The LRB and LFB were prepared from sodium sulfate, and the LFB and LFM were spiked with a mixture of the target analytes to yield a final GC extract concentration of 15 ng/mL. Percent recovery for each analyte was calculated in the LFB and LFM samples. Acceptable target recoveries for all analytes ranged from 70% to 130% for LFB, and from 50% to 150% with a 30% RPD for the duplicate LFM samples. Any analytes or samples which did not meet the predetermined quality criteria of the LFB and LFM samples were reanalyzed, or reported as estimated. Samples were also re-extracted and analyzed if an analyte was present in the LRB above the QL and the analyte was present in the sample above the QL but less than 10 times the detected blank concentration. Standard reference material (SRM) 1947, purchased from National Institute of Standard and Technology (NIST), was used as a certified second source standard to monitor extraction efficiency and instrument quantitation, and was extracted and analyzed with each extraction batch. All data reported in this study were reviewed and validated against the project requirements by a third party not involved in the data generation. Only valid results were used in the statistical analyses. Measured concentrations and qualifiers for each of the 50 contaminants analyzed in the 540 fillet samples are included as (SI Table S1–S3).

2.4 Data Analysis

National and Subpopulation Estimates—The NRSA survey design and results provide national and regional estimates of fish tissue contaminant concentrations, and as a result, reported results are expressed as population estimates, and not site or sample summary data. The population estimates are based on weighted analytical results from sampling sites. The weights are based on the survey design and are the inverse of the probability of selecting a sampling site. The probability of selecting a site depends on the stratification and unequal probability of selection associated with the site. The weights are the total river length represented by the sample site. Percentiles and mean population estimates of fish tissue analyte concentrations were calculated from the weighted data, using routines developed by the U.S. EPA in the statistical calculation package *spsurvey R*(47) for the R statistical computing environment.(48) In addition to the urban and nonurban subgroups, statistical parameters were generated for three aggregated ecoregions (EHIGH, PLNLOW, and WMTS) used in U.S. EPA National Aquatic Resource Surveys.(49) A standard normal Z-test was used to investigate fish tissue analyte concentration differences between the urban and nonurban subgroups, and between ecoregions:

$$z = \frac{meanA - meanB}{\sqrt{\left(\text{stderr}A^2 + \text{stderr}B^2\right)}}$$

where mean$A$ is the weighted mean estimate, stderr$A$ is the standard error estimate for mean$A$ for subgroup A, and similarly for subgroup B. Compounds were evaluated by summed groups, including summed PCB and PBDE congeners, total DDT (the five DDT compounds and degradates listed in Table 1), and total chlordane (cis- and trans-chlordane, cis- and trans-nonachlor, and oxychlordane). Prior to summing data into groups, any detections below the MDLs were considered nondetects and the results were treated as zeros during the statistical analysis.

Human Health Risk Estimate—The two categories of human health risk that are reflected in selected SVs are cancer and noncancer (Table 2). The survey design provides national estimates of river kilometers that are expected to contain fish with fillet concentrations that would exceed those screening value thresholds. A cancer SV(37) is based on a level of excess cancer risk from exposure to a carcinogenic substance ranging from 10^{-4} to 10^{-6}. The noncancer SVs are based on a reference dose (RfD) for a toxicant, which is the level of exposure over a lifetime at which no observable adverse effects will occur. Cancer SVs are generally lower than noncancer SVs (Table 2), but the ratio can vary. For this assessment, a cancer SV reflecting 10^{-5} cancer risk has been applied to allow comparison of all the frequently detected compounds across regions, and to be comparable to previous U.S. EPA assessments.(22, 26) A noncancer SV was also applied to provide context for the concentrations reported here in terms of thresholds widely used in fish consumption advisories.(37) For the PBDEs, no cancer SV was applied, since the PBDE congeners represented in this study have not been classified as carcinogenic.(4) Limited RfDs are available for PBDE congeners, however, the California EPA has published fish tissue advisory levels based on noncancer risk.(38)

Wildlife Risk Estimate—The fish tissue study conducted under the 2008–2009 NRSA was designed to assess human health impacts from fish consumption, which is why fillets were targeted for analysis. To estimate the possible risk to wildlife that may be consuming fish collected in this survey, wildlife values (WVs) for mink, kingfisher, and larger birds (if available) that represent thresholds for toxic effects (such as reproductive or development success, organismal viability or growth, effect on population dynamics) were used.(50, 51) However, these previously reported WVs represent whole-fish tissue concentrations, so a conversion factor had to be applied to convert whole-fish tissue WVs to fillet WVs. For total PCBs, a conversion factor was averaged from across three references;(28, 52, 53) one conversion factor was used for DDT and chlordane;(52) and another was identified for PBDEs.(51) All of these conversion factors are listed in Table 2, along with the resulting fillet WVs. Dieldrin WVs were not converted from whole body to fillets, since it has been reported that dieldrin was present in roughly the same average concentrations in game-fish fillets as in whole-body bottom-feeders. No other publications were found that provided information comparing whole fish dieldrin to fillet concentrations from the same fish species.(54) Since a conversion factor for the wildlife values from fillets to whole fish had to be applied, limited inferences can be made for the wildlife results. However, because different species can display very different sensitivities to the same chemicals, the results of the wildlife risk estimate do provide valuable insight into the relative risks of exposure for human, mammalian, and avian species across the different contaminant groups.

