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Contaminants in fish tissue from US lakes and reservoirs: a national probabilistic study

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Abstract An unequal probability design was used to develop national estimates for 268 persistent, bioaccumulative, and toxic chemicals in fish tissue from lakes and reservoirs of the conterminous United States (excluding the Laurentian Great Lakes and Great Salt Lake). Predator (fillet) and bottom-dweller (whole body) composites were collected from 500 lakes selected randomly from the target population of 147,343 lakes in the lower 48 states. Each of these composite types comprised nationally representative samples whose results were extrapolated to the sampled population of an estimated 76,559 lakes for predators and 46,190 lakes for bottom dwellers. Mercury and PCBs were detected in all fish samples. Dioxins and furans were detected in 81% and 99% of predator and bottom-dweller samples, respectively. Cumulative frequency distributions showed that mercury concentrations exceeded the EPA 300 ppb mercury fish tissue criterion at nearly half of the lakes in the sampled population. Total PCB concentrations exceeded a 12 ppb human health risk-based consumption limit at nearly 17% of lakes, and dioxins and furans exceeded a 0.15 ppt (toxic equivalent or TEQ) risk-based threshold at nearly 8% of lakes in the sampled population. In contrast, 43 target chemicals were not detected in any samples. No detections were reported for nine organophosphate pesticides, one PCB congener, 16 polycyclic aromatic hydrocarbons, or 17 other semivolatile organic chemicals.

Keywords Fish tissue · Contaminants · Lakes · Probabilistic survey · Mercury · PCB · Dioxin · Furan · DDT · Chlordane

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Introduction

Investigating the bioaccumulation of contaminants in fish is an important area of research because tissue residues have implications for human and ecological health (Bruggeman et al. 1984; McCarty and Mackay 1993; Yeardley et al. 1998). Contaminants in fish pose risks to human consumers (e.g., recreational and subsistence anglers) and to piscivorous wildlife. Fish acquire contaminants and concentrate them in their tissues by uptake from water (bioconcentration) and through dietary routes (bioaccumulation) (Branson et al. 1985; Kucklick et al. 1996). They may bioaccumulate chemicals to more than one million times the concentration detected in the water column (USEPA 1992). Many investigations have used fish as a time-integrating indicator of persistent pollutant contamination (Larsson et al. 1993). Most studies have been spatially focused on a particular waterbody (Sloan et al. 1985; Cope et al. 1990; Lange et al. 1993; Stow and Carpenter 1994; Madenjan et al. 1999) or analytically focused on a specific chemical or group of chemicals (Isensee and Jones 1975; USEPA 1987, 1999a; Gardner and White 1990; Glassmeyer et al. 1997; Stafford and Haines 1997). Earlier studies involving a larger spatial scale include surveys of elemental fish tissue contamination in northeastern US lakes (Yeardley et al. 1998) and mercury contamination in streams and rivers of the western US (Peterson et al. 2007). The National Study of Chemical Residues in Fish (USEPA 1992) is an example of a freshwater study with broad-ranging spatial coverage and a diverse list of target chemicals; however, it applied a targeted design to investigate areas of known contamination. A review of existing studies and programs demonstrated a need for a comprehensive characterization of chemical contamination in freshwater fish tissue and identification of the extent of that contamination in US lakes and reservoirs. The following study, the National Lake Fish Tissue Study, was designed to address that national data gap.

This study was a priority activity sponsored under EPA's Persistent, Bioaccumulative, and Toxic (PBT) Chemical Program. PBT pollutants are toxic chemicals that do not readily break down in the environment, are not easily metabolized and can accumulate in the food web through consumption or uptake, and can be hazardous to human or ecosystem health. In 1998, EPA developed a Multimedia Strategy for PBT Pollutants (USEPA 1998). Its purpose was to identify actions to address risks posed by PBT chemicals in the environment, and a priority action was to evaluate the occurrence of PBT chemicals in fish from US waters. EPA's Office of Water conducted this comprehensive study to determine the extent of PBT chemical contamination in freshwater fish from lakes and reservoirs as a part of that strategy.

Study planning began in 1998 by convening a workshop involving scientists from state, federal, and tribal agencies to obtain technical recommendations related to sampling design, target chemicals, sampling methods, and data management. Lakes and reservoirs were chosen as the focus of the study because they are environments where contamination may be more likely to accumulate than in rivers or streams. They occur in a variety of landscapes, provide important sport fisheries and other recreational opportunities, and can receive contaminants from several sources, including direct discharges into the water, air deposition, and agricultural or urban runoff. The 1998 update of the National Listing of Fish Advisories (USEPA 1999b) provided support for the value of monitoring fish contamination as one way to protect human health when it reported that 15.8% of the Nation's total lake acres were under fish consumption advisories at the end of 1998.

This study is unique in two respects. It is the first national freshwater fish contamination study with sampling sites selected according to a probabilistic (random) sampling design, and it includes data on the largest set of PBT chemicals (268) studied in fish to date. The objective of this study was to estimate the national distribution of the mean levels (i.e., composite average or lake mean concentrations) of selected PBT chemical residues in fish tissue from lakes and reservoirs of the conterminous United States (excluding the Laurentian Great Lakes and Great Salt Lake). EPA worked with partner agencies in states, Tribes, and other federal organizations over a four-year period (2000–2003) to collect fish from 500 lakes and reservoirs selected randomly throughout the lower 48 states. The study design afforded the first opportunity to develop national estimates of the median concentrations of PBT chemicals in lake fish, to estimate the percentage of lakes and reservoirs with fish tissue concentrations above a specified human health threshold, and to define a national baseline for tracking changes in PBT chemical concentrations in freshwater fish as a result of pollution control activities.

Based on regional surveys conducted by EPA's Environmental Monitoring and Assessment Program (EMAP), a probabilistic survey design was developed to address the study objective. This study is one in a series of probabilistic surveys undertaken by EPA since the late 1990s to provide statistically based data to characterize the national condition of lakes, streams, rivers, and coastal waters (USEPA 2006a, 2007a). Probability sampling provides the basis for estimating resource extent and condition, for characterizing trends in extent or condition, and for representing spatial pattern, all with known certainty (Olsen et al. 1998). A probabilistic survey design has some inherent characteristics that distinguish it from other sampling designs. First, the population being sampled is explicitly described. Second, every element in the population has the opportunity to be sampled with known probability. Third, the selection process includes an explicit random element. An unequal probability survey design was an essential component of the study since the specific purpose was to describe the condition of resources on a national basis. Design specifics (including discussions of statistical selection of lakes, sample weight calculations, and development of target population and sampled population estimates) are detailed in Olsen et al. (2008).

Methods and materials

Lake selection

Lakes and reservoirs were selected using a probability-based approach (Olsen et al. 2008). For this study, lakes were defined as permanent bodies of water with permanent fish populations of predators and/or bottom dwellers and a surface area of at least one hectare, a depth of at least one meter, and at least 1,000 square meters of open (unvegetated) water. Lake selection was limited to the lower 48 states, which contain an estimated target population of 147,343 lakes and reservoirs that met the study criteria. A candidate list of lakes was randomly selected, and 500 sites were identified from the target population (via mapping and field reconnaissance) that could be accessed for fish collection.

Target species selection

Twelve predator species (e.g., largemouth bass) and six bottom-dwelling species (e.g., common carp) were targeted for the study (USEPA 2000a). Preferred target species were limited to reduce variability. Including these two distinct ecological groups of fish allowed monitoring of a variety of habitats, feeding strategies, and physiological factors that could result in differences in the bioaccumulation of chemicals. Target species were selected based on USEPA (2000b) recommendations that the species should be ubiquitous, abundant, easily identified, commonly consumed by humans, able to accumulate high concentrations of chemicals, and sufficiently large (i.e., adult specimens) to provide adequate tissue for analysis.

