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Assessment of Contaminant Bioaccumulation in Aquatic Biota on and Adjacent to the Oak Ridge Reservation—2015

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OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

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Environmental Sciences Division

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ACRONYMS

BCK	Bear Creek kilometer
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
CRK	Clinch River kilometer
CVAFS	cold vapor atomic fluorescence spectrometry
DOE	US Department of Energy
EFK	East Fork Poplar Creek kilometer
EFPC	East Fork Poplar Creek
EPA	US Environmental Protection Agency
ERM	Emory River mile
ETTP	East Tennessee Technology Park
FYR	five-year review
HCK	Hinds Creek kilometer
LEFPC	Lower East Fork Poplar Creek
LWBR	Lower Watts Bar Reservoir
MCK	McCoy Branch kilometer
MDL	method detection limit
MEK	Melton Branch kilometer
MIK	Mitchell Branch kilometer
ND	not detected
NPDES	National Pollutant Discharge Elimination System
NRWQC	National Recommended Water Quality Criterion
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reservation
PCBs	polychlorinated biphenyls
PCB_T	total PCB concentration
PCK	Poplar Creek kilometer
RER	Remediation Effectiveness Report
TEF	toxic equivalency factor
TEQ	toxic equivalent
TMDL	total maximum daily load
TRM	Tennessee River mile
TWRA	Tennessee Wildlife Resources Agency
UEFPC	Upper East Fork Poplar Creek
WCK	White Oak Creek kilometer
WOC	White Oak Creek
WOL	White Oak Lake
Y-12	Y-12 National Security Complex

1. INTRODUCTION

This report provides information on contaminant concentrations in multiple wildlife prey species inhabiting or associated with water bodies on and downstream from the Oak Ridge Reservation (ORR), including regional reference sites. This information can be used to understand the nature and extent of contaminant exposure and transfer through the food chain and to provide a baseline for evaluating temporal trends in contaminant exposure and accumulation. This information was gathered as part of the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) Five-Year Review (FYR) process, and it also addresses regulator comments on the annual Remediation Effectiveness Report (RER).

This report summarizes the results of biological sampling completed in the spring and summer of 2015, prior to the 2016 FYR. Sampled locations included key sites upstream from the point sources of contaminants, along with watershed integration points on and off the ORR. Integration points are spatially the most important sampling sites because they capture biological responses to upstream contaminant inputs from both aqueous and groundwater sources, as well as various remedial actions. These sites are most often located at a watershed administration unit boundary and may have regulatory significance for performance measures. In many cases, there are long-term biological and other monitoring data associated with these sites; these data generally fall within the areas of exposure and effects.

To assess human health risks to exposure to contaminants in aquatic systems, fish fillets are typically monitored because this is the edible portion for humans. To assess ecological risks of contaminant exposure, however, researchers must use a different strategy. Aquatic birds, fish, and other wildlife consume whole body fish as well as a host of invertebrate prey. Benthic macroinvertebrates and emergent insects often comprise a large proportion of the diets of many fish and terrestrial animals that live adjacent to bodies of water (Christensen and Moore 2007; Duffield and Nelson 1998; Suter and Cormier 2015). As such, these prey may be important vectors of contaminants through the food chain (Smith et al. 2016; Nagle et al. 2001; Reinfelder et al. 1998). Spiders that live in riparian habitat and prey on aquatic insects may serve as an important link for contaminant transfer between aquatic and terrestrial ecosystems (Burdon and Harding 2008; Cristol et al. 2008; Walters et al. 2010; Otter et al. 2013). Therefore, as in the 2011 FYR (Mathews et al. 2011), aquatic invertebrates, whole body forage fish, and turtle tissues were included in this assessment to provide data needed for a more complete ecological risk assessment.

The primary contaminants of concern in the evaluation include mercury (Hg), methylmercury (MeHg), polychlorinated biphenyls (PCBs), silver (Ag), arsenic (As), beryllium (Be), cadmium (Cd), chromium (Cr), copper (Cu), lithium (Li), molybdenum (Mo), nickel (Ni), lead (Pb), antimony (Sb), selenium (Se), thallium (Tl), uranium (U), and zinc (Zn). Additionally, a suite of dioxins and furans was measured in fish from White Oak Creek (WOC), as these compounds have been found to be elevated in the past (BJC 2005; Mathews et al. 2011).

2. SITE DESCRIPTION

The ORR, a US Department of Energy (DOE) facility, comprises 33,500 acres in East Tennessee. The site was developed as part of the Manhattan Project in the 1940s, and many areas within the ORR are still used for industrial and/or national security purposes. As a result, many of the streams and reservoirs on and adjacent to the ORR have received significant contaminant inputs over the years. Here we consider contaminant concentrations in biota in five major watersheds on the ORR that are operationally defined as East Fork Poplar Creek (EFPC), WOC watershed, Bear Creek, Chestnut Ridge, and East Tennessee Technology Park (ETTP). We also examine contaminant concentrations in the reservoirs downstream of the ORR, which we define as "off-site," as well as at reference sites with no contaminant inputs from DOE facilities (Table 2.1, Figs. 2.1, 2.2; BJC 2005).

Based on historical knowledge of contaminant concentrations in surface waters and sediments on the ORR, over 20 stream and reservoir sites were included in this study, representative of sites down-gradient of contamination sources (i.e., integration points) as well as upstream locations and reference sites. Integration points on the ORR included two stream locations each on EFPC and WOC watershed, one location each on Bear Creek and Mitchell Branch, and three ponds at ETTP (Fig. 2.1, Table 2.1). Integration points at off-site locations included two on the Clinch River and one each on Poplar Creek and the Tennessee River (Fig. 2.2, Table 2.1). Upstream locations were selected to isolate areas of water bodies potentially affected by remedial actions, thus providing information on the effects of these actions. Upstream sites on the ORR included one each on EFPC and McCoy Branch and two in the WOC watershed (Fig. 2.1, Table 2.1).

Four reference sites were used in the assessment (Figs. 2.1 and 2.2): (1) Hinds Creek (Hinds Creek kilometer [HCK] 20.6) was used as a reference site to assess contaminant accumulation in both stream fish and invertebrates; (2) an inlet on Melton Hill Reservoir located on the southeastern boundary of the ORR at Freels Bend (approximately Clinch River kilometer 65) was used as a reference site for invertebrates for the ETTP ponds and the Lower Watts Bar Reservoir (LWBR) site on the Tennessee River (Tennessee River mile [TRM] 531); (3) the Emory River at river mile (ERM) 6.0 was used as a fish and invertebrate reference site for the ETTP ponds and larger river systems; and (4) the Clinch River at kilometer (CRK) 38.4 was used as a turtle reference site for CRK 15 and CRK 32.

Table 2.1. Biological sampling sites, locations, and contaminants analyzed for the 2015 Oak Ridge Reservation Five-Year Review

Watershed	Water body	Site[a]	Site type	Contaminants of concern	Assessment group
EFPC	EFPC	EFK 24.4	Upstream	Hg, metals, PCBs	Fish and invertebrates
	EFPC	EFK 23.4	Integration	Hg, metals, PCBs	Fish and invertebrates
	EFPC	EFK 6.3	Integration	Hg, metals, PCBs	Fish and invertebrates
WOC watershed	WOC	WCK 3.9	Upstream	Hg, metals, PCBs	Fish and invertebrates
	WOC	WCK 2.3	Integration	Hg, metals, PCBs, dioxins/furans	Fish and invertebrates[b]
	WOC	WCK 1.5	Integration	Hg, metals, PCBs, dioxins/furans	Fish
	Melton Branch	MEK 0.2	Upstream	Hg, metals, PCBs, dioxins/furans	Fish and invertebrates[b]
Bear Creek	Bear Creek	BCK 9.9	Integration	Hg, metals, PCBs	Fish and invertebrates
Chestnut Ridge (McCoy Branch)	McCoy Branch	MCK 1.4	Upstream	Hg, metals, PCBs	Fish and invertebrates
ETTP	Mitchell Branch	MIK 0.2	Integration	Hg, metals, PCBs	Fish and invertebrates
	K-1007-P1 Pond	P1 Pond	Integration	Hg, metals, PCBs	Fish and invertebrates
	K-901-A Pond	K-901	Integration	Hg, metals, PCBs	Fish and invertebrates
	K-720 Slough	K-720	Integration	Hg, metals, PCBs	Fish and invertebrates
Off-site	Clinch River	CRK 15	Integration	Hg, metals, PCBs	Fish and invertebrates
	Clinch River	CRK 15	Integration	Hg, PCBs, gamma	Turtles
	Clinch River	CRK 32.2	Integration	Hg, metals, PCBs	Fish and invertebrates
	Clinch River	CRK 32.2	Integration	Hg, PCBs, gamma	Turtles
	Poplar Creek	PCK 1.6	Integration	Hg, metals, PCBs	Fish and invertebrates
	Tennessee River	TRM 531[c]	Integration	Hg, metals, PCBs	Fish and invertebrates
Reference	Emory River	ERM 6.0[c]	Reference	Hg, metals, PCBs	Fish and invertebrates
	Hinds Creek	HCK 20.6	Reference	Hg, metals, PCBs, dioxins/furans	Fish and invertebrates[b]
	Clinch River	CRK 38.4	Reference	Hg, PCBs, gamma	Turtles
	Freels Bend	Freels	Reference	PCBs	Invertebrates

[a]BCK = Bear Creek kilometer, CRK = Clinch River kilometer, ERM = Emory River mile, EFK = East Fork Poplar Creek kilometer, HCK = Hinds Creek kilometer, MCK = McCoy Branch kilometer, MEK = Melton Branch kilometer, MIK = Mitchell Branch kilometer, PCK = Poplar Creek kilometer, TRM = Tennessee River mile, WCK = White Oak Creek kilometer.
[b]Invertebrates were not analyzed for dioxins/furans.
[c]Names used for the Tennessee and Emory River sites are based on miles from their confluence with the Ohio and Clinch Rivers, respectively, to provide consistency with long-term historical use and data sets.

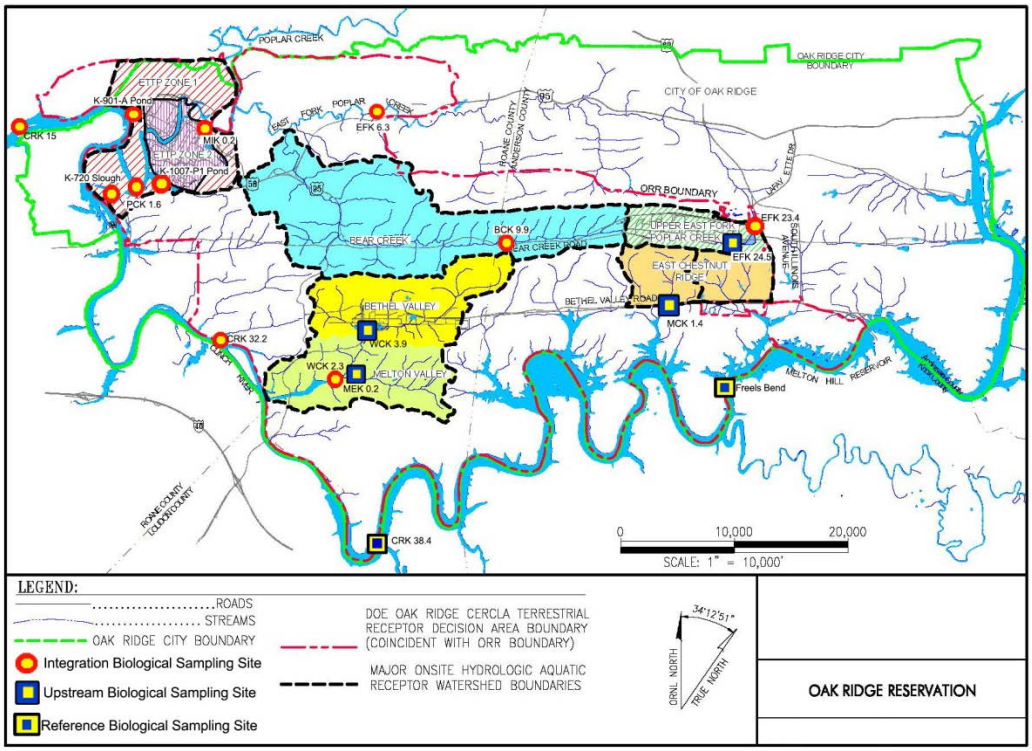

Fig. 2.1. Map of onsite biological sampling sites, off-site sampling sites on the Clinch River and Poplar Creek, and the Freels Bend reference site. See Table 2.1 for site abbreviations.

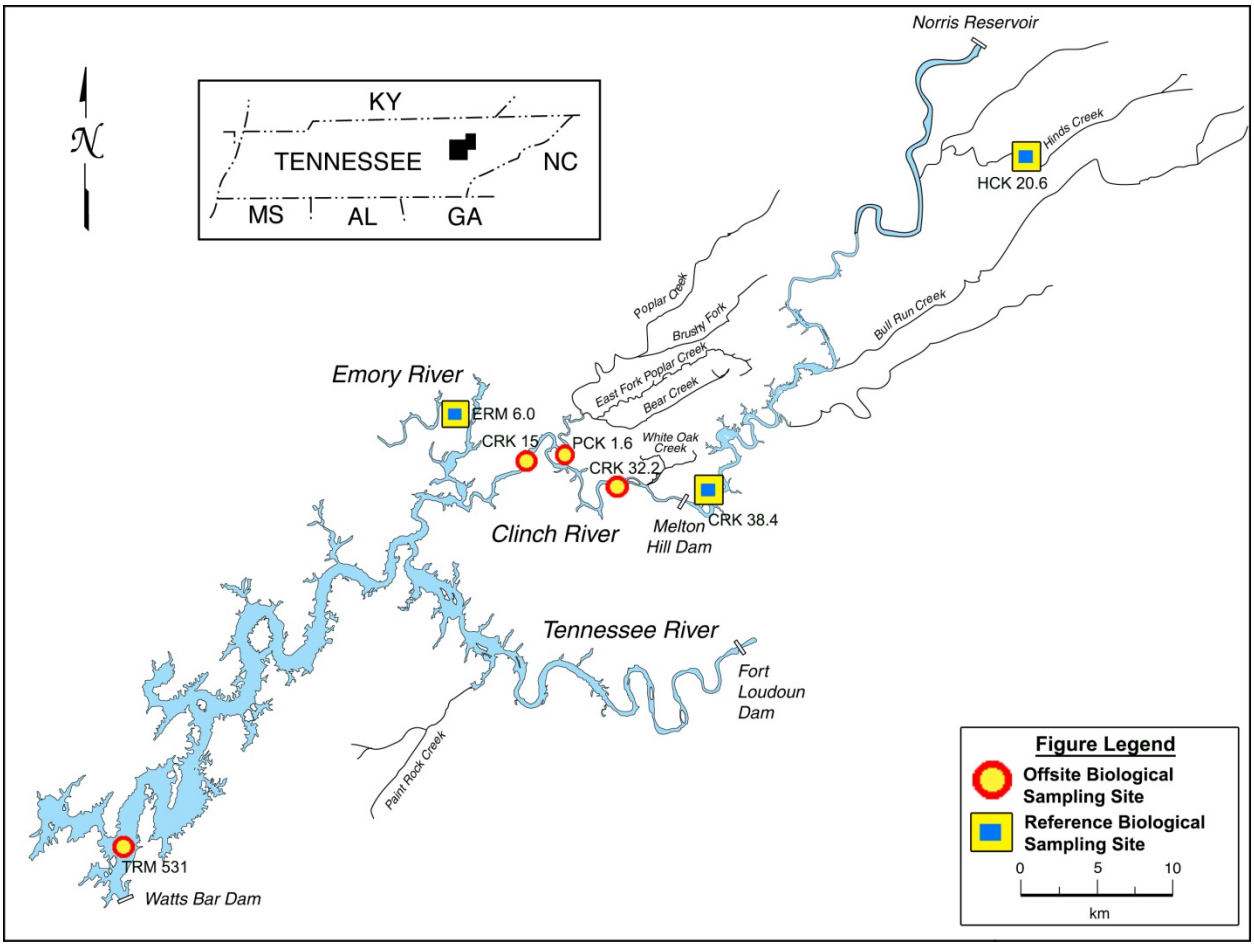

Fig. 2.2. Map of off-site and reference biological sampling sites. See Table 2.1 for site abbreviations.

3. MATERIALS AND METHODS

3.1 TARGET SPECIES

3.1.1 Invertebrate Target Taxa

Sampling locations for invertebrates presented a wide variety of physical and chemical conditions (flowing and non-flowing waters, various levels of chemically and physically altered conditions), making it necessary to select a number of target species representative of the different field conditions. The choice of target species in 2015 was based on results from the 2010 assessment (Mathews et al. 2011), the stated preference for collecting emerging aquatic insects to provide information on transfer of contaminants to potential terrestrial consumers, and species availability. However, as in 2010 (Mathews et al. 2011), collection of emerging insects proved extremely challenging, especially given the limitation of low biomass for some of the most common species and the unpredictability in the emergence of the adult insects. For this reason, invertebrates had to be collected opportunistically based on availability of sufficient biomass, similarity to species or species groups used in 2010, and similarity in species or species groups at sites with similar habitat types (e.g., ponds/standing water, large river sites, and streams). Thus, depending upon site, collections of invertebrates included adult insects (terrestrial stage of aquatic insects); immature insects in their aquatic stage (nymphs or larvae); or non-insect invertebrates (e.g., snails). A list of the species and species groups included in samples from each site and for the contaminants of concern is presented in Table A.1.

In 2010 the adult damselfly *Argia apicalis* was sufficiently abundant at all ETTP pond sites (i.e., K-901-A Pond, K-720 Slough, and K-1007-P1 Pond) and at the Freels Bend reference site to use as the target species for those sites, but in 2015, they were only abundant enough for use at K-901-A Pond, PCK 1.6, and Freels Bend. As a substitute, adult dragonflies (*Erythemis simplicicollis*) were collected at K-1007-P1 Pond and K-720 Slough. Like *Argia*, *E. simplicicollis* is an insect in the order Odonata. The nymphs of both species are predators that live primarily in shallow standing water habitats. Both species emerge as adults throughout the summer, with the adult stage lasting 2–4 weeks. As adults, both species continue to feed primarily on small emerging aquatic insects.

Adult mayflies (*Hexagenia bilineata*) were collected from CRK 15, TRM 531, and ERM 6.0 in 2015; adult mayflies were found only at CRK 15 in 2010. As in 2010, large aquatic snails (*Pleurocera canaliculatum*) were again collected at CRK 32.2; neither adult mayflies nor damselflies were found in sufficient numbers for analysis.

As in 2010, multiple attempts to collect adult caddisflies and mayflies at multiple stream sites on the ORR resulted in limited success using battery-powered light traps (Kovats and Ciborowski 1989). Therefore, the most readily available and/or common species of immature insects and/or snails were collected at all stream sites (Table A.1). The majority of samples consisted of Megaloptera larvae (fishflies or hellgrammites), Trichoptera larvae (caddisflies), Odonata nymphs (dragonflies and damselflies), or a mixture of two or more of those insect taxa. The most commonly used caddisflies were filter-feeding species in the family Hydropsychidae (*Cheumatopsyche* and *Hydropsyche*), but the caddisflies *Chimarra* and *Pycnopsyche* were included in samples from McCoy Branch (MCK 1.4) and Bear Creek (BCK 9.9) respectively. Mayfly nymphs (*Hexagenia limbata*) were also collected at HCK 20.6 in sufficient quantities for analysis of metals and Hg. Finally, snails (*Pleurocera* sp. or *Elimia claviformes*, depending on which species was available) were collected for analysis of all or some analytes at WCK 3.9, BCK 9.9, and MIK 0.2 because of low abundances/biomass of immature insects.

Fishing spiders (or nursery-web spiders, family Pisauridae) were collected from EFK 6.3, EFK 23.4, WCK 2.3, and HCK 20.6 in 2015. As in 2010, the species collected included *Dolomedes scriptus* and *Dolomedes vittatus*, although only *D. vittatus* were used in samples from EFK 23.4 (Table A.1).

3.1.2 Vertebrate Target Taxa

Target species for mercury, metals, and PCB bioaccumulation monitoring in fish on and off the ORR included forage fish, sunfish, and upper trophic level predator species. Forage fish were analyzed as whole body composites to evaluate ecological exposure, whereas predator fillets were analyzed to evaluate potential human health exposure.

Central stonerollers (*Campostoma anomalum*) are common forage fish in East Tennessee streams and were the target species for all onsite locations except MCK 1.4 and the ponds at ETTP. A lack of sufficient numbers of stonerollers at MCK 1.4 required that an alternative minnow species be used: western blacknose dace (*Rhinichthys atratulus*). Both minnow species are abundant, widespread, and relatively short-lived and sedentary, so they are used as biosentinels to monitor short-term changes in contaminant exposure at a given site. They are also important prey for larger fish, so they serve as an indicator of wildlife exposure and risk. In larger waterways and reservoirs, gizzard shad (*Dorosoma cepedianum*) are an abundant forage fish species and are important prey for larger game fish. Gizzard shad were therefore the target species at large off-site locations and the K-901-A Pond and K-720 Slough at ETTP (Table A.1).

