Occurrence and Distribution of Organochlorine Pesticides, Polychlorinated Biphenyls, and Trace Elements in Fish Tissue in the Lower Tennessee River Basin, 1980-98

Water-Resources Investigations Report 01-4184
National Water-Quality Assessment Program
Occurrence and Distribution of Organochlorine Pesticides, Polychlorinated Biphenyls, and Trace Elements in Fish Tissue in the Lower Tennessee River Basin, 1980-98

By RODNEY R. KNIGHT and JEFFREY R. POWELL

U.S. GEOLOGICAL SURVEY

Water-Resources Investigations Report 01-4184

Nashville, Tennessee
2001
The U.S. Geological Survey (USGS) is committed to serve the Nation with accurate and timely scientific information that helps enhance and protect the overall quality of life, and facilitates effective management of water, biological, energy, and mineral resources. Information on the quality of the Nation’s water resources is of critical interest to the USGS because it is so integrally linked to the long-term availability of water that is clean and safe for drinking and recreation and that is suitable for industry, irrigation, and habitat for fish and wildlife. Escalating population growth and increasing demands for the multiple water uses make water availability, now measured in terms of quantity and quality, even more critical to the long-term sustainability of our communities and ecosystems.

The USGS implemented the National Water-Quality Assessment (NAWQA) Program to support national, regional, and local information needs and decisions related to water-quality management and policy. Shaped by and coordinated with ongoing efforts of other Federal, State, and local agencies, the NAWQA Program is designed to answer: What is the condition of our Nation’s streams and ground water? How are the conditions changing over time? How do natural features and human activities affect the quality of streams and ground water, and where are those effects most pronounced? By combining information on water chemistry, physical characteristics, stream habitat, and aquatic life, the NAWQA Program aims to provide science-based insights for current and emerging water issues and priorities. NAWQA results can contribute to informed decisions that result in practical and effective water-resource management and strategies that protect and restore water quality.

Since 1991, the NAWQA Program has implemented interdisciplinary assessments in more than 50 of the Nation’s most important river basins and aquifers, referred to as Study Units. Collectively, these Study Units account for more than 60 percent of the overall water use and population served by public water supply, and are representative of the Nation’s major hydrologic landscapes, priority ecological resources, and agricultural, urban, and natural sources of contamination.

Each assessment is guided by a nationally consistent study design and methods of sampling and analysis. The assessments thereby build local knowledge about water-quality issues and trends in a particular stream or aquifer while providing an understanding of how and why water quality varies regionally and nationally. The consistent, multi-scale approach helps to determine if certain types of water-quality issues are isolated or pervasive, and allows direct comparisons of how human activities and natural processes affect water quality and ecological health in the Nation’s diverse geographic and environmental settings. Comprehensive assessments on pesticides, nutrients, volatile organic compounds, trace metals, and aquatic ecology are developed at the national scale through comparative analysis of the Study-Unit findings.

The USGS places high value on the communication and dissemination of credible, timely, and relevant science so that the most recent and available knowledge about water resources can be applied in management and policy decisions. We hope this NAWQA publication will provide you the needed insights and information to meet your needs, and thereby foster increased awareness and involvement in the protection and restoration of our Nation’s waters.

The NAWQA Program recognizes that a national assessment by a single program cannot address all water-resource issues of interest. External coordination at all levels is critical for a fully integrated understanding of watersheds and for cost-effective management, regulation, and conservation of our Nation’s water resources. The Program, therefore, depends extensively on the advice, cooperation, and information from other Federal, State, interstate, Tribal, and local agencies, non-government organizations, industry, academia, and other stakeholder groups. The assistance and suggestions of all are greatly appreciated.

Robert M. Hirsch
Associate Director for Water
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Conversion Factors, Vertical Datum, Water-Quality Units, and List of Acronyms and Abbreviations

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Sea level: In this report, “sea level” refers to the National Geodetic Vertical Datum of 1929—a geodetic datum derived from a general adjustment of the first-order level nets of the United States and Canada, formerly called Sea Level Datum of 1929.

Water-quality units

µg/g micrograms per gram
mg/kg milligrams per kilogram

Abbreviations and Acronyms

ADEM Alabama Department of Environmental Management
BHC Benzene hexachloride
DCPA Dimethyl 2,3,5,6-tetrachloroterephthalate
DDD Dichlorodiphenyldichloroethane
DDE Dichlorodiphenyldichloroethylene
DDT Dichlorodiphenyltrichloroethane
FDA U.S. Food and Drug Administration
L Tenn Lower Tennessee River Basin
NAS/NAE National Academy of Sciences and National Academy of Engineering
NAWQA National Water-Quality Assessment Program
NCBP National Contaminant Biomonitoring Program
NYDEC New York Department of Environmental Conservation
PCB Polychlorinated biphenyl
TDEC Tennessee Department of Environment and Conservation
TVA Tennessee Valley Authority
U.S. EPA U.S. Environmental Protection Agency
USGS U.S. Geological Survey
Occurrence and Distribution of Organochlorine Pesticides, Polychlorinated Biphenyls, and Trace Elements in Fish Tissue in the Lower Tennessee River Basin, 1980-98

By Rodney R. Knight and Jeffrey R. Powell

ABSTRACT

The U.S. Geological Survey, as part of the National Water-Quality Assessment Program, evaluated the occurrence and distribution of organochlorine pesticides, polychlorinated biphenyls, and trace elements in fish tissue in samples collected in the lower Tennessee River Basin study unit. Fish tissue analysis provides a time-averaged measurement of contaminants as well as a direct measurement of the contaminants that bioaccumulate in fish tissue. Bioaccumulation of contaminants in fish tissue may result in concentrations that can affect human, wildlife, or aquatic health. Data for two types of tissue analyses were evaluated to assess the occurrence and distribution of contaminants: whole fish for organochlorine pesticides and polychlorinated biphenyls and fish fillets for organochlorine pesticides, polychlorinated biphenyls, and trace elements. The fish tissue data analyzed for this study cover an 18-year span including data collected in 1998 by the U.S. Geological Survey as part of the National Water-Quality Assessment Program; data collected from 1980 through 1997 by the Tennessee Valley Authority; and data collected from 1992 through 1997 by the Tennessee Department of Environment and Conservation. Concentration data for constituents that are on the U.S. Environmental Protection Agency Priority Pollutant List were summarized and compared against existing action levels or guidelines.