Sample collection

Fish composite samples were collected annually from about 125 different lakes and reservoirs across the country over a period of four years (2000–2003). Field teams used active (e.g., electrofishing) and passive (e.g., gill netting) sampling methods to collect separate composites of predators and bottom dwellers from each of the 500 lakes, primarily during the summer and fall months. Each composite consisted of five adult fish of the same species and similar size (i.e., the smallest individual in a composite was not less than 75% of the total length of the largest individual). Every sampling team applied study-specific standard operating procedures (SOPs) developed to establish consistent methods for fish collection, handling, and shipping (USEPA 2000a). Field teams measured total body length of each fish, wrapped whole fish in aluminum foil (foil rinsed with methylene chloride and oven dried), sealed the fish in food-grade polyethylene tubing, and completed standard field record forms. All fish were packed on dry ice and shipped overnight to the laboratory.

Target chemical selection

Candidate chemicals were derived from two primary sources: an EPA list of 451 persistent, bioaccumulative, and toxic (PBT) chemicals and a list of 130 chemicals referenced in several contemporary fish tissue and bioaccumulation studies (USEPA 1992, 1995, 1997, 1998; NOAA 1993). Chemical selection was based on the availability of detailed chemical-specific information, and chemicals were chosen based on their known tendency to accumulate and their importance in one or more EPA programs. A target list of 268 chemicals was compiled that included mercury, five forms of inorganic and organic arsenic, 17 dioxins and furans, the full complement of 209 polychlorinated biphenyl (PCB) congeners (yielding 159 measurements), 46 organochlorine (OC) and organophosphate (OP) pesticides, and 40 semivolatile organic compounds.

Sample analysis

Fish tissue samples were analyzed for 268 chemicals (Table 1). Fillets were analyzed for predator composites and whole bodies were analyzed for bottom-dweller composites to generate data relevant to both human and ecosystem health. All tissue samples were prepared by a single laboratory in a strictly controlled, contamination-free environment (USEPA 2005) prior to distribution to one of four commercial laboratories for analysis. Variability among sample results was minimized by using the same laboratory and method for analysis of each chemical for the duration of the study.

Standard EPA methods were applied for analysis of metals (Methods 1631B for total mercury and 1632A for arsenic), pesticides (Methods 1656A for OC pesticides and 1657A for OP pesticides), semivolatile organic chemicals (Method 1625C), PCBs (Method 1668A), and dioxins/furans (Method 1613B) to determine tissue concentrations (reported on a wet weight basis) (USEPA 2005). Each method specified procedures for analysis, QA/QC requirements, and reporting limits (USEPA 1989, 1994, 1999c, d, 2000c, d, 2001a, b). Study-specific modifications were made to two of the methods to achieve lower detection limits. Method 1613B for dioxins and furans was modified to increase tissue sample size used for analysis from 10 to 100 g, and a sixth calibration solution was added that contained all method-specified chemicals (at levels lower than specified in the method) to verify linearity at the targeted lower concentrations (i.e., ten times lower than specified in the method). Modifications of Method 1656A for OC pesticides included concentrating the tissue sample extracts before instrumental analysis by a factor of five beyond method-specified levels to ensure that all target pesticides could be quantified at levels less than or equal to the human health risk-based thresholds applied for this study.

Statistical methods

Statistical analysis of the fish tissue data was conducted using R statistical software (R Development Core Team 2007). Data were analyzed for each target chemical or chemical group by fish composite type. Elements of the probabilistic survey design, along with field data and laboratory results, were incorporated into analysis of the tissue data. The statistical analysis process consisted of the following steps: determining the status of each lake using site reconnaissance information; adjusting the survey design (sample) weights based on lake status; estimating the target population (i.e., number of lakes that met the definition of a lake); estimating the number and proportion of lakes in the sampled population (i.e., accessible target lakes); and developing estimates of percentiles and cumulative distribution of tissue concentrations by chemical and composite type for the sampled population of lakes (Olsen et al. 2008).

Data analysis focused on the study objective to develop estimates of the national distribution of mean levels (i.e., lake means or composite average concentrations) of the target chemicals in fish tissue. These national distributions were described by percentiles for target chemicals that dominated in frequency of occurrence. Cumulative distribution functions (CDFs; Zar 1999) were used to characterize the probability distribution of the concentration for selected chemicals in fish tissue. Probability distributions were presented for commonly detected chemicals as plots of chemical concentrations (x-axis) versus the cumulative number and percentage of lakes (y-axis).

Table 1 Target chemicals and associated analytical methods

Analytical method	Target chemical	
Total mercury by oxidation, purge and trap, and cold vapor atomic fluorescence spectrometry (Method 1631, Revision B with Appendix A—Digestion procedures for total mercury in tissue, sludge, sediment, and soil)	Mercury	
Arsenic speciation by arsine generation, chromatography, and atomic absorption spectrometry (Method 1632, Revision A)	Arsenic (III)	Dimethylarsinic acid (DMA)
	Arsenic (V)	Monomethylarsonic acid (MMA)
	Total inorganic arsenic	
Polychlorinated biphenyls by isotope dilution high-resolution gas chromatography/mass spectrometry (GC/MS) (Method 1668, Revision A)	209 congeners, including the following 12 “dioxin-like” congeners:	
	3,3′,4,4′-TeCB	3,3′,4,4′,5-PeCB
	3,4,4′,5-TeCB	2,3,3′,4,4′,5-HxCB
	2,3,3′,4,4′-PeCB	2,3,3′,4,4′,5′-HxCB
	2,3,4,4′,5-PeCB	2,3′,4,4′,5,5′-HxCB
	2,3′,4,4′,5-PeCB	3,3′,4,4′,5,5′-HxCB
	2′,3,4,4′,5-PeCB	2,3,3′,4,4′,5,5′-HpCB
Dioxins and furans by isotope dilution high-resolution GC/MS (Method 1613, Revision B)	2,3,7,8-TCDD	2,3,7,8-TCDF
	1,2,3,7,8-PeCDD	1,2,3,7,8-PeCDF
	1,2,3,4,7,8-HxCDD	2,3,4,7,8-PeCDF
	1,2,3,6,7,8-HxCDD	1,2,3,4,7,8-HxCDF
	1,2,3,7,8,9-HxCDD	1,2,3,6,7,8-HxCDF
	1,2,3,4,6,7,8-HpCDD	1,2,3,7,8,9-HxCDF
	OCDD	2,3,4,6,7,8-HxCDF
		1,2,3,4,6,7,8-HpCDF
		1,2,3,4,7,8,9-HpCDF
		OCDF
Organochlorine pesticides by gas chromatography/halide specific detector (GC/HSD) (Method 1656, Revision A)[a]	2,4′-DDD (TDE)[b]	Kepone (Chlordecone)
	2,4′-DDE[b]	Methoxychlor
	2,4′-DDT[b]	Mirex
	4,4′-DDD (TDE)	*cis*- and *trans*-Nonachlor
	4,4′-DDE	Octachlorostyrene
	4,4′-DDT	Oxychlordane
	Aldrin	Pendimethalin (Prowl)
	cis- and *trans*-Chlordane	Pentachloronitrobenzene (PCNB)
	Dicofol	*cis*-Permethrin and *trans*-Permethrin
	Dieldrin	Toxaphene
	Endosulfan sulfate	Trifluralin
	Endosulfan I	$\alpha-$BHC
	Endosulfan II	$\beta-$BHC
	Endrin	$\gamma-$BHC (Lindane)
	Ethalfluralin (Sonalan)	$\delta-$BHC
	Heptachlor	Pentachloroanisole
	Heptachlor epoxide	
	Isodrin	
Organophosphorus pesticides by gas chromatography/flame photometric detector (GC/FPD; Method 1657, Revision A)	Chlorpyrifos	Paraoxon
	Diazinon	Parathion (ethyl)
	Disulfoton	Terbufos
	Disulfoton sulfone	Terbufos sulfone
	Ethion	

Table 1 (continued)