In addition to assessing bioaccumulation of metals, mercury, and PCBs, bioaccumulation of dioxins and furans in Melton Branch (MEK 0.2) and White Oak Lake (WCK 1.5) was also assessed in whole body forage fish and fillets of largemouth bass (*Micropterus salmoides*). Stonerollers were collected from MEK 0.2, and gizzard shad and largemouth bass were collected from WCK 1.5 for dioxin/furan analysis. Largemouth bass are large, upper trophic level predatory fish and are therefore susceptible to contaminant bioaccumulation. Because they are common game fish, fillet concentrations of contaminants are relevant to assessing human health risks, and they often represent the maximum potential dose of many contaminants to humans. In addition to being used in the evaluation of dioxin and furan bioaccumulation in White Oak Lake, largemouth bass were also collected in the K-1007-P1 Pond as part of the FYR to evaluate PCB bioaccumulation in the 5 years after major remediation activities in this pond.

Because there were two main target species for fish bioaccumulation (stonerollers and gizzard shad), fish were collected from two different reference locations. Stonerollers were collected from Hinds Creek (HCK 20.6), a stream site that has been used as a reference site for bioaccumulation monitoring for over 20 years. Gizzard shad were collected from the Emory River (ERM 6.0).

In addition to fish collections, common snapping turtles (*Chelydra serpentina*) were collected from three off-site locations (CRK 15, CRK 32.2, and CRK 38.4) to evaluate contaminant bioaccumulation in a long-lived reptilian game species. Contaminant accumulation was evaluated in three tissue types in turtles— muscle, liver, and fat—to address human health and ecological risk concerns.

3.2 SAMPLE COLLECTION

3.2.1 Invertebrate Sampling

Sampling for invertebrates began in early June and continued through early September. Three replicate composite samples were collected from each site for analysis of each of the three contaminants of concern. Table A.1 summarizes the species content of each sample. All equipment and supplies were either certified clean or acid washed (e.g., bottles and vials), or they were washed with Liquinox and

rinsed with distilled water (e.g., stainless forceps, collection nets) between sites. Fresh nitrile gloves were worn during collections.

Adult damselflies (*Argia apicalis*) and dragonflies (*Erythemis simplicicollis*) were collected with clean insect sweep nets along the shoreline at the ETTP ponds, Freels Bend, and PCK 1.6. As specimens were collected, they were transferred from the net to new plastic sample bags and placed into a cooler of ice within 30–60 min for storage and transport to the laboratory.

Adult mayflies (*Hexagenia bilineata*) were collected at CRK 15, TRM 531, and ERM 6.0 along the shore from a boat with clean sweep nets. Collected mayflies were transferred to clean, 1 L glass sample bottles and stored in a cooler on ice for transport to the laboratory. To limit possible bias due to potential sex differences in contaminant accumulation, only males were included in samples that were analyzed.

Snails were collected by hand (using nitrile gloves) from submerged rocks in shallow water or along the shoreline at BCK 9.9, MIK 0.2, and CRK 32.2 (See Table A.1 for the specific species used at each site). Collected snails were placed in clean glass sample bottles partially filled with water from the site and were stored in a cooler on ice for transport to the laboratory.

All samples of insect nymphs and larvae were collected with aquatic kick and dip nets. Caddisfly and fishfly larvae were primarily collected from riffles (i.e., fast-flowing, shallow water), while dragonfly and damselfly nymphs were primarily collected from large submerged balls of tree roots located in the water next to the shore. Nymphs of the burrowing mayfly *Hexagenia limbata* were collected with a dip net from soft sediments at HCK 20.6, the only stream site where this species of mayfly was found. The sample bottles were placed on ice in a cooler for transfer back to the laboratory.

Spiders were collected with an insect sweep net from overhanging vegetation, accumulations of woody debris in the water, and tree trucks next to the water while wading in the water. Netted spiders were transferred to clean 1 L glass sample bottles and placed on ice in a cooler within 30 min after collection. Samples were kept on ice for transport to the laboratory.

3.2.2 Vertebrate Sampling

All fish were collected by electrofishing, a technique that uses submerged conducting probes to pass electric current through water to temporarily stun or incapacitate fish. Electrofishing equipment differs in design depending upon the size and depth of the body of water being sampled. Backpack electrofishers (Smith-Root, Vancouver, WA) were used to collect fish from streams, while electrofishing boats were used in the ETTP ponds, White Oak Lake (WOL), and large off-site locations. Each electrofishing unit has a self-contained gasoline- or battery-powered generator that delivers up to 1,200 volts of pulsed direct current. The pulse frequency, output voltage, and waveform are varied, depending upon the specific conductance of water, which affects the size and strength of the electrical field in the water. Stunned fish are generally attracted to and swim toward the conducting probes, facilitating their capture. Collected fish were placed on ice and brought back to the lab for processing.

In contrast to the invertebrate sampling, which was expressly done in 2015 for this FYR, bioaccumulation monitoring in fish is routinely conducted for various contaminants at most of the sites included in this FYR. Although the target fish species and contaminants considered in this FYR may differ from the routine biomonitoring program, an effort was made to leverage routine sample collection activities wherever possible. Because contaminant concentrations can vary significantly in small fish over short periods, and such short-term variation can have significant implications for long-term biomonitoring programs (Eagles-Smith and Ackerman 2009), every effort was made to preserve the routine bioaccumulation monitoring sampling schedule.

Sampling for forage fish began in mid-November of 2014 and continued through late June 2015. Thirty fish of similar size and weight were collected from each site, with the goal of obtaining three replicate composite samples of ten fish each. As with the invertebrate sample collection, alternative species had to be collected at several locations where the target species were not present. In McCoy Branch (MCK 1.4), stonerollers were not present, so 24 western blacknose dace (*Rhinichthys obtusus*) were collected instead, producing 3 composites of 8 fish each. In Mitchell Branch, the collection site (MIK 0.2) was a slow-flowing, deep, muddy site that is not ideal habitat for stonerollers, so only 21 fish were collected, producing 3 composites of 7 fish each.

In 2009, remediation of the K-1007-P1 Pond included removal of forage and predatory fish, and then restocking the pond with smaller, lower trophic level fish. The restocked species are not expected to accumulate PCBs as readily as previous species. Bluegill (*Lepomis macrochirus*) was among the restocked species, so it was collected as the target species in the K-1007-P1 Pond. Bluegill are short-lived and relatively sedentary, so contaminant concentrations in these fish are representative of exposure at the site of collection. Damage to the pond weir grate during a significant flood in May 2010 allowed largemouth bass (*Micropterus salmoides*) to reenter the pond. Thus, in 2015, largemouth bass were also collected from the K-1007-P1 Pond for analysis.

Three turtles were collected with hoop nets from each of three off-site locations (CRK 15, CRK 32.2, and CRK 38.4). Commercially available bait was placed in each hoop net, and up to six hoop nets were placed at each site. Hoop nets were left in place for approximately 16 hours in habitats known to be frequented by turtles (e.g., large logs, undercut banks). One end of the nets was left protruding above the water line to provide captured turtles with access to air. When nets were retrieved, non-target species were released.

3.3 LABORATORY PROCESSING

3.3.1 Invertebrate processing

The laboratory procedures used to process invertebrate samples varied by species and species group. All specimens were handled with clean or new supplies and equipment by researchers wearing nitrile gloves. All plastic and glass lab supplies with the potential of coming into contact with specimens were first cleaned with Liquinox soap, soaked in 10% nitric acid for 24 h, and then triple-rinsed with distilled water. The same cleaning sequence was followed for stainless-steel equipment, but these items were not soaked in nitric acid. After processing was complete, samples were stored in a secure freezer (~-20°C) until being shipped for analysis. All samples were shipped to analytical laboratories on dry ice.

Samples of adult damselflies, dragonflies, and spiders arriving at the laboratory were immediately transferred to a secure freezer (~-20°C) and held for ≥1 h before further processing. Samples of these taxa were later removed from the freezer and sorted by species (spiders only) and sex. Adult mayflies were allowed to thaw (~1 h) after removal from the freezer, and only the male imagos were included in samples. Samples of adult damselflies included a mixture of equal numbers of males and females (~40 : 60 male to female mass). Both species of spiders (*D. vittatus* and *D. scriptus*) were collected at EFK 23.4, but because of the low biomass of *D. scriptus*, samples from that site included only *D. vittatus.* Samples from the other three sites included both *D. vittatus* and *D. scriptus.* When possible, similar proportions of males and females of each species were included in samples for a given contaminant of concern. Segregating samples by sex and species helps limit potential bias that can be associated with physiological differences between species and gender that can differentially affect assimilation and accumulation of contaminants. Specimens were added to clean, pre-weighed, and tared 40 mL sample bottles with forceps until the target biomass was reached; weights were determined to the nearest 0.1 mg with an analytical balance. A label with a unique six-digit sample identification number was affixed to the side of each sample, and the samples were returned to the secure freezer.

Samples of aquatic insect nymphs and larvae were removed from the cooler and thoroughly rinsed with distilled water to remove any adhering silt and other debris. Once rinsed, the specimens were placed in clean sample bottles and stored in a secure freezer until further processing could be completed. Organisms were removed from the freezer on a later date, placed in a clean plastic Petri plate, and sorted by taxonomic group. Specimens were placed on clean, dry Kimwipes to remove excess external moisture, and then they were added to a clean pre-weighed and tared 40 mL sample bottle until the target biomass was reached. Weights were determined to the nearest 0.1 mg. A label with a unique six-digit sample identification number was affixed to the side of each sample, and the samples were returned to the secure freezer.

After arriving at the laboratory, snails were removed from the ice, and the surface of their shells was scrubbed with a small acid-washed brush to remove attached debris (e.g., algae, silt, clay). The scrubbed snails were then rinsed with distilled water, placed in clean sample bottles, and held in a freezer until further processed. Snails were later removed from the freezer and allowed to thaw for ~30 min. Once thawed, specimens were briefly blotted on clean Kimwipes to remove excess external moisture. The shell on the snails was then carefully cut away from the tissue with fine stainless steel wire cutters (prewashed with Liquinox soap and triple-rinsed with distilled water), starting at the juncture between the aperture and the beginning of the spire and continuing along the edge of the spire until reaching the long fibrous muscle that connects the animal to the shell. The muscle was detached from the inner shell wall with clean stainless steel forceps, and all tissue was removed and placed in a clean, pre-weighed, and tared 40 mL sample bottle. This was repeated until the target biomass was obtained. A label with a unique sample identification number was affixed to the side of each sample bottle before returning the processed samples to the secure freezer.

3.3.2 Vertebrate Processing

Fish were processed immediately upon arriving at the laboratory after collection. Each largemouth bass collected was given a unique sample identification number, measured for total length and weight, and scaled and filleted with clean stainless steel dissecting tools. All tools were rinsed thoroughly with distilled water between individual fish from the same site. When fish from multiple sites were processed in the same day, the tools were washed with Liquinox detergent and rinsed with distilled water. Fillets from each fish were wrapped in separate aluminum foil pouches and stored in a locked freezer (-20°C) until they were shipped to analytical laboratories. All samples were shipped on dry ice, with signed chain-of-custody documentation.

Forage fish were separated into three 10-fish composite samples in the laboratory. Each composite sample was given a unique sample identification number, and each fish that was part of a composite sample was measured and weighed before being frozen whole in an aluminum foil pouch (-20°C). Frozen fish were later thawed slightly before being triple-processed through a stainless steel meat grinder to obtain a homogenous composite sample of the whole body fish. The meat grinder was thoroughly rinsed in between each composite sample and was washed with Liquinox detergent in between samples from different sites. Composite samples were then divided into separate aliquots for each contaminant of concern: 20 g for PCBs, 5 g for Hg, 5 g for metals, 10 g for gamma, and 5 g for dioxin/furans. Each aliquot was wrapped in a separate aluminum foil pouch and stored in a secure, locked freezer (-20°C) until being shipped to analytical laboratories. All samples were shipped on dry ice, with signed chain-of-custody documentation.

Turtles were brought back to the laboratory in a large covered tote. They were euthanized following approved Animal Care and Use Committee protocols before dissection. Equal aliquots of muscle, liver, and fat were taken from each turtle and were composited by site and tissue type.

3.4 ANALYTICAL METHODS

All fish and invertebrate samples were sent to accredited laboratories and were prepared and analyzed using clean techniques and procedures specified in appropriate US Environmental Protection Agency (EPA) methods. Samples for total mercury (Hg_T) and MeHg analysis were shipped to Brooks Applied Labs (Bothell, WA). For Hg_T, samples were analyzed by EPA method 1631 (cold vapor atomic fluorescence spectrometry [CVAFS]) following digestion in nitric acid and bromine chloride dilution (EPA 2001b). MeHg samples were analyzed by EPA method 1630 (distillation, aqueous ethylation, and CVAFS) following alkaline digestion using potassium hydroxide (Liang et al. 1996, EPA 2001a). Samples collected for metal analysis were sent to ALS Analytical Laboratory (Fort Collins, Colorado). Metal samples were analyzed by inductively coupled plasma mass spectrometry using EPA method SW846-6020A (EPA 2007) following digestion in nitric acid (EPA 1996a). PCB samples were shipped to TestAmerica (Knoxville, Tennessee), where they were prepared using solvent extractions (EPA 1996b) and analyzed by gas chromatography (EPA method SW846-8082) (EPA 1996c). Dioxin and furan samples were sent to Southwest Research Institute (San Antonio, Texas) and were extracted and analyzed by high resolution gas chromatography/mass spectrometry (EPA method SW846-8290) (EPA 1994).

3.5 DATA ANALYSIS

Descriptive statistics (means and standard deviations) were calculated (in most cases, n = 3) for the results and presented in tables and figures. If an analyte was not detected in one or more of the samples used to calculate the mean (i.e., the concentration in a given sample was below the method detection limit [MDL]), the result used for the non-detect was one-half of the MDL (EPA 1998). If the analyte was not detected in any of the samples from a given site, the analyte was reported as less than (<) the highest MDL reported for the replicates. When calculating sums of analytes (total Aroclors, total Heptafurans, etc.), if none of the analytes used to calculate the sum was above detection limits, this sum was reported as not detected (ND).

4. RESULTS

4.1 POLYCHLORINATED BIPHYENLS

On the ORR, the Aroclors most often detected in environmental (and especially biota) samples are Aroclors 1248, 1254, and 1260. For this reason, total PCB concentrations (PCB_T) are operationally defined here as the sum of concentrations of these three Aroclors. In general, across the ORR, PCB_T concentrations were higher in fish than in invertebrates collected from the same location (Fig. 4.1), likely because fish tissue contains higher lipid content than invertebrate tissue. At all sites on the ORR, PCB_T concentrations in fish and spiders were elevated with respect to the Hinds Creek reference site, where none of the Aroclors were found to be above detection limits in fish. The same was true of invertebrates at many of the stream sites, but PCB concentrations in invertebrates collected from the ponds on the ORR and at off-site locations were considerably lower than those collected from stream sites. PCB_T concentrations in fish collected from off-site locations decreased as distance from the ORR increased, as is typical of point source contamination.

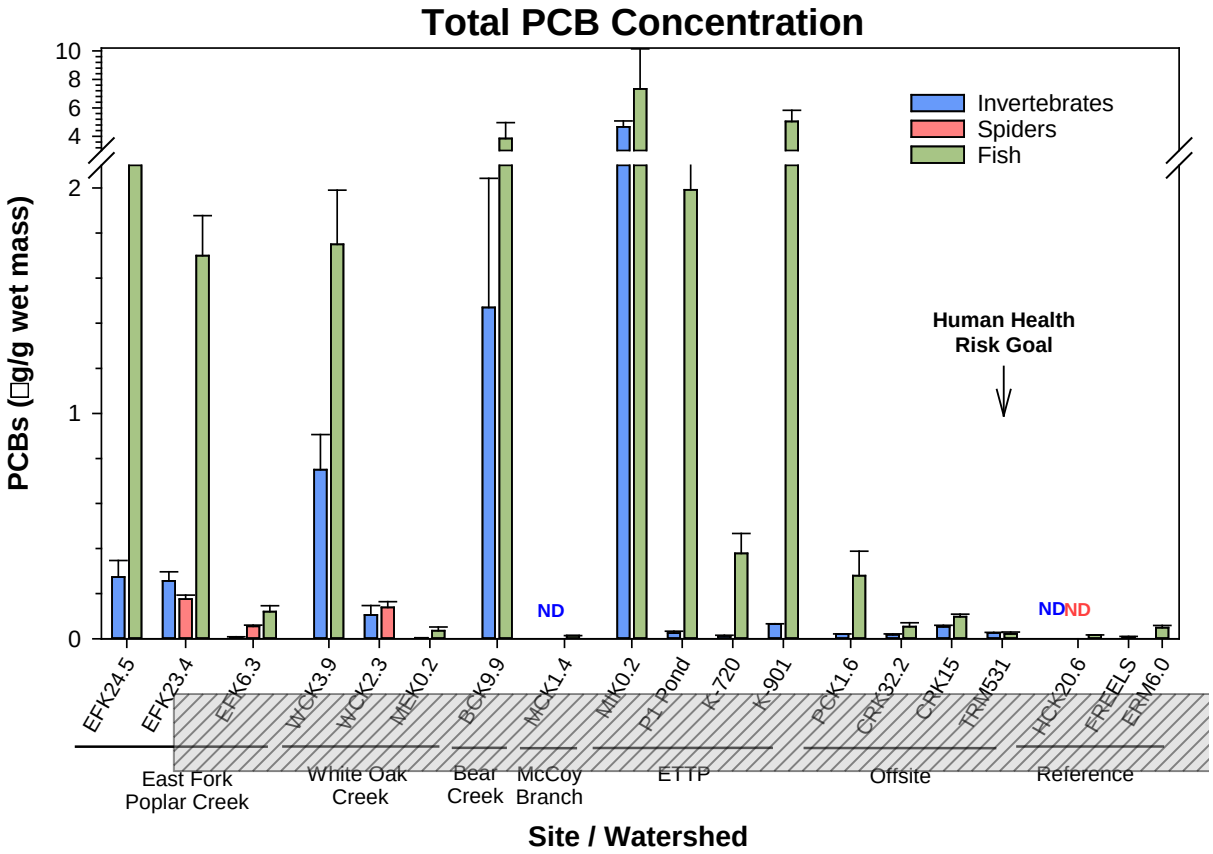

Fig. 4.1. Polychlorinated biphenyl (PCB) concentrations in biota collected from the Oak Ridge Reservation and off-site locations and reference locations, 2015. Values are means ± 1 standard deviation. The human health risk goal is the concentration at which fishing restriction signs are posted. ND = not detected and color-coded by biota grouping. Note the change in scale after the break in the y-axis.

The highest PCB_T concentrations were observed in both the fish (stonerollers) and invertebrates (snails) collected from MIK 0.2 (7.33 and 4.67 μg/g wet wt., respectively; Fig 4.1, Table B.1). Concentrations in whole body gizzard shad collected from the K-901-A Pond were also elevated (5.05 μg/g) with respect to

the remainder of the sites monitored, though the concentrations in invertebrates collected from this site were considerably lower than those collected in Mitchell Branch, but this is likely because of the difference in species collected (damselflies from the K-901-A Pond vs. snails at MIK 0.2). Indeed, the other sites where PCB_T concentrations in invertebrates were elevated were BCK 9.9 (1.47 μg/g) and WCK 3.9 (0.75 μg/g)—the other sites on the ORR at which snails were the representative invertebrate species.

Species difference among fish sampled at the different sites also undoubtedly played a role in spatial patterns observed in PCB_T concentrations. For example, while concentrations in whole body composites of bluegill collected from the K-1007-P1 Pond were lower (1.99 μg/g) than those in gizzard shad from the K-901-A Pond (5.05 μg/g), largemouth bass fillets collected from the K-1007-P1 Pond (5.23 μg/g) were >5 times higher than those in the same species collected from the K-901-A Pond (0.66 μg/g) (Peterson et al. 2016). PCB_T concentrations in gizzard shad in K-901-A Pond were 25 times higher than the concentrations observed in the same species collected from the K-720 Slough (5.05 μg/g wet mass, versus 0.38 μg/g wet mass). PCB_T concentrations in adult dragonflies from K-1007-P1 Pond and adult damselflies from K-901-A Pond were elevated with respect to concentrations at the Freels Bend reference site, whereas PCB_T at K-720 Slough was similar to concentrations at Freels Bend (Fig. 4.1, Table B.1).

Similar to patterns observed in the 2010 FYR (Mathews et al. 2011), within the stream sections of EFPC and WOC, PCB_T concentrations were highest in biota collected from upstream sections and decreased with distance downstream. From upstream to downstream in EFPC, concentrations in central stonerollers decreased by about a third from 2.4 μg/g at EFK 24.4 to 1.7 μg/g at EFK 23.4, and then by over an order of magnitude at EFK 6.3 (0.12 μg/g). Similar patterns were observed in invertebrates and spiders in EFPC, though the spatial gradients were not as marked as for fish. Concentrations in aquatic invertebrates were similar at EFK 24.5 (0.27 μg/g wet mass) and EFK 23.4 (0.26 μg/g wet mass) in upper EFPC (UEFPC; Fig. 4.1, Appendix B, Table B.1), but they were considerably lower at EFK 6.3 (0.008 μg/g wet mass). PCB_T concentrations in spiders dropped from 0.18 μg/g at EFK 23.4 to 0.06 μg/g at EFK 6.3.