From the list of organochlorine pesticide compounds analyzed, \( p,p' \)-dichlorodiphenyltrichloroethylene (\( p,p' \)-DDE), a breakdown product of dichlorodiphenyltrichloroethane (DDT), was the most commonly detected compound with detections at 83 percent of the sites sampled. Eleven \( p,p' \)-DDE samples exceeded action levels or guidelines with concentrations ranging from 0.20 to 12.8 milligrams per kilogram. Five other organochlorine compounds, \( p,p' \)-dichlorodiphenyldichloroethane (\( p,p' \)-DDD), dieldrin, endrin, chlordane, and polychlorinated biphenyls, also exceeded action levels and guidelines, but the detection frequencies at sampling sites generally were less than 70 percent.

Mercury, the only trace element to exceed a guideline, was detected at 51 of 102 sites sampled for trace elements. Selenium was detected in fish fillet samples from 70 of 102 sites sampled, which was more sites than for any other trace element; however, selenium did not exceed the 50 micrograms per gram U.S. Environmental Protection Agency screening criteria. Arsenic and cadmium also were detected at 44 and 54 percent of the sampling sites, respectively.

INTRODUCTION

Water-quality assessment activities in the lower Tennessee (LTEN) River Basin study unit started in 1997. The LTEN River Basin (fig. 1) is one of 59 river basins selected for water-quality assessment as part of the U.S. Geological Survey (USGS) National Water-Quality Assessment (NAWQA) Program, which began in 1991. Initial assessment efforts in the LTEN River Basin included describing the occurrence and distribution of organochlorine pesticides, polychlorinated biphenyls (PCB’s), and trace elements in fish tissue.
Figure 1. Location of the lower Tennessee River Basin study unit.
According to Crawford and Luoma (1993), four advantages exist for using fish tissues in water-quality assessments when compared to using standard water chemistry sampling. First, tissue analysis can increase the probability of detecting trace amounts of some contaminants that tend to bioaccumulate in tissue. Second, tissues provide a time-averaged assessment or record of the presence of contaminants in the environment. Third, tissue analyses provide direct measurements of bioavailability of contaminants to other living organisms. Finally, by integrating tissue, water, and sediment analyses, complementary or multiple lines of evidence are provided to assist in the understanding of contaminant fate, distribution, and effects.

Industrial and municipal effluents, nonpoint-source runoff, and atmospheric deposition are all potential sources of organochlorine pesticides, PCB’s, and trace elements that may accumulate in fish tissue. Accumulation of organochlorine pesticides and PCB’s in fish tissue is a direct result of human activity because these compounds are synthetic. While dichlorodiphenyltrichloroethane (DDT) use was banned by the U.S. Environmental Protection Agency (U.S. EPA) in 1973, production and use of most organochlorine pesticides were discontinued in the United States by the mid-1980’s; nevertheless, many of the organochlorine pesticides and their metabolites (breakdown products) can still be found in water, sediment, and fish tissue. Trace elements occur naturally in the environment and reflect soil type, geology, and land use in the watershed.

**Purpose and Scope**

The purpose of this report is to summarize the occurrence and distribution of organochlorine pesticides, PCB’s, and trace elements in fish tissue in the LTEN River Basin. Existing fish tissue data were compared with action levels and guidelines from the Niagara River Biota Contamination Project (Newell and others, 1987), National Academy of Sciences and National Academy of Engineering (1973), U.S. Food and Drug Administration (1989), and U.S. EPA (1995) to place the data in a human and aquatic health context. This report is limited to the occurrence and distribution of organic compounds and trace elements that are listed on the U.S. EPA Priority Pollutant List (U.S. Environmental Protection Agency, 1994) and that have an action level or guideline available. Analyses presented in this report were based on fish tissue data collected by the USGS during August and September 1998, by the Tennessee Valley Authority (TVA) from 1980 through 1997, and by the Tennessee Department of Environment and Conservation (TDEC) from 1992 through 1997. This report summarizes existing data for organochlorine pesticides, PCB’s, and trace elements in fish tissue collected from sampling sites primarily located along the main stem of the Tennessee River within the LTEN River Basin, and does not supersede fish consumption advisories issued by State agencies.

**Consumption Advisories**

The LTEN River Basin contains approximately 26,700 stream miles and 382,500 lake acres. In 1998, less than 4 percent of the lake acres and less than 1 percent of the stream miles in the LTEN River Basin had consumption advisories posted for the consumption of fish by humans. Currently (2000), advisories issued by TDEC exist for Woods and Nickajack Lakes in Tennessee because of elevated concentrations of PCB’s (Freeman and Denton, 1997). The Alabama Department of Environmental Management (ADEM) has current consumption advisories for the Tennessee River, Indian Creek, and Huntsville Spring Branch in Alabama because of elevated DDT concentrations (Alabama Department of Environmental Management, 1996). Current fish advisories exist for streams, rivers, and reservoirs in Tennessee (Tennessee Department of Environment and Conservation, 2000) and Alabama (Alabama Department of Public Health, 2000).

**Description of the Lower Tennessee River Basin Study Unit**

The LTEN River Basin study unit extends from Chattanooga, Tennessee, to near Paducah, Kentucky, at the confluence of the Tennessee and Ohio Rivers and encompasses approximately 19,500 square miles. Most of the study unit is in Middle Tennessee and northern Alabama, with smaller parts in southwestern Kentucky, northeastern Mississippi, and northwestern Georgia. Approximately 57 percent of the study unit is in Tennessee, 35 percent in Alabama, 5 percent in Kentucky, 2 percent in Mississippi, and 1 percent in Georgia. Population in the study unit is about 1.5 million (U.S. Department of Commerce, 1997). The most populated cities in the study unit are Huntsville, Alabama (population 160,000), Chattanooga, Tennessee...
(population 152,000), and Decatur, Alabama (population 52,000) (Woodside and Mitchell, 1998). Land-surface elevations in the study unit range from about 300 feet above sea level near Paducah, Kentucky, to more than 2,900 feet above sea level along the eastern edge of the Sequatchie River Basin. Annual precipitation varies from 47 to 63 inches, with higher precipitation amounts recorded in the northeastern area of the study unit. Average runoff ranges from 18 inches in the northwest to 30 inches in the southeast. The study unit has a temperate climate with an average annual temperature of about 58 °F.