Analytical method	Target chemical	
Semivolatile organic chemicals by isotope dilution GC/MS (Method 1625, Revision C with modifications for tissue)	3,3′-dichlorobenzidine	Butyl benzyl phthalate
	1,2,4,5-Tetrachlorobenzene	Chrysene (PAH)
	1,2,4-Trichlorobenzene	Dibenzo(a,h)anthrancene (PAH)
	1,2-Dichlorobenzene	Di-*n*-butyl phthalate
	1,3-Dichlorobenzene	Diethylstilbestrol (DES)
	1,4-Dichlorobenzene	Fluoranthene (PAH)
	2,4,5-Trichlorophenol	Fluorene (PAH)
	4,4′-Methylenebis (2-chloroaniline)	Hexachlorobenzene
	4-Bromophenyl phenyl ether	Hexachlorobutadiene
	4-Nonylphenol	Indeno(1,2,3-cd)pyrene (PAH)
	Acenaphthene (PAH)	Naphthalene (PAH)
	Acenaphthylene	Nitrobenzene
	Anthracene (PAH)	Pentachlorobenzene
	Benzo(*a*)anthracene (PAH)	Pentachlorophenol
	Benzo(*a*)pyrene (PAH)	Perylene (PAH)
	Benzo(*b*)fluoranthene (PAH)	Phenanthrene (PAH)
	Benzo(*g*,*h*,*i*)perylene (PAH)	Phenol
	Benzo(*j*)fluoranthene (PAH)	Phenol, 2,4,6-tris(1,1 dimethylethyl)-
	Benzo(*k*)fluoranthene (PAH)	Pyrene (PAH)
	Bis(2-ethylhexyl) phthalate	Tetrabromobisphenol A

[a]PCB Aroclors were not target chemicals for this study, but seven Aroclors were analyzed incidentally using Method 1656, Revision A, including Aroclors 1016, 1221, 1232, 1242, 1248, 1254, 1260

[b]Chemicals were added to the target chemical list after year 1 of the study

Results

An unequal probability design (Stevens and Olsen 2004) was used to develop national estimates of predator and bottom-dweller fish tissue concentrations for 268 target chemicals. This element of the study design required application of sample weights to the fish tissue data, which were derived from the various inclusion probabilities assigned to each of six lake size categories (Olsen et al. 2008). Assigning different probabilities to each category prevented small lakes from dominating the group of lakes selected for sampling and allowed a similar number of lakes to be sampled in each size category. Two statistical sets of lakes, the target population and the sampled population, define how broadly the fish tissue concentration results apply to lakes and reservoirs in the United States. Potential sampling locations were limited to the target population of an estimated 147,343 lakes in the lower 48 states that met the study definition of a lake (Alaska and Hawaii were not included due to resource and study design constraints). The sampled population consisted of all target lakes that were accessible for fish collection. Under ideal circumstances the target and sampled populations would coincide. Due to accessibility issues (e.g., private property access denial), the sampled population is a subset of the target population.

Sampling results

Fish were collected from 500 lakes (Fig. 1) selected randomly from the target population. Sampling locations included both private and public access lakes that ranged in size from one to 384,615 hectares (or from 2.5 to 950,000 surface acres). Sampling sites occurred in 47 of the lower 48 states (no lakes or reservoirs were randomly drawn in Delaware), and all sites are listed in USEPA (2007b). A total of 486 predator composites and 395 bottom-dweller composites were collected from the 500 lakes. Each of these composite types comprised nationally representative samples whose results can be extrapolated to the sampled population of an estimated 76,559 lakes for predators and 46,190 lakes for bottom dwellers.

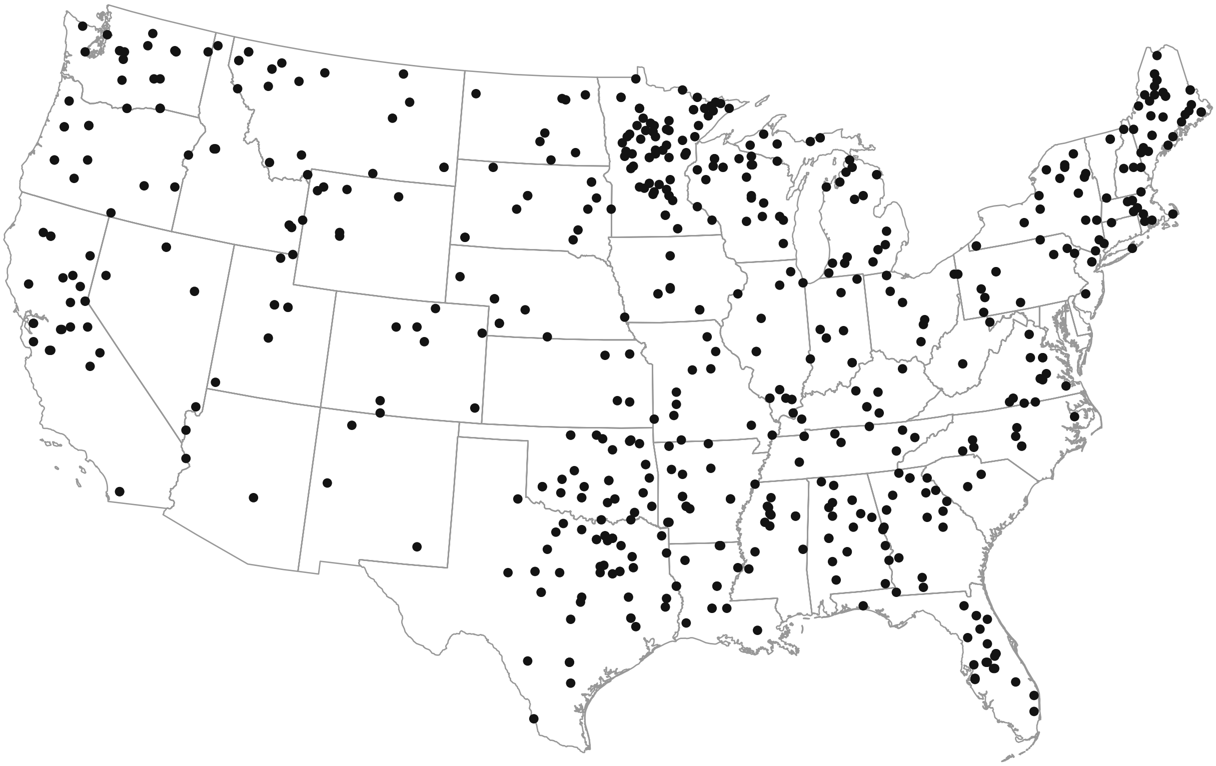

Fig. 1 Map of fish tissue sampling locations (500 lakes)

All results apply to these sampled populations of lakes.

Target fish species and size recommendations were based on guidance in USEPA (2000b), and were clearly identified in study-specific SOPs. Adherence to those procedures controlled the number of species submitted for analysis; however, the outcome of sampling efforts ultimately depended on the natural diversity and abundance of fish in the target lakes. All fish were adult specimens, and length ranges within each composite were strictly controlled by SOP requirements. For predator composites, a total of 31 species were collected nationwide, with four of the recommended target species representing 72% of the total. Largemouth bass (*Micropterus salmoides*), walleye (*Sander vitreus*), northern pike (*Esox lucius*), and smallmouth bass (*Micropterus dolomieu*) accounted for 50%, 10%, 7%, and 5% of all predator composites, respectively. Common carp (*Cyprinus carpio*), white sucker (*Catostomus commersoni*), and channel catfish (*Ictalurus punctatus*) accounted for 62% of all bottom-dweller composites.

Chemical detections

All fish tissue results were reported as wet weight concentrations and were expressed as the mass of the chemical per unit mass of fish tissue. Each analytical method specified the reporting units for a particular chemical or chemical group. In reporting the analytical results, it was important to distinguish between detection and presence of a chemical in fish tissue samples. Estimates of fish tissue concentrations ranging from the method detection limit (MDL) to the minimum level (ML) or quantitation limit were reported as being present with a 99% level of confidence. Study-specific information on MDLs and MLs is provided in USEPA (2007b). In cases where a chemical was reported as not detected at the MDL level, there is a 50% possibility that the chemical may be present. Therefore, results for chemicals not detected in fish tissue samples are reported here as less than the MDL rather than zero.

Forty-three target chemicals were not detected in any fish samples (Table 2). No detections were reported for the nine organophosphate pesticides (e.g., chlorpyriphos and diazinon), for one of the 209 PCB congeners (PCB-161), or for 16 of the 17 polycyclic aromatic hydrocarbons (PAHs) analyzed as semivolatile organic chemicals. Seventeen other semivolatile organic chemicals were not detected in fish tissue, including hexachlorobenzene, which was a priority chemical under EPA's PBT Chemical Program. Thirty-four target chemicals were detected infrequently in samples, including three forms of arsenic, 25 organochlorine pesticides, and six semivolatile organic compounds.