Within the WOC watershed, the highest concentration (PCB_T) in invertebrates (0.75 μg/g) was found at WCK 3.9 (Table B.1, Fig. 4.1). Concentrations of PCBs in invertebrates from WCK 2.3 were lower than at WCK 3.9 by almost fivefold (0.1 μg/g wet mass and 0.75 μg/g wet mass, respectively). In contrast, total PCB concentrations in invertebrates from lower Melton Branch (MEK 0.2), a tributary to WOC just upstream of WCK 2.3, were very low (0.004 μg/g wet mass). Concentrations of PCBs in spiders at WCK 2.3 (0.14 μg/g wet mass) were similar to concentrations found in invertebrates from this site. (Table B.1, Fig. 4.1). Concentrations of PCBs were elevated in fish from WCK 3.9 (1.75 μg/g wet mass), but they were only slightly elevated at MEK 0.2 (0.04 μg/g wet mass).

PCB_T concentrations in both stonerollers and snails collected from upper Bear Creek were among the highest on the ORR (Fig 4.1, Table B.1). Of all the stream locations on the ORR, McCoy Branch had the lowest PCB_T concentrations in biota, with the invertebrate concentration below the detection limit and the concentration in blacknose dace (0.014 μg/g) comparable to reference site concentrations. Elevated concentrations of PCBs were found in fish at all off-site locations (Table B.1, Fig. 4.1) with respect to reference site concentrations. As at other sites assessed in this study, concentrations of PCBs were higher in fish than in invertebrates. Concentrations of PCBs in invertebrates at all off-site locations were relatively low (Table B.1; Fig. 4.1). PCB concentrations in adult damselflies from PCK 1.6 (0.021 μg/g wet mass) were comparable to those found in adult dragonflies in nearby K-1007-P1 Pond (0.025 μg/g wet mass).

The composition of the different Aroclors and/or congeners can sometimes provide clues as to the ultimate source of PCBs, because different Aroclors were sometimes used for different purposes. Across

the ORR and across taxa sampled, Aroclors 1254 and 1260 were the predominant Aroclors detected in the present study. The relative concentrations of the different Aroclors within the environment may provide clues to the persistence of each of the Aroclors in the mixture. For example, in invertebrates collected from the two UEFPC sites, EFK 24.5 and EFK 23.4, both Aroclors 1254 and 1260 were detected, whereas only Aroclor 1260 was detected in the invertebrate samples from EFK 6.3. However, in contrast to the insects, Aroclor 1260 was dominant in EFK 23.4 spiders, whereas Aroclor 1254 and 1260 were present in similar concentrations at EFK 6.3 (Fig. 4.1, Table B.1) and in fish. All three Aroclors were detected in fish from EFK 24.5, only Aroclors 1254 and 1260 were detected in fish at EFK 23.4, and only Aroclor 1260 was detected at EFK 6.3.

4.2 MERCURY

Because MeHg is so bioaccumulative, typically >95% of the Hg_T found in fish fillets is MeHg. Total Hg, being easier and cheaper to measure, is thus often used as a proxy for MeHg in fish fillets. However, when considering biota at lower trophic levels to assess ecological risks, it is clear that the proportion of MeHg is much more variable depending both on the aqueous Hg_T concentrations and on the trophic position of the animal. In general, the greater the exposure to aqueous Hg_T, the lower the percent MeHg in the animal, and the higher the trophic level, the higher the MeHg concentration. Mercury concentrations in stonerollers were generally lower than in invertebrates across the ORR, likely because these forage fish are lower in the food chain, feeding more on periphyton and detritus on rocks than the invertebrates collected, many of which are predators.

These patterns are apparent in EFPC, which had by far the highest concentrations of Hg (both Hg_T and MeHg) in resident biota on the ORR (Fig. 4.2). Aqueous Hg_T concentrations were highest in UEFPC closer to the Y-12 National Security Complex (Y-12), where the source of Hg originates and decreases with distance downstream (Watson et al. 2016). Invertebrates and forage fish followed the same spatial patterns. Total Hg concentrations in caddisfly larvae were highest at EFK 24.5 (2.52 μg/g), decreased to 0.67 μg/g at EFK 23.4, and were 0.25 μg/g in damselfly and dragonfly larvae from EFK 6.3 (Fig. 4.2, B.2). Hg_T in fishing spiders was higher in UEFPC at EFK 23.4 (0.60 μg/g) than at EFK 6.3 (0.31 μg/g), and Hg_T in stonerollers decreased from 0.76 μg/g at EFK 24.5 to 0.40 μg/g at EFK 23.4 and to 0.17 μg/g at EFK 6.3. Mean Hg_T concentrations in forage fish collected in the two UEFPC sites were an order of magnitude higher than at any other sites in this study.

Methylmercury concentrations in biota collected from EFPC, as well as the proportion of MeHg, increased with distance downstream in invertebrates and in forage fish. Mean MeHg concentrations in stonerollers increased from 0.05–0.08 μg/g (6–20% MeHg) in UEFPC to 0.12 μg/g at EFK 6.3 (72% MeHg) and mean MeHg concentrations in aquatic insects increased from 0.03–0.04 μg/g in UEFPC (1–6% MeHg) to 0.24 μg/g further downstream at EFK 6.3 (96% MeHg). The fishing spiders collected for this study are predators and feed at a higher trophic level than the forage fish or the other invertebrates, and as a result, the %MeHg was much higher (72–74% in UEFPC and 96% in lower EFPC [LEFPC]). The concentration of Hg_T in spiders at the reference site, HCK 20.6, was dramatically lower than at EFPC (0.025 μg/g wet mass), whereas the proportion of MeHg was slightly higher (~81%).

Mean Hg_T concentrations in invertebrates collected in WOC were elevated with respect to reference sites, and they decreased from upstream to downstream from 0.15 μg/g at WCK 3.9 to 0.05 μg/g at WCK 2.3. Concentrations in invertebrates collected at MEK 0.2 (0.03 μg/g) in the WOC watershed were comparable to reference sites. The reported analytical results for invertebrate MeHg samples for both WOC sites and MEK 0.2 were higher than the concentrations reported for Hg_T. While the percentage of MeHg may near 100% in some animals under the right conditions (e.g., predators), MeHg concentrations exceeding total Hg concentrations is analytically impossible. In all likelihood, this discrepancy resulted

because separate aliquots from those samples were used for extracting Hg and MeHg, in which case, the differences were caused by variable distribution of Hg and/or MeHg in the samples. Even though total Hg and MeHg were over- or underestimated in those samples, based on results from all sites in this study, it is reasonable to assume that the majority (i.e., > 50%) of the Hg in the total Hg results for the affected invertebrate samples was MeHg. The concentration of Hg in spiders from WCK 2.3 was higher than in insects from the same site (0.087 µg/g wet mass and 0.053 µg/g wet mass, respectively) and more than 3 times higher than the concentration found in spiders at HCK 20.6. However, the concentration of Hg in spiders was substantially lower at WCK 2.3 than that at either EFK 23.4 or EFK 6.3 (0.597 µg/g wet mass and 0.251 µg/g wet mass, respectively). Concentrations and the spatial pattern of whole body Hg_T and MeHg in fish from the WOC watershed were similar to those found for invertebrates, although the proportion of MeHg was higher in invertebrates (Table B.2, Fig. 4.2). Hg_T concentrations in fish at MEK 0.2 were similar to concentrations in fish from the reference site HCK 20.6.

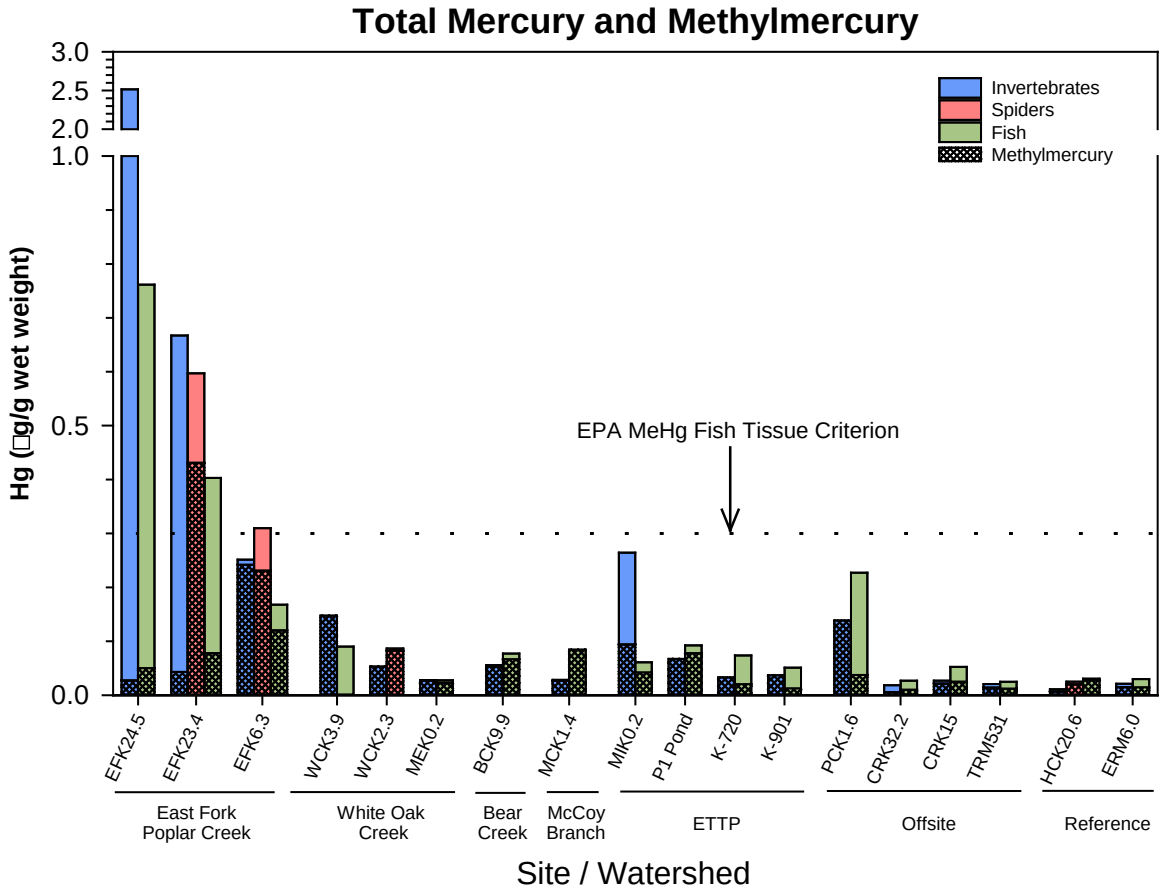

Fig. 4.2. Total mercury (HgT) and methyl mercury (MeHg) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Each bar represents means for Hg_T (color-coded by biota group), and the mean concentration of MeHg comprising total concentration is represented by black hashes for each bar. The dashed line represents the Environmental Protection Agency's guideline for MeHg in fish tissue (0.3 µg/g). Note the change in scale after the break in the y-axis.

Across the rest of the ORR, Hg_T concentrations in biota were much lower than those in EFPC (and in most cases were lower than WOC), and the %MeHg was much higher, ranging from a minimum of 36% in snails collected from MIK 0.2 to 100% in samples across the ORR. Incidentally, the snails collected at MIK 0.2, which had the lowest percentage of MeHg of all samples outside of EFPC, also had the highest

Hg_T concentrations of all the biota outside of EFPC (0.26 μg/g). Concentrations of Hg_T were only moderately elevated in invertebrates and fish from BCK 9.9 and MCK 1.4 compared with the reference site HCK 20.6 (Table B.2, Fig. 4.2), while the percentage of MeHg was high at both sites (87–95%). Species differences likely played an important part in the differences observed in the percentage of MeHg in invertebrates from MIK 0.2 and the other two streams. Snails, which graze on periphyton (and are largely herbivores), were used for Hg/MeHg analyses for MIK 0.2, while insects from the order Megaloptera (fishfly larvae), which are predators feeding at a higher level in the food chain, were used for Hg/MeHg analysis for the other two sites; a higher proportion of MeHg would be expected in predators due to their higher trophic level.

Mean Hg_T concentrations in the ETTP ponds were relatively low and were only slightly elevated with respect to the reference sites. Mean Hg_T concentrations in adult damselflies/dragonflies from all three ETTP ponds ranged from 0.03 μg/g at the K-720 Slough to 0.07 μg/g at the K-1007-P1 Pond (Table B.2, Fig. 4.2). Mean Hg_T concentrations in whole body fish were slightly elevated with respect to the reference sites, ranging from 0.05 μg/g in gizzard shad collected from the K-901-A Pond to 0.1 μg/g in bluegill from the K-1007-P1 Pond, compared with 0.03 μg/g at the reference sites. Methylmercury accounted for 65–71% of the Hg_T in invertebrates collected from the reference sites. As found in some invertebrate samples from the WOC watershed (see above results), estimates of %MeHg at the three ETTP ponds were ≥100%. Methylmercury concentrations in fish ranged between 0.013 and 0.067 μg/g wet mass, but these concentrations accounted for 26% of Hg_T in gizzard shad from K-901-A Pond and 67% of Hg_T in bluegill from K-1007-P1 Pond. This difference in the relative MeHg content may be due to species differences because gizzard shad and bluegill have different dietary preferences. Methylmercury concentrations were higher in invertebrates than in fish in all three ETTP ponds.

Mean Hg_T concentrations in biota collected off-site from PCK 1.6 (0.14–0.23 μg/g) were comparable to concentrations seen in LEFPC (0.2–0.25 μg/g), but concentrations decreased with increasing distance downstream from the ORR so that concentrations at all other off-site locations were comparable to the reference sites (0.01–0.03 μg/g). The highest concentration of Hg_T found in invertebrates at an off-site location was at PCK 1.6 (0.139 μg/g wet mass; Table B.2; Fig. 4.2), and as with many sites mentioned previously, the analytical results for invertebrates at PCK 1.6 indicated that 100% of the Hg_T was MeHg, but as noted in previous sections above, this is likely due to different aliquots used for the two analyses. However, it does suggest that a relatively large proportion of the Hg_T in the damselflies from that site is in the form of MeHg (Table B.2, Fig. 4.2). Concentrations of Hg_T in invertebrates at other off-site locations were much lower (0.018–0.027 μg/g wet mass), and the proportion of MeHg ranged from 32 to 80%. The highest Hg_T and MeHg concentrations in fish at off-site locations were at PCK 1.6 (0.227 and 0.038 μg/g wet mass, respectively; Table B.2, Fig. 4.2). Concentrations in fish from the Clinch and Tennessee Rivers were slightly higher or comparable to concentrations observed at reference sites.

4.3 METALS

4.3.1 Antimony

Antimony (Sb) was at or below detection limits in most samples, and where it was detected, concentrations were low, with no apparent spatial or species-specific trends (Table B.3). Antimony concentrations in biota collected on and downstream of the ORR were comparably low, and in many cases, they were lower than concentrations in biota collected from the reference sites. Antimony was generally highest in biota collected in the WOC watershed, where it was detected in all biota samples; all other watersheds had samples below detection limits. The highest mean Sb concentration was measured in stonerollers collected from MEK 0.2 (0.034 μg/g), which was higher than the concentration in stonerollers collected at the HCK 20.6 reference site, but comparable to the mean concentration in gizzard

shad collected at the ERM 6.0 reference site (0.031 µg/g). Spiders collected at WCK 2.3 were the only spiders with measurable Sb concentrations (mean 0.02 µg/g). With the exception of snails collected at CRK 32.2 (mean concentration 0.01 µg/g) and gizzard shad collected at TRM 531 (mean concentration 0.008 µg/g), all other biota samples collected from off-site locations had Sb concentrations below detection limits. Emergent insects collected at the ETTP ponds all had detectable Sb concentrations (ranging from 0.011 to 0.015 µg/g), whereas Sb concentrations were below detection limits at reference locations.

4.3.2 Arsenic

Mean arsenic (As) concentrations were generally higher in invertebrates than in fish or spiders, with the exceptions of the three ETTP ponds and most off-site locations (Fig. 4.3, Table B.3). The sites with the highest As concentrations in invertebrates were WCK 3.9, MCK 1.4, MIK 0.2, and CRK 32.2. The elevated As concentrations (2.43 µg/g) in caddisfly larvae collected at MCK 1.4 are likely due to elevated exposure concentrations. McCoy Branch and Rogers Quarry have historically been impacted by coal ash deposits from Y-12 operations, and As is one of the most important coal ash–associated contaminants. The elevated As concentrations in invertebrates at the other three sites (1.7 µg/g at WCK 3.9, 3.4 µg/g at MIK 0.2, and 2.77 at CRK 32.2) may be due to the species collected; these were the only three sites where snails were used as the representative invertebrates in this study.

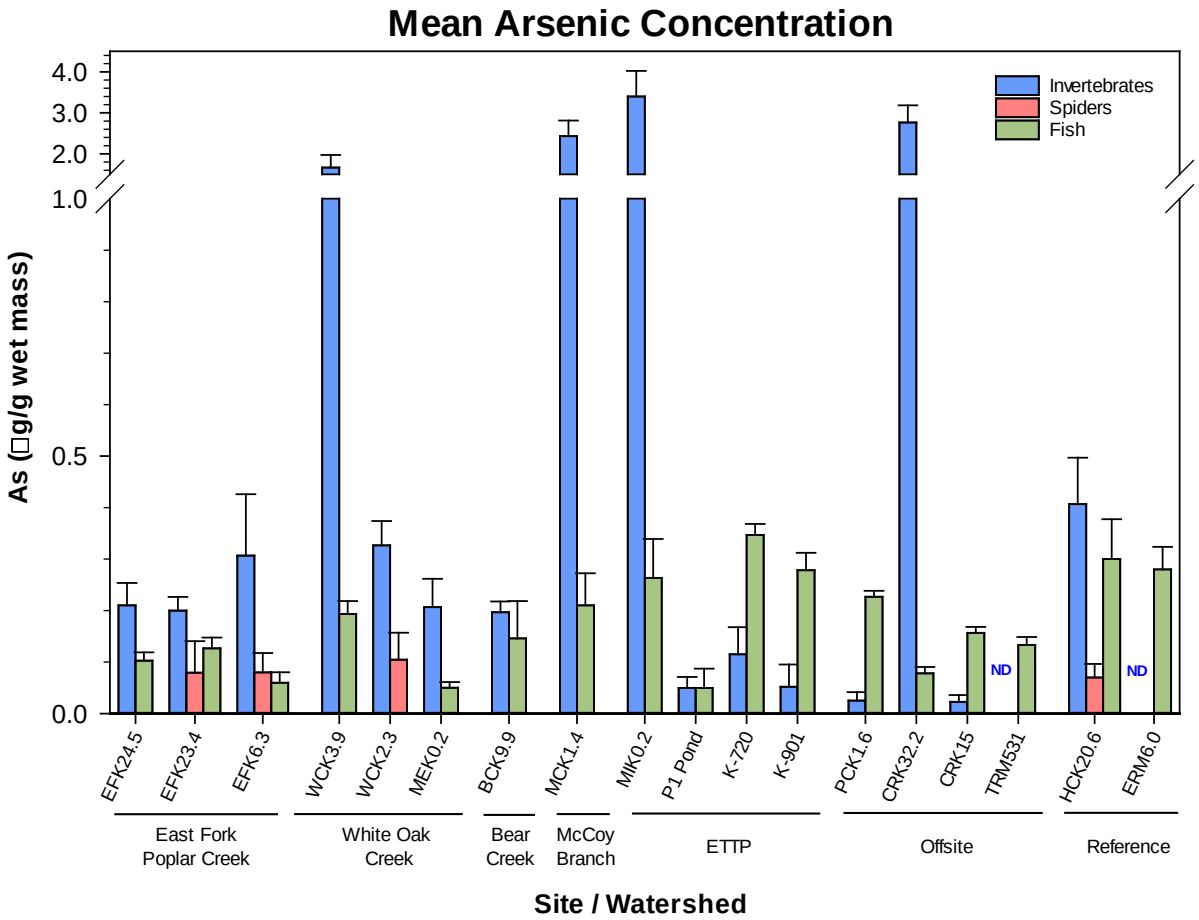

Fig. 4.3. Total arsenic (As) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ±1 standard deviation and color-coded by biota grouping. ND = not detected and color-coded by biota grouping.

Concentrations across all other sites on the ORR and off-site were comparable or lower than those measured in biota collected at the reference sites. In particular, at sites where emergent insects (adults) were collected, As concentrations were particularly low: 0.05 µg/g in dragonfly adults at K-1007-P1 Pond, 0.12 µg/g in dragonfly adults in K-720 Slough, 0.05 µg/g in damselfly adults at the K-901-A Pond, 0.02 µg/g in mayfly adults at CRK 15, 0.03 µg/g in damselfly adults at PCK 1.6, and below detection limits in mayfly adults collected from TRM 531 and the ERM 6.0 reference site. Arsenic concentrations in fish were generally lower at sites where stonerollers or bluegill were collected and higher where gizzard shad were collected. Mean As concentrations in stonerollers were < 0.13 µg/g in EFPC, < 0.2 µg/g in the WOC watershed, and < 0.3 µg/g in Mitchell Branch, all lower than the mean concentration in stonerollers collected at the HCK 20.6 reference site (0.3 µg/g). Mean As concentrations in gizzard shad were < 0.3 µg/g at the K-901-A Pond and PCK 1.6, < 0.2 µg/g at CRK 15, < 0.1 µg/g at CRK 32.2, and 0.13 at TRM 531, with all being similar to the concentrations in gizzard shad from the ERM 6.0 reference site (0.28 µg/g).