The main stem of the lower Tennessee River is highly regulated with few free-flowing stream reaches. Six major reservoirs were constructed along the main stem from the 1920’s through the 1940’s for purposes of power generation, navigation, and flood control. Today, these reservoirs also are used extensively for sources of drinking water and recreational activities such as fishing, swimming, and boating.

Land use varies in the study unit. Forest covers about 51 percent of the study unit (Tennessee Valley Authority, written commun., 1992). Additional land uses include row crops and pasture land (40 percent), urban (1 percent), and other land uses (8 percent), such as wetlands, water, and barren land. Row-crop cultivation is present predominantly along the main stem and tributaries of the Tennessee River in northern Alabama and along the western edge of the study area. Cotton, corn, and soybeans are the primary row crops. Areas of confined-animal feeding operations are concentrated primarily in northern Alabama.

Kingsbury and others (1999) describe three hydrogeologic regions in the LTEN River Basin study unit. These hydrogeologic regions lie within three physiographic provinces (Kingsbury and others, 1999, fig. 1). Along the western edge of the study unit, the Coastal Plain Physiographic Province encompasses about 18 percent of the LTEN River Basin area and generally consists of a thick series of unconsolidated sands, gravels, silts, and clays. Shallow ground water occurs primarily in these gravels and sands and is an important source of drinking water. The Interior Low Plateaus Physiographic Province is centrally located and encompasses about 59 percent of the LTEN River Basin. In this province, ground water moves through an overburden consisting of 10 to more than 200 feet of regolith, a mixture of soil and weathered rock, and subsequently into an underlying carbonate aquifer. Carbonate aquifers are important sources of drinking water in many areas throughout the LTEN River Basin study unit. Thin regolith, caves, and sinkholes in this province increase the susceptibility of ground-water contamination from surface water. The Cumberland Plateau section of the Appalachian Plateaus Physiographic Province is located along the eastern edge of the study unit and encompasses about 23 percent of the LTEN River Basin. Most of this area is underlain by sandstones with ground water occurring primarily in interconnected fractures.

Acknowledgments

The authors thank Wade Bryant of the USGS for field assistance as well as technical guidance; Lisa Nowell and Rod DeWeese of the USGS for their help in locating action levels and guidelines for comparisons; Don Dycus, Tyler Baker, and Kenny Gardner of the Tennessee Valley Authority for providing fish tissue data and information about sample methodology; and Greg Denton of the Tennessee Department of Environment and Conservation for providing assistance concerning fish tissue information.

METHODS

Various field collection and sampling protocols were used by the USGS, TVA, and TDEC. In addition, different action levels and guidelines were used as they apply to the analysis of whole fish and fish fillets. The analysis of organochlorine pesticides in fish tissue samples collected by the USGS involved the homogenization of whole fish and the use of gas chromatography and mass spectrometry. Analytical methods used for the determination of concentrations of organochlorine pesticides can be found in Crawford and Luoma (1993) and Leiker and others (1995). Methods used by TVA and TDEC can be found in Williams and Dycus (1993) and Freeman and Denton (1997), respectively.

Field Collection

The USGS NAWQA Program recommends several fish species for fish tissue contaminant analysis. Species selection for this study was based on a targeted taxa list of recommended fish species, which was developed by Crawford and Luoma (1993). Because of geographic distribution of some fish species on the targeted taxa list, some species were
Methods

Whole fish were analyzed to determine what pesticides were present and the areal distribution of their occurrence, and to assess the concentration levels of certain pesticides that wildlife may ingest as a result of eating fish. Approximately 10 fish of the same species were collected at each site to gain a representative sample. Once collected, both external and internal observations were recorded (for example, sex, coloration of organs, mesentaric fat, tumors, lesions, and parasites). In addition, scales or pectoral spines and length and weight data were collected to determine the

A variety of stream habitats were sampled (pools, undercut banks, logjams) to obtain a sufficient number of fish from the targeted species.

unavailable or impractical to collect. Top predator species, such as largemouth bass (*Micropterus salmoides*), smallmouth bass (*Micropterus dolomieu*), redeye bass (*Micropterus coosae*), spotted bass (*Micropterus punctatus*), and rock bass (*Ambloplites rupestris*), were the indicator species collected for analysis. Fish were collected using electrofishing techniques in accordance with NAWQA sampling protocols as described by Crawford and Luoma (1993).

Prior to dissection, the exterior, entrails, and gills of fish were visually examined for discoloration, tumors, parasites, or other anomalies that might be indicative of degraded water-quality conditions.
age and size of individual specimens. Whole fish were shipped frozen to the USGS National Water-Quality Laboratory in Lakewood, Colorado, for analysis.

The primary indicator species collected by the TVA was channel catfish (*Ictalurus punctatus*). The channel catfish is highly sought by commercial and sport fishermen and was selected by the TVA because individual channel catfish tend to concentrate contaminants more so than other fish species due to the higher lipid content in catfish. Other important species collected by TVA included largemouth bass, striped bass (*Morone saxitilis*), buffalo (*Ictiobus spp.*), and crappie (*Pomoxis spp.*). Fish fillets, including the “belly flap” (a layer of fat associated with the fillet), were sampled in an effort to define which species are affected by contaminants, the geographical boundaries of contamination, and to document trends in contaminant concentrations. The information collected by TVA is shared with public health agencies such as TDEC and ADEM for their determination of any possible restrictions or consumption advisories that may need to be issued on fish consumption. Five specimens were collected by TVA using electrofishing equipment and commercial fishing gear such as seines and other nets. Black bass (*Micropterus spp.*) collected for analysis had to be at least 10 inches in length; channel catfish collected for analysis had to weigh at least 1 pound, and striped bass had to weigh at least 2 pounds. Upon collection, the fish were examined both internally and externally for physical abnormalities. Any fish having tissue unsuitable for human consumption was not included in a sample. The fish were placed on wet ice and shipped to the TVA laboratory for analysis (Williams and Dycus, 1993).