Cumulative distribution functions

Cumulative distribution function (CDF) plots were used to display chemical concentrations versus the cumulative number or percentage of lakes from the sampled population. Since many target chemicals occurred infrequently, they lacked sufficient data to develop a CDF with adequate resolution. Probability distributions were plotted only for chemicals with at least 50 data points (i.e., tissue concentrations above the MDL) and a published human health criterion or risk-based consumption limit. Five chemicals or chemical groups met these criteria, including total mercury, PCBs (sum of congeners), dioxins and furans (TEQ), total DDT, and total chlordane.

Application of human health thresholds (Table 3) on CDF plots allowed EPA to estimate the number of lakes with tissue concentrations above a level protective of human health. The value applied for mercury was the EPA fish tissue criterion for mercury (USEPA 2001c). Since EPA does not have fish tissue criteria for the other chemicals (PCBs, dioxins/furans, DDT, and chlordane), all other values were risk-based consumption limits published in USEPA (2000e). Specifically, these risk-based values were the upper limit of the four-meal-per-month concentration range for the conservative consumption limit where tissue concentrations were available for both cancer and noncancer health endpoints. Predator results are highlighted because the human health criterion or risk-

Table 2 Chemicals not detected in fish tissue composite samples

Chemical group	Chemical	
PCBs	PCB-161	
Organophosphate pesticides	Chlorpyrifos	Ethyl parathion
	Diazinon	Paraoxon
	Disulfoton	Terbufos
	Disulfoton sulfone	Terbufos sulfone
	Ethion	
PAHs	Acenaphthene	Benzo(*k*)fluoranthene
	Acenaphthylene	Chrysene
	Anthracene	Dibenzo(*a*,*h*)anthracene
	Benzo(*a*)anthracene	Fluoranthene
	Benzo(*a*)pyrene	Fluorene
	Benzo(*b*)fluoranthene	Indeno (1,2,3-cd) pyrene
	Benzo(*g*,*h*,*i*)perylene	Perylene
	Benzo(*j*)fluoranthene	Phenanthrene
Other semivolatile organic chemicals	1,2,4,5-Tetrachlorobenzene	4-Bromophenyl phenyl ether
	1,2,4-Trichlorobenzene	Diethylstilbestrol
	1,2-Dichlorobenzene	Hexachlorobenzene
	1,3-Dichlorobenzene	Hexachlorobutadiene
	1,4-Dichlorobenzene	Nitrobenzene
	2,4,5-Trichlorophenol	Pentachlorobenzene
	2,4,6-Tris(1,1-dimethylethyl) phenol	Pentachlorophenol
	3,3′-dichlorobenzidine	Tetrabromobisphenol A
	4,4′-Methylenebis (2-chloroaniline)	

Table 3 Values for interpreting predator results

Chemical	Value type	Health endpoint	Fish tissue concentration	Units
Mercury	Criterion[a]	Noncancer	300	ppb
PCBs	Risk-based threshold[b]	Cancer	12	ppb
Dioxins/Furans	Risk-based threshold[b]	Cancer	0.15	ppt
DDT	Risk-based threshold[b]	Cancer	69	ppb
Chlordane	Risk-based threshold[b]	Cancer	67	ppb

[a]Value for mercury is the tissue-based water quality criterion (WQC) published by EPA in January 2001 (USEPA 2001c)

[b]Taken from *EPA's guidance for assessing chemical contaminant data for use in fish advisories, volume 2: Risk assessment and fish consumption limits, 3rd ed.* (USEPA 2000e)

based consumption limits were readily available to interpret the fillet data for chemicals that occurred frequently in edible tissue.

Mercury results

Freshwater fish contamination studies have shown that methylmercury can account for (on average) more than 90% of the mercury concentration in predator fish tissue (Bloom 1992; USEPA 2006b). USEPA (2000b) and USEPA (2006b) recommended monitoring for total mercury concentrations (rather than methylmercury) in fish contaminant screening studies, applying the conservative assumption that all mercury is present in fish tissue as methylmercury. The fish tissue criterion for methylmercury was applied to mercury results to identify the number (and percentage) of the sampled population of the Nation's lakes that exceed this threshold. The "Water Quality Criterion for the Protection of Human Health: Methylmercury" (USEPA 2001c) guidance identified a fish tissue criterion of 0.3 mg methylmercury per kilogram (300 ppb) of fish tissue (wet weight). This represents the concentration in fish tissue that should not be exceeded based on a total consumption-weighted rate of 0.0175 kg of fish/day (assuming a human adult body weight default value of 70 kg and a reference dose of 0.0001 mg/kg day).

Mercury was detected in 100% of the composite samples collected for this study at concentrations above the quantitation limit of 2 ng/g (ppb). Concentrations in predators ranged from 23 ppb to a maximum of 6,605 ppb, and the median concentration was 285 ppb (Table 4). Bottom-dweller concentrations ranged from 5 to 596 ppb, with a median concentration of 69 ppb (Table 5). The mean mercury concentration was 352 ppb for predators and 96 ppb for bottom dwellers. National means were calculated for mercury only. A statistically valid mean could be calculated for mercury because it was found in every sample above the quantitation limit. A CDF graph (Fig. 2) was used to illustrate the predator mercury concentrations that correspond to the percentage and number of the sampled population of lakes. The mercury fish tissue criterion was overlaid on the CDF to identify the number and percentage of lakes with fish tissue mercury concentrations above or below the threshold. The CDF (Fig. 2) showed that edible portions (fillets) of predators in 48.8% of the sampled population of lakes had tissue concentrations that exceeded the EPA 300 ppb fish tissue criterion for mercury, representing a total of 36,422 lakes nationwide.

Table 4 Percentiles for concentrations of five commonly detected chemicals in predator composites

Chemical	10th	25th	50th	75th	90th	95th	Maximum
Mercury	89.33	176.67	284.60	432.08	561.79	833.41	6605 ppb
PCB (sum of congeners)	0.49	1.00	2.16	8.13	18.17	33.16	704.92 ppb
Dioxin and Furan (TEQ)	<MDL	<MDL	0.006	0.05	0.11	0.32	7.54 ppt
Total DDT	<MDL	<MDL	1.47	6.95	19.68	30.57	1481.40 ppb
Total Chlordane	<MDL	<MDL	<MDL	<MDL	3.62	8.27	99.99 ppb

Table 5 Percentiles for concentrations of five commonly detected chemicals in bottom-dweller composites

Chemical	10th	25th	50th	75th	90th	95th	Maximum
Mercury	20.10	39.27	68.56	124.17	219.58	247.31	596 ppb
PCB (sum of congeners)	2.31	5.15	13.90	70.87	130.79	198.32	1266.25 ppb
Dioxin and Furan (TEQ)	0.06	0.16	0.41	1.07	1.77	2.01	23.81 ppt
Total DDT	1.82	4.23	12.68	35.35	153.92	218.63	1760.57 ppb
Total Chlordane	<MDL	<MDL	1.65	9.31	25.96	30.93	377.98 ppb

Total PCB results

All samples were analyzed for the full complement of 209 PCB congeners, which produced 159 congener measurements. These measurements included results for 126 individual congeners and 33 groups of two to six co-eluting congeners. USEPA (2000b) recommends the reporting of total PCB concentrations (calculated as the sum of the concentrations of the congeners or homologues, i.e., co-eluting groups), since Aroclor analysis does not adequately represent bioconcentrated PCB mixtures found in fish tissue. A risk-based tissue residue health endpoint for total PCBs was applied to predator results to identify the number (and percentage) of the sampled population of the Nation's lakes that exceed the risk-based threshold. USEPA (2000e) lists a cancer health endpoint of 0.012 ppm (12 ppb) PCBs (wet weight) in fish tissue. This threshold represents the fish tissue concentration that should not be exceeded based on a total consumption-weighted rate of four 8-oz (0.227 kg) fish meals per month [assuming a human adult body weight default value of 70 kg, a cancer slope factor of 2 (mg/kg day)$^{-1}$, and a 1 in 100,000 risk level] (USEPA 2000e).