4.3.3 Beryllium

Beryllium (Be) was at or below detection limits in most samples, and where it was detected, the concentrations were low and exhibited no apparent spatial or species-specific trends (Table B.3). The highest mean Be concentration was in caddisfly larvae from MCK 1.4 (0.033 µg/g); Be was not detectable in blacknose dace collected at this site. Beryllium was also detected in snails collected at CRK 32.2 (0.016 µg/g) and in stonerollers at EFK 24.5 (0.005 µg/g) and WCK 3.9 (0.014 µg/g), but concentrations were low and comparable to concentrations measured in stonerollers at the HCK 20.6 reference site. Beryllium was also detected in gizzard shad collected from the K-720 Slough (0.011 µg/g) and at off-site locations PCK 1.6 (0.014 µg/g) and TRM 531 (0.005 µg/g), but concentrations were comparable to those measured in the same species from the ERM 6.0 reference site.

4.3.4 Cadmium

By far, the highest cadmium (Cd) concentrations in this study were observed in spiders. Mean Cd concentrations in spiders from EFPC (2.56 µg/g at EFK 23.4 and 3.26 µg/g at EFK 6.3) were about an order of magnitude higher than concentrations in spiders at the HCK 20.6 reference site (0.30 µg/g) and were orders of magnitude higher than concentrations seen in fish and invertebrates at most other sites on the ORR (Fig. 4.4). Mean Cd concentrations in spiders collected at WCK 2.3 (0.75 µg/g) were lower than in EFPC, but they were still elevated with respect to spiders at the reference site. As for many other contaminants, the highest Cd concentrations in (non-spider) invertebrate samples were seen at sites where snails were the representative invertebrate collected: 0.50 µg/g at MIK 0.2, 0.34 µg/g at WCK 3.9, and 0.24 µg/g at CRK 32.2. Except for caddisfly larvae from BCK 9.9 (0.35 µg/g), invertebrates at all other sites had Cd concentrations < 0.20 µg/g. Stonerollers collected at BCK 9.9 had the highest Cd concentrations in fish in this study (0.21 µg/g). All other fish collected on or off the ORR had concentrations < 0.04 µg/g, and many sites had Cd concentrations below detection limits: MEK 0.2, K-1007-P1 Pond, K-901-A Pond, CRK 32.2, CRK 15, and the HCK 20.6 reference site.

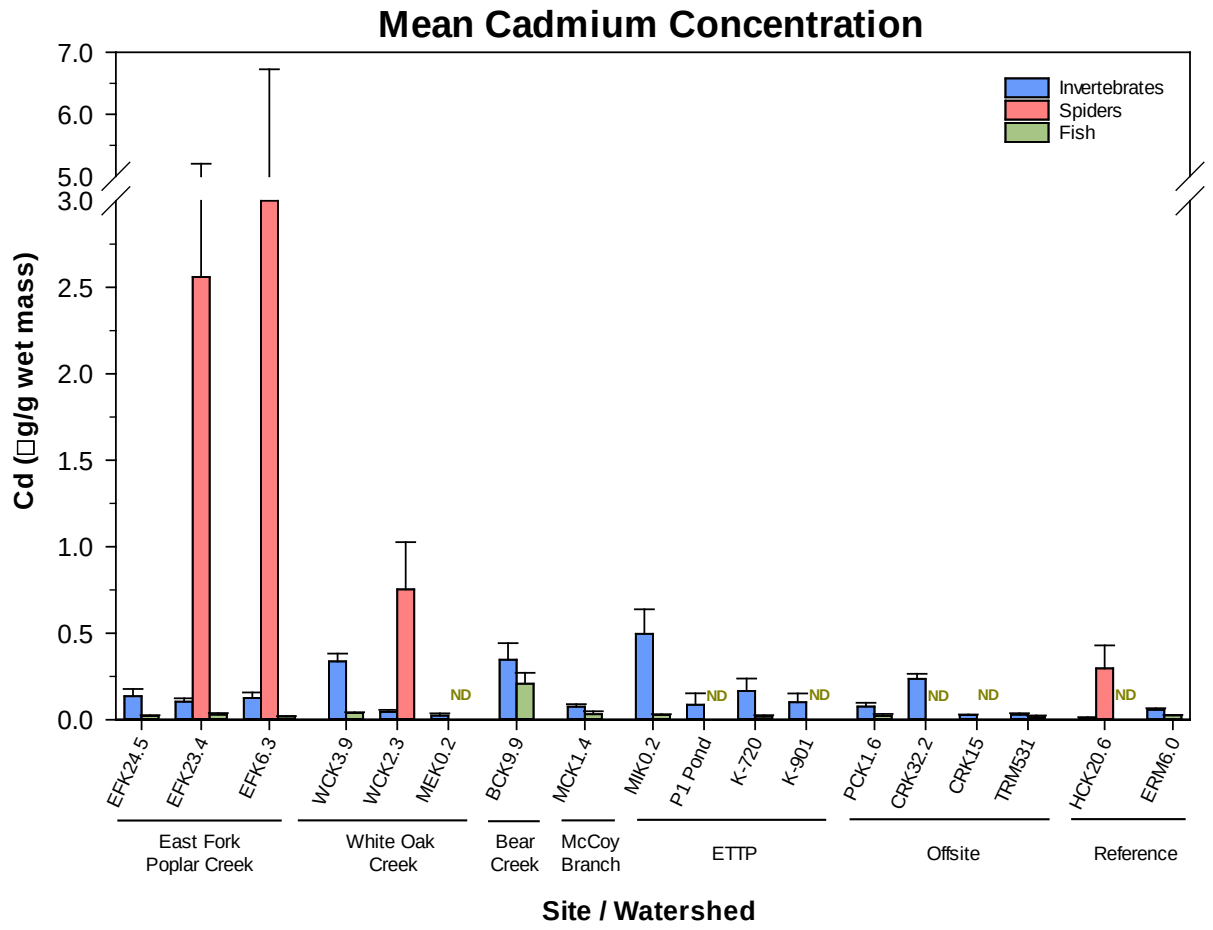

Fig. 4.4. Total cadmium (Cd) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ±1 standard deviation. ND = not detected and color-coded by biota grouping.

4.3.5 Chromium

Chromium (Cr) concentrations in biota were variable by site and species (Fig. 4.5, Table B.3). The highest concentration of Cr in invertebrates was found in snails from MIK 0.2 (2.3 µg/g). Other than modestly elevated concentrations of Cr in invertebrates from WCK 3.9, MEK 0.2, MCK 1.4, and CRK 32.2, concentrations in invertebrates from other sites and reference sites were comparable. The highest mean Cr concentrations were found in gizzard shad from the K-901-A Pond (7.37 µg/g), where concentrations were over threefold higher than concentrations in the same species collected from the ERM 6.0 reference site (1.97 µg/g). All other gizzard shad collected on and off the ORR had lower Cr concentrations than the reference fish, ranging from a low of 0.1 µg/g at CRK 32.2 to 0.86 µg/g at PCK 1.6. In general, Cr concentrations in gizzard shad appear to be higher than in stonerollers, as the mean concentration in stonerollers collected from the HCK 20.6 reference site (0.55 µg/g) was > threefold lower than the mean concentration in gizzard shad from the ERM 6.0 reference site. Chromium concentrations in stonerollers appeared to increase with distance downstream in EFPC, so that concentrations in UEFPC (~0.2–0.4 µg/g) were comparable to or lower than the concentration seen in stonerollers from the reference site, but the mean concentration in stonerollers from EFK 6.3 was elevated (1.33 µg/g) with respect to reference fish.

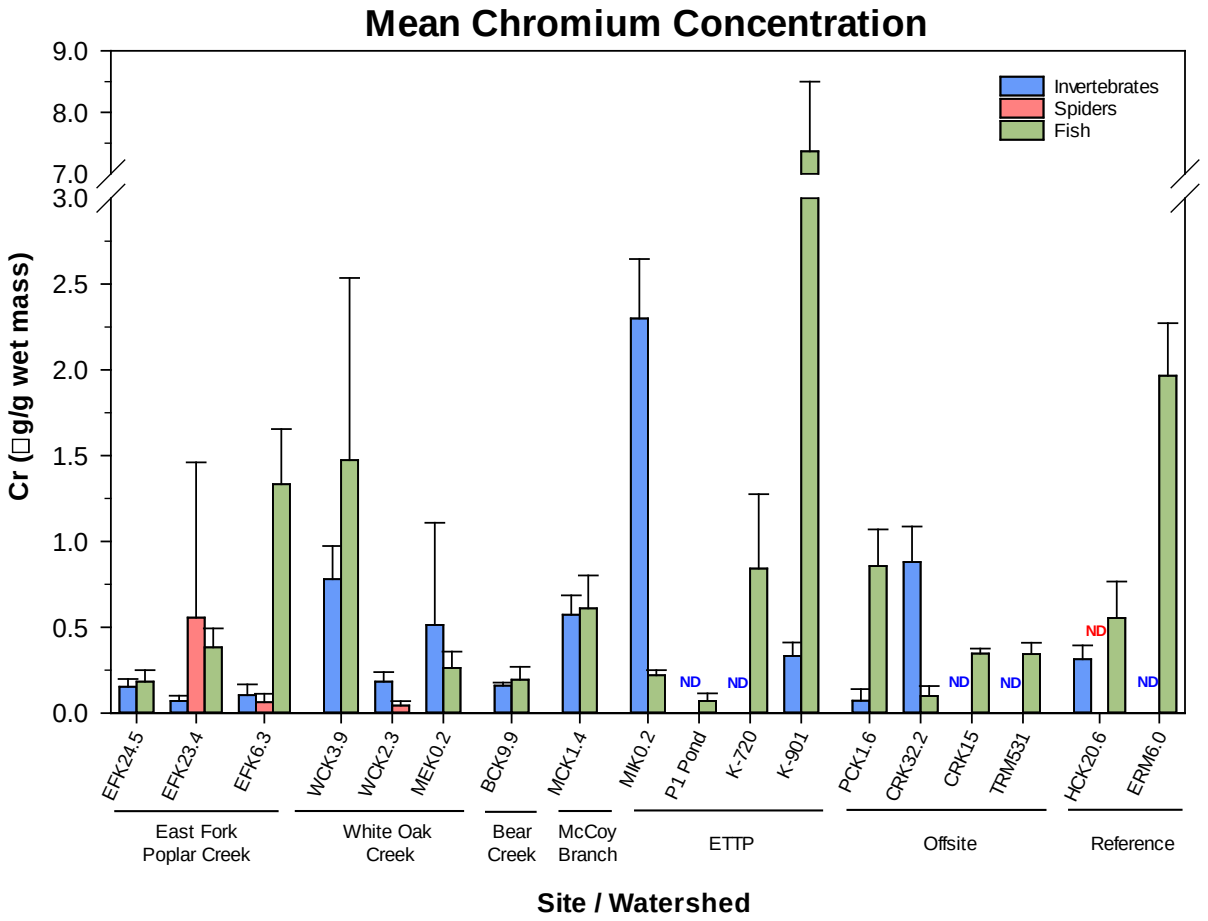

Fig. 4.5. Total chromium (Cr) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ± 1 standard deviation. ND = not detected and color-coded by biota grouping.

4.3.6 Copper

At all sites, mean copper (Cu) concentrations were higher in invertebrates than in fish, and of the invertebrate groups, concentrations were higher in spiders were than in insects from the same site (Fig. 4.6, Table B.3). Copper concentrations in spiders were highest at EFK 6.3 (mean 47.7 µg/g); Cu concentrations in spiders from EFK 23.4 (27.3 µg/g) and WCK 2.3 (25.3 µg/g) were comparable to the concentrations in spiders from HCK 20.6 (26.7 µg/g).

As with many other metals, the highest Cu concentrations in invertebrates were found in snails and spiders (snails: 44.7 µg/g at WCK 3.9, 29.3 µg/g at MIK 0.2, and 22.7 µg/g at CRK 32.2; spiders: 25.3 µg/g at WCK 2.3, 26.7 µg/g at HCK 20.6, 27.3 µg/g at EFK 23.4, and 47.67 µg/g at EFK 6.3). At sites where the representative invertebrate taxa were insects, Cu concentrations were generally higher in emergent insects than in immature insects (i.e., larvae and nymphs) at all biota sites including references. Compared with emergent insects from ERM 6.0, Cu concentrations in emergent insects from ETTP ponds were slightly elevated (range of 8.6 µg/g in adult damselflies at the K-901-A Pond to 12.7 µg/g in adult dragonflies at K-720 Slough compared with 6.8 µg/g in mayfly adults from ERM 6.0).

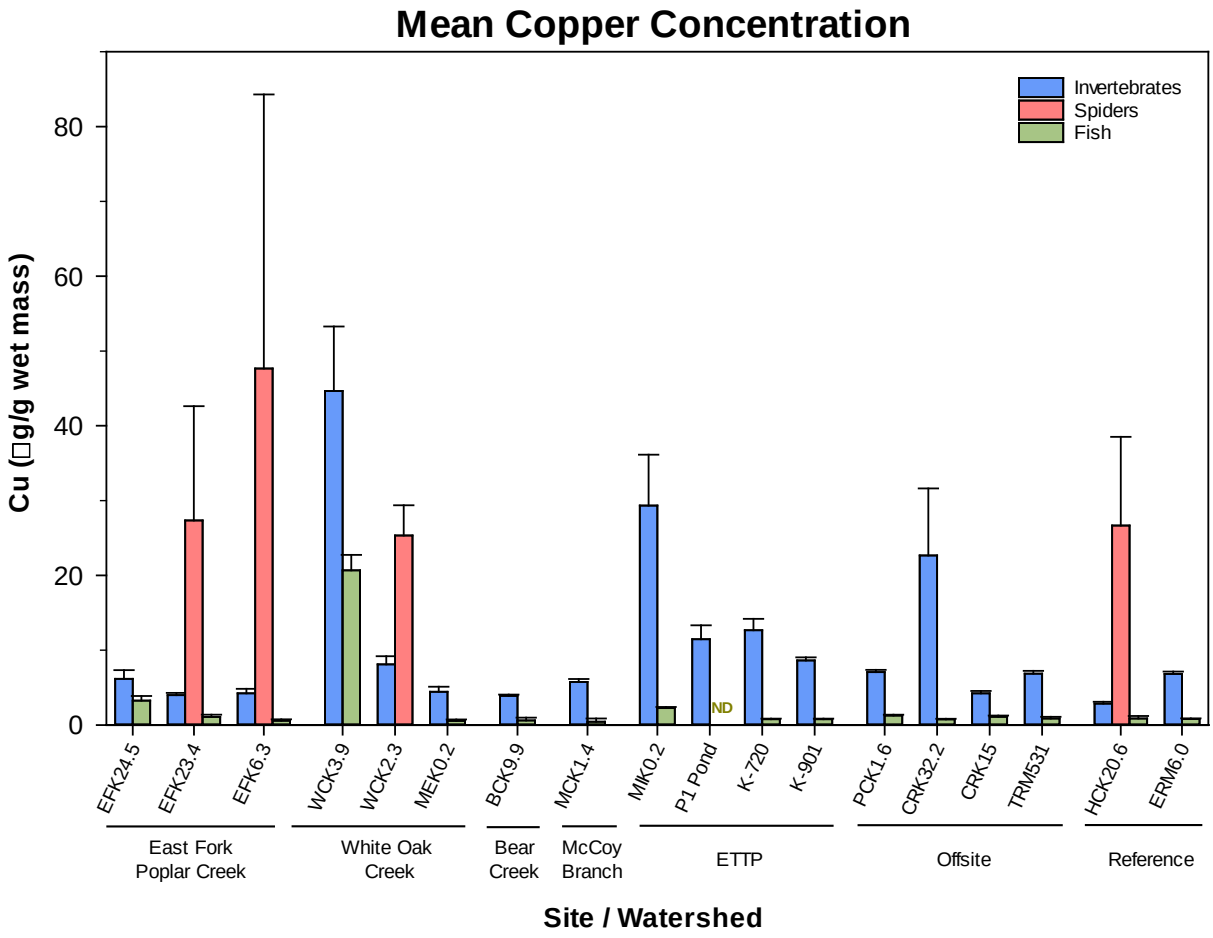

Fig. 4.6. Total copper (Cu) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ± 1 standard deviation. ND = not detected and color-coded by biota grouping.

Similar to invertebrates, Cu concentration were highest in fish from WCK 3.9 (mean concentration 20.7 µg/g in stonerollers compared with 0.9 µg/g in stonerollers collected from the HCK 20.6 reference site). Copper concentrations in stonerollers collected from EFPC were highest at EFK 24.5 (3.3 µg/g), decreasing with distance downstream to concentrations similar to those in the same species at the reference site (1.1 µg/g at EFK 23.4 and 0.6 µg/g at EFK 6.3). Copper concentrations in fish at all other sites were comparable to reference site concentrations.

4.3.7 Lithium

Lithium (Li) was at or below detection limits in most samples, and where it was detected, concentrations were low with no apparent spatial or species-specific trends (Table B.3). The highest mean Li concentration was observed in caddisfly larvae collected at MCK 1.4 (0.8 µg/g), where concentrations were about double those seen in various insect larvae collected at the HCK 20.6 reference site (0.25 µg/g). Lithium concentrations in spiders were below detection limits at all sites except at EFK 23.4, where the mean concentration was 0.14 µg/g. Lithium was also below detection limits in all samples composed of emergent insects. Lithium concentrations in fish collected on and downstream of the ORR were lower than in fish collected from the reference sites (0.44 µg/g in stonerollers at HCK 20.6 and 0.38 µg/g in gizzard shad at ERM 6.0).

4.3.8 Lead

Mean lead (Pb) concentrations were generally higher in invertebrates than in fish, except where emergent insects were collected as the representative invertebrate (Fig. 4.7, Table B.3). Lead concentrations in emergent insects from the K-1007-P1 Pond, CRK 15, TRM 531, and the ERM 6.0 reference site were below detection limits. Lead was detected in adult dragonflies from the K-720 Slough (0.04 µg/g), in damselflies collected from the K-901-A Pond (0.03 µg/g), and in damselflies from PCK 1.6 (0.03 µg/g), but concentrations were low. Lead concentrations in spiders at all sites were low and comparable to concentrations measured in emergent insects.

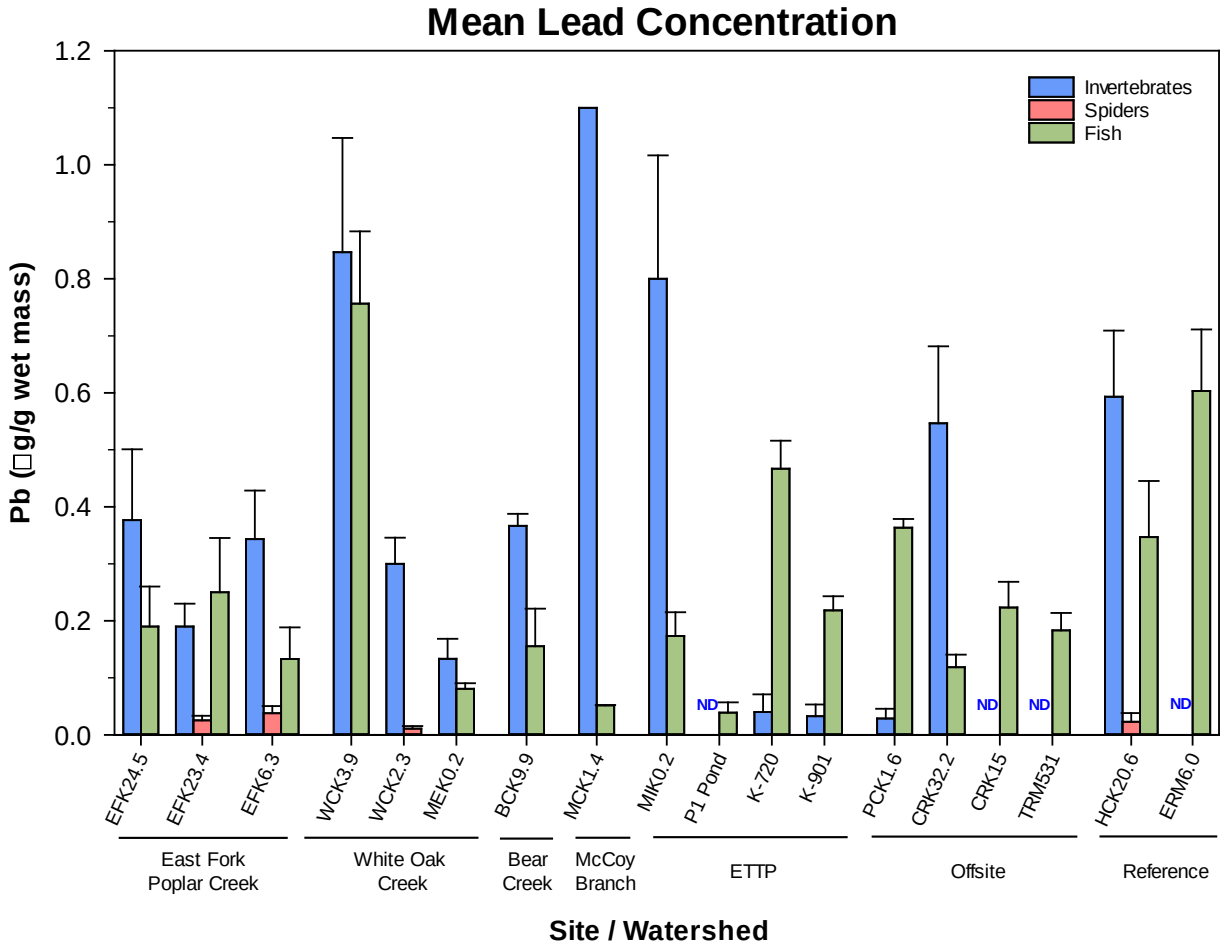

Fig. 4.7. Total lead (Pb) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ± 1 standard deviation. ND = not detected and color-coded by biota grouping.