Chemical Co-Occurrence—Co-occurrence of the four main chemical groupings was examined using a previously described procedure,(55) Samples that exceeded the weighted median concentrations for any of the four major contaminant groups (PCBs, PBDEs, total chlordane, and total DDT) were identified. Samples that exceeded the respective median concentrations were then compared to calculate the percentage of the samples with concentrations above the median of any one compound group that occurred: (1) singly; (2) with a second contaminant group also above its median; (3) as a combination of any three contaminant groups; or 4) with all four compound groups occurring together above their median concentrations.

3.0 Results and Discussion

PCBs in Fish Tissue

PCBs were detected in 93.5% of the fish fillet samples, which results in 48% (40 030 river km (±2432 km)) of the national sampled population of rivers having fillet tissue concentrations that exceed the cancer SV of 12 μg/kg, as shown in Table 3. By comparison, nearly 70% of the sampled population of urban rivers had fillet tissue concentrations that exceeded the PCB cancer SV. The maximum summed PCB concentration measured in urban river samples was 857 μg/kg. Among the three ecoregions, PCB detections dominated in the EHIGH, where the mean summed PCB concentration was 47.1 μg/kg. In this ecoregion, fillet tissue concentrations exceeded the PCB cancer SV of 12 μg/kg in 54.3% of the sampled population of EHIGH rivers. The mean summed PCB concentrations in fillet samples from the PLNLOW and WMTS were 30.7 μg/kg and 11.9 μg/kg, respectively. The PCB SV exceedances were lower in these two ecoregions. In the PLNLOW, 50.8% of the

sampled population of PLNLOW rivers had fillet concentrations that exceeded the 12 μg/kg SV. By contrast, 23.6% of the WMTS sampled population of rivers exceeded the cancer SV.

The national, ecoregion, urban river, and nonurban river population percentile estimates for the summed PCB concentrations are shown in Figure 2. The national estimate of the median summed PCB concentration is 11.3 μg/kg, which is almost the same as the cancer SV of 12 μg/kg. All of the 75th percentile estimates of fillet concentrations lie between the cancer SV (12 μg/kg, which corresponds to the WMTS estimate) and noncancer SV (47 μg/kg, which corresponds to the urban river estimate). All of the 90th percentile estimates exceed 47 μg/kg except the estimate for the WMTS ecoregion. For the wildlife estimates, fish tissue concentrations in 9.3% of the national sampled population exceed the fillet WV for mink, whereas fish tissue concentrations in only 1.3% of the sampled population exceeded the kingfisher fillet WV. Human SVs for both cancer and noncancer are lower than both PCB WV values, and therefore, in general, the human health SVs are protective of wildlife risks as well.

The relative abundance of the 21 PCB congeners found in fish fillet tissue from nonurban vs urban rivers is shown in SI Figure S-1. All mean PCB congener concentrations are higher in samples from urban rivers than from nonurban rivers, but the magnitude of the difference varies. Congeners 138 and 153 are the most abundant PCBs in both nonurban and urban river fish fillets, which has also been the case in other environmental samples.(16) The coplanar PCBs 77, 126, and 169 (indicated by the arrows in SI Figure S-1) are present only at low concentrations in fillet tissue from both subpopulations. In this study, 21 PCB congeners were quantified out of the full suite of 209 PCB congeners. Since there is additional PCB mass in the samples that was not measured for this study, the summed 21 PCB congeners underestimate the total mass present in the samples. Although this study underestimates the total PCB mass, the congeners analyzed include 10 of the 11 congeners with the highest frequency of environmental occurrence, as well as three dioxin-like congeners.56) Therefore, the results of this study are still applicable to the human health and wildlife exposure analysis.