Largemouth bass (*Micropterus salmoides*), channel catfish (*Ictalurus punctatus*), and common carp (*Cyprinus carpio*) were the preferred indicator species collected for analysis by TDEC. At least five fish from each of these species were preferred, but when these fish were not available, smallmouth bass (*Micropterus dolomieu*), sunfish (*Ambloplites rupestris* and *Lepomis spp.*), suckers (*Moxostoma spp.* and *Hypentelium nigricans*), buffalo (*Ictiobus spp.*), flathead catfish (*Pylodictis olivaris*), and bullhead catfish (*Ameiurus spp.*) were substituted. Samples were collected with either electrofishing equipment or nets. Fish fillets with the belly flap (edible portion) were analyzed by TDEC to assist in the development of fish consumption advisories.

### Action Levels and Guidelines

Fish tissue data from over 200 sites in the LTEN River Basin were compared with action levels and guidelines in order to place the fish tissue data in a human and aquatic health context. Different action levels or guidelines are available for analyzing tissue from fillets and whole fish. The most recent fish tissue sample concentration data from sites that were sampled multiple times were used to depict the areal distribution of organochlorine compounds and trace elements in the study unit. By allowing for the most recent concentration data to be used in action level and guideline comparisons, maps illustrating sampling sites can be presented as the most recent information available for each site for each organochlorine compound and trace element. Whole fish organochlorine pesticide and PCB concentrations were compared to guidelines developed by the National Academy of Sciences and National Academy of Engineering (NAS/NAE) in 1973. The NAS/NAE guidelines were developed to provide a measure of nonlethal levels of contaminants. Additionally, organochlorine pesticide and PCB concentrations were compared to criteria developed by the New York Department of Environmental Conservation (NYDEC). The criteria presented by NYDEC were designed to protect piscivorous (fish-eating) wildlife and are based on studies of 18 different species of mammals, reptiles, and birds living along the Niagara River (Newell and others, 1987). Fish fillet data were compared to action levels developed by the U.S. Food and Drug Administration (FDA) (1989). FDA action levels for pesticides in fish fillets are available only for those pesticides that are either no longer registered for use or are severely limited in their application, but are persistent in the environment and may be unavoidable in fish tissue. Some of the action levels and guidelines used for comparison in this report were developed for use with the sum of concentrations for members within a particular group. For example, the action levels for aldrin concentrations in fish fillets are meant to be used for the sum of the concentrations of aldrin and dieldrin. Guidelines for aldrin concentrations in whole fish are meant to be used for the sum of concentrations for aldrin, chlordane, dieldrin, endosulfan, endrin, heptachlor, heptachlor epoxide, lindane, toxaphene, and benzene hexachloride (BHC). This same use of the guidelines also is true for the diphenyl aliphatic pesticides (Nowell and Resek, 1994). In this report, the action levels and guidelines are compared to pesticides singly, not
to the additive sum of the other compounds of the group. Application of the action levels and guidelines to single pesticides was chosen because of the abundance of records available for the LTEN River Basin. The application of the action level to the sum of concentrations for a group of pesticides was completed for a limited number of sites and resulted in no change in the detection frequency.

Trace element data from fish fillets were compared against FDA action levels for mercury (U.S. Food and Drug Administration, 1989). These same data were compared to U.S. EPA screening values for the protection of human health for mercury, cadmium, selenium, and arsenic (U.S. Environmental Protection Agency, 1995).

**OCCURRENCE AND DISTRIBUTION OF ORGANOCHLORINE PESTICIDES IN FISH TISSUE**

Organochlorine pesticides listed on the U.S. EPA Priority Pollutant List that have an action level or guideline available for either whole fish or fish fillets are discussed in this section. The organochlorine pesticides discussed are divided into five subgroups: cyclodienes, chlorinated benzene derivatives, polychloroterpenes, diphenyl aliphatics, and polychlorinated biphenyls. Each of the five subgroups is presented with background information about past uses and properties as well as the occurrence of the compound in the study unit. Throughout the study unit, 101 sites were sampled for organochlorine pesticides.

The five organochlorine pesticide subgroups are further divided into three subgroups: (1) cyclodienes, chlorinated benzene derivatives, polychloroterpenes, diphenyl aliphatics, and polychlorinated biphenyls; (2) diphenyl aliphatics (DDT and breakdown products); and (3) polychlorinated biphenyls (PCB’s) for discussion in this report. The 13 organochlorine pesticides that have action levels or guidelines and that are listed on the U.S. EPA Priority Pollutant List are aldrin, chlordane, dieldrin, endrin, mirex, heptachlor, heptachlor epoxide, hexachlorobenzene, and toxaphene; (2) diphenyl aliphatics (DDT and breakdown products); and (3) polychlorinated biphenyls (PCB’s) for discussion in this report. The 13 organochlorine pesticides that have action levels or guidelines and that are listed on the U.S. EPA Priority Pollutant List are aldrin, chlordane, dieldrin, endrin, mirex, heptachlor, heptachlor epoxide, hexachlorobenzene, and toxaphene. No method exists to measure the amount of dieldrin that originated as aldrin for the initial application. Dieldrin, heptachlor epoxide, and endrin (figs. 5, 6, and 7, respectively) each were detected in whole fish and fish fillet samples. Approximately 300 samples were collected and analyzed for the 6 cyclodiene pesticides discussed here. Only 2 of the 300 samples had concentrations that equaled or exceeded the action levels or guidelines used herein for comparison.
Table 1. Summary of organochlorine compounds in fish fillet and whole fish samples collected in the lower Tennessee River Basin, 1980-98