In invertebrates, spatial patterns in Pb concentrations were similar to those for As, with the highest concentrations at MCK 1.4 and the three sites where snails were analyzed: WCK 3.9, MIK 0.2, and CRK 32.2. Lead concentrations were highest in caddisfly larvae from MCK 1.4 (1.1 µg/g), but concentrations in blacknose dace from the same site were very low (0.052 µg/g) with respect to concentrations in stonerollers from the HCK 20.6 reference site (0.35 µg/g). Lead concentrations in both snails (0.85 µg/g) and stonerollers (0.76 µg/g) from WCK 3.9 were elevated with respect to reference site concentrations. Lead concentrations in snails from CRK 32.2 (0.55 µg/g) were lower than those in snails

collected at sites on the ORR. Concentrations in insect larvae from the streams on the ORR were lower than those in insect larvae from HCK 20.6.

The highest Pb concentrations in fish were found in stonerollers from WCK 3.9. Lead concentrations in stonerollers collected at all other sites on the ORR were lower than those in the same species from HCK 20.6. Lead concentrations in gizzard shad collected at all sites on and downstream of the ORR were lower than mean Pb concentrations in the same species from ERM 6.0 (mean 0.6 µg/g).

4.3.9 Molybdenum

Molybdenum (Mo) concentrations were generally low (and in many cases below detection limits; Table B.3, Fig. 4.8). However, concentrations were elevated in insect larvae collected at EFK 24.5 (mean 0.78 µg/g), BCK 9.9 (mean 0.42 µg/g), and MCK 1.4 (mean 0.63 µg/g). Concentrations in fish collected from these three sites and most other sites in this study were low (≤ 0.07 µg/g), and comparable to those measured in fish from reference sites (0.07 µg/g); concentrations in fish only from EFK 23.4 (0.095 µg/g) and WCK 3.9 (0.109 µg/g) were slightly elevated relative to reference sites. Concentrations in snails from MIK 0.2, WCK 3.9, and CRK 32.2 were slightly elevated with respect to concentrations in other invertebrates from the reference sites. Molybdenum concentrations were slightly elevated in spiders from EFK 23.4 (mean 0.2 µg/g); all other spiders had Mo concentrations comparable to those at HCK 20.6 (0.06 µg/g).

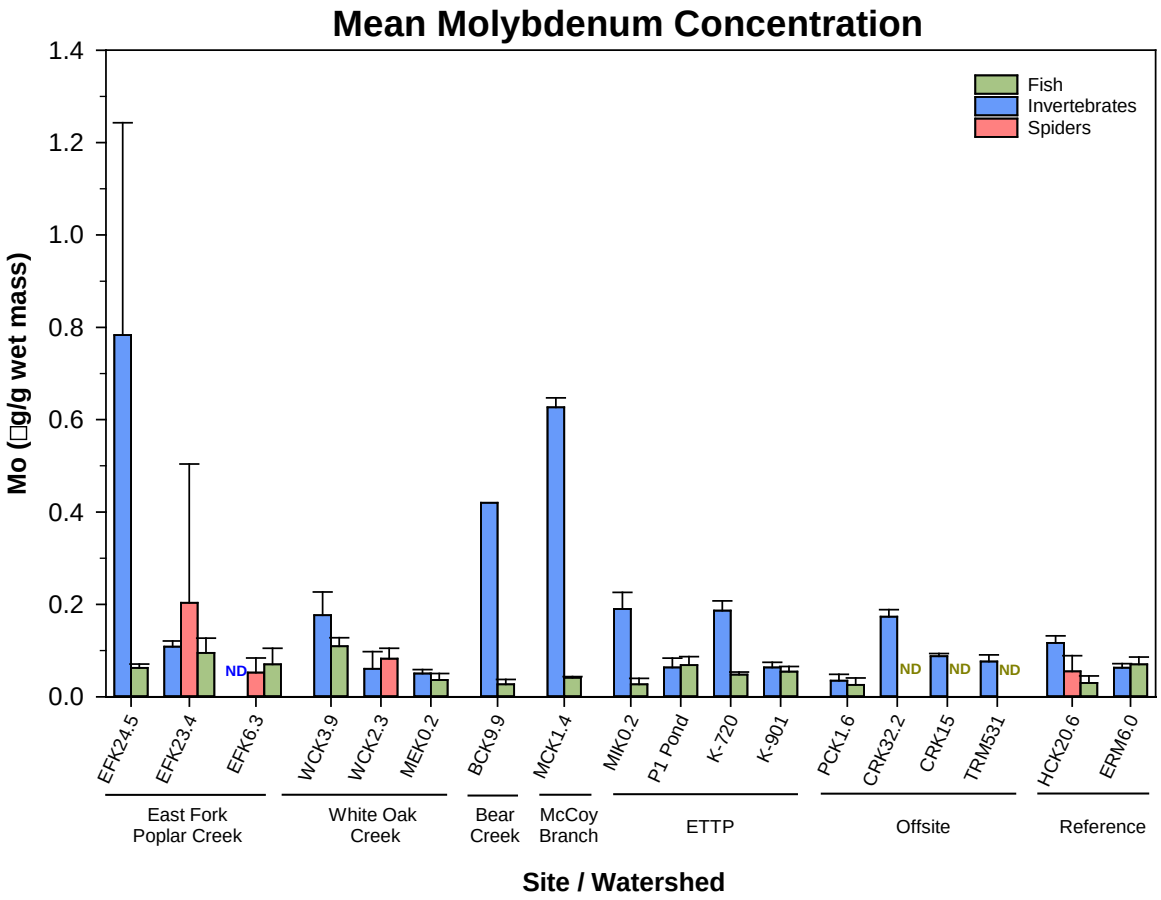

Fig. 4.8. Total molybdenum (Mo) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ± 1 standard deviation. ND = not detected and color-coded by biota grouping.

4.3.10 Nickel

Nickel (Ni) concentrations were generally low in all species and at all sites, with many samples having concentrations below the detection limits (Table B.3, Fig. 4.9). However, snails from MIK 0.2 (mean concentration 18.0 µg/g) and CRK 32.2 (mean concentration 5.1 µg/g) had Ni concentrations orders of magnitude higher than those in other biota at other sites. Notably, Ni concentrations in snails from WCK 3.9 (mean concentration 0.88 µg/g) were not similarly elevated. Nickel concentrations in fish were remarkably similar across all species and at all sites both on and off the ORR. Nickel concentrations were below detection limits in all emergent insects. The mean Ni concentration in spiders from EFK 23.4 was low (0.55 µg/g) and comparable to Ni concentrations in fish and insect larvae from the same site. Nickel concentrations at the other three sites were below detection limits.

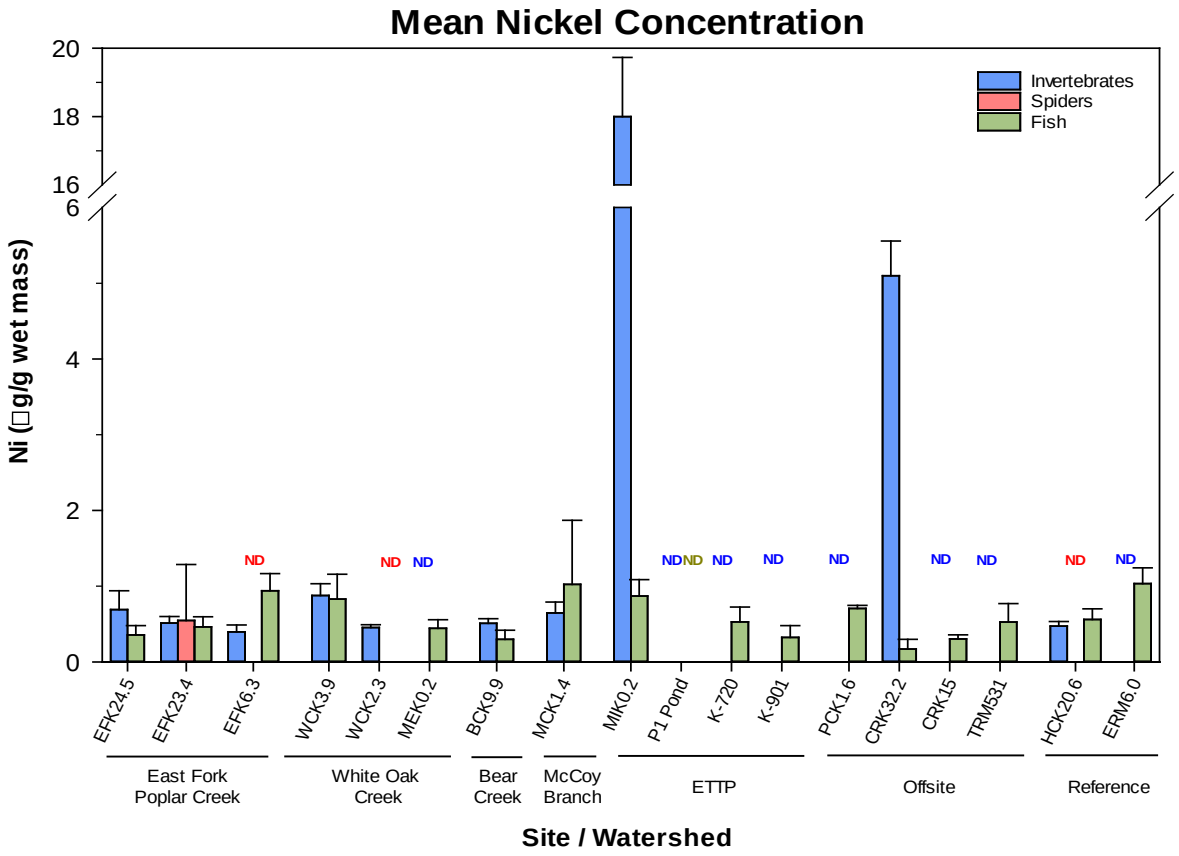

Fig. 4.9. Total nickel (Ni) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ± 1 standard deviation. ND = not detected and color-coded by biota grouping.

4.3.11 Selenium

The highest Selenium (Se) concentrations in both invertebrates and fish were found in the snails (mean concentration 1.4 µg/g) and blacknose dace (1.4 µg/g) from MCK 1.4 (Table B.3, Fig. 4.10). Like As, Se is a common contaminant in fly ash; thus, this was likely related to the historical discharges of fly ash into the McCoy Branch watershed. The Se concentrations in biota at MCK 1.4 were elevated with respect to biota from the reference sites (ranging from 0.47 to 0.8 µg/g, depending on the site and species). Selenium concentrations at all other sites on and downstream of the ORR were comparable to those in biota from the reference sites. Selenium concentrations were generally comparable between invertebrates

and fish from the same site except for spiders and snails, which had elevated concentrations with respect to other biota. The mean Se concentration in spiders from EFK 6.3 was 0.88 µg/g compared with ~0.48 µg/g in both insect larvae and fish from this site. The mean Se concentration in spiders from WCK 2.3 was 0.97 µg/g, compared with 0.47 µg/g in insect larvae from the same site. Snails at MIK 0.2 (mean concentration 0.68 µg/g) and CRK 32.2 (mean concentration 0.84 µg/g) had higher Se concentrations than fish from those sites.

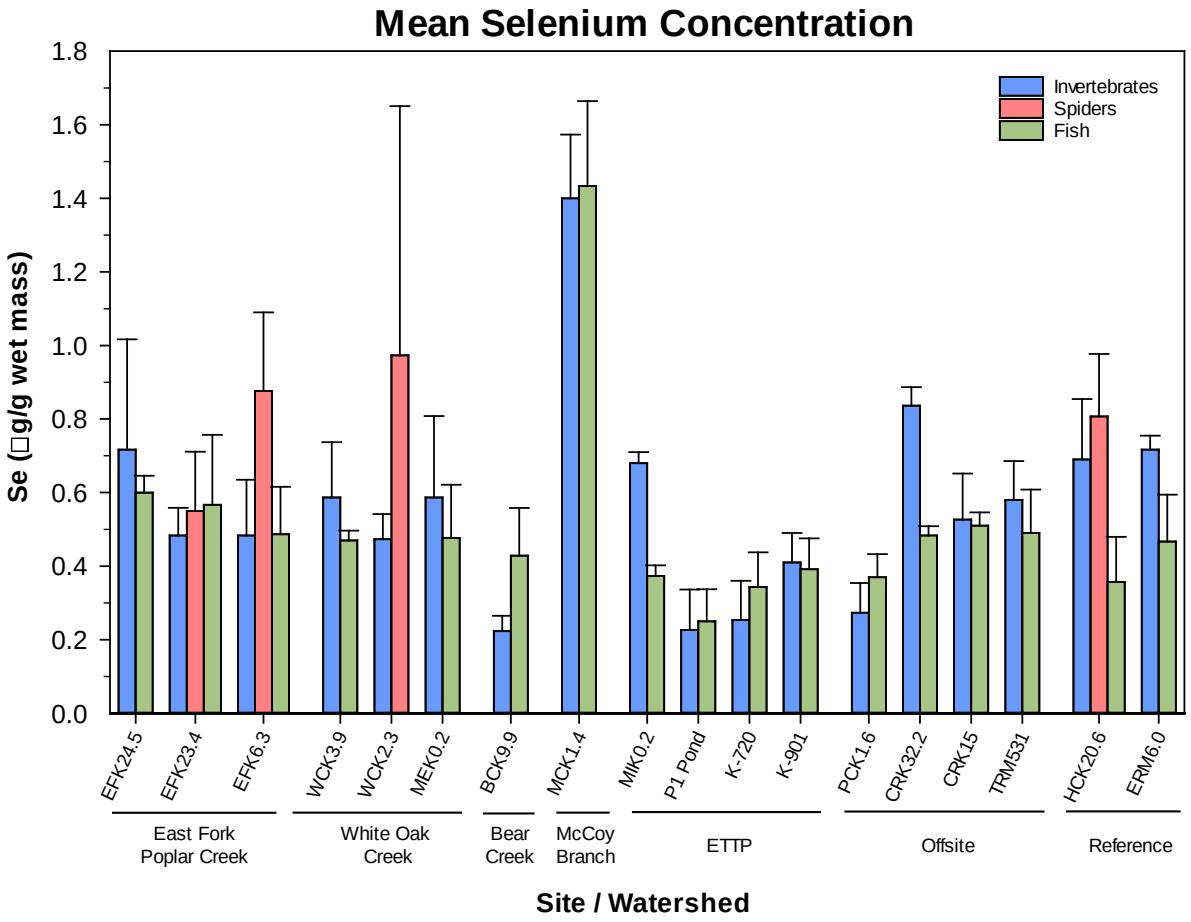

Fig. 4.10. Total selenium (Se) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ± 1 standard deviation.

4.3.12 Thallium

Thallium (Tl) concentrations were at or below detection limits in most samples, and where detected, concentrations were low with no apparent spatial or species-specific trends (Table B.3). The highest mean Tl concentration was found in dragonflies from the K-720 Slough (0.04 µg/g), where concentrations were an order of magnitude higher than those in biota from the reference sites (<0.008 µg/g), as well as in most other biota in this study. Thallium concentrations in gizzard shad collected from the K-720 Slough were also relatively elevated (mean concentration 0.02 µg/g) compared to reference site biota. As with many other metals, Tl concentrations were elevated in invertebrates from MIK 0.2 and CRK 32.2 (mean concentration 0.03 µg/g at both sites), but this may be due in part to interspecies differences in metal accumulation, because snails were collected as the representative invertebrate species at these two sites.

4.3.13 Uranium

Uranium (U) was at or below detection limits in most samples, and where U was detected, concentrations were low and showed no apparent spatial or species-specific trends (Table B.3; Fig. 4.11). The highest mean U concentration (0.97 µg/g) was found in snails from MIK 0.2. The other two sites where snails were collected, WCK 3.9 and CRK 32.2 (mean U concentration 0.12 µg/g at both sites), had slightly elevated concentrations in invertebrates, but the concentrations were about an order of magnitude less than at MIK 0.2. Uranium concentrations in fish from these three sites were low (mean concentration ~0.03 µg/g each at MIK 0.2, WCK 3.9, and CRK 32.2). Uranium concentrations were also elevated in both caddisfly larvae (0.75 µg/g) and stonerollers (0.28 µg/g) from BCK 9.9, which was the only site where U concentrations in fish were elevated with respect to the reference site. Uranium was not detected in emergent insects and was below detection limits in insect larvae from most other sites except those in EFPC, where concentrations were highest at EFK 24.5 (0.12 µg/g) and decreased with distance downstream (0.05 µg/g at EFK 23.4 and 0.03 µg/g at EFK 6.3). The same trends were found in stonerollers from EFPC, where concentrations were highest at EFK 24.5 (0.13 µg/g) and decreased with distance downstream (0.11 µg/g at EFK 23.4 and 0.04 µg/g at EFK 6.3). Uranium concentrations in spiders were low at all ORR sites (<0.03 µg/g) but were below detection limits at HCK 20.6.

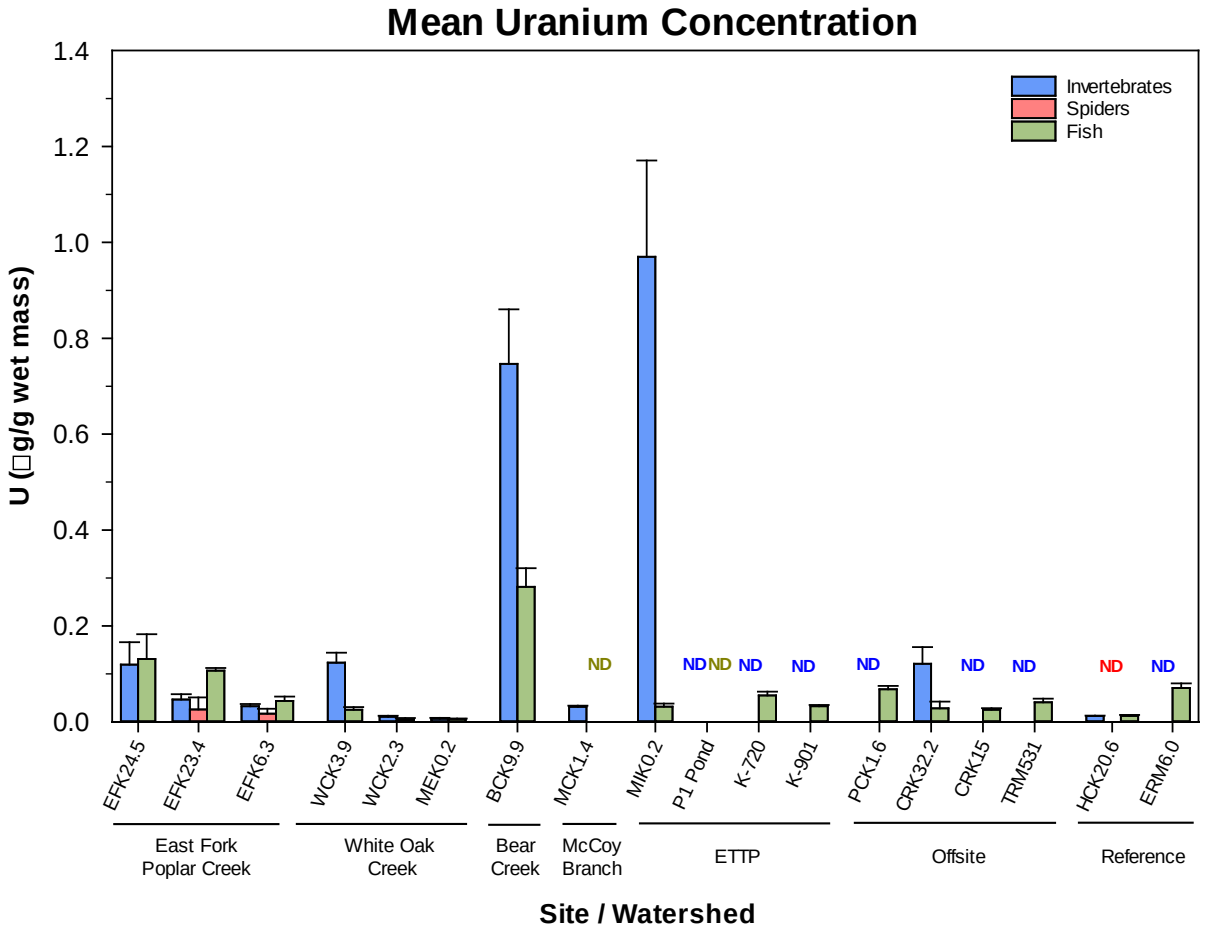

Fig. 4.11. Total uranium (U) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ± 1 standard deviation. ND = not detected and color-coded by biota grouping.