PBDEs in Fish Tissue

Similar to PCBs, the eight congeners of PBDEs are commonly detected in fish tissue samples. All of the results presented in Table 3 and SI Figure S-2 indicate higher concentrations in urban river samples than in samples from nonurban rivers. In contrast, PBDE results among the three ecoregions show (Table 3) no clear distinctions. SI Figure S-3 shows the relative abundance of individual PBDE congeners in fillet tissue samples from nonurban and urban rivers. PBDE concentrations in fish from U.S. rivers are dominated by BDE 47, followed by BDEs 100 and 99. Only 0.3% of the national sampled population had fillet concentrations that exceeded the 210 μg/kg human health SV for PBDEs (Table 3). The higher noncancer SV for PBDEs relative to PCBs and DDT is a reflection of their lower relative toxicity, but again, limited RfD values are available for PBDEs. The lower toxicity and shorter half-life of PBDEs(57) compared to PCBs(58) could be contributing factors to the lower percentage of the national sampled population having fillet tissue concentrations that exceed the SV for PBDEs. In contrast to PCBs, the applied PBDE WVs are lower than

the human health SVs. Summed PBDE results for the wildlife risk estimation showed that fish tissue concentrations in 33.4% of the national sampled population exceed the converted fillet WV for kestrels, and for mink, fish tissue concentrations in 14.5% of the national sampled population exceeded the WV (Table 3).

OCPs in Fish Tissue

Lower concentrations were observed for the organochlorine pesticides (OCPs) chlordane and total DDT relative to the summed PCBs. Even 28 years after U.S. EPA withdrew approval for its use for underground termite control around the foundations of homes,(59) chlordane was detected in 88.5% of all fish tissue samples and 93.9% of the urban river samples (Table 3). Concentrations exceeded the cancer-based human health SV of 67 μg/kg (44) in fish fillet samples from less than 1% of the sampled population of rivers. When WVs were applied for chlordane, fish tissue concentrations in 36.9% of the national sampled population exceeded the kingfisher fillet WV and fish tissue concentrations in 0% of the national sampled population exceeded the mink fillet WV (Table 3).

Although banned in 1972 (for most uses) in the U.S.,(6) DDT and its metabolites (total DDT) were detected in 100% of the fish samples in this study (Table 3). Concentrations of total DDT were found to be elevated in a few samples. However, the mean concentration (13.8 μg/kg) is well below the human health cancer-based SV of 69 ug/kg for total DDT, and both the percentage of samples and the river length they represent are low (1947 km, ±511 km, Table 3). Only the 95th percentile estimates for total DDT concentrations from urban rivers exceed the cancer-based SV (SI Figure S-4). Additionally, the tissue concentrations for 4,4′-DDT (mean 0.75, median 0.18, maximum 36.3 μg/kg) are much lower than total DDT, which indicates that new sources of DDT are unlikely. The main contributor to total DDT was 4,4′-DDE (mean 10.7, median 4.7 μg/kg). Fish tissue concentrations in 31.5% of the national sampled population exceeded the WV for total DDT for kingfishers, and fish tissue concentrations in less than 1% of the national sampled population exceeded the WV for mink. Like chlordane, the converted total DDT WV for Kingfishers was lower than that of minks and the human health cancer SVs listed in Table 3, indicating that these organochlorine pesticides may pose more of a risk for avian species.

Analytical results for dieldrin also showed widespread occurrence with a 71% frequency of detection, and dieldrin has a relatively lower cancer SV (1.5 μg/kg). Dieldrin concentrations in fillet samples exceeded this SV in 31.2% of the sampled population of rivers, including 41.2% of urban rivers and 40.4% of PLNLOW rivers (Table 3). Other OCPs detected in this study included aldrin, alpha-BHC, endrin, lindane, endrin ketone, heptachlor epoxide, hexachlorobenzene, and mirex. These other OCPs had detection frequencies of less than 50%, and none of the fillet concentrations exceeded human health SVs for these compounds.

Subpopulation Differences

Concentrations of mercury in fish tissue from these same samples were previously compared across the three ecoregions and between nonurban and urban sites.(34) Those results revealed no significant differences across the subpopulations of rivers, however, different results were observed for the organo-halogen compounds. Figure 3 compares the weighted

mean concentrations of the organo-halogens in fish from urban versus nonurban subpopulations and from ecoregions, and the significance of any differences as determined using the Z test are listed in SI Table S4. The means for all chemical groups were higher for samples from urban rivers than those from nonurban rivers. The means for summed PCBs, summed PBDEs, and total DDT were significantly higher for urban river samples, which reflects the greater extent of chemical use and release of a variety of contaminants into rivers in populated areas where they can bioaccumulate in fish. Higher organochlorine concentrations in aquatic environments in urban areas have been documented previously in urban bed sediments.(60) Not only could urban sources from chemical manufacturing, industrial use of chemicals, and domestic application of pesticides contribute to enhanced concentrations in fish tissue from urban waters, but proximity to wastewater treatment plant (WWTP) discharges could also be a factor, since WWTPs have been demonstrated to be an important point source of PCB and PBDE contamination.(61)