[Compounds in bold were detected at or above established action levels or guidelines; F, fish fillets; W, whole fish; mg/kg, milligram per kilogram; <, less than; ---, no information; DCPA, dimethyl 2,3,5,6-tetrachloroterephthalate; BHC, benzene hexachloride; PCB, polychlorinated biphenyl; DDT, dichlorodiphenyltrichloroethane; DDD, dichlorodiphenyldichloroethane; DDE, dichlorodiphenyldichloroethylene]

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<td>.01 - .26</td>
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<td>W</td>
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<td></td>
<td>W</td>
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<td>10</td>
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<td>10</td>
<td>0</td>
<td>&lt;.005</td>
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bNiagara River Biota Contamination Project, New York State Department of Environmental Conservation (Newell and others, 1987).
cCompilation of legal limits for hazardous substances in fish and fishery products (Nauen, 1983).
dFDA tolerance level (U.S. Food and Drug Administration, 1989).
<table>
<thead>
<tr>
<th>Compound</th>
<th>Tissue type</th>
<th>Number of samples</th>
<th>Number of detections</th>
<th>Number of sites sampled for contaminant</th>
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<th>Range of data (mg/kg)</th>
<th>Action level (mg/kg)</th>
<th>Guideline (mg/kg)</th>
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<td>d,5.0</td>
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<td>Mirex</td>
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<td>46</td>
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<td>0</td>
<td>0</td>
<td>&lt;0.005</td>
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<td>Delta-BHC</td>
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### Diphenyl aliphatics

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<th>Range of data (mg/kg)</th>
<th>Action level (mg/kg)</th>
<th>Guideline (mg/kg)</th>
<th>Number of sites with detections above lowest action level</th>
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<td>.01 - 6.7</td>
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<td>5</td>
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<td>1</td>
<td>10</td>
<td>1</td>
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<td>&lt;0.005 - .0058</td>
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### Polychlorinated biphenyls

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<td>&lt;0.05 - .30</td>
<td>---</td>
<td>a,5b,13</td>
<td>2</td>
</tr>
</tbody>
</table>

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*b Niagara River Biota Contamination Project, New York State Department of Environmental Conservation (Newell and others, 1987).

*c Compilation of legal limits for hazardous substances in fish and fishery products (Nauen, 1983).

*d FDA tolerance level (U.S. Food and Drug Administration, 1989).
Figure 2. Location of sampling sites with detections of aldrin in fish tissue in the lower Tennessee River Basin, 1980-98.
Figure 3. Location of sampling sites with detections of heptachlor in fish tissue in the lower Tennessee River Basin, 1980-98.
Figure 4. Location of sampling sites with detections of mirex in fish tissue in the lower Tennessee River Basin, 1980-98.
Figure 5. Location of sampling sites with detections of dieldrin in fish tissue in the lower Tennessee River Basin, 1980-98.
EXPLANATION
Action level or guideline reported in milligrams per kilogram (mg/kg)

△ FISH FILLET SAMPLE BELOW 0.3 mg/kg ACTION LEVEL
〇 WHOLE FISH SAMPLE BELOW 0.1 mg/kg GUIDELINE
● LOCATION OF SAMPLING SITE

Figure 6. Location of sampling sites with detections of heptachlor epoxide in fish tissue in the lower Tennessee River Basin, 1980-98.
Figure 7. Location of sampling sites with detections of endrin in fish tissue in the lower Tennessee River Basin, 1980-98.
Dieldrin equaled the 0.10 milligram per kilogram (mg/kg) guideline set by the NAS/NAE (1973) for whole fish. Endrin was detected at a concentration of 0.027 mg/kg, which exceeded the 0.025 mg/kg criteria established by the Niagara River Biota Contamination Project (Newell and others, 1987) for whole fish. According to Schmidt and others (1990), geometric mean concentrations for dieldrin, heptachlor epoxide, and endrin have remained steady across the Nation. This information was the result of the National Contaminant Biomonitoring Program (NCBP) that collected fish tissue samples at 117 locations across the Nation from 1976 until 1984.

Total chlordane (chlordane) is represented by the sum of the concentrations of cis- and trans-chlordane, cis- and trans-nonachlor, and oxychlor dane, the five major components of technical chlordane (L.H. Nowell, U.S. Geological Survey, written commun., 2000). In August 1998, chlordane was detected at only two whole fish sampling sites (fig. 8) with one site having a concentration of 0.149 mg/kg, above the 0.1 mg/kg NAS/NAE guideline for chlordane for whole fish. Chlordane was detected at 26 fish fillet sampling sites. These sites were sampled between 1985 and 1994, and one sample exceeded the 0.3 mg/kg FDA action level for fish fillets. This sample was collected in November 1990 with a concentration of 0.36 mg/kg; the sampling site was Wheeler Lake at Tennessee River mile 300. Results from the NCBP revealed that chlordane concentrations declined throughout the late 1970’s, but then remained steady for a period of time nationwide (Schmidt and others, 1990). Chlordane, like other organochlorine pesticides, is transferred up the food chain to fish. Chlordane can remain active in soils for 20 years and, like other cyclodiene pesticides, was used as a deterrent to termites.

Hexachlorobenzene is a chlorinated benzene derivative that possesses fungicidal properties. This compound was first used in 1945 as a treatment to control fungal activity on seeds (Ware, 1989). No detections of hexachlorobenzene were found in whole fish tissue samples in the LTEN River Basin.