PCBs were detected in 100% of the composite samples. Total PCB concentrations (sum of the congeners) in predators ranged from 0.06 to 704.92 ppb, and the median concentration was 2.16 ppb (Table 4). Bottom-dweller concentrations ranged from 0.60 to 1,266.25 ppb, and the median concentration was 13.90 ppb (Table 5). Figure 3 indicates that edible portions of predators in 16.8% of the sampled population of lakes had total PCB tissue concentrations that exceeded the 12 ppb human health risk-based threshold, representing a total of 12,886 lakes nationwide.

Total dioxin and furan results

Samples were analyzed for 17 dioxins and furans, and results were reported as toxicity-weighted total concentrations derived by multiplying results for each chemical by an individual toxicity equivalency factor (TEF), then summing

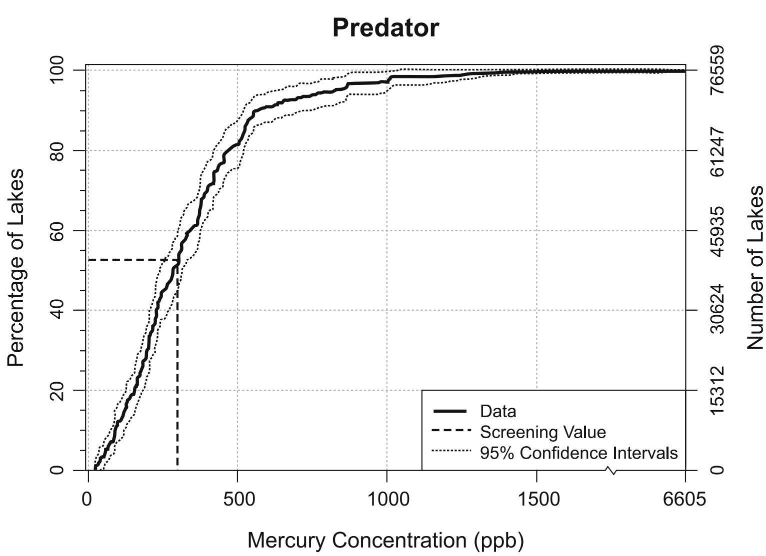

Fig. 2 Cumulative distribution function of mercury concentrations in predator samples

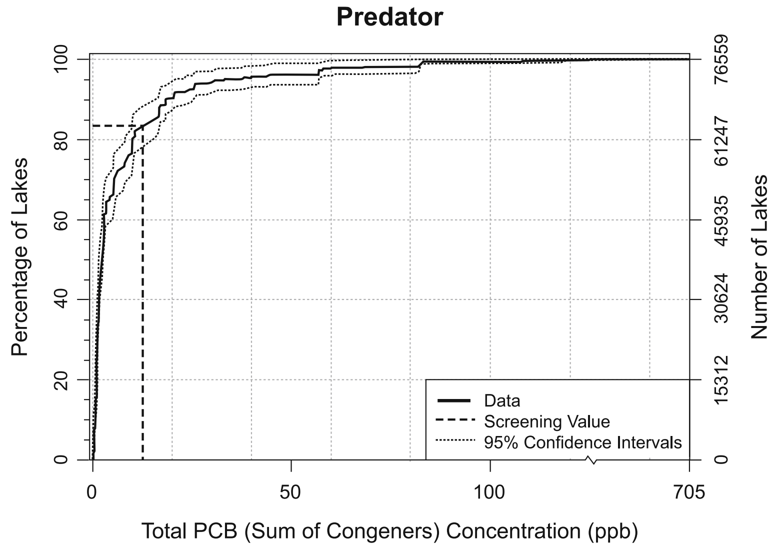

Fig. 3 Cumulative distribution function of total PCB concentrations in predator samples

the 17 results (for a TEQ concentration). USEPA (2000e) recommends analyzing the 17 dioxins and furans together as a simplifying and interim approach until further guidance is available on this chemical group. A risk-based tissue residue health endpoint of 0.15 ppt for dioxins and furans (TEQ-wet weight) (USEPA 2000e) was applied to predator dioxin and furan results. This represents the concentration in fish tissue that should not be exceeded based on a total consumption-weighted rate of four 8-oz (0.227 kg) fish meals per month [assuming a human adult body weight default value of 70 kg, a cancer slope factor of 1.56×10^{-5} (mg/kg day)$^{-1}$, and a 1 in 100,000 risk level] (USEPA 2000e).

Dioxins and furans were detected in 81% of the predator samples and 99% of the bottom-dweller samples. Concentrations in predators ranged from 0.00002 ppt (TEQ) to 7.54 ppt (TEQ), and the median concentration was 0.006 ppt (TEQ) (Table 4). Bottom-dweller concentrations ranged from 0.0008 to 23.81 ppt (TEQ), with a median concentration of 0.41 ppt (TEQ; Table 5). Figure 4 shows that edible portions of predators in 7.6%

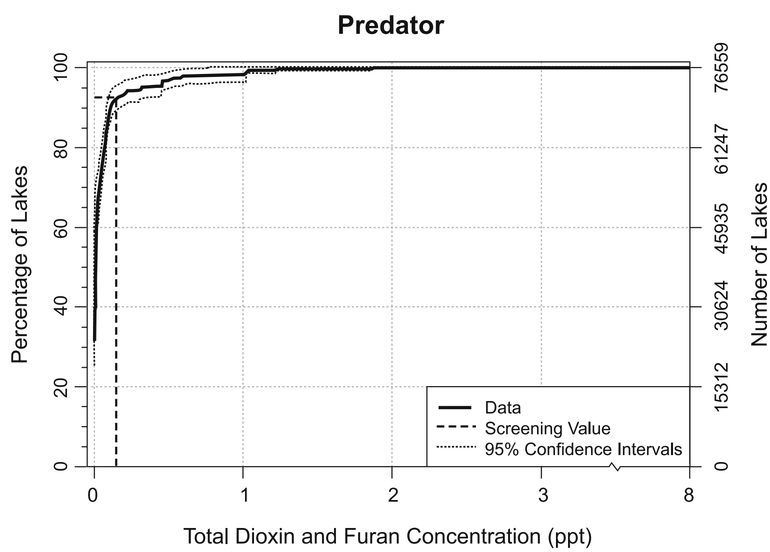

Fig. 4 Cumulative distribution function of dioxin and furan (TEQ) concentrations in predator samples

of the sampled population of lakes had tissue concentrations that exceeded the 0.15 ppt (TEQ) human health risk-based threshold for dioxins and furans, representing a total of 5,856 lakes across the country.

Total DDT results

Fish are typically exposed to a mixture of DDT, DDE, DDD, and their degradation and metabolic products. USEPA (2000e) recommends reporting total DDT fish tissue results, based on the sum of the 4,4′- and 2,4′ isomers of DDT, DDE, and DDD. A cancer health endpoint of 0.069 ppm (69 ppb) DDT (wet weight) in fish tissue (USEPA 2000e) was applied to predator results. This represents the concentration in fish tissue that should not be exceeded based on a total consumption-weighted rate of four 8-oz (0.227 kg) fish meals per month [assuming a human adult body weight default value of 70 kg, a cancer slope factor of 0.34 (mg/kg day)$^{-1}$, and a 1 in 100,000 risk level] (USEPA 2000e).

DDT was detected in 78% of the predator samples and 98% of bottom-dweller samples. Total DDT concentrations in predators ranged from 0.77 to 1,481.4 ppb, and the median concentration was 1.47 ppb (Table 4). Bottom-dweller concentrations ranged from 0.82 to 1,760.57 ppb, with a median concentration of 12.68 ppb (Table 5). Edible portions of predators in 1.7% of the sampled population of lakes had tissue concentrations that exceeded the 69 ppb human health risk-based threshold for DDT, representing a total of 1,329 lakes across the country (Fig. 5).

