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The National Study of Chemical Residues in Lake Fish Tissue

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U.S. Environmental Protection Agency
Office of Water
Office of Science and Technology

September 2009

Notice

This report was prepared by the U.S. Environmental Protection Agency (EPA), Office of Water, Office of Science and Technology. It has been subjected to the Agency's peer review and administrative review processes. The EPA Project Manager for preparation of this document was Leanne Stahl who provided overall project coordination and technical direction. Tetra Tech, Inc. provided primary support for the development of this document under Contract Numbers EP-C-04-030 and EP-C-09-019. Blaine Snyder was the Tetra Tech Project Manager. Additional support was provided by Computer Sciences Corporation under Contract Number ET-W-06-046.

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www.epa.gov/waterscience/fishstudy/

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Table of Contents

Figures

Tables

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List of Acronyms and Abbreviations

ATSDR	Agency for Toxic Substances and Disease Registry
BHC	benzene hexachloride
CAS	Chemical Abstracts Service
CD	compact disc
CDF	cumulative distribution function
CEC	containment of emerging concern
CFR	Code of Federal Regulations
COC	chain of custody
DDD	dichlorodiphenyldichloroethane
DDE	dichlorodiphenyldichloroethylene
DDT	dichlorodiphenyltrichloroethane
DMA	dimethylarsinic acid
EMAP	Environmental Monitoring and Assessment Program
EPA	Environmental Protection Agency (U.S.)
FDA	Food and Drug Administration
GC/FPD	gas chromatography/flame photometric detector
GC/HSD	gas chromatography/halide specific detector
GC/MS	gas chromatography/mass spectrometry
GIS	geographic information system
HpCB	heptachlorobiphenyl
HpCDD	heptachlorodibenzodioxin
HpCDF	heptachlorodibenzofuran
HxCB	hexachlorobiphenyl
HxCDD	hexachlorodibenzodioxin
HxCDF	hexachlorodibenzofuran
IRIS	Integrated Risk Information System
MDL	method detection limit
ML	minimum level
MMA	monomethylarsonic acid
NOAA	National Oceanic and Atmospheric Administration
OCDD	octachlorodibenzodioxin
OCDF	octachlorodibenzofuran

OPPTS	Office of Pesticides, Prevention, and Toxic Substances
ORD	Office of Research and Development
OST	Office of Science and Technology
OW	Office of Water
PAH	polycyclic aromatic hydrocarbon
PBT	persistent, bioaccumulative, and toxic
PCB	polychlorinated biphenyl
PeCB	pentachlorobiphenyl
PeCDD	pentachlorodibenzodioxin
PeCDF	pentachlorodibenzofuran
QA/QC	quality assurance/quality control
QAPP	quality assurance project plan
RF3	River Reach File Version 3
SCC	sample control center
SOP	standard operating procedure
SV	screening value
TeCB	tetrachlorobiphenyl
TEF	toxicity equivalency factor
TEQ	toxic equivalency concentration
TCDD	tetrachlorodibenzodioxin
TCDF	tetrachlorodibenzofuran
USEPA	United States Environmental Protection Agency
WQC	water quality criterion

Executive Summary

Environmental monitoring provides crucial data for describing the condition of the environment and for assessing the effectiveness of pollution control activities. In the 1990s, EPA identified a lack of information necessary to accurately characterize the condition of the Nation's surface waters and responded by designing a series of statistically-based surveys to produce information on the condition of lakes, streams, rivers, and coastal waters in the United States. The National Study of Chemical Residues in Lake Fish Tissue (or National Lake Fish Tissue Study) is one of the statistically-based surveys conducted by EPA since the late 1990s. This study is a national screening-level survey of chemical residues in fish tissue from lakes and reservoirs in the conterminous United States (lower 48 states), excluding the Laurentian Great Lakes and Great Salt Lake. It is unique among earlier fish monitoring efforts in the United States because the sampling sites were selected according to a statistical (random) design. Study results allow EPA to estimate the percentage of lakes and reservoirs in the United States with chemical concentrations in fish tissue that are above levels of potential concern for humans or for wildlife that eat fish. This survey also includes the largest set of chemicals ever studied in fish. Whole fish and fillets were analyzed for 268 persistent, bioaccumulative, and toxic (PBT) chemicals, including mercury, arsenic, dioxins and furans, the full complement of polychlorinated biphenyl (PCB) congeners, and a large number of pesticides and semivolatile organic compounds.