4.3.14 Zinc

Zinc (Zn) concentrations on and downstream of the ORR were generally comparable to Zn concentrations in biota collected from reference sites (Table B.3, Fig. 4.12). Zinc concentrations were highest in spiders, both on the ORR (mean concentrations ranging from 88.3 µg/g at EFK 23.4 to 95.0 µg/g at WCK 2.3) and at the HCK 20.6 reference site (mean concentration 75.3 µg/g). The highest Zn concentrations in other invertebrates were found in caddisfly larvae from BCK 9.9 (mean concentration 63.0 µg/g). In EFPC, zinc concentrations in invertebrates followed a spatial pattern similar to other metals in that concentrations in invertebrates were highest upstream (mean 55.3 µg/g at EFK 24.5) and decreased with distance downstream (mean 40.0 µg/g at EFK 23.4 and 29.0 µg/g at EFK 6.3). Most often, Zn concentrations in invertebrates were comparable or slightly higher in invertebrates than in fish collected from the same site. This was especially true of the ETTP ponds and off-site locations where samples of emergent insects and gizzard shad were analyzed. Emergent insects had lower Zn concentrations at both off-site locations as well as at the ERM 6.0 reference site than insect larvae on the ORR and at the HCK 20.6 reference site. The same was true of gizzard shad from off-site locations and ERM 6.0 where concentrations lower than those in stonerollers collected on the ORR and HCK 20.6.

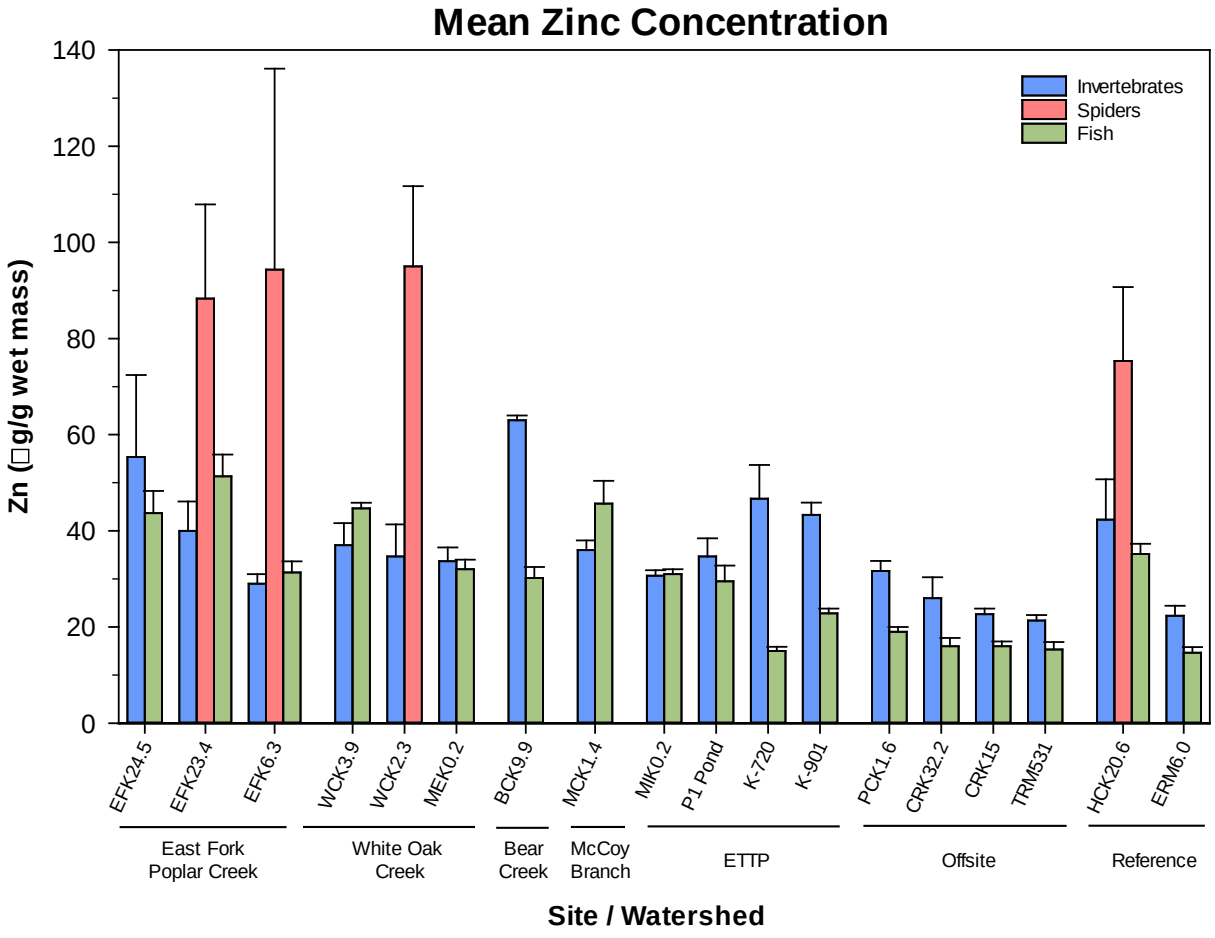

Fig. 4.12. Total zinc (Zn) concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. Values are means ± 1 standard deviation.

4.4 DIOXINS AND FURANS

Mean concentrations of dioxins and furans were highest in whole body gizzard shad from WCK 1.5 (Table B.4). Concentrations of all the dioxin and furan compounds were higher in whole body gizzard shad than in fillets of largemouth bass from the same site. The difference between whole body fish and fish fillets in concentrations could be due in part to differences in the lipid content in the different tissue types because these compounds are lipophilic. The lipid content was higher in whole body gizzard shad (averaging 2.17%) than in largemouth bass fillets (averaging 0.78%). For comparison, the average lipid content in gizzard shad from ERM 6.0 was 2.67%, whereas that in stonerollers from MEK 0.2 was 9.97%. Concentrations of dioxin and furan compounds in stonerollers from MEK 0.2 were considerably lower than in gizzard shad from WCK 1.5 (i.e., White Oak Lake), but they were still slightly elevated with respect to gizzard shad from ERM 6.0 (Table B.4). Across all species and sites, concentrations of Octachloro-dibenzo[b,e][1,4]dioxin were highest of all the dioxin and furan compounds, averaging 1,064 pg/g in whole body gizzard shad and 7.04 pg/g in fillets of largemouth bass from WCK 1.5, 31.1 pg/g in stoneroller minnows from MEK 0.2, and 23.8 pg/g in gizzard shad from ERM 6.0. In general, concentrations of dioxins were highest for the most chlorinated compounds and decreased with degree of chlorination. Toxic equivalents (TEQs) were calculated using toxic equivalency factors (TEFs) that have been adopted by the World Health Organization for human health risk assessments for dioxins and furans (Van den Berg et al. 1998). These calculated TEQs are shown in Table B.4 and were highest for gizzard shad and largemouth bass from WCK 1.5 and only slightly elevated in fish from MEK 0.2 relative to the reference site.

4.5 TURTLES

The highest Hg and MeHg concentrations in all tissues from turtles were found at CRK 15 in the Clinch River just downstream of Poplar Creek (Table B.5). Mean Hg concentrations in muscle (0.11 µg/g) and fat (0.02 µg/g) from turtles at CRK 15 were about double the concentrations in these tissue types in turtles collected further upstream in the Clinch. Methylmercury concentrations in all tissue types followed the same spatial patterns, but the difference in concentration between sites was not as marked as with Hg. Concentrations of both Hg and MeHg in turtles at all sampling locations were highest in liver tissue and lowest in fat tissue. Concentrations of MeHg in all turtle tissues from all sampling locations were below the EPA guideline for MeHg in fish fillets (0.3 µg/g). Almost all of the mercury in liver tissue was inorganic mercury, and concentrations of total Hg in liver tissue from all sites were above the guidelines for MeHg in fish fillet. Concentrations of Hg in all tissues at all sites were lower in 2015 than they were in the same tissues of turtles collected in 2010. Methylmercury concentrations, on the other hand, were higher in 2015 than in turtles collected in 2010 (Table B.5).

Aroclor 1260 was the only detectable PCB Aroclor in turtle tissues, so PCB results do not include Aroclors 1248 and 1254. PCB concentrations were more similar across sites than Hg, with some evidence of higher exposure downstream of major DOE inputs. PCB concentrations in liver and fat tissues were lower in 2015 than those in 2010, but muscle tissue concentrations were higher in 2015. Not surprisingly, PCBs, which are lipophilic, were found in higher concentrations in fat and liver tissues than in muscle. Cesium-137 was below detection limits at all sites and in all tissues.

5. DISCUSSION

5.1 PCBS

Polychlorinated biphenyls are a group of 209 related chlorinated organic compounds that were produced and widely used between 1929 and 1979 because of their stability under extreme thermal, abrasive, radioactive, and corrosive environments (Beyer and Biziuk 2009; Ross 2004). They were used extensively at ETTP, Y-12, and ORNL in a variety of products (e.g., plastics, paints) and as a flame retardant, insulator, and/or coolant in transformers and other electrical or mechanical equipment. In 1978, the Toxic Substances Control Act banned the manufacture of PCBs and placed requirements on continued use of PCBs, but because of their slow rate of degradation, legacy sources in and around the ORR have contributed to elevated PCB concentrations in stream water and sediments. PCBs are lipophilic (lipid seeking) and tend to bioaccumulate in aquatic food chains even though water concentrations may be below detection limits. Results for the FYR confirmed that high concentrations persist in aquatic biota at several locations on the ORR.

In the 2015 FYR, as in the 2010 FYR (Mathews et al. 2011), the highest concentrations of PCBs were found in fish, likely due to higher lipid content in some fish (e.g., stonerollers) and higher trophic levels in other fish. For bioaccumulative contaminants such as PCBs, fish bioaccumulation data have become important measures of compliance for the Clean Water Act and CERCLA. Regulatory guidance and human health risk levels have varied more widely for PCBs, depending on the regulatory program and the assumptions used in the risk analysis. The Tennessee water quality criteria for individual Aroclors and total PCBs are both 0.00064 µg/L under the recreation designated use classification and are the target for PCB-focused total maximum daily loads (TMDLs), including for local reservoirs (Melton Hill, Watts Bar, and Fort Loudon) (TDEC 2010a, 2010b, 2010c). However, most conventional PCB water analyses have detection limits much higher than the PCB Ambient Water Quality Criteria. Therefore, in Tennessee as well as many other states, assessments of impairment for water body segments as well as public fishing advisories for PCBs are based on fish tissue concentrations.

Historically, the US Food and Drug Administration threshold limit of 2.0 µg/g in fish fillet was used for PCB advisories; then for many years in Tennessee, an approximate range of 0.8 to 1 µg/g was used, depending on the data available and factors such as the fish species and size. We have compared our results to this range of concentrations in Fig. 4.1. The remediation goal at the ETTP K-1007-P1 Pond is 1.0 µg/g for fish fillet and 2.3 µg/g for whole body fish. Most recently, the water quality criterion has been used by the Tennessee Department of Environment and Conservation to calculate the fish tissue concentration triggering a determination of impairment and a TMDL, and this concentration is 0.02 µg/g in fish fillet (TDEC 2010a, 2010b, 2010c). The fish and invertebrate PCB concentrations across the ORR are well above this most conservative concentration, but in many cases are below the target concentrations set for fillet and whole body fish in the K-1007-P1 Pond.

Although many of the whole body fish collected across the ORR exceed the 0.8–1.0 µg/g used for fish fillet advisories in Tennessee, only those from upper EFPC, upper Bear Creek, Mitchell Branch, and the K-901-A Pond exceed the 2.3 µg/g target set at ETTP for whole body fish (Fig. 4.1). For the first time since major remediation and fish removal actions took place at the K-1007-P1 Pond in 2009, mean whole body PCB_T concentrations in bluegill (1.99 µg/g) were below the 2.3 µg/g remediation goal at this site (Fig. 5.1). Largemouth bass, which were collected in 2015 for the first time since the 2009 remediation actions as part of the FYR, had fillet PCB concentrations above the target of 1 µg/g in fillets. However, over the 5 years since remediation, concentrations have decreased more than fivefold, from 15.3 µg/g in 2009 to 5.33 µg/g in similarly sized fish collected in 2015 (Fig. 5.1). PCB concentrations in both water and dragonflies collected at the K-1007-P1 Pond have also decreased significantly (> threefold) since 2010 (Fig. 5.1, Peterson et al. 2016).

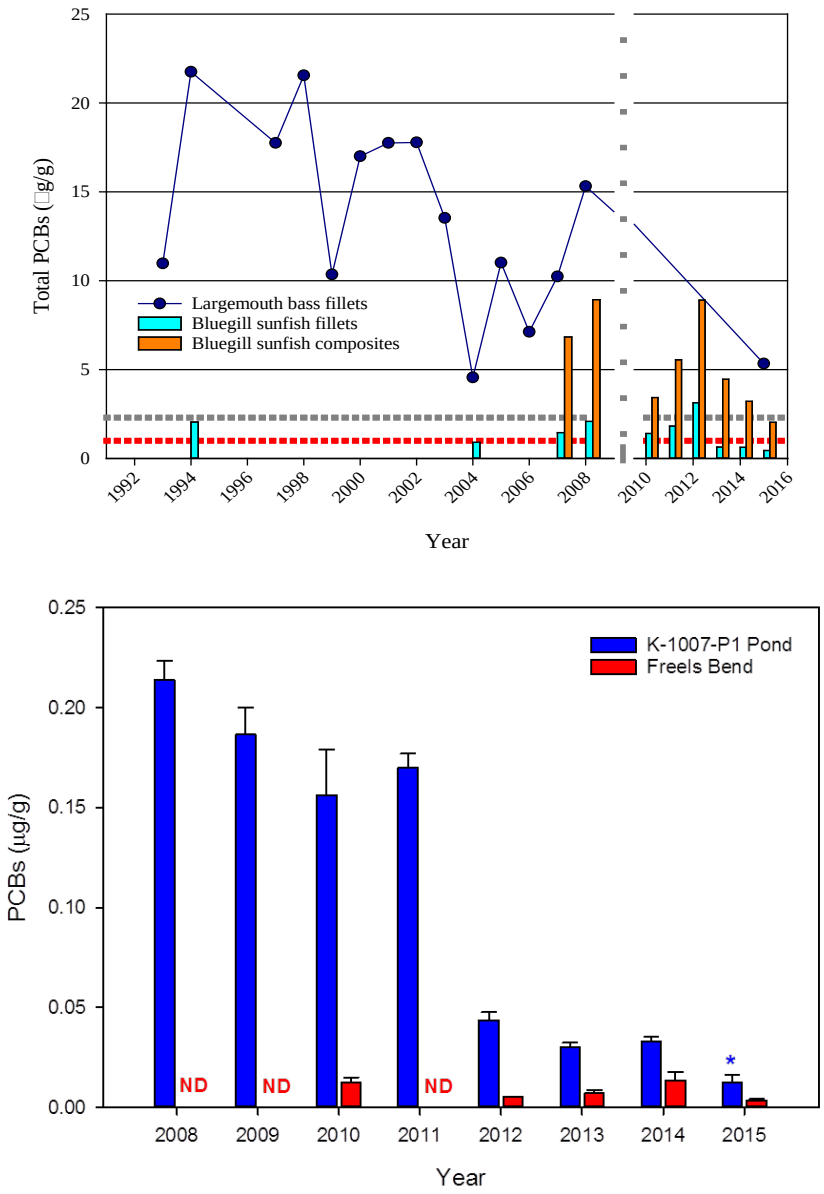

Fig. 5.1. Mean total polychlorinated biphenyl (PCB) concentrations in largemouth bass and bluegill (fillets and whole body composites) from K-1007-P1 Pond (µg/g, top panel) and mean total PCB concentrations (± standard deviation) in adult damselflies (*Argia apicalis*) from K-1007-P1 Pond and the Freels Bend reference site on the Melton Hill Reservoir (µg/g, bottom panel). ND = Not detected. * = Adult dragonflies (*Erythemis simplicicollis*) used for analysis at K-1007-P1 Pond in 2015. Historical data are included in both graphs for comparison. Vertical gray dashed line in the top graph depicts remedial actions (fish removal) in June 2009. The horizontal dotted red line depicts target PCB concentrations for fish fillets (1 µg/g), and the dotted gray line depicts target PCB concentrations for whole body composites (2.3 µg/g).

The highest concentrations of PCBs continue to be found in waterbodies around ETTP, with all but the K-720 Slough having concentrations in fish that greatly exceeded the human health risk goal of 0.8–1.0 µg/g wet mass in fish tissue. As reported in the 2010 FYR, fish from Mitchell Branch site MIK 0.2 had the highest concentration of PCBs of all biota and sites in 2015, though concentrations in 2015 were slightly lower than in 2010 (7.33 µg/g in 2015 vs. 9.0 µg/g in 2010; Fig. 4.1; Mathews et al. 2011). In contrast, PCB concentrations in invertebrates at the same site were several orders of magnitude higher in

2015 than in 2010, but this is likely due to changes in the species used (dragonfly and damselfly nymphs in 2010 vs. snails in 2015). PCB concentrations in invertebrates also were notably higher in 2015 at other sites where snails were used, including BCK 9.9 and WCK 3.9. Total PCB concentrations in fish from those three sites were relatively similar in 2010 and 2015, strongly suggesting that the apparent increase in PCB concentrations at snail sites was due to species differences rather than actual changes in environmental concentrations.

Another notable difference in invertebrate PCB concentrations between the 2010 and 2015 FYRs was in UEFPC. In 2010, the only site where the total PCB concentration in invertebrates exceeded the human health risk goal was EFK 24.5. However, in 2015, the total PCB concentration in invertebrates at this site was much lower than the risk goal. Though the PCB concentrations in fish collected at this site exceeded the human health risk goal of 0.8–1.0 μg/g, concentrations were ~50% lower in 2015 than in 2010. Similar species of invertebrates were used in both periods, so given that the invertebrate species were used and the fish concentrations declined as well, these results suggest a likely decline in PCBs in upper EFPC. This could be due to lower flows in this portion of the stream because the cessation of flow augmentation occurred near Outfall 200 inside the Y-12 National Security Complex in April 2014 (Watson et al. 2016).

Other ORR sites where concentrations of PCBs exceeded the human health risk goal (0.8–1 μg/g) in 2015 included fish from EFK 23.4 in the EFPC watershed, fish from WCK 3.9 in the WOC watershed, and invertebrates and fish from BCK 9.9 in the Bear Creek watershed. Although PCB concentrations were elevated in several other sites included in the 2015 FYR (i.e., EFK 6.3 and PCK 1.6), the concentrations at all of these sites were well below the human health risk goal. At other sites, concentrations for invertebrates and fish were either near or below the analytical detection limit (WCK 2.3, MEK 0.2, MCK 1.4, CRK 32.2, CRK 15.0, and TRM 531).

Spatially, concentrations of PCBs in fish showed a distinct decrease with distance from the highest concentrations in ORR streams to the lowest concentration at the most distant site TRM 531 (Fig. 4.1). Spatial trends in invertebrate PCB concentrations were similar but much less pronounced, which may in part be due to the use of different species at several sites.

5.2 MERCURY

Mercury contamination on the ORR has a long history that dates back to the 1950s in some streams. The greatest amount of Hg use was at the Y-12 Complex for isotope separation from the mid-1950s through the early 1960s (Brooks and Southworth 2011). Mercury was also used at ORNL in the 1950s in small-scale pilot tests for the isotopic separation process (Taylor 1989). Spills and subsequent discharges of Hg-contaminated water to adjoining streams led to significant contamination of the sediment and biota. Despite various remedial efforts, adjacent streams at these facilities continue to receive residual loading from contaminated drains from within the facilities, as well as legacy inputs from exchange with previously contaminated sediments.

As for PCBs, fish bioaccumulation data for Hg have become important measures of compliance for both the Clean Water Act and CERCLA. Because the largest dose of Hg to humans is from the consumption of contaminated fish, the EPA's National Recommended Water Quality Criterion (NRWQC) for MeHg in fish fillets (0.3 μg/g) is used as the trigger point for fish consumption advisories in Tennessee, the target concentration for National Pollutant Discharge Elimination System (NPDES) permit compliance, and the threshold for impairment designations that require a TMDL assessment. This trigger point is shown in Fig. 4.2 for reference. With the exception of spiders collected from EFK 23.4, all MeHg concentrations in biota were below the trigger point. In fact, except for the biota collected from EFPC, Hg_T concentrations in biota across the reservation were below the trigger point for MeHg. This is not surprising because the

present study by design considers animals that are at the base of aquatic food chains with less potential for MeHg bioaccumulation. Spiders have been shown, through previous studies (Cristol et al. 2008) and from the 2010 FYR (Mathews et al. 2011), to have exceptionally high concentrations and proportions of MeHg compared with other invertebrates.