Total chlordane results

All samples were analyzed for *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, and oxychlordane. Results for these five degradation products of chlordane were summed to yield values for total chlordane. USEPA (2000b) recommends monitoring for total chlordane concentrations in fish contaminant and health advisory surveys. A cancer health endpoint of 67 ppb chlordane (wet weight) in fish tissue (USEPA 2000e) was applied to predator chlordane results. This risk-based endpoint represents the concentration in fish tissue that should not be exceeded based on a total consumption-weighted rate of four 8-oz (0.227 kg) fish meals per month [assuming a human adult body weight default value of 70 kg, a cancer slope factor of 0.35 (mg/kg day)$^{-1}$, and a 1 in 100,000 risk level] (USEPA 2000e).

Chlordane was detected in 20% of the predator samples and 50% of bottom-dweller samples. Concentrations in predators ranged from 0.59 to 99.99 ppb, and the median concentration was less than the MDL for total chlordane

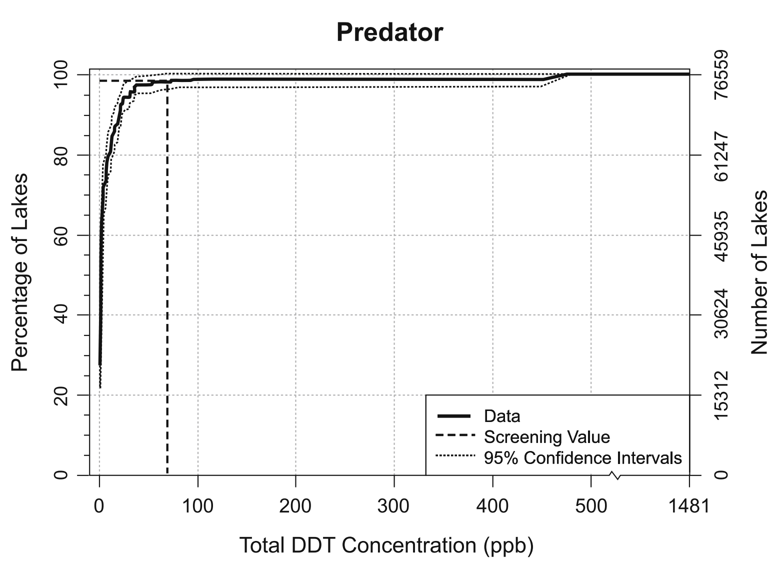

Fig. 5 Cumulative distribution function of total DDT concentrations in predator samples

(Table 4). Bottom-dweller concentrations ranged from 0.50 to 377.98 ppb, with a median concentration of 1.65 ppb (Table 5). Predator fillets in 0.3% of the sampled population of lakes had fish tissue concentrations that exceeded the 67 ppb human health risk-based threshold for total chlordane, representing a total of 235 lakes nationwide (Fig. 6).

Sampling variability

A representative set of lakes was selected for collection of replicate samples. Field teams collected predator and bottom-dweller replicates from 70 and 52 of these lakes, respectively. Replicate and standard composites were collected simultaneously to provide data for estimating sampling variability, and they were compared using detection limits to evaluate agreement between sample pairs. The paired sample results were divided into three categories for comparison: both pairs below detection, both pairs above detection, and pairs with one below and one above detection.

Sample pair comparisons showed 100% agreement for the 43 chemicals not detected in any of the fish samples (i.e., those chemicals absent in standard samples were also absent in their paired replicate samples). Detections were also in complete agreement for predator and bottom-dweller mercury results, in that mercury occurred in all sample pairs above detection. Detection results varied for the other commonly detected chemicals. For the 159 PCB congener results, 75% of the paired predator results and 80% of the paired bottom-dweller results showed 90% to 100% agreement between detections. Agreement between dioxin and furan detections ranged from 71% to 100% for both the predator and bottom-dweller sample pairs. Predator detection agreement ranged from 87% to 100% for DDT and 94% to 100% for chlordane, while agreement in detections for bottom dwellers ranged from 75% to 100% for DDT and 88% to 96% for chlordane.

Discussion

The survey design incorporated statistical techniques (e.g., quartering) to ensure that sampling sites were well distributed throughout the lower 48 states (Olsen et al. 2008). Clusters of sites in a few geographic areas (e.g., Minnesota and Texas) reflected the greater availability of candidate lakes and reservoirs for selection (Fig. 1). Selection of a nationally representative sample of lakes for each of the four sampling years (2000–2003) was another important component of the survey design. It was included to allow development of national estimates of the distribution of PBT chemicals in lake fish (with wider confidence intervals) if the study was terminated before 2003.

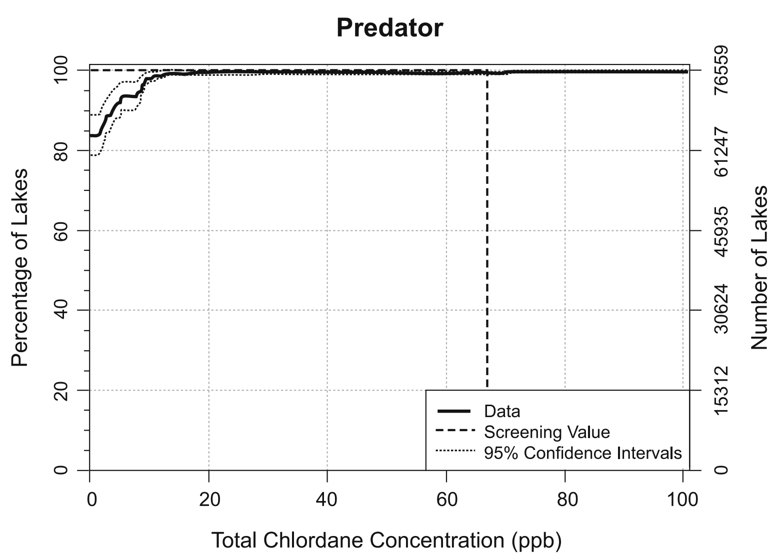

Fig. 6 Cumulative distribution function of total chlordane concentrations in predator samples

According to the National Listing of Fish Advisories (USEPA 2007c), mercury, PCBs, dioxins and furans, DDT, and chlordane accounted for nearly 98% of the advisories in effect at the end of 2006. This study confirms that those same chemicals were commonly detected in fish from lakes and reservoirs of the conterminous United States. Results indicated that mercury, PCBs, and dioxins/furans, in particular, were widely distributed.

Mercury contamination is ubiquitous because atmospheric deposition can carry inorganic mercury far from anthropogenic or natural point sources (Fitzgerald et al. 1998). A major source of atmospheric mercury is the natural degassing of the earth's crust; however, mercury releases are also attributable to anthropogenic sources such as mining and smelting, industrial processes, and the combustion of fossil fuels (ATSDR 1999). In the aquatic environment, microorganisms convert deposited (inorganic) mercury to toxic methylmercury that accumulates in fish. Nearly all fish and shellfish contain traces of mercury (USEPA 2007c), and methylmercury in fish is known to bind to amino acids in fish muscle tissue (Bloom 1992; Leaner and Mason 2004). Elevated mercury concentrations in fish are the leading cause of fish consumption advisories, accounting for 80% of the advisories listed in EPA's 2005/2006 National Listing of Fish Advisories. In 2006, 35% of the total lake acres in the US were under advisory for mercury (USEPA 2007c). Results from this study confirm the wide distribution of mercury since it was detected at quantifiable levels in every fish sample from all 500 sampling locations. Mercury concentrations were generally lower in bottom dwellers than in predators. This is consistent with the findings of Rose et al. (1999) and Peterson et al. (2007) where piscivorous predator species typically accumulated higher concentrations of mercury than non-piscivores. Statistical analysis showed that mercury concentrations in predators occurred above the EPA 300 ppb mercury fish tissue criterion at nearly half of the lakes in the sampled population. These elevated mercury concentrations in predators apply to an estimated 36,422 lakes (or 48.8% of the sampled population) in the lower 48 states. Overall, the results from this probability-based study underscore the pervasive nature of mercury deposition on lakes in the conterminous US.