Partnerships made this study possible. Prior to beginning field sampling for the study, EPA built a national network of partners that included 47 states, three tribes, and two other federal agencies. Fisheries staff from more than 50 agencies worked for nearly five years to evaluate the suitability of lakes for sampling and to collect fish for the study. This study provides an excellent example of state, federal, and tribal collaboration. Participating agencies made a critical contribution to the success of this study through their voluntary commitments and dedicated efforts.

Fire Lake, Michigan - (Target Lake Number 0309)

Study Objective

The objective of the National Lake Fish Tissue Study is to estimate the national distribution of selected persistent, bioaccumulative, and toxic (PBT) chemical residues in fish tissue from lakes and reservoirs of the conterminous United States. Results from this study provided EPA with the first opportunity to:

- Develop national estimates of the median concentrations of PBT chemicals in lake fish;
- Estimate the percentage of lakes and reservoirs with fish tissue concentrations above specified thresholds related to human health; and
- Define national baseline information for tracking changes in concentrations of PBT chemicals in freshwater fish as a result of the combined effects of pollution control activities and natural degradation.

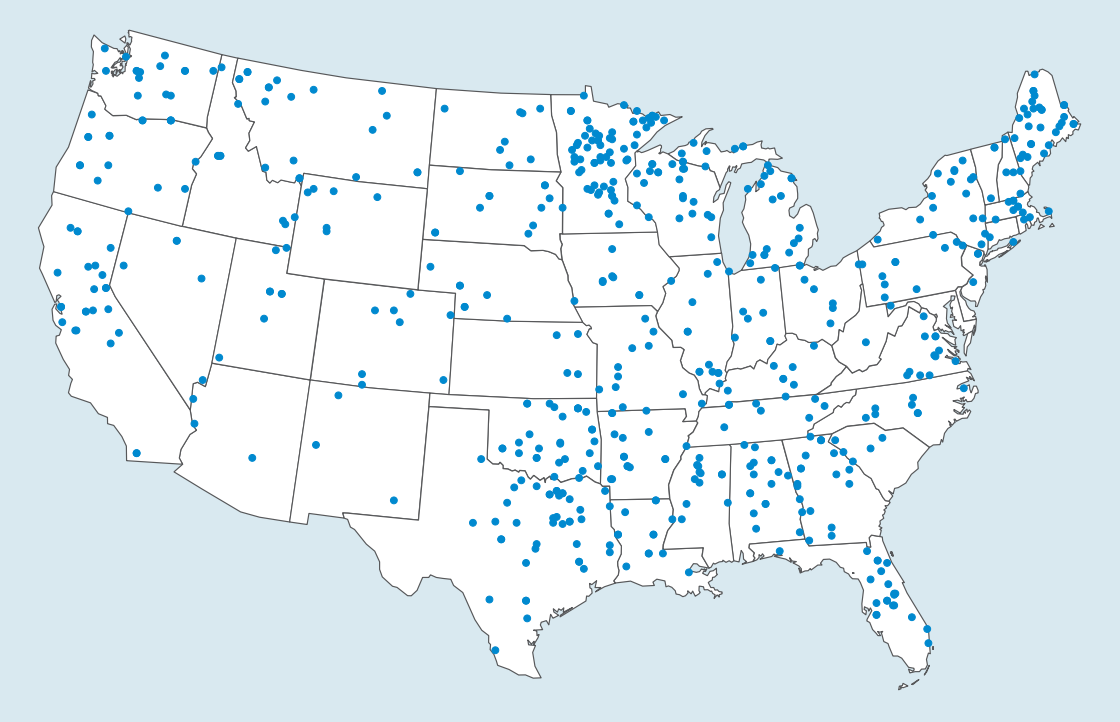
Study Design and Approach

The National Lake Fish Tissue Study focused on lakes and reservoirs (hereafter referred to collectively as lakes) for two reasons: they occur in a variety of landscapes where they can receive and accumulate contaminants from several sources (including direct discharges into water, air deposition, and agricultural or urban runoff) and there is usually limited dilution of contaminants compared to flowing streams and rivers. Monitoring fish for chemical contamination in lakes is also important because these areas are frequently used for sport fishing. According to EPA's National Listing of Fish Advisories, 43% of the Nation's lake acres are under fish consumption advisories.