Among the ecoregions, summed PCB concentrations in river fish samples from the EHIGH were significantly higher than in samples from both the PLNLOW and WMTS (Figure 3, SI Table S4). Summed PCB concentrations in fish from the PLNLOW were also significantly higher than those from the WMTS. Although summed PBDE concentrations were significantly higher in samples from urban rivers relative to nonurban rivers, there was no significant difference in summed PBDE concentrations among ecoregions. Total chlordane concentrations were found to be significantly higher in fillet tissue samples from rivers in both EHIGH and PLNLOW ecoregions relative to the WMTS ecoregion, but the difference in EHIGH and PLNLOW total chlordane concentrations was not significant ((Figure 3, SI Table S4). Total DDT concentrations in samples from waters in the PLNLOW were significantly higher relative to those from the EHIGH, but unlike total chlordane, the difference in total DDT concentrations among the EHIGH, PLNLOW, and WMTS ecoregions was not significant.

Differences observed in the chemical concentrations relate to various factors, including their historical use and the geographic distribution of these chemical contaminants. Summed PCB concentrations are significantly higher in the EHIGH region relative to both other ecoregions, which reflects the industrial and urban history of this region where PCBs were most predominately used and released into the environment.(60) The ubiquity of PBDEs is consistent with its discharge pathway from dust in homes, through wastewater treatment systems,(21) and then to rivers. Similarly, the predominance of DDT in fish tissue from the PLNLOW ecoregion is consistent with the agricultural character of the middle of the country. The lack of a significant difference between the PLNLOW and WMTS total DDT results is also consistent with the high agricultural use of some states in the WMTS ecoregion. Chlordane, last labeled for use for underground application against termites,(59) persists in the EHIGH and PLNLOW regions but not in the WMTS.

Co-Occurring Contaminants

Co-occurrence of these four chemical groups was examined with the procedure outlined by Thompson and Boekelheide,(55) in which fish tissue concentrations that exceeded their respective median values are used to determine co-occurrence at elevated concentrations.

This analysis (Figure 4) revealed that the most common mode of co-occurrence (fish from 32% of river sites with concentrations above the median for any compound) was for all four compound groups to co-occur at concentrations above their respective weighted medians. The next highest category of co-occurrence (16%) was for any three compound groups to co-occur above their median concentrations. PCBs co-occurred with PBDEs in 11% of sampled populations; chlordane with total DDT and PCBs with total DDT each co-occurred in 10% of the sampled population above the medians for these compound groups. The rarity was for these contaminants to occur singly above the median concentrations as was the case for PCBs in 4%, PBDEs and chlordane each in 5%, and total DDT in 7% of samples from the sampled population of rivers.

The extent of co-occurrence of these compounds at relatively elevated concentrations underscores the exposure risk that is posed by their presence in fish tissue. The U.S. EPA has provided approaches to quantify the toxicity of combinations of contaminants based on whether the contaminants are known to have similar or dissimilar modes of toxicity.(62) For compounds with similar modes of toxicity, the doses are considered additive. For dissimilar responses, the risks are considered separately and then combined. Additive risk, however, has been poorly defined and infrequently studied, especially for legacy contaminants, such as PCBs and OCPs no longer in use, that are perceived to be a diminishing threat. Accordingly, the background environmental chemical load and the potential for such additive effects should be taken into consideration when new chemicals, including organo-halogen compounds such as brominated flame retardants or chlorinated antimicrobials, are discharged to surface waters.