DDT is a member of the diphenyl-aliphatic chemical class, which was widely used as an insecticide in the United States until 1973 when it was banned except for use in public health emergencies. Initial uses of DDT in the Tennessee River Valley area were for insect control around reservoirs and other wet areas. Technical DDT is composed of three isomers, para-para (p,p’), ortho-para (o,p’), and ortho-ortho (o,o’). The p,p’ isomer makes up approximately 80% of technical DDT. DDT proved to be extremely effective against flies and mosquitoes, with malaria control being DDT’s most beneficial public-health use. More than 4 billion pounds of DDT have been used throughout the world for insect control since 1940 (Ware, 1989). Photo courtesy of the Tennessee Valley Authority Historic Collection.
Figure 8. Location of sampling sites with detections of total chlordane in fish tissue in the lower Tennessee River Basin, 1980-98.
Figure 9. Location of sampling sites with detections of toxaphene in fish tissue in the lower Tennessee River Basin, 1980-98.
77 percent of the total DDT compound and is typically the most frequently analyzed and detected isomer. Although use of DDT has been banned for over 25 years in the United States, concentrations of DDT and its breakdown products are still detected in whole fish and fish fillet samples. The persistence of DDT in fish tissue may be attributed to the fact that this synthesized product is insoluble in water, and extremely soluble in fatty tissue as well as resistant to metabolism (Ware, 1989).

The first breakdown product of DDT is DDD, which subsequently breaks down into DDE. Of these three compounds, the most commonly detected was the DDE isomer, \( p,p' \)-DDE, which was detected at approximately 80 percent of the sites sampled. The compound \( p,p' \)-DDE composed approximately 80 percent of the total DDT concentration detected in whole fish and approximately 48 percent detected in fish fillet samples. Because DDE is one of the final breakdown products of DDT, higher percentages of DDE were expected in whole fish sample results for two reasons. First, whole fish samples were collected in the summer of 1998 whereas fish fillet samples were collected from 1985 to 1994 allowing more time for DDT metabolization in the whole fish samples. Secondly, whole fish have a higher fat content compared to fish fillets. Because organochlorine compounds such as DDE are more soluble in fatty tissue, higher concentrations of DDE were expected to be found in whole fish. Because DDE was present in a greater percentage of the samples than either of the parent compounds, these results may indicate that the breakdown of DDT is in the final stages. This finding is parallel with the finding made by Schmidt and others (1990) using the NCBP concentration data. Schmidt and others (1990) noted that the proportional composition of the DDT mixture changed little from the mid-1970’s through 1981 and consisted of about 70 percent \( p,p' \)-DDE, 20 percent \( p,p' \)-DDD, and 10 percent \( p,p' \)-DDT. When the data collected by the NCBP from 1984 is included, the proportional composition of \( p,p' \)-DDE of the total DDT mixture increases to 73 percent, which is similar to data for the LTEN River Basin whole fish samples and may indicate the same transition from parent to metabolite noted in the LTEN River Basin (Schmidt and others, 1990). Concentrations of \( p,p' \)-DDE were as high as 1.2 mg/kg in whole fish and 12.8 mg/kg in fish fillets (table 1). No detections of \( p,p' \)-DDT were above action levels or guidelines (fig. 10); however, concentrations of \( p,p' \)-DDD in fish fillet samples and \( p,p' \)-DDE in whole fish and fish fillet samples exceeded action levels and guidelines, respectively (figs. 11 and 12, respectively). Fish fillet samples with concentrations of these compounds that exceeded the action levels were collected between October 1991 and October 1993. Fish fillet samples collected after this time period had concentrations that were below action levels for \( p,p' \)-DDD and \( p,p' \)-DDE.

Although concentrations of \( p,p' \)-DDT, \( p,p' \)-DDD, and \( p,p' \)-DDE were frequently detected in fish tissue across the LTEN River Basin, one area in particular had concentrations that were above action levels and guidelines. The area surrounding Indian Creek, located just west of Huntsville, Alabama, and the confluence of Indian Creek with the main stem of the Tennessee River, had 10 of 17 \( p,p' \)-DDD and \( p,p' \)-DDE sample concentrations that equaled or exceeded the action levels or guidelines established by the NAS/NAE, NYDEC, or FDA. Fish tissue samples that exceeded the action levels and guidelines were collected between 1991 and 1998. The highest concentrations of \( p,p' \)-DDT, \( p,p' \)-DDD, and \( p,p' \)-DDE were collected in October 1991 in Wheeler Lake at Tennessee River mile 320. The most recent sample collected in this area was spotted bass (Micropterus punctulatus) in August 1998, which had a \( p,p' \)-DDE concentration of 1.2 mg/kg. This sample was collected in Indian Creek upstream from the posted fish consumption advisory section of the creek. In September 2000, fish advisories recommended that no fish be consumed that are taken from Indian Creek between Redstone Arsenal and the Tennessee River as a result of elevated concentrations of DDT (Alabama Department of Public Health, 2000).

**Polychlorinated Biphenyls**

Polychlorinated biphenyls (PCB’s) are synthetically halogenated aromatic hydrocarbons that were first developed in 1881. Total PCB’s refers to the sum of 209 individual chlorinated chemicals known as congeners. By 1930, PCB’s were in general use as lubricants, heat transfer agents, flame retardants, and plasticizers. By July 1979, the use of PCB’s was restricted by the U.S. EPA as a result of contamination concerns. These restrictions essentially stopped the use, production, and distribution of PCB-containing products except in totally enclosed systems and when special exemption was granted by the U.S. EPA.
Figure 10. Location of sampling sites with detections of $p,p'$-DDT in fish tissue in the lower Tennessee River Basin, 1980-98.
Figure 11. Location of sampling sites with detections of \( p,p' \)-DDD in fish tissue in the lower Tennessee River Basin, 1980-98.
Figure 12. Location of sampling sites with detections of $p,p'$-DDE in fish tissue in the lower Tennessee River Basin, 1980-98.
PCB’s reach the environment in several ways including industrial effluents, landfills, and equipment storage sites. PCB’s bond tightly with particulate matter, which enables PCB’s to be resuspended in lakes and streams. This resuspension has been documented to occur for up to 15 years and is indicative of the slow degradation of PCB’s (Eisler, 1986).