This study applied a statistical or probability-based sampling approach so that results could be used to describe fish tissue contaminant concentrations in lakes on a national basis. The Nation's lakes were divided into six size categories based on surface area. Assigning different probabilities to each category prevented small lakes from dominating the group of lakes selected for sampling. It also allowed a similar number of lakes to be selected in each size category.

For this study, a lake is defined as a permanent body of water with a permanent fish population that has a surface area of at least one hectare (2.47 acres), a depth of at least one meter (3.28 feet), and at least 1,000 square meters of open, unvegetated water. The lower 48 states contain an estimated 147,000 lakes meeting these criteria (i.e., the target population). A list of candidate lakes was randomly selected from the target population for this study. From this list, EPA identified 500 sites that were accessible and appropriate for fish collection.

The target population consists of all lakes in the lower 48 states that met the study definition of a lake (147,000 lakes). The sampled population consists of all target lakes that were accessible for fish collection. Under ideal circumstances, the target and sampled populations



National Lake Fish Tissue Study Sampling Sites (500 lakes)

should coincide. However, for this study, the sampled population is a subset of the target population. A large number of target lakes were not accessible to field sampling teams because the lakes were either located in remote wilderness areas or on private property where landowners denied EPA permission to sample them. There is a different sampled population for each composite type based on differences in the occurrence of predators and bottom dwellers at the 500 sampling locations. The sampled population for predators is an estimated 76,559 lakes, and the sampled population for bottom dwellers is an estimated 46,190 lakes. All predator and bottom-dweller results presented in this report apply to these sampled populations of lakes.

Target Chemical Selection

The National Lake Fish Tissue Study includes the largest number of chemicals ever studied in fish. EPA developed the list of target chemicals for this study from the agency's multimedia list of 451 PBT chemicals, along with a list of 130 chemicals referenced in several contemporary fish tissue and bioaccumulation studies. EPA selected chemicals that had detailed information available, were known to accumulate, and were identified as important in one or more EPA programs. The final list contains 268 chemicals, including mercury, five forms of arsenic, 17 dioxins and furans, 159 PCB congener measurements, 46 pesticides, and 40 semivolatile organic compounds.

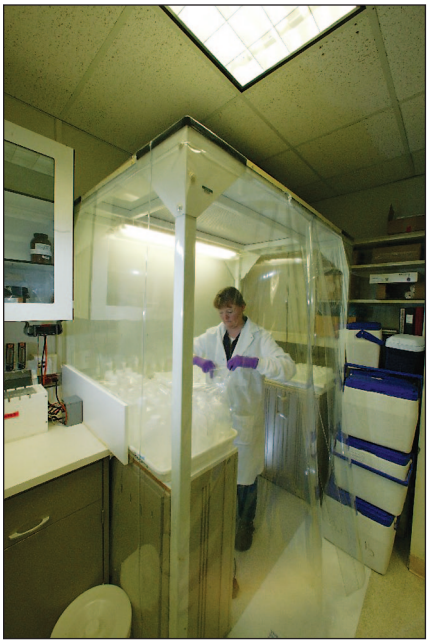
Sample Collection

EPA planned four years of field sampling for the National Lake Fish Tissue Study. After a brief pilot in the fall of 1999 to test sampling logistics, EPA and its partners began full-scale fish sampling in 2000 and continued sampling annually through 2003. Each year of the study, field sampling teams collected fish from about 125 different lakes distributed across the lower 48 states. These teams applied consistent methods nationwide to collect composite samples of a predator fish species (e.g., bass or trout) and a bottom-dwelling species (e.g., carp or catfish) from each lake or reservoir. EPA identified twelve target predator species and six target bottom-dwelling species to limit the number of species included in the study. Predator and bottom-dweller composites each consisted of five adult fish of the same species and similar size (i.e., the smallest individual in a composite was no less than 75% of the total length of the largest individual). Field teams re-sampled more than 10% of the lakes to allow EPA to evaluate any possible sampling variability.