Biota in EFPC continue to have the highest Hg concentrations of all locations on the ORR. East Fork Poplar Creek sites EFK 24.5 and 23.4 had the highest concentrations of Hg in all groups of biota in 2015, which in some cases were more than several orders of magnitude higher than other locations on the ORR. Furthermore, only invertebrates from Mitchell Branch (MIK 0.2) and fish from Poplar Creek (PCK 1.6) had Hg concentrations similar to concentrations in invertebrates and fish from EFK 6.3. Concentrations of Hg and MeHg in fish and aquatic invertebrates from EFK 24.5 and EFK 23.4 were similar to those found in 2010, but Hg_T was about 50% lower in spiders at EFK 23.4 in 2015, whereas the difference in %MeHg between 2010 and 2015 was much less (85% in 2010 vs. 72% in 2015; Mathews et al. 2011).

Except for Melton Branch (MEK 0.2), biota of at least one target group from the remaining sites assessed in 2015 had concentrations of Hg higher than those at the reference sites. However, the concentrations at all of these sites except WCK 3.9, MIK 0.2, K-1007-P1 Pond, and PCK 1.6 were <0.1 µg/g wet mass, which is well below the EPA's criterion for MeHg in fish of 0.3 µg/g wet mass. As in 2010, PCK 1.6 had some of the highest Hg concentrations outside of the EFPC watershed (Mathews et al. 2011). As in 2010, Hg concentrations in fish and invertebrates were notably higher than at reference sites, and concentrations in the current and 2010 studies were similar. As observed at EFK 23.4, the Hg concentration in spiders from WCK 2.3 was about 50% lower than in 2010, although the %MeHg was still well above 80%. The Hg concentration in invertebrates at MIK 0.2 was more than twofold that in 2010, but a mixture of dragonfly and damselfly nymphs were analyzed for Hg in 2010, and snails were analyzed in 2015. As alluded to above for PCBs, the difference in invertebrate concentration is more likely due to species differences in Hg bioaccumulation rather than changes in exposure.

Longitudinally, there was a distinct trend of decreasing concentrations with distance from the main sources of Hg. This was evident in fish and invertebrates in EFPC and in invertebrates in WOC, with the most precipitous decrease evident in EFPC. Although there was little difference between EFK 6.3 and PCK 1.6 in Hg concentrations, concentration at CRK15 and TRM531 were similar to those at the reference sites. A similar longitudinal trend was observed in 2010, as well (Mathews et al. 2011).

Except for MeHg in fish from PCK 1.6, where it was ~17% of the Hg_T, the lowest proportion of MeHg was found in fish and aquatic invertebrates at EFKs 24.5 and 23.4. This trend was apparent in 2010 as well (Mathews et al. 2011). The %MeHg in invertebrates at other sites was well above 50% except CRK 32.2, where it was 32% of the Hg_T. It was notable that at sites where gizzard shad were used for Hg analysis, the %MeHg was 50% or less (i.e., K-720 Pond, K-901-A Pond, CRK 32.2, CRK 15, PCK 1.6, TRM 531, and ERM 6.0), whereas at sites where other species were used, MeHg comprised >65% of the Hg_T.

5.3 METALS

Most of the metals considered in this study were elevated at one or more of the study sites relative to reference sites. Many of these metals are particle-reactive and remain bound to sediment and other particles. Because many aquatic invertebrates incidentally ingest sediment particles as they feed, their body burdens for particle-reactive metals may be elevated relative to other biota. However, because a large proportion of sediment-bound metal is not bioavailable, it is not assimilated into the tissues of the organism. In this study, due to concerns with the ecological risks associated with exposure of wildlife to aquatic insects or whole body fish, the whole bodies of the organisms were analyzed as they were collected in the field, with no attempt to remove the unassimilated gut material from the animals.

However, studies from 2009 to 2015 on contaminant bioaccumulation in aquatic invertebrates downstream of a major fly ash spill at the Tennessee Valley Authority Kingston Fossil Plant in Kingston, Tennessee, have shown that depuration of invertebrates (i.e., allowing animals to empty their digestive tracts) before they are analyzed removes between 30 and 70% of metal (depending on the metal and invertebrate species) from their body burden (J. G. Smith, ORNL, unpublished data).

5.3.1 Antimony, Beryllium, Lithium, Thallium

Concentrations of Sb, Be, Li, and Tl were generally near or below their analytical detection limits in a large number of invertebrate samples (53% to 89% non-detects), and concentrations of Sb, Be, and Li were near or below their analytical detection limits in a high percentage of fish samples (38% to 71%), as well. At locations where these four metals were detected, their concentrations were usually higher in invertebrates than in fish. This was likely related to the position in the food chain and mode of feeding of invertebrates, which contribute to higher incidental intake of inorganic sediment particles that are likely to be enriched with metals. In fish, there was little difference between reference sites and other sites in concentrations of any of these four metals. If the species of the analyzed sample is taken into account, then Sb was slightly elevated in stoneroller minnows from MEK 0.2 and WCK 3.9 and slightly elevated in invertebrates from MEK 0.2. In addition to MEK 0.2, Sb was moderately elevated in invertebrates from MIK 0.2, WCK 2.3, and MCK 1.4. Thallium was slightly to moderately elevated in fish (gizzard shad) and invertebrates (adult dragonflies) from K-720 Slough, but Tl also was moderately elevated in invertebrates from MIK 0.2, CRK 32.2, and MCK 1.4.

Putting the concentrations of Sb, Be, Li, and Tl in perspective with concentrations of potential ecological concern is difficult given the dearth of published results on their bioaccumulation potential in aquatic animals. Much of the published information on these metals is based on laboratory toxicity tests, and bioaccumulation studies often include results from contaminated sites only. In a study of a stream receiving coal combustion waste, the concentration of Be in depurated clams (*Corbicula fluminea*) inhabiting the stream was ~0.046 µg/g wet mass (converted from dry mass assuming a dry mass to wet mass conversion of 15%), but the authors provided no information on background concentrations (Fletcher et al. 2014). Barber et al. (2006) analyzed samples of whole body *Gambusia affinis* samples collected from a wetland created by discharged effluent from a wastewater treatment facility in Arizona for a suite of trace elements, including Be and Li. Beryllium was not detected in any samples, and the concentration of Li was ~0.09 µg/g wet mass (converted from dry mass assuming a dry mass to wet mass conversion for whole body fish of 30%). Given the low frequency of detection of Be and the presence of relatively similar concentrations between reference and other sites, Be is not likely an ecological concern in any of the waterbodies assessed for the FYR. Whereas Li was moderately elevated in invertebrates from MCK 1.4, the fact that Li had a relatively low frequency of detection in invertebrate samples and the highest concentrations found in fish were at reference sites would suggest that Li, like Be, is not likely an ecological concern in the waterbodies assessed.

Fu et al. (2010) collected fish for Sb analysis from several waterbodies in China near that country's largest Sb mine and Sb smelter. They found an average Sb concentration of 0.025 µg/g wet mass in samples of fish muscle (converted from dry mass assuming a dry mass to wet mass conversion for fish filets of 20%). Culioli et al. (2009) analyzed samples of invertebrates (depurated) and rainbow trout (*Salmo trutta*, whole body and filets) in a watershed near an As mine in France. Antimony was not detected in samples of whole body fish from a reference site, but downstream of the mine mean concentrations of Sb ranged from 0.003 to 0.135 µg/g wet mass (converted from dry mass assuming a dry mass to wet mass conversion for whole body fish of 30%). Culioli et al. (2009) only analyzed invertebrate samples collected from a single location downstream of the former mine. They detected Sb in 18 of 20 invertebrate species, and the concentrations found in samples from which Sb was detected ranged from 2.17 to 49.4 µg/g wet mass (converted from dry mass assuming a dry mass to wet mass conversion 25%).

Based on the results of the Fu et al. (2010) and Culioli et al. (2009) studies and the concentrations found in invertebrates at FYR sites where detected, Sb is not likely an ecological concern in waterbodies assessed in this FYR.

In a study on the toxicity of Tl to the amphipod *Hyalella azteca*, Borgmann et al. (1998) included an evaluation of the uptake of Tl from Lake Ontario, Canada, sediment. From their study, they were able to derive estimates of body concentrations at which Tl produced 25% mortality (LBC25). After an exposure period of 4 weeks, the estimated LBC25 was 17.78 µg/g wet mass (estimated from a dry mass concentration of 59.3 µg/g dry mass assuming a dry mass to wet mass conversion of 30%). In a study of Tl exposure of lake trout from Lake Michigan, Lin et al. (2001) found concentrations of Tl ranging from 9.8 to 496 µg/g wet mass (mean 140.8 µg/g wet mass). While concentrations of Tl may have been elevated relative to reference sites in this FYR, based on results from published studies, the concentrations of Tl found in invertebrates and fish in this FYR appear to be well below any threshold of potential ecological concern.

5.3.2 Arsenic

Concentrations of As were elevated at MCK 1.4, MIK 0.2, WCK 3.9, and CRK 32.2, but only in invertebrates. Arsenic concentrations in fish at all sites and invertebrates at the remaining sites were <1.0 µg/g wet mass. Fish concentrations exceeded those of invertebrates only at sites where the terrestrial adult stage of aquatic insects was used (i.e., dragonflies or damselflies at K-720 Slough, K-901-A Pond, and PCK 1.6; adult mayflies at CRK 15 and TRM 531). Arsenic concentrations were generally higher in 2015 than in 2010, and as in 2010, the highest concentrations of As in invertebrates in 2015 also were at MCK 1.4, MIK 0.2 and CRK 32.2 (Mathews et al. 2011). High concentrations of As at these sites and between years at some sites may in part be related to the use of different invertebrate species from different trophic levels. At MCK 1.4 and CRK 32.2, where similar (MCK 1.4) or the same (CRK32.2) taxa were used in 2010 and 2015, there was less difference in concentrations between the studies. At WCK 3.9 and MIK 0.2, in contrast, samples consisting of a mixture of predominantly immature predatory insects were analyzed in 2010, while snails, which are grazers, were used in 2015. Concentrations of trace elements are often highest in invertebrates that feed by grazing on periphyton attached to solid substrates (Newman et al. 1985, Wayland and Crosley 2006). Not only does the periphyton include algae and other microorganisms, but it also accumulates detritus and fine sediment particles that are often enriched with metals; the abiotic particles are consumed incidentally along with the biotic components of the periphyton.

Background concentrations of As in immature aquatic insects in some western states have been reported to range from about 0.015 µg/g wet mass to 4 µg/g wet mass (dry to wet mass conversion factor of 0.25), (Cain et al. 1992; Farag et al. 1998; Wayland and Crosley 2006). Background concentrations of As in undepurated mayfly nymphs (*Hexagenia bilineata*) from the upper Watts Bar watershed (Tennessee) range from about 0.46 µg/g to 1.06 µg/g wet mass (dry to wet mass conversion factor of 0.2), but background concentrations of As in adult mayflies of the same species are much lower (0.042 µg/g wet mass to 0.048 µg/g wet mass; Smith et al. 2016; J. G. Smith, ORNL unpublished data). Background concentrations of As in undepurated snails (*Pleurocera canaliculatum*, same species at CRK 32.2) in the upper Watts Bar watershed are somewhat higher than in the mayfly nymphs (1.54 µg/g wet mass to 1.87 µg/g wet mass based on dry to wet mass conversion of 0.2; J. G. Smith, ORNL unpublished data). Rainbow et al. (2012) used results on an assemblage of metal-intolerant mayflies and metal bioaccumulation in co-occurring caddisflies to derive toxicological thresholds for streams in the United Kingdom. From their effort, they derived a tissue threshold of ~21.3 µg/g wet mass, above which ecotoxicological effects could occur in mayflies (wet mass estimated from a dry mass concentration of 85 µg/g dry mass assuming dry mass to wet mass conversion of 25%). Using growth, feeding behavior, and several histopathological responses in juvenile rainbow trout (*Oncorhynchus mykiss*), Cockell et al.

(1991) conducted a chronic toxicity study of dietary arsenic (as disodium arsenate heptahydrate) and estimated a maximum acceptable dietary concentration between 13 and 33 µg/g. However, in another toxicity study of dietary arsenic using lake white fish (*Coregonus clupeaformis*), Pedlar et al. (2002) reported liver and gallbladder damage at As concentrations as low as 1 µg/g. The range of As concentrations and associated responses reported in the literature clearly show that negative effects from exposure to As are going to depend on many factors that will vary from one location to another (e.g., species present, feeding habits of the species present, water quality conditions). Based on the range of As concentrations that have reportedly been linked to negative effects, only the concentrations found in invertebrates from WCK 3.9, MCK 1.4, MIK 0.2, and CRK 32.2 in 2015 could potentially be of ecological concern. Except for WCK 3.9, there was little difference between the As concentrations found at these sites in 2015 and 2010 (Mathews et al. 2011). Because the samples collected in 2010 and 2015 from WCK 3.9 were composites of different species, and snails appear to accumulate more As than other invertebrate species used in this study, it is not clear if the concentration differences between studies at this site represent a real change or just an artifact caused by species differences in As bioaccumulation. Finally, emerging aquatic insects appear to retain only a small fraction of the As accumulated in their immature aquatic stage. Thus, significant As exposure to terrestrial consumers from emerging aquatic insects is unlikely to occur.

5.3.3 Cadmium

Cadmium is a non-essential metal that can be extremely toxic, especially in freshwater systems. Although it is not known to biomagnify in aquatic food chains, in invertebrates it follows similar bioaccumulation patterns as Zn (Poteat et al. 2013) and can be toxic to fish through aqueous exposure by inhibiting calcium uptake at the gills (Alsop and Wood 2011). Cadmium concentrations were generally low across all sites in all biota but were elevated in spiders on the ORR, especially in EFPC (Fig. 4.4). The sources of Cd on the ORR are not known.

5.3.4 Chromium

Chromium is a metal used for electroplating, municipal treatment plants, oil drilling, cooling towers, and other industrial purposes. Though it can be found in the environment in several different chemical forms, hexavalent chromium (Cr^{+6}) is the most biologically active. It is known to be essential to humans and some animals, but the chemistry of chromium is not well understood. Its toxicity, behavior, and bioavailability depend on its speciation. In 2007, a plume of hexavalent Cr was found in groundwater at ETTP, and elevated concentrations were found in storm drain 170, which discharges into Mitchell Branch, prompting toxicity concerns. Elevated concentrations of this metal were observed in the snails collected at MIK 0.2 in this study and also in the fish collected in the K-901-A Pond, which was also known to have elevated Cr concentrations. Aqueous Cr concentrations in Mitchell Branch have decreased significantly since 2007, and Cr concentrations in stonerollers collected for the 2015 FYR had lower concentrations than those in the 2010 FYR study. The invertebrate Cr concentrations were higher in 2015, but this is likely due to species differences—snails were collected in 2015, whereas insect larvae were collected in 2010. Chromium concentrations in shad collected from the K-901-A Pond were comparable in 2010 and 2015.

At other sites on and off the ORR, Cr concentrations were variable and did not follow any distinct patterns. Concentrations of Cr were elevated in upper WOC, likely due to cooling tower discharges. In contrast to many other metals in this study, concentrations of Cr were generally higher in fish than in invertebrates except at sites where snails were analyzed. Chromium concentrations in gizzard shad from the ERM 6.0 reference site were elevated with respect to all other fish besides shad from the K-901-A Pond.

5.3.5 Copper

Like many other trace metals, Cu is an essential micronutrient at low concentrations but becomes toxic at elevated concentrations. Copper can enter the environment by leaching from wastes or in wastewater, combustion of fossil fuels, phosphate fertilizer production, and other industrial uses. Copper is also used to prevent biofouling and is found in high concentrations in cooling water. Once released into natural waters, Cu soon attaches to particles and can chelate with dissolved organic compounds, affecting the bioavailability and toxicity of Cu.

Copper bioaccumulation in fish is actively regulated to maintain a supply that is adequate to meet biological demands but not high enough to be toxic. Copper concentrations in fish across the ORR were very low, with the exception of WCK 3.9, which also had the highest Cu concentration in invertebrates. Upper WOC receives blowdown from several cooling towers on the ORNL campus, and elevated Cu concentrations have been measured in water and biota in this creek, giving rise to toxicity concerns.

Across the rest of the ORR, the differences in invertebrate Cu concentrations between sites are likely due to differences in the way various species process Cu. As with many other metals, Cu concentrations were highest in spiders and snails. Among the insects, emergent insects generally had higher Cu concentrations than insect larvae. Cu is a known cofactor for many biochemical functions, including energy generation (Nose et al. 2006). Because emergent insects require energetic stores for flight, it follows that emergent insects would need more Cu stores than nymphs.

5.3.6 Molybdenum

Molybdenum is an essential micronutrient for both plants and animals, and thus, is required in low concentrations (Eisler 1989). Molybdenum has many industrial uses and is present with coal deposits. During the combustion process, some Mo remains with coal ash byproducts and some enters the atmosphere. One of the uses of Mo on the ORR is as a corrosion inhibitor in cooling towers. There is very little information on the toxicity or bioaccumulation of Mo, but it is generally considered to be of little toxicological concern except in fairly high concentrations.

Like some other metals, the highest concentrations of Mo in the FYR were in invertebrates. This is most likely because bioconcentration of Mo through the food chain is negligible (Regoli et al. 2012). The highest concentrations of Mo are generally found in plants (Eisler 1989), and thus, animals at the base of the food chain are likely to have the highest accumulated concentrations. Locations where Mo was elevated notably above background concentrations in the FYR included EFK 24.5, BCK 9.9, and MCK 1.4, but the concentrations were elevated only in invertebrates. The concentrations found at EFK 24.5 and MCK 1.4 in 2015 were generally similar to those found in 2010 (Mathews et al. 2011). Elevated concentrations also were found at EFK 23.4 and WCK 3.9 in 2010, but in the current FYR, the concentrations at those sites were similar to those at the reference sites. The likely source of Mo at EFK 24.5 is the discharge of cooling tower blowdown into the stream, whereas the likely source of Mo in McCoy Branch is from the coal ash deposits remaining in the watershed that were discharged into the stream's headwaters from the Y-12 Plant's coal-fired power plant from the mid-1950s through the mid-1990s.

Published accounts of Mo bioaccumulation from field studies are limited. In a study of the Gunnison River watershed in Colorado, Colborn (1982) measured Mo in samples of immature aquatic insects from sites upstream and downstream of mining and/or milling operations for coal, Mo, and uranium. At background locations, mean concentrations of Mo ranged from 0.018 µg/g wet mass to 0.17 µg/g wet mass (wet mass estimated from dry mass concentrations of 0.09 µg/g to 0.85 µg/g based on a dry mass to wet mass conversion of 20%), and at a downstream location the maximum concentration they reported

was 0.33 µg/g wet mass (wet mass estimated from dry mass concentrations of 1.65 µg/g based on a dry mass to wet mass conversion of 20%). Regoli et al. (2012) determined Mo concentrations in fish (*Gasterosteus aculeatus*) collected from wastewater collection tanks and a canal at a Mo processing plant. The mean whole body concentration of Mo they found in the fish was 2.99 µg/g wet mass. From their literature review for their study, Regoli et al. (2012) determined that whole body concentrations of Mo in fish at background locations were generally < 0.25 µg/g wet mass (wet mass estimated from dry mass concentration of 1.0 µg/g based on a dry mass to wet mass conversion of 25%), and at heavily contaminated sites, Mo concentrations were generally < 2.5 µg/g wet mass (wet mass estimated from dry mass concentration of 10.0 µg/g based on a dry mass to wet mass conversion of 25%). Based on these reported values, concentrations of Mo found in this FYR appear to be only modestly elevated in invertebrates at EFK 24.5, BCK 9.9, and MCK 1.4.

5.3.7 Nickel

As in 2010, the highest concentrations of Ni were found in invertebrates at ETTP in Mitchell Branch (MIK 0.2) and off-site at CRK 32.2 (Mathews et al. 2011). Other than a 50% lower concentration of Ni in fish from MIK 0.2 in 2015 compared with 2010, concentrations of Ni at the remaining non-reference sites in 2015 were relatively similar to those in 2010 in invertebrates, spiders, and fish. The concentration of Ni in invertebrates at MIK 0.2 in 2015 was five times higher than in 2010. However, different species of invertebrates from MIK 0.2 were analyzed in 2010 (composite samples of dragonfly and damselfly nymphs) and 2015 (composite samples of snails). Given the tendency for concentrations of some contaminants to be higher in snails (e.g., see Sections 5.3.2 and 5.3.5), the difference between the two studies was probably due to species differences in bioaccumulation and assimilation of Ni, and not to actual differences in environmental concentrations of Ni. Even though different species were analyzed from MIK 0.2 in 2010, the Ni concentrations found at that time also were elevated; thus, the results from 2015 continue to indicate that Ni concentrations in invertebrates at MIK 0.2 are abnormally high.