PCB’s were detected at 78 percent of the sites sampled (fig. 13) between 1985 and 1998. Concentrations of PCB’s ranged from 0.1 to 85 mg/kg in fish fillet samples and were detected at 63 of the 75 sampling sites in the LTEN River Basin (table 1). Fourteen samples that exceeded the 2.0 mg/kg action level were collected between August 1985 and March 1987. Fish fillet samples collected at some sites after this time period had levels of PCB’s that were below the action level. Concentrations of PCB’s in whole fish ranged from less than 0.05 to 0.30 mg/kg. Whole fish samples did not have the high detection percentage of fish fillets, but were detected at 3 of the 10 whole fish sampling stations. Two samples exceeded the NYDEC criteria of 0.13 mg/kg (Newell and others, 1987). When compared to the data collected by Schmidt and others (1990), the detection percentage for total PCB’s is similar with 90 percent of the 117 NCBP sampling sites having detections of PCB residues. According to Schmidt and others (1990), this percentage has remained steady since the mid-1970’s. Mean concentrations of total PCB’s for the same NCBP data, however, have declined significantly when comparing data from the mid-1970’s to data of the early-1980’s. Currently (2000), fish consumption advisories are in effect for Woods Lake and Nickajack Lake as a result of PCB contamination (Tennessee Department of Environment and Conservation, 2000).

### OCCURRENCE AND DISTRIBUTION OF TRACE ELEMENTS IN FISH TISSUE

Trace elements in the aquatic environment are derived from human activities and from natural geologic processes. Human activities that lead to the introduction of trace elements in the water column, and subsequently to aquatic organisms, include fossil-fuel combustion (coal-fired generating plants), discharges from point and non-point sources, emissions from automobiles, mineral and strip mining, textile and smelting operations, and agricultural application of fertilizers and pesticides. Coal-fired power generation contributes trace elements to the environment when fly ash containing concentrations of trace elements becomes airborne and then settles on the land, rivers, streams, and reservoirs to become part of the water column and the sediment layer. In addition to airborne precipitate, fly ash has historically been used to surface roads, make soap, and produce charcoal (Sorenson, 1991). Stream channels cutting through formations that contain high amounts of trace elements, as well as eroding rocks and soils, can transport trace elements into the aquatic environment.

Fish fillet samples were analyzed for 10 trace elements at 102 sites in the LTEN River Basin between 1980 and 1996. Trace elements without an available action level for comparison include chromium, copper, lead, nickel, silver, and zinc. The information used for comparison included FDA action levels (U.S. Food and Drug Administration, 1989) and U.S. EPA screening criteria (U.S. Environmental Protection Agency, 1995). The trace element data discussed in this report are based on fish fillet samples collected by the TVA from 1980 through 1996.

Arsenic, mercury, cadmium, and selenium are the only trace elements listed on the U.S. EPA Priority Pollutant List (U.S. Environmental Protection Agency, 1994) that have existing action levels (table 2). Generally, trace elements were detected in the highest concentrations in reservoirs, such as Wheeler and Wilson, along the main stem of the Tennessee River.

Application of herbicides used as defoliants also can contribute to the presence of trace elements. Defoliants are used to desiccate leaves from plants prior to harvesting to reduce the amount of leaf debris in harvesting equipment. Arsenic used in defoliants is usually in the form of arsenic acid (H₃AsO₄). In the 1940’s, arsenic was used not only to desiccate crops for harvesting but also to reduce the amount of vegetation (water lilies, bulrushes, and other rooted plants) in reservoirs. Studies have shown that arsenic was still present in reservoirs as many as 13 years after application was discontinued (Sorenson, 1991). Concentrations of arsenic in fish fillet samples (fig. 14) ranged from 0.02 to 0.38 µg/g with no sample having a concentration higher than the 3.0 µg/g screening criteria (table 2). Arsenic was detected in fish fillets at 45 of the 102 sampling sites.

Mercury is a naturally occurring element present in the Earth’s crust throughout the world. Various manufacturing processes and agricultural chemicals have increased the mercury levels in surface water, and subsequently, in the tissue of fish that feed...
Figure 13. Location of sampling sites with detections of PCB's in fish tissue in the lower Tennessee River Basin, 1980-98.
Table 2. Summary of trace elements in fish fillet samples collected in the lower Tennessee River Basin, 1980-96
[Trace elements in **bold** were detected at or above established action levels; µg/g, microgram per gram; ---, no information]

<table>
<thead>
<tr>
<th>Compound</th>
<th>Number of samples</th>
<th>Number of detections</th>
<th>Number of sites sampled for contaminant</th>
<th>Number of sites with detections</th>
<th>Range of data (µg/g)</th>
<th>Action level (µg/g)</th>
<th>Number of sites with detections above lowest action level</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arsenic</td>
<td>397</td>
<td>71</td>
<td>102</td>
<td>45</td>
<td>0.02 - 0.38</td>
<td>3.0</td>
<td>0</td>
</tr>
<tr>
<td>Cadmium</td>
<td>397</td>
<td>138</td>
<td>102</td>
<td>55</td>
<td>.002 - .5</td>
<td>10.0</td>
<td>0</td>
</tr>
<tr>
<td>Chromium</td>
<td>397</td>
<td>153</td>
<td>102</td>
<td>56</td>
<td>.02 - 2.6</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>Copper</td>
<td>397</td>
<td>103</td>
<td>102</td>
<td>55</td>
<td>.2 - 5.6</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>Lead</td>
<td>397</td>
<td>147</td>
<td>102</td>
<td>69</td>
<td>.02 - 3.0</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td><strong>Mercury</strong></td>
<td><strong>397</strong></td>
<td><strong>108</strong></td>
<td><strong>102</strong></td>
<td><strong>51</strong></td>
<td><strong>.11 - 5.0</strong></td>
<td><strong>0.6</strong></td>
<td><strong>1.0</strong></td>
</tr>
<tr>
<td>Nickel</td>
<td>397</td>
<td>10</td>
<td>102</td>
<td>7</td>
<td>1.0 - 9.0</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>Selenium</td>
<td>397</td>
<td>185</td>
<td>102</td>
<td>70</td>
<td>.05 - .71</td>
<td>50.0</td>
<td>0</td>
</tr>
<tr>
<td>Silver</td>
<td>397</td>
<td>0</td>
<td>102</td>
<td>0</td>
<td>---</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>Zinc</td>
<td>397</td>
<td>220</td>
<td>102</td>
<td>79</td>
<td>3.0 - 46.0</td>
<td>---</td>
<td>---</td>
</tr>
</tbody>
</table>

*a*U.S. Environmental Protection Agency screening criteria for protection of human health. Based on $1 \times 10^{-5}$ risk factor for an average-sized adult (70 kg) and a consumption of 6.5 grams of fish per day (U.S. Environmental Protection Agency, 1995).