Nationally representative data on the occurrence of organic contaminants in fillet tissue of fish from U.S. rivers indicate that PCBs, PBDEs, chlordane, and DDT are still pervasive. PCBs, PBDEs, and total DDT occur at significantly higher concentrations in fish from urban rivers; however, the distribution of chemical groups varies more among the ecoregions. Co-occurrence in fish tissue at concentrations above the medians is typically observed, most frequently (32%) with all four of the chemical classes. This indicates that monitoring of fish tissue for assessment, fish consumption advisories, and the protection of aquatic life continues to be important for both new and legacy organic compounds. Individual organo-halogen compounds seldom occur alone in fish tissue. Therefore, these analytical results should be viewed in the context of co-occurring compounds and assessments of risk to human health and aquatic life that reflect the integrated chemical burden in fish. Any new organo-halogen compounds introduced to the environment will likely add to the existing overall burden of such compounds in fish tissue, since this work has demonstrated that POPs concentrations measured in fish potentially consumed by humans and representative avian and mammalian species still exceed human health SVs and WVs in many U.S. waters today.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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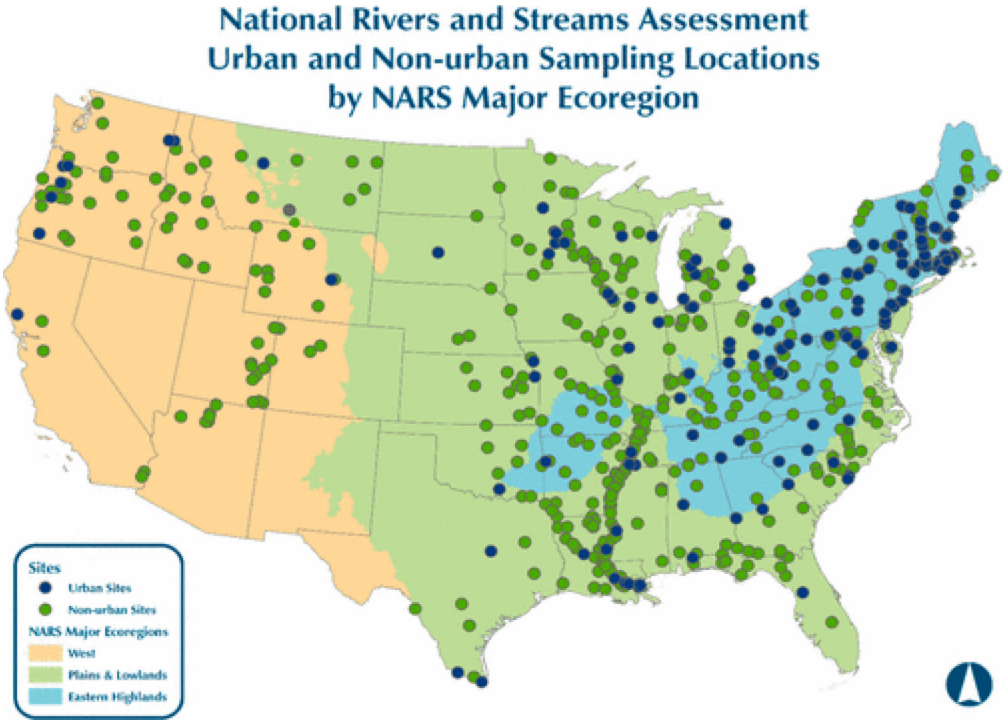

Figure 1.
National map of NRSA 2008–2009 sampling locations (n = 540) within national aquatic resource survey ecoregions.

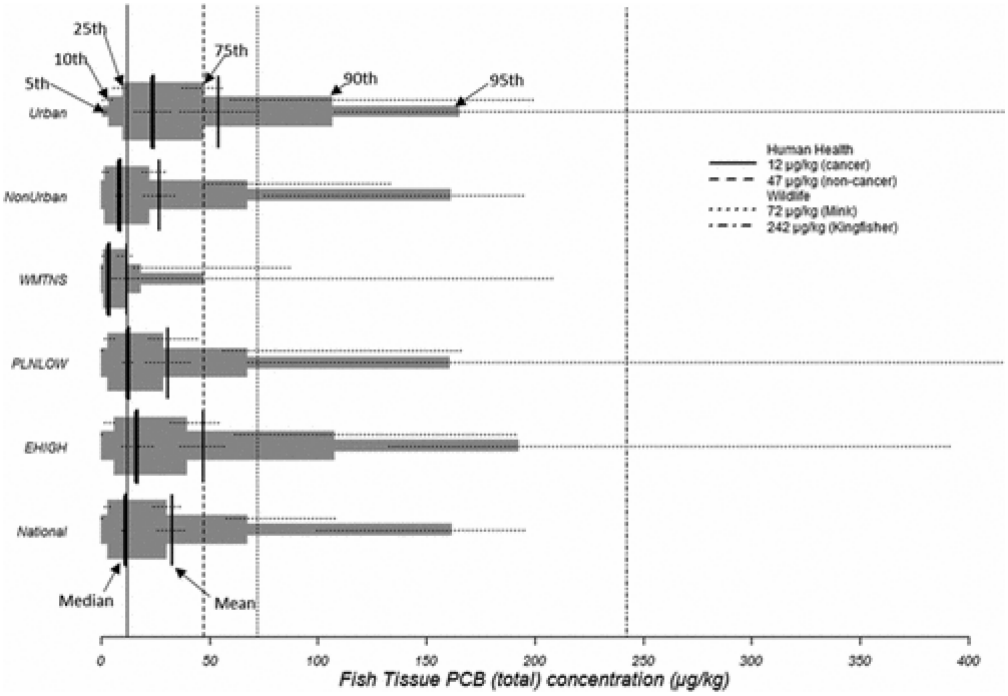

Figure 2.

Estimated population percentile distribution for summed PCBs in fish tissue from U.S. river sampling sites, nonurban and urban, and by ecoregion. Confidence intervals for each percentile, mean, and median are represented by the horizontal dashed lines.