Sampling at Norvell Lake, Michigan – (Target Lake Number 0664)

Sample Analysis

EPA analyzed different tissue fractions for predator composites (fillets) and bottom-dweller composites (whole bodies) to obtain chemical residue data for the 268 target chemicals. Analyzing fish fillets provides information for human health, while whole-body analysis produces information for ecosystem health. A single laboratory prepared all fish samples in a strictly-controlled, contamination-free environment. This laboratory distributed fish tissue samples to four laboratories that specialize in analysis of metals, pesticides, semivolatile organic chemicals, and PCBs, dioxins, and furans. To minimize variability among sample results, EPA used the same laboratory for each type of analysis, and these laboratories applied the same analytical method for each chemical for the duration of the study. Resulting fish tissue concentrations were reported on a wet weight basis.



Analytical Chemistry Laboratory Clean Room

Statistical Analysis

The data analyzed for the National Lake Fish Tissue Study include tissue concentrations for each target chemical (e.g., mercury) or chemical group (e.g., PCBs) and fish composite type (i.e., predator and bottom-dweller composites). Analysis of the fish tissue data incorporated elements of the survey design, along with information from the field sampling operations and laboratory analyses. Statistical analysis included evaluating information to determine the status of each lake, adjusting the sample weights based on the lake status, estimating the number and proportion of lakes in the sampled population, and estimating the cumulative distribution and percentile concentrations of the target chemicals in fillets for predators and whole bodies for bottom dwellers.

Results

The National Lake Fish Tissue Study is the first national assessment of freshwater fish contamination in the United States for which sampling sites were selected according to a statistical (random) design. To interpret the results, it is essential to understand the following important points about this design:

Lake 79, Wyoming – (Target Lake Number 0052)

- During the four-year sampling period, field teams collected 486 predator composites and 395 bottom-dweller composites from the 500 sampling locations. Predator and bottom-dweller species did not occur together at every sampling site; however, if either a predator or bottom-dweller species was present, the target lake was sampled.
- The 486 predator composites and the 395 bottom-dweller composites collected during the study each comprise nationally-representative samples for the lower 48 states whose results can be extrapolated to an estimated 76,559 lakes for predators and an estimated 46,190 lakes for bottom-dwellers.
- The unequal probability design makes it necessary to apply sample weights (derived from the various probabilities assigned to each of the lake size categories) to the fish tissue data to develop national estimates of fillet (predator) and whole-body (bottom-dweller) fish tissue concentrations for each of the 268 target chemicals.

Results from the National Lake Fish Tissue Study indicate that mercury, PCBs, and dioxins and furans are widely distributed in lakes and reservoirs in the lower 48 states. Mercury and PCBs were detected in all the fish samples collected from the 500 sampling sites. Dioxins and furans were detected in 81% of the predator samples (fillet composites) and 99% of the bottom-dweller samples (whole-fish composites). In contrast, there were a number of chemicals that were not detected in any of the fish samples collected during the study. Forty-three of the 268 target chemicals were not detected in any samples, including all nine organophosphate pesticides (e.g., chlorpyriphos and diazinon), one PCB congener (PCB-161), and 16 of the 17 polycyclic aromatic hydrocarbons (PAHs) analyzed as semivolatile organic chemicals. There were also seventeen other semivolatile organic chemicals that were not detected.