Although the US banned the production and use of PCBs in 1979, study results show that they are still widely distributed in the environment. PCBs can still be released into the environment from illegal or improper disposal of industrial wastes, leaks from hazardous waste sites, and burning of some wastes in incinerators (ATSDR 2000). PCBs are extremely persistent in the environment, and they can be widely dispersed by atmospheric transport. In water, some PCBs may remain dissolved, but most partition into bottom sediments and adsorb onto organic particles. USEPA (2007c) identified 1,023 fish consumption advisories for PCBs in 2006 that affected 11.6% of the Nation's total lake acres. Like mercury, PCBs were detected in 100% of both the predator and bottom-dweller samples. Concentrations in bottom dwellers were generally higher than levels detected in predators. This may be linked to the lipophilic nature of PCBs, which tend to accumulate in fatty tissues and organs (Bruggeman et al. 1984; Larsson et al. 1993). Total PCB concentrations exceeded the human health risk-based threshold of 12 ppb at nearly 17% of the lakes in the sampled population, or an estimated 12,886 lakes in the lower 48 states.

Dioxins and furans commonly refer to a group of synthetic organic chemicals that includes 210 structurally related chlorinated dibenzo-*p*-dioxins and chlorinated dibenzofurans (USEPA 2000e). The generic term dioxin is used here to refer to the aggregate of this group of compounds. Dioxins are not produced commercially, but rather are unintentional byproducts of combustion and certain industrial chemical processes (ATSDR 1998). In 2006, a total of 38,181 lake acres or 1% of the Nation's total lake acres were under a consumption advisory for dioxins (USEPA 2007c). Dioxin levels in the environment have been declining since the early 1970s (USEPA 2006c); however, this study confirms that they remain widely distributed. Dioxins and furans were detected in 99% of the bottom-dweller samples and in 81% of the predator samples. Higher concentrations occurred in the whole-body tissue of bottom dwellers than in the fillets of predators, which may be linked to their lipophilic nature (Bruggeman et al. 1984;

Jones et al. 2001). Total dioxin and furan concentrations in predator samples exceeded the 0.15 ppt (TEQ) human health risk-based threshold at nearly 8% of the lakes in the sampled population or an estimated 5,856 lakes.

During the 1970s and 1980s, the US banned the manufacture and use of most of the organochlorine pesticides included in the study. The low percentages of detections for the majority of these pesticides suggest that they are being effectively sequestered in lake environments. Two notable exceptions were DDT and chlordane. DDT was detected in 98% of the bottom-dweller samples and 78% of the predator samples. Chlordane was detected in 50% and 20% of bottom-dwellers and predators, respectively. Both pesticides had higher concentrations in the whole-body tissue of the bottom dwellers than in the predator fillets, consistent with the findings of Bruggeman et al. (1984), Larsson et al. (1993), and Ruus et al. (2002). Total concentrations of DDT and chlordane rarely exceeded their respective human health risk-based thresholds of 69 ppb and 67 ppb. Total DDT in predator samples exceeded the threshold at less than 2% of the lakes in the sampled population (an estimated 1,329 lakes). Total chlordane concentrations in predators occurred above the threshold value at less than 1% of the lakes in the sampled population (an estimated 230 lakes).

Conclusions

This study generated the first national freshwater fish contamination dataset to allow a statistically valid evaluation of PBT chemicals in lakes and reservoirs of the conterminous United States. It produced complete and high quality data, and set a high quality assurance/quality control (QA/QC) standard for fish tissue studies (USEPA 2005). A study of this magnitude could not be conducted without a large network of partners. Participation of fisheries experts from more than 50 agencies ensured success and provided an effective model of state, federal, and tribal collaboration. The full complement of data from this study defined a national baseline of fish contamination in lakes and reservoirs of the lower 48 states. Future monitoring will be necessary to determine contaminant trends in lake fish tissue and to track changes in PBT chemical concentrations resulting from pollution control activities.

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References

Agency for Toxic Substances and Disease Registry (ATSDR). (1998). *Toxicological profile for chlorinated dibenzo-p-dioxins (CDDs).* Atlanta, GA: US Department of Health and Human Services, Public Health Service.

Agency for Toxic Substances and Disease Registry (ATSDR). (1999). *Toxicological profile for mercury.* Atlanta, GA: US Department of Health and Human Services, Public Health Service.

Agency for Toxic Substances and Disease Registry (ATSDR). (2000). *Toxicological profile for polychlorinated biphenyls.* Atlanta, GA: US Department of Health and Human Services, Public Health Service.

Bloom, N. (1992). On the chemical form of mercury in edible fish and marine invertebrate tissue. *Canadian Journal of Fisheries and Aquatic Sciences, 49,* 1010–1017.

Branson, D. R., Takahashi, I. T., Parker, W. M., & Blau, G. E. (1985). Bioconcentration kinetics of 2,3,7,8-tetrachlorodibenzo-p-dioxin in rainbow trout. *Environmental Toxicology and Chemistry, 4,* 779–788.

Bruggeman, W. A., Opperhuizen, A., Wijbenga, A., & Hutzinger, O. (1984). Bioaccumulation of super-lipophilic chemicals in fish. *Toxicological and Environmental Chemistry, 7,* 173–189.

Cope, W. G., Wiener, J. G., & Rada, R. G. (1990). Mercury accumulation in yellow perch in Wisconsin seepage lakes—relation to lake characteristics. *Environmental Toxicology and Chemistry, 9,* 931–940.

Fitzgerald, W. F., Engstrom, D. R., Mason, R. P., & Nater, E. A. (1998). The case for atmospheric mercury contamination in remote areas. *Environmental Science and Technology, 32,* 1–7.

Gardner, A. M., & White, K. D. (1990). Polychlorinated dibenzofurans in the edible portion of selected fish. *Chemosphere, 21*(1–2), 215–222.

Glassmeyer, S. T., Myers, T. R., DeVault, D. S., & Hites, R. A. (1997). Toxaphene in Great Lakes fish: A temporal, spatial, and trophic study. *Environmental Science and Technology, 31,* 84–88.

Isensee, A. R., & Jones, G. E. (1975). Distribution of TCDD in aquatic organisms. *Environmental Science and Technology, 9,* 668–672.

Jones, P. D., Kannan, K., Newsted, J. L., Tillitt, D. E., Williams, L. L., & Giesy, J. P. (2001). Accumulation of 2,3,7,8-tetrachlorodibenzo-p-dioxin by rainbow trout (*Onchorhynchus mykiss*) at environmentally relevant dietary concentrations. *Environmental Toxicology and Chemistry, 20*(2), 344–350.

Kucklick, J. R., Harvey, H. R., Ostrom, P. H., Ostrom, N. E., & Baker, J. E. (1996). Organochlorine dynamics in the pelagic food web of Lake Baikal. *Environmental Toxicology and Chemistry, 15*(8), 1388–1400.

Lange, T. R., Royals, H. E., & Connor, L. L. (1993). Influence of water chemistry on mercury concentration in largemouth bass from Florida lakes. *Transactions of the American Fisheries Society, 122,* 78–84.

Larsson, P., Okla, L., & Collvin, L. (1993). Reproductive status and lipid content as factors in PCB, DDT, and HCH contamination of a population of pike (*Esox lucius* L.). *Environmental Toxicology and Chemistry, 12,* 855–861.

Leaner, J. J., & Mason, R. P. (2004). Methylmercury uptake and distribution kinetics in sheepshead minnows, *Cyprinodon variegatus,* after exposure to Ch_3Hg-spiked food. *Environmental Toxicology and Chemistry, 23*(9), 2138–2146.

Madenjan, C. P., Desorcie, T. J., Stedman, R. M., Brown, E. H., Jr., Eck, G. W., Schmidt, L. J., et al. (1999). Spatial patterns in PCB concentrations of Lake Michigan lake trout. *Journal of Great Lakes Research, 25,* 149–159.

McCarty, L. S., & Mackay, D. (1993). Enhancing ecotoxicological modeling and assessment. *Environmental Science and Technology, 27,* 1718–1728.

National Oceanic and Atmospheric Administration (NOAA) (1993). *Sampling and analytical methods of the national status and trends program-national benthic surveillance and mussel watch projects 1984–1992.* NOAA Technical Memorandum NOS ORCA 71. July, 1993.

Olsen, A. R., Stevens, D. L., Jr., & White, D. (1998). Application of global grids in environmental sampling. *Computing Science and Statistics, 30*, 279–284.

Olsen, A. R., Snyder, B. D., Stahl, L. L., & Pitt, J. L. (2008). Survey design for lakes and reservoirs in the United States to assess contaminants in fish tissue. *Environmental Monitoring and Assessment* doi:10.1007/s10661-008-0685-8.