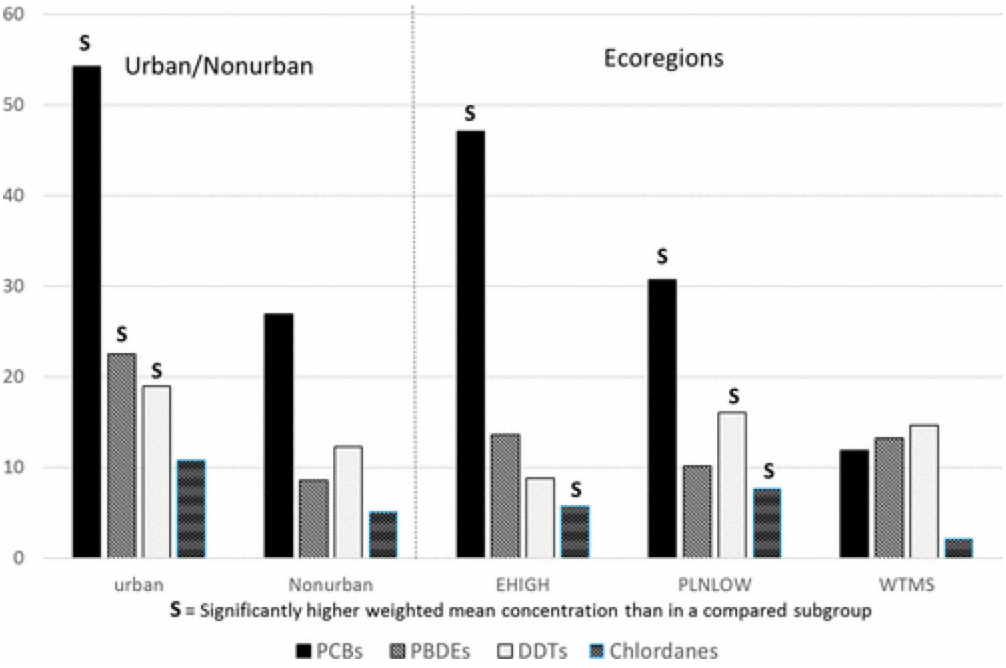

Figure 3.

Comparison of weighted mean contaminant concentrations between subgroups in μg/kg (*y* axis).

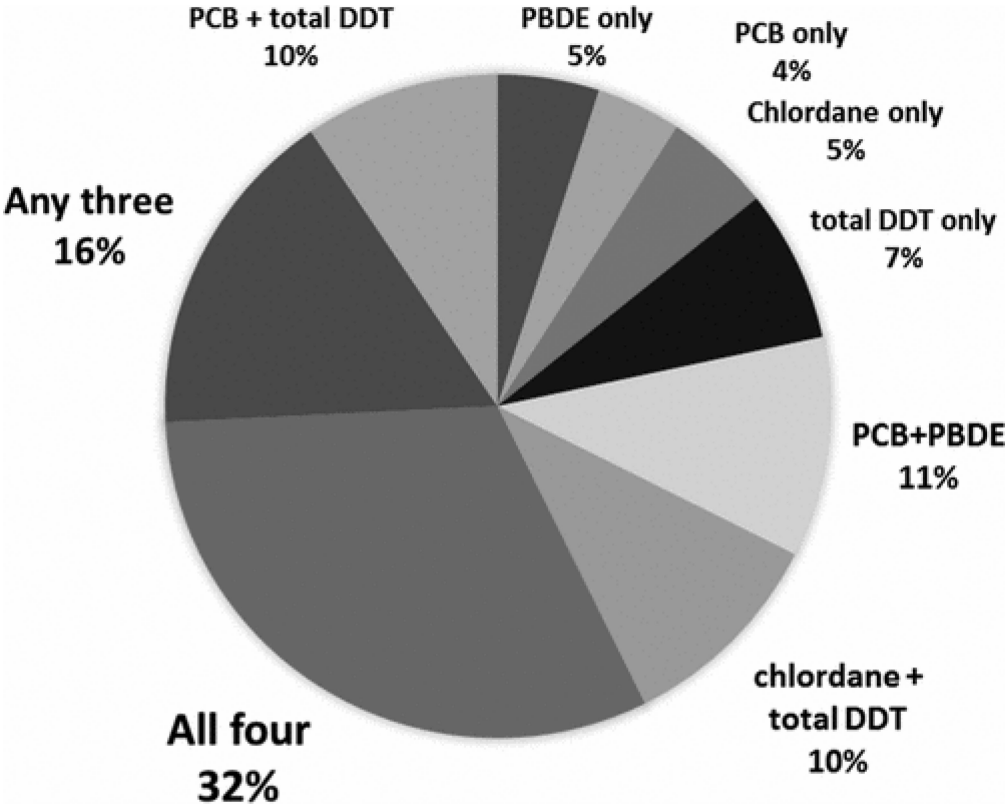

Figure 4.
Co-occurrence of PCBs, PBDEs, chlordane, and DDT above respective median values in fish tissue samples from major U.S. Rivers.