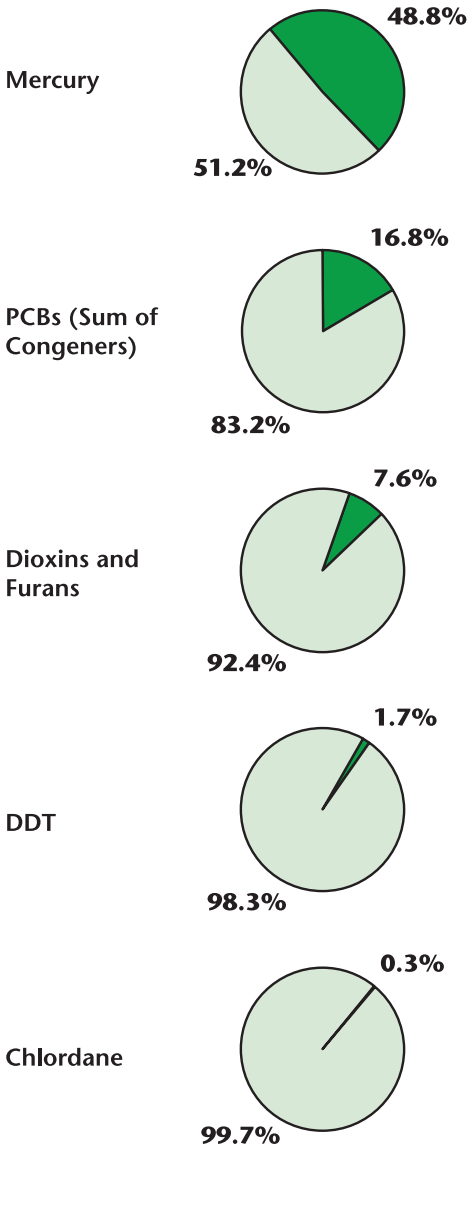
In reporting the analytical results for this study, it is important to distinguish between detection and presence of a chemical in a fish tissue sample. Estimates of fish tissue concentrations ranging from the method detection limit (MDL) to the minimum level of quantitation (ML) are reported as being present with a 99% level of confidence. However, if a chemical is 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 the fish tissue samples are reported as less than the MDL rather than zero. In interpreting the results, it is also important to know the MDL for each chemical (Appendix B).

According to EPA's 2008 Biennial National Listing of Fish Advisories, mercury, PCBs, dioxins and furans, DDT, and chlordane accounted for 97% of the advisories in effect at the end of 2008. These five chemicals were also commonly detected in fish samples collected for the National Lake Fish Tissue Study. Since human health screening values (SVs) were readily available, they were applied to total concentrations of mercury, PCBs,

Snowbank Lake, Minnesota – (Target Lake Number 0235)

Percentage of Lakes Above or Below Human Health Screening Values

(Predator Results: Sampled Population = 76,559 Lakes)



dioxins and furans, DDT, and chlordane found in predator fillets. The mercury SV is the tissue-based water quality criterion published by EPA in 2001. All other SVs are risk-based consumption limits published in 2000 in EPA's *Guidance for Assessing Chemical Contaminant Data for Use in Fish Consumption Limits, Third Edition*. Specifically, the applied SVs are the upper limit of the four-meal-per-month concentration range for the conservative consumption limit (where tissue concentrations are available for both cancer and noncancer health endpoints). If available, wildlife criteria could be applied in the same manner to interpret the whole-body data from analysis of bottom-dweller samples.

Predator results for the five commonly-detected chemicals indicate that:

- 48.8% of the sampled population of lakes had mercury tissue concentrations that exceeded the 300 ppb (0.3 ppm) human health SV for mercury, which represents a total of 36,422 lakes.
- 16.8% of the sampled population of lakes had total PCB tissue concentrations that exceeded the 12 ppb human health SV, which represents a total of 12,886 lakes.
- 7.6% of the sampled population of lakes had dioxin and furan tissue concentrations that exceeded the 0.15 ppt [toxic equivalency or TEQ] human health SV, which represents a total of 5,856 lakes.
- 1.7% of the sampled population of lakes had DDT tissue concentrations that exceeded the 69 ppb human health SV, which represents a total of 1,329 lakes.
- 0.3% of the sampled population of lakes had fish tissue concentrations that exceeded the 67 ppb human health SV for chlordane, which represents a total of 235 lakes.

This report provides national baseline information to track changes in PBT chemical concentrations in freshwater fish resulting from pollution control

activities and chemical degradation. The results of the National Lake Fish Tissue Study are presented to inform the public, water quality specialists, natural resource managers, and government officials of the distribution and prevalence of selected PBT chemicals in fish tissue from lakes and reservoirs of the conterminous United States. The findings should also be useful to aid in the design and focus of future fish tissue contaminant studies. National Lake Fish Tissue Study sampling design information and results have also been published in peer-reviewed journal articles by Olsen et al. (2009) and Stahl et al. (2009). Additional information, including instructions for obtaining study data and links to related study literature, is posted on the Internet at ***http://www.epa.gov/waterscience/fishstudy/***.