*b*Food and Drug Administration legal limit for removing fish from marketplace (U.S. Environmental Protection Agency, 1992).
Figure 14. Location of sampling sites with detections of arsenic in fish tissue in the lower Tennessee River Basin, 1980-96.
in our streams and rivers. Until the early 1970’s, the process of electrolysis was one of the main sources of mercury in surface water. After the use of electrolysis diminished, the primary source of mercury was the burning of fossil fuels and smelting. This latter source may have lead to the increased background levels of mercury worldwide (Hem, 1989). Other possible sources of mercury include slimicides used to control fouling of mechanical pumps, preservation of wood fiber at paper plants, the manufacture of plastics and electronics, and biocide use to control mold on seed grain. Elevated mercury concentrations in fish tissue in northwestern Alabama (fig. 15) are the result of industries located along the main stem of the Tennessee River (Don Dycus, Tennessee Valley Authority, oral commun., 2000). Methylated mercury, considered to have the highest toxicity of mercury compounds, is the most bioavailable form of mercury (Eisler, 1987); however, data presented in this report describe total mercury concentrations. Mercury was detected in fish fillets at 51 of 102 sampling locations with concentrations ranging from 0.11 to 5.0 µg/g (table 2). Mercury concentrations were higher than the 0.6 µg/g U.S. EPA screening criteria in 62 samples and at 8 sites (fig. 15 and table 2).

Concentrations of cadmium in fish tissue varies and depends on fish species. In a study conducted by Murphy and others (1978), 11 fish species from an industrially polluted lake showed that piscivorous fish (black crappie and largemouth bass) tended to have lower cadmium levels than did omnivorous fish (warmouth and bluegill). Higher levels of cadmium were found in redear sunfish (*Lepomis microlophus*), which consumed considerably higher amounts of benthic macroinvertebrates from the cadmium-rich sediment than do other omnivores (Murphy and others, 1978). In the LTEN River Basin, cadmium (fig. 16) was detected at 55 of the 102 sampling sites and in 35 percent of the fish fillet samples; however, all detections were below the 10 µg/g screening criteria (table 2).

Selenium availability is almost entirely attributed to human activity. Coal and petroleum combustion provide approximately 70 percent of available selenium to the air, whereas glass manufacturing provides about 20 percent, and metallurgical processes provide 8 percent. Electronics, xerography, and mining provide the remaining 2 percent. Fly ash from fossil-fuel combustion, which contains 300 to 1,100 times more coal-derived selenium than any other element, is responsible for most of the selenium in the environment (Sorenson, 1991). Selenium had the highest detection frequency in fish tissue of the 10 trace elements analyzed and approximately 70 percent of the sampling sites (fig. 17) had a measurable concentration. Selenium concentrations varied from 0.05 to 0.71 µg/g (table 2); however, selenium did not exceed the U.S. EPA screening criteria of 50 µg/g.

**SUMMARY**

Over 200 sites were sampled for the presence of organochlorine pesticides, polychlorinated biphenyls, and trace elements in fish tissue. The sampling sites presented in this report are located primarily along the main stem of the Tennessee River, a small spatial coverage when compared to the entire LTEN River Basin study unit, which encompasses approximately 19,500 square miles.

Between 1980 and 1998, 101 sites in the LTEN River Basin study unit were sampled to determine the presence of 28 organochlorine compounds in fish tissue. The compounds *p,p*′-DDT, *p,p*′-DDD, and *p,p*′-DDE were detected at 60, 70, and 84 of the 101 sampling sites, respectively. The compound *p,p*′-DDD was detected above action levels and guidelines at two fish tissue sampling sites. The compound *p,p*′-DDE was the most frequently detected compound in fish tissue with detections at 83 percent of the sites sampled. Fish tissue samples from five sites exceeded *p,p*′-DDE action levels and guidelines. PCB’s were detected in fish tissue at 66 of 85 sampling sites, but only samples from 8 sites had concentrations exceeding action levels or guidelines. PCB and *p,p*′-DDE concentrations were detected above action levels and guidelines in both whole fish and in fish fillet samples. Dieldrin was detected at 13 of 85 fish tissue sampling sites with 1 whole fish sample having a detection equal to the 0.1 mg/kg guideline. Endrin was detected at 30 of 92 fish tissue sampling sites with 1 whole fish sample exceeding the 0.025 mg/kg guideline. Total chlordane was detected at 28 of 96 fish tissue sampling sites. Concentrations above action levels and guidelines occurred in one fish fillet sample and one whole fish sample.

Fish fillet samples were analyzed for 10 trace elements at 102 sites in the LTEN River Basin between 1980 and 1996. Mercury, a naturally occurring element, was detected in fish fillet samples at 51 of 102 sampling sites and was the only trace element to exceed an action level. Eight sampling sites had fish
Figure 15. Location of sampling sites with detections of mercury in fish tissue in the lower Tennessee River Basin, 1980-96.
Figure 16. Location of sampling sites with detections of cadmium in fish tissue in the lower Tennessee River Basin, 1980-96.
Figure 17. Location of sampling sites with detections of selenium in fish tissue in the lower Tennessee River Basin, 1980-96.
tissue samples with mercury concentrations higher than the action level; however, sampling sites where concentrations exceeded the action level were relatively localized. Generally, the highest concentrations of trace elements were found at sites located on reservoirs (Wheeler and Wilson) along the main stem of the Tennessee River. Selenium was detected in fish fillet samples from 70 of 102 sites sampled but did not exceed the 50 micrograms per gram U.S. Environmental Protection Agency screening criteria. Arsenic and cadmium were detected at 45 and 55 of the 102 sampling sites, respectively.

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