Table 1.

Target analytes with their respective maximum observed method detection limit (MDL) and quantitation limit (QL) (μg/kg wet weight), as described in Method Section 2.3

analyte	CAS number	max MDL	max QL
PCB-8	34883–43–7	0.37	1.99
PCB-18	37680–65–2	0.38	1.23
PCB-28	7012–37–5	0.48	1.53
PCB-44	41464–39–5	1.30	4.13
PCB-52	35693–99–3	0.86	2.73
PCB-66	32598–10–0	1.25	3.97
PCB-77	32598–13–3	0.61	1.93
PCB-101	37680–73–2	0.54	1.72
PCB-105	32598–14–4	0.30	0.95
PCB-118	31508–00–6	0.39	1.24
PCB-126	57465–28–8	0.35	1.11
PCB-128	38380–07–03	0.41	1.30
PCB-138	35065–28–2	2.68	8.52
PCB-153	35065–27–1	0.51	1.63
PCB-169	32774–16–6	0.36	1.14
PCB-170	35065–30–6	0.39	1.24
PCB-180	35065–29–3	0.33	1.05
PCB-187	52663–68–0	0.83	2.62
PCB-195	52663–78–2	0.83	2.63
PCB-206	40186–72–9	0.55	1.74
PCB-209	2051–24–3	0.45	1.43
summed PCBs	a sum of the above 21 congeners		
PBDE-47	5436–43–1	0.37	1.23
PBDE-66	189084–61–5	0.26	0.86
PBDE-99	60348–60–9	0.31	0.99
PBDE-100	189084–64–8	0.52	1.64
PBDE-138	182677–30–1	0.59	1.97
PBDE-153	68631–49–2	0.58	1.93
PBDE-154	207122–16–5	0.55	1.84
PBDE-183	207122–15–4	0.82	2.74
summed PBDE	a sum of the above 8 congeners		
2,4′-DDD	53–19–0	0.37	1.18
4,4′-DDD	72–54–8	0.28	0.89
4,4′-DDE	72–55–9	0.28	0.91
2,4′-DDT	789–02–6	0.30	0.95
4,4′-DDT	55–29–3	0.32	1.02

analyte	CAS number	max MDL	max QL
summed DDT	a sum of the above 5 DDT compounds		
alpha-chlordane (cis-chlordane)	5103–71–9	0.29	0.92
gamma-chlordane (trans-chlordane)	5103–74–2	0.38	1.22
oxychlordane	27304–13–8	0.37	1.18
summed chlordane	a sum of the above 3 chlordane compounds		
dieldrin	60–57–1	0.93	2.95
aldrin	309–00–2	0.30	0.97
alpha-BHC	319–84–6	0.31	1.04
gamma-BHC (lLindane)	58–89–9	0.23	0.73
endosulfan II	33213–65–9	0.46	1.53
endrin	72–20–8	0.37	1.18
endrin ketone	53494–70–5	0.34	1.08
heptachlor	76–44–8	0.28	0.89
heptachlor epoxide	1024–57–3	0.30	0.95
hexachlorobenzene	118–74–1	0.30	0.95
mirex	2385–85–5	0.37	1.18
cis-nonachlor	5103–73–1	0.29	0.99
trans-nonachlor	39765–80–5	0.29	0.92

Table 2.

Summary of the Human Health Screening Values (SVs) and Wildlife Values (WV), with All Values Being Presented in μg/kg Wet Weight (ppb); and the Applied Fillet to Whole Fish Conversion Factors[a]

compound	cancer SV	noncancer SV	mink WV	kingfisher WV	fillet to whole fish conversion factor
PCBs	12	47	72	242	1.83
PBDEs	N/A	210	21	8.7[b]	1.50
DDT	69	120	216	12.0	1.66
chlordane	67	1200	573	3.1	1.44
dieldrin	1.5	120	20	360	

[A] HH SVs are based on the upper estimates of consuming one 8 oz meal of fish per week. See Methods Section 2.4 for a description of Cancer and Non-Cancer SVs, and WV.

[B] Kestril WV instead of Kingfisher, taken from Canadian Environmental Protection Act(38)

Table 3.