Peterson, S. A., VanSickle, J., Herlihy, A. T., & Hughes, R. M. (2007). Mercury concentration in fish from streams and rivers throughout the western United States. *Environmental Science and Technology, 41*(1), 58–65.

R Development Core Team. (2007). *The R project for statistical computing.* R Development Core Team, Vienna, Austria (website). http://www.R-project.org.

Rose, J., Hutcheson, M. S., West, C. R., Pancorbo, O., Hulme, K., Cooperman, A., et al. (1999). Fish mercury distribution in Massachusetts, USA lakes. *Environmental Toxicology and Chemistry, 18*(7), 1370–1379.

Ruus, A., Ugland, K. I., & Skaare, J. U. (2002). Influence of trophic position on organochlorine concentrations and compositional patterns in a marine food web. *Environmental Toxicology and Chemistry, 21*(11), 2356–2364.

Sloan, R., Brown, M., Brandt, R., & Barnes, C. (1985). Hudson River PCB relationships between resident fish, water, and sediment. *Northeastern Environmental Science, 3,* 137–151.

Stafford, C. P., & Haines, T. A. (1997). Mercury concentration in Maine sport fish. *Transactions of the American Fisheries Society, 126,* 144–152.

Stevens, D. L., & Olsen, A. R. (2004). Spatially balanced sampling of natural resources. *Journal of the American Statistical Association, 99,* 262–278.

Stow, C. A., & Carpenter, S. R. (1994). PCB concentration trends in Lake Michigan coho (*Oncorhynchus kisutch*) and chinook salmon (*O. tshawytscha*). *Canadian Journal of Fisheries and Aquatic Sciences, 51,* 1384–1390.

US Environmental Protection Agency (USEPA) (1987). *National dioxin study: Tiers 3, 5, 6, and 7.* US EPA, Office of Water Regulations and Standards, Washington, DC. EPA-440/4-87-003.

US Environmental Protection Agency (USEPA) (1989). *Method 1625C: Semivolatile organic compounds by isotope dilution GC/MS.* US EPA, Office of Science and Technology, Engineering and Analysis Division. Washington, DC. Region 1#01A0006089.

US Environmental Protection Agency (USEPA) (1992). *National study of chemical residues in fish.* US EPA, Office of Science and Technology, Washington, DC. EPA 823-R-92-008a.

US Environmental Protection Agency (USEPA) (1994). *Method 1613: Tetra- through octa-chlorinated dioxins and furans by isotope dilution HRGC/HRMS. Revision B.* US EPA, Office of Water, Washington, DC. EPA 821-B-94-005.

US Environmental Protection Agency (USEPA) (1995). *Guidance for assessing chemical contaminant data* *for use in fish advisories. volume 1: Fish sampling and analysis, 2nd ed.* US EPA, Office of Water, Washington, DC. EPA 823-R-95-007.

US Environmental Protection Agency (USEPA) (1997). *Guidance for assessing chemical contaminant data for use in fish advisories. volume 2: Risk assessment and fish consumption limits, 2nd ed.* US EPA, Office of Water, Washington, DC. EPA 823-B-97-009.

US Environmental Protection Agency (USEPA) (1998). *A multimedia strategy for priority persistent, bioaccumulative, and toxic (PBT) pollutants [DRAFT].* US EPA, Office of Pollution Prevention and Toxics, Washington, DC.

US Environmental Protection Agency (USEPA) (1999a). *The national survey of mercury concentrations in fish—Database summary 1990–1995.* US EPA, Office of Water, Washington, DC. EPA-823-R-99-014.

US Environmental Protection Agency (USEPA) (1999b). *Update: National listing of fish and wildlife advisories.* US EPA, Office of Water, Washington, DC. EPA-823-R-99-005.

US Environmental Protection Agency (USEPA) (1999c). *Method 1631, Revision B: Mercury in water by oxidation, purge and trap, and cold vapor atomic fluorescence spectrometry.* US EPA, Office of Water, Washington, DC. EPA 821-R-99-005.

US Environmental Protection Agency (USEPA) (1999d). *Method 1668, Revision A: Chlorinated biphenyl congeners in water, soil, sediment, and tissue by HRGC/HRMS.* US EPA, Office of Water, Washington, DC. EPA-821-R-00-002.

US Environmental Protection Agency (USEPA) (2000a). *Field sampling plan for the national study of chemical residues in lake fish tissue.* US EPA, Office of Water, Office of Science and Technology, Washington, DC. EPA-823-R-02-004.

US Environmental Protection Agency (USEPA) (2000b). *Guidance for assessing chemical contaminant data for use in fish advisories, volume 1: Fish sampling and analysis, 3rd ed.* US EPA, Office of Water, Washington, DC. EPA-823-B-00-007.

US Environmental Protection Agency (USEPA) (2000c). *Method 1656, Revision A: Organo-halide pesticides in wastewater, soil, sludge, sediment, and tissue by GC/HSD.* US EPA, Office of Water, Washington, DC. EPA-821-R-00-017.

US Environmental Protection Agency (USEPA) (2000d). *Method 1657, Revision A: Organo-phosphorus pesticides in wastewater, soil, sludge, sediment, and tissue by GC/FPD.* US EPA, Office of Water, Washington, DC. EPA-821-R-00-018.

US Environmental Protection Agency (USEPA) (2000e). *Guidance for assessing chemical contaminant data for use in fish advisories, volume 2: Risk assessment and fish consumption limits, 3rd ed.* US EPA, Office of Water, Office of Science and Technology, Washington, DC. EPA 823-B-00-008.

US Environmental Protection Agency (USEPA) (2001a). *Appendix to Method 1631: Total mercury in tissue, sludge, sediment, and soil by acid digestion and BrCl oxidation.* US EPA, Office of Water, Washington DC. EPA-821-R-01-013.

US Environmental Protection Agency (USEPA) (2001b). *Method 1632, Revision A: Chemical speciation of arsenic in water and tissue by hydride generation quartz furnace atomic absorption spectrometry.* US EPA, Office of Water, Washington, DC. EPA-821-R-01-006.

US Environmental Protection Agency (USEPA) (2001c). *Water quality criterion for the protection of human health: methylmercury.* US EPA, Office of Water, Office of Science and Technology, Washington, DC. EPA-823-R-01-001.

US Environmental Protection Agency (USEPA) (2005). *Quality assurance report for the national study of chemical residues in lake fish tissue: Analytical data for years 1 through 4.* US EPA, Office of Water, Washington, DC. EPA-823-R-05-005.

US Environmental Protection Agency (USEPA) (2006a). *Wadeable stream assessment: A collaborative survey of the Nation's streams.* US EPA, Office of Water, Washington, DC. EPA 841-B-06-002.

US Environmental Protection Agency (USEPA) (2006b). *Draft guidance for implementing the January 2001 methylmercury water quality criterion.* US EPA, Office of Water, Washington, DC. EPA 823-B-04-001.

US Environmental Protection Agency (USEPA) (2006c). *Persistent, bioaccumulative, and toxic (PBT) chemical program: Dioxins and furans.* US EPA (website). http://www.epa.gov/pbt/pubs/dioxins.htm.

US Environmental Protection Agency (USEPA) (2007a). *National Estuary Program coastal condition report.* US EPA, Office of Water, Washington, DC. EPA-842-B-06-001.

US Environmental Protection Agency (USEPA) (2007b). *National lake fish tissue study: results.* US EPA (website). http://www.epa.gov/waterscience/fish/study/results.htm.

US Environmental Protection Agency (USEPA) (2007c). *2005/2006 National listing of fish advisories fact sheet.* US EPA, Office of Water, Washington, DC. EPA-823-F-07-003.

Yeardley, R. B., Jr., Lazorchak, J. M., & Paulsen, S. G. (1998). Elemental fish tissue contamination in northeastern U.S. lakes: Evaluation of an approach to regional assessment. *Environmental Toxicology and Chemistry, 17*(9), 1875–1884.

Zar, J. H. (1999). *Biostatistical analysis* (4th ed.). Upper Saddle River, NJ: Prentice Hall.