Weighted POPs Fish Tissue Concentration Results by Site Type and Ecoregion, And Percent River km Exceeding Human Cancer Screening Values (CSV), Noncancer Screening Values, And Wildlife Values (WV)

PCB summed congeners			
statistic	**national**	**nonurban**	**urban**
detects	505/540 sites (93.5%)	343/377 sites (91.0%)	162/163 sites (99.4%)
mean	32.7 ug/kg	26.9 ug/kg	54.2 ug/kg
median	11.3 ug/kg	8.6 ug/kg	23.8 ug/kg
max	857 ug/kg	412 ug/kg	8567 ug/kg
%>CSV (12 ug/kg)	48.0%	42.0%	69.8%
%>non-CSV (47 ug/kg)	16.7%	14.6%	25.7%
%>mink WV (72 ug/kg)	9.3%	8.9%	10.9%
%>KF WV(242 ug/kg)	1.4%	0.7%	3.8%
statistic	EHIGH	PLNLOW	WMTS
sites	189	280	71
mean	47.1 ug/kg	30.7 ug/kg	11.9 ug/kg
median	16.7 ug/kg	12.6 ug/kg	3.8 ug/kg
%>CSV (12 ug/kg)	54.3%	50.8%	23.6%
%>non-CSV (47 ug/kg)	20.2%	18.0%	6.1%
%>mink WV (72 ug/kg)	13.2%	8.5%	4.6%
%>KF WV(242 ug/kg)	3.8%	0.5%	0%
PBDE summed congeners			
statistic	**national**	**nonurban**	**urban**
detects	497/540 sites (92.0%)	340/377 sites (90.2%)	157/163 sites (96.3%)
mean	11.6 ug/kg	8.6 ug/kg	22.5 ug/kg
median	4.7 ug/kg	3.6 ug/kg	8.0 ug/kg
maximum	311 ug/kg	151 ug/kg	310 ug/kg
%>non-CSV (210 ug/kg)	0.3%	0%	1.2%
%>mink WV(21 ug/kg)	14.6%	10.6%	29.1%
%>kestral WV(8.7 ug/kg)	33.4%	29.5%	47.8%
statistic	EHIGH	PLNLOW	WMTS
sites	189	280	71
mean	13.6 ug/kg	10.2 ug/kg	13.2 ug/kg
median	6.8 ug/kg	3.5 ug/kg	4.8 ug/kg
%>non-CSV (210 ug/kg)	0%	0.5%	0%
%>mink WV(21 ug/kg)	17.5%	13.3%	13.9%
%>kestral WV(8.7 ug/kg)	42.0%	31.5%	23.8%
total DDT			
statistic	**national**	**nonurban**	**urban**
detects	533/540 sites (98.7%)	370/377 sites (98.1%)	163/163 sites (100%)

PCB summed congeners			
statistic	**national**	**nonurban**	**urban**
mean	13.8 ug/kg	12.3 ug/kg	19.0 ug/kg
median	6.3 ug/kg	5.7 ug/kg	9.5 ug/kg
maximum	294 ug/kg	170 ug/kg	294 ug/kg
%>CSV (69 ug/kg)	2.3%	1.6%	5.1%
%>mink WV(216 ug/kg)	0.1%	0%	0.3%
%>KF WV(12 ug/kg)	31.6%	28.3%	43.7%
statistic	EHIGH	PLNLOW	WMTS
samples	189	280	71
mean	8.8 ug/kg	16.0 ug/kg	14.6 ug/kg
median	3.6 ug/kg	7.5 ug/kg	4.5 ug/kg
%>CSV (69 ug/kg)	1.0%	2.7%	3.5%
%>mink WV(216 ug/kg)	0.2%	0.03%	0%
%>KF WV(12 ug/kg)	21.0%	36.9%	31.4%
total chlordane			
statistic	**national**	**nonurban**	**urban**
detects	478/540 sites (88.5%)	325/377 sites (86.2%)	153/163 sites (93.9%)
mean	6.3 ug/kg	5.1 ug/kg	10.8 ug/kg
median	2.0 ug/kg	1.6 ug/kg	2.7 ug/kg
maximum	311 ug/kg	87.1 ug/kg	311 ug/kg
%>CSV (67 ug/kg)	0.6%	0.3%	1.6%
%>mink WV(573 ug/kg)	0%	0%	0%
%> KF WV(3.1 ug/kg)	36.9%	34.1 0%	47.2%
statistic	EHIGH	PLNLOW	WMTS
samples	189	280	71
mean	5.7 ug/kg	7.6 ug/kg	2.1 ug/kg
median	2.5 ug/kg	2.2 ug/kg	0.8 ug/kg
%>CSV (67 ug/kg)	1.0%	0.5%	0%
%>mink WV(573 ug/kg)	0%	0%	0%
%> KF WV(3.1 ug/kg)	39.9%	40.9%	14.5%
dieldrin			
statistic	**national**	**nonurban**	**urban**
%>CSV (1.5 ug/kg)	31.2%	28.5	41.2%
statistic	EHIGH	PLNLOW	WMTS
%>CSV (1.5 ug/kg)	24.1%	40.4%	7.8%