Tissue concentrations for Ni reported in the literature vary widely. In a study of nine streams near phosphate mining operations in the Blackfoot River watershed in Idaho, Hamilton et al. (2002) reported Ni concentrations in composite samples of mixed invertebrate species ranging from 0.36 µg/g wet mass (a reference site) to 9.2 µg/g wet mass (mine-affected site) (estimated from dry mass concentrations of 1.8 µg/g dry mass and 46 µg/g dry mass, respectively, assuming a dry mass to wet mass conversion of 20%). The highest concentration found at a reference site was 1.38 µg/g wet mass. Mean Ni concentrations of Ni in whole body fish (multiple species) were generally much lower than those in invertebrates, with mean concentrations ranging from 0.48 µg/g wet mass to 0.96 µg/g wet mass (estimated from dry mass concentrations of 1.8 µg/g dry mass and 46.0 µg/g dry mass, respectively, assuming a dry mass to wet mass conversion of 30%). Concentrations at reference sites were no different than those at sites affected by mine activities. In a study of three rivers in Belgium, Bervoets et al. (2004) found concentrations ranging from <0.25 µg/g wet mass to ~31.25 µg/g wet mass in chironomid larvae (*Chironomus riparius*) (estimated from dry mass concentrations of 1.0 µg/g dry mass and ~125 µg/g dry mass, respectively, assuming a dry mass to wet mass conversion of 25%). Finally, Borgmann (2003) estimated an LBC_{25} of 38.8 µg/g wet mass (i.e., the body concentration at which 25% mortality occurs; estimated from dry mass concentrations of 194.0 µg/g dry mass, assuming a dry mass to wet mass conversion of 20%) for Ni in the amphipod *Hyalella azteca.* Based on these published results, the concentration in snails from MIK 0.2 is probably very high, whereas the concentrations found at CRK 32.2 in 2015 and at MIK 0.2 and CRK 32.2 in 2010 were moderately high. The concentration at MIK 0.2 in 2015 would appear to be high enough to be of potential ecological concern.

5.3.8 Lead

Like many other metals, once lead enters the aquatic environment, it tends to adsorb to sediment particles. Thus, for example, Pb concentrations in some invertebrates often closely follow Pb concentrations in the sediment (Cain and Luoma 1998). Because of this close relationship with sediment, higher concentrations could be expected in invertebrate and fish species with feeding habits that are likely to increase the incidental consumption of sediment particles.

A large volume of literature exists on the bioaccumulation and toxicity of Pb to aquatic organisms. Lead concentrations ranging from below the analytical detection limit to concentrations in excess of 700 µg/g wet mass (estimated from a concentration of 3,893 µg/g dry mass and assuming a wet to dry mass conversion of 20%) have been reported for invertebrates (e.g., Farag et al. 1998). Farag et al. (1998) also measured mean Pb concentrations of 2.0 µg/g wet mass (estimated from a concentration of 10 µg/g dry mass and assuming a wet to dry mass conversion of 20%) in invertebrates from reference sites in a metal-impacted watershed in Idaho. There also are published efforts to estimate whole body concentrations at which toxicity may be expected.to occur. For example, in a laboratory bioassay in which amphipods (*Hyalella azteca*) were exposed to sediment spiked with Pb, the mean LBC_{50} (i.e., body concentration at which 50% of the test animals die) estimated by Borgmann and Norwood (1999) was 8.06 µg/g wet mass (estimated from a concentration of 709 nmol/g dry mass and assuming a wet to dry mass conversion of 20%). Although there was some indication that Pb was slightly elevated in invertebrates from MCK 1.4, the concentration at that site was considerably lower than the LBC_{50} concentration estimated by Borgmann and Norwood (1999) and some of the values reported in the literature for background locations.

5.3.9 Selenium

Selenium is a naturally occurring metalloid that is an essential micronutrient and necessary to all living organisms in small quantities, but can be toxic at elevated concentrations. However, it has the smallest range between essentiality and toxicity of any of the essential elements. It has two modes of toxicity in the aquatic environment—acute toxicity via water exposure at relatively high concentrations (e.g., > 100 µg/L), and chronic toxicity to egg-laying animals via dietary exposure at lower concentrations (< 10 µg/L) (Stewart et al. 2004). Though Se can occur naturally in soils, it is also found in elevated concentrations in coal ash, and thus, elevated concentrations are often associated with coal ash discharges or spills (Chapman et al. 2010). In contrast to most other trace elements, Se may biomagnify in aquatic food chains, becoming increasingly concentrated as it is transferred from the base of the food chain (e.g., periphyton, algae) to upper trophic level predators (e.g., fish, aquatic birds), potentially posing ecological and human health risks (Chapman et al. 2010).

As with Hg, accumulation of Se in aquatic animals occurs predominantly through dietary rather than aqueous exposure (Xu and Wang 2002; Luoma et al. 1992; Mathews et al. 2014), but many factors affect the bioavailability of Se to aquatic organisms including speciation (Turner and Swick 1983), watershed flow regime (i.e., lentic vs. lotic systems; Adams et al. 2000), and food chain pathway (Stewart et al. 2004). For these reasons, as for Hg, tissue-based toxicity thresholds were recently proposed for Se because tissue concentrations integrate the route, duration, and magnitude of exposure and are therefore considered to be a more consistent indicator of selenium exposure and risk. The new tissue thresholds include a guideline for concentrations in whole body fish of 8 µg/g dry mass, which corresponds to ~1.6 µg/g wet mass (EPA 2015).

Though none of the mean Se concentrations in this study exceeded 1.6 µg/g, the mean Se concentration in the invertebrates and fish from MCK 1.4 approached this value, and one of the three samples of invertebrates at this site measured 1.6 µg/g, while one fish sample measured 1.7 µg/g. McCoy Branch and

Rogers Quarry have historically had elevated Se concentrations in biota due to discharges of coal ash into the headwaters of McCoy Branch from the Y-12 Complex. Interestingly, Se and Hg have been found to be inversely correlated in fish tissue, suggesting that there may be an antagonistic effect of Se on Hg toxicity. The Se concentrations in MCK 1.4 were the highest in the present study (Fig. 4.10), and the Hg concentrations in biota at this site were among the lowest in this study (Fig. 4.2). Selenium concentrations in biota collected from reference sites were among the higher Se concentrations measured in the study, and Hg concentrations in reference site biota were among the lowest.

Selenium concentrations at all sites were generally comparable in 2010 and 2015, though there were some slight differences in concentration between studies. Invertebrate concentrations of Se were generally higher at sites where snails were collected; thus, the species analyzed could play a role in concentration differences between sites and years.

5.3.10 Silver

Silver is placed in the same class as Cd, Cr (IV), Cu, and Hg in terms of toxicity (Ratte 1999). Like other metals, the high toxicity of Ag is related to its individual species and not the total concentration, with the Ag^{+} ion being the most toxic. Once in the aquatic environment, most of the Ag is absorbed to sediments and suspended particles, thus reducing its toxicity and bioavailability (Ratte 1999; Shafer et al. 1998). These characteristics of Ag may explain in part the trends found in the present FYR. Except for spiders at EFPC sites and WCK 3.9 and adult dragonflies at K-1007-P1 Pond, the highest concentrations were found at sites where grazing invertebrates (i.e., snails) and fish (i.e., stonerollers) were analyzed. Because animals were not depurated before analysis, much of the Ag in these groups probably was associated with inorganic particles present in their digestive tracts. By grazing the periphyton growing on the substrate, they incidentally consume inorganic particles that settle on and become entrapped in the periphyton matrix. The notably elevated concentration of Ag in adult dragonflies at K-1007-P1 Pond is similar to what was observed at K-1007-P1 Pond in 2010 when the Ag concentration in adult damselflies was notably elevated (Mathews et al. 2011). The concentrations of Ag in spiders from lower WOC at WCK 2.3 and the HCK 20.6 reference site were similar to the concentrations in spiders from those sites in 2011. The concentrations of Ag in spiders at all sites where they were collected were several times higher than those in invertebrate samples from those sites (either dragonfly and damselfly nymphs or a mixture of immature insects were analyzed from all four sites). Because spiders are strictly predators, this indicates that Ag was in a form readily transformed up the food chain.

Published studies on Ag bioaccumulation in field-collected biota are largely restricted to estuarine and marine species (Eisler 1996; Ratte 1999). The results of several laboratory toxicity tests of Ag in fish have been reported in which concentrations of Ag also were reported, although in most cases the reported concentrations were for specific organs and tissues such as the liver and gills (e.g., Coleman and Cearley 1974; Galvez et al. 1998; Morgan et al. 2005; Rose-Janes and Playle 2000). The tissue concentrations reported from such studies are not necessarily comparable to field-based studies such as ours. Since a large coal ash spill occurred in late 2008 at the Kingston Fossil Plant, the bioaccumulation of potential metal contaminants in invertebrates from the upper Watts Bar Watershed has been monitored. At reference sites, mean Ag concentrations in undepurated snails have been ~0.26 µg/g wet mass; 0.02 µg/g wet mass in undepurated mayfly nymphs; and ~0.004 µg/g wet mass in adult mayflies (J. G. Smith, ORNL unpublished data). Based on a comparison among sites included in the current study, Ag appears to be elevated at all three sites in EFPC, both sites in WOC, MIK 0.2, K-1007-P1 Pond, and CRK 32.2. However, based on published and unpublished studies, the concentrations of Ag found in biota in the current study would appear to be near background for most sites except WCK 3.9, K-1007-P1 Pond, and CRK 32.2, where Ag concentrations appear to be slightly elevated.

5.3.11 Uranium

Uranium behaves much like many other metals when it enters a body of water. The fate of U is governed by factors such as the pH, presence/absence of clays, the amount of dissolved and particulate organic matter present, and the concentration of ions such as calcium (Crawford and Liber 2015; Kraemer and Evans 2012). Once in the water, U usually becomes associated with sediments, in which case, aquatic animals that feed by grazing or gathering loose particles from the substrate are more likely to ingest uranium-laden sediment particles incidentally with their food. This may in part explain the spatial trends observed in this FYR. Uranium concentrations were clearly elevated in invertebrates at BCK 9.9 and MIK 0.2 and in fish at BCK 9.9 in 2015. The species of caddisfly larvae (*Pycnopsyche luculenta*) analyzed from BCK 9.9 feed on decaying leaves that enter the water from streamside trees. As the leaves decay, they can become coated with fine sediment particles that can be consumed incidentally with the target food of leaves. The fish from BCK 9.9 and MIK 0.2 (stonerollers) and the snails from MIK 0.2 graze algae from rock surfaces. Because no biota were depurated before analysis, a high content of inorganic particles in the digestive tracts may be a major factor in the higher U concentrations at those sites. However, in contrast to what was found at BCK 9.9, U in stonerollers from MIK 0.2 was not elevated. The reason for this difference is unknown. Uranium also was elevated at BCK 9.9 and MIK 0.2 in 2010 (Mathews et al. 2011), although there were some minor concentration differences between that study and the current study. Even so, the concentration of U was clearly higher at those two sites in both studies compared with the reference sites and most other sites.

Concentrations of U also appeared to be slightly elevated in fish from EFKs 24.5 and 23.4 and invertebrates from EFK 24.5. This is generally consistent with what was found in 2010, except that the concentration of U in invertebrates from EFK 23.4 in 2015 was much lower than what was found in 2010 (Mathews et al. 2011). Again, this difference may have been related to differences in feeding habits of the invertebrates that were used. In 2010, samples of filter-feeding caddisflies from EFK 23.4 were analyzed, whereas samples consisting of a mixture of dragonfly and damselfly nymphs were used in 2015. Filter-feeding caddisflies generally feed on suspended particulate organic matter, in which case they may incidentally consume organic and inorganic particles enriched with U. Dragonfly and damselfly nymphs, on the other hand, are predators and therefore are likely to ingest less inorganic particles.

Like some of the other lesser studied metals (e.g., Ag), U bioaccumulation reporting from field studies is rare. Most publications report results from laboratory studies in which exposures to U and the water quality conditions where the exposure take place are strictly controlled (e.g., Crawford and Liber 2015; Kraemer and Evans 2012); thus, any reported tissue or whole body concentrations are not directly comparable with those reported in field studies. In one study in which U concentrations were reported, Barber et al. (2006) analyzed whole body *Gambusia affinis* (mosquitofish) from a wetland created by the discharge from a wastewater treatment facility. The median concentration they reported for U was 0.035 µg/g (assumed to be wet mass based on the sample handling description in the Methods section of Barber et al. 2006). Uranium was one of 26 metals included in an analysis of invertebrate bioaccumulation downstream of the Kingston Fossil Plant coal ash spill in late 2008 (J. G. Smith, ORNL, unpublished data). The average concentration of U found in depurated snails (which therefore should result in much lower concentrations than in non-depurated snails [Muscatello and Liber 2010]) was 0.14 µg/g wet mass; the mean concentration found in non-depurated mayfly nymphs was 0.063 µg/g wet mass; and the mean concentration found in adult mayflies was 0.009 µg/g wet mass. Only the U concentrations in invertebrates from MIK 0.2 and BCK 9.9 and fish from BCK 9.9 exceeded the concentration in snails from the Kingston study. The concentration found in snails from the Kingston study also was higher than the concentrations found in snails from WCK 3.9 and CRK 32.2 in the current study, which suggests the concentrations in snails from those sites were within the range of background levels.

5.3.12 Zinc

Zinc is an essential micronutrient that is crucial for cell division, protein synthesis, immune functions, and growth, and is therefore tightly regulated. Like many other trace elements, it can be toxic at elevated concentrations, and many factors including pH, alkalinity, dissolved organic matter, and other cations can affect its bioavailability and therefore toxicity. Zinc is used to galvanize steel, protecting against corrosion, so it is widely used to protect buildings, vehicles, and other steel structures and is ubiquitous in the environment. Because Zn is so tightly regulated in biota, concentrations were generally comparable at all sites and were not elevated with respect to reference site locations, with the exception of megalopteran larvae at BCK 9.9, which had slightly higher Zn concentrations than at other sites. As with many other metals, concentrations were higher in spiders than in other species.

5.4 DIOXINS AND FURANS

Dioxins and furans are highly toxic and can cause reproductive, developmental, and immunological problems. The most potent of these toxic compounds is 2,3,7,8 Tetrachlorodibenzodioxin (TCDD)—all other compounds are related to TCDD using toxic equivalency factors (TEFs) to determine the overall bioaccumulation of this class of compounds, expressed as toxic equivalents (TEQs) (Van den Berg et al. 1998). Dioxins and furans are not routinely monitored in biota on the ORR, so historical trend analysis was not possible. Figure 5.2 shows the TEQs calculated for the fish in this study and compares these results for the previous 5 years. The risk-based threshold for dioxins and furans is 0.15 pg/g in fish fillets (EPA 2000), which is for human health risks and assumes the consumption of four meals of fish per month. As can be seen from Table B.4, all fish collected for this study exceeded the risk-based threshold, with the highest risk in White Oak Lake (WOL). TEQs in WOL gizzard shad have been decreasing over the past 5 years but remain elevated with respect to all other sites.

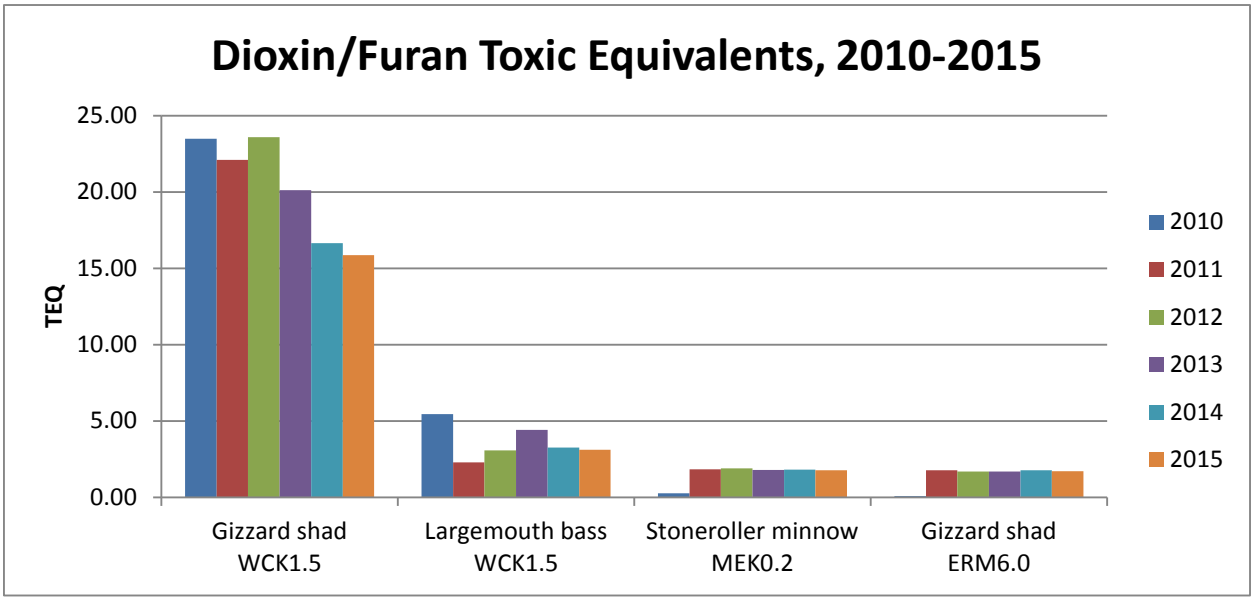

Fig. 5.5. Toxic equivalents (TEQs) for dioxins and furans in fish collected in the White Oak Creek watershed from 2010 to 2015. TEQs calculated as described in Van den Berg et al. (1998). All gizzard shad and stoneroller samples were whole body composites, whereas largemouth bass were fillets.

The source for dioxins and furans to the WOC watershed is not currently known. These compounds are often byproducts of various industrial activities and are most often produced when organic compounds are combusted in the presence of chlorine. They are therefore produced in waste incinerators, chemical

manufacturing, etc. Besides combustion, other processes (bleaching, application of herbicides, etc.) can be sources of these compounds to the environment. More monitoring is needed to determine the sources and risks of dioxins on the ORR.

5.5 TURTLES

According to the Tennessee Wildlife Resources Agency (TWRA), turtles are commonly sought by local recreational fishermen, and common snapping turtles are the only legal reptilian game species in Tennessee. Contaminant concentrations in these long-lived animals are therefore relevant to assessing both human health and ecological risk. Contaminant concentrations in muscle tissues of turtles were generally higher downstream of DOE inputs, but concentrations were low with respect to risk thresholds. At all sites, concentrations of Hg and MeHg in turtle muscle were < 0.3 μg/g risk threshold for MeHg in fish, and concentrations of PCBs in turtle muscle were well below the 0.8–1.0 μg/g risk threshold for PCBs in fish (Table B.5). Not surprisingly, PCBs, which are lipophilic, were found in higher concentrations in fat and liver tissues than in muscle. Cesium-137 was below detection limits at all sites and in all tissues. Though contaminant concentrations were similar in 2010 and 2015 in most tissues and most sites, PCB concentrations in fat tissue in turtles have steadily decreased over the past 10 years at the three sites monitored, from 66 μg/g in 2005 to 21.7 μg/g in 2015 at CRK 15, from 38 μg/g to 18 μg/g at CRK 32.2, and from 84 μg/g to 5 μg/g at CRK 38.4 (Table B.5). This pattern follows the general pattern of decreasing PCB concentrations in fish from offsite locations (UCOR 2014).

5.6 SUMMARY AND CONCLUSIONS

Several potential contaminants of concern to wildlife and humans continue to be elevated in biota at locations on and off the ORR. The major findings of this study include the following:

1. The highest concentrations of PCBs were generally found at sites closest to the three DOE facilities and the K-1007-P1 and K-901-A Ponds at ETTP. A noteworthy decline in total PCB concentrations, however, was apparent at K-1007-P1 Pond in 2015, with concentrations ~2.5-fold lower than in 2010.

2. The highest concentrations of Hg continue to exist in EFPC. Maximum concentrations occur at the most upstream site, but then decrease considerably with downstream distance. The %MeHg, in contrast, tends to increase with downstream distance.

3. The concentrations of Hg and MeHg (but not %MeHg) in spiders from EFK 23.4 were about 2 times lower in 2015 than in 2010, but there was little change in concentrations at EFK 6.3.

4. The major contaminant of concern in the ETTP ponds, particularly the K-1007-P1 and K-901-A Ponds, continues to be PCBs.

5. In addition to PCBs, Cr levels continue to be elevated at the K-901-A Pond, exceeding concentrations found at most other locations by a factor of ~threefold.

6. Concentrations of PCBs and Cr were generally much higher in fish than invertebrates.

7. Concentrations of most metals were generally higher in invertebrates than in fish, most likely because their mode of feeding makes them more likely to incidentally consume metal-laden sediment particles.

8. There were generally fewer differences between invertebrates and fish in concentrations of the essential metals Se and Zn.

9. In contrast to Se and Zn, the essential metal Cu was higher in invertebrates than fish, most likely due to the greater role it plays in various invertebrate physiological processes. Highest Cu concentrations in fish and invertebrates were found in WOC at WCK 3.9.

10. The highest concentrations of U were in Bear Creek (BCK 9.9) and Mitchell Branch (MIK 0.2). Given the history of waste disposal practices and facility operations in those watersheds, this result is not surprising.

11. There were strong indications in the trends of some contaminants in invertebrates and fish that spatial differences were controlled more by species differences in contaminant processing than in actual spatial differences in the environmental quantities present (e.g., Ag, Cu, and Ni in snails; Ag, Cu, and Zn in spiders; and PCBs in fish).

12. Molybdenum concentrations were generally highest at sites closest to main operations at facilities (EFK 24.5 at the Y-12 Complex and WCK 3.9 at ORNL) or locations in the watershed where historic releases of coal ash occurred (MCK 1.4).

13. Cadmium concentrations were highest in spiders in EFPC, exceeding concentrations of all taxa at other sites by a factor of at least sixfold. Spiders from EFK 23.4 and EFK 6.3 were not analyzed for metals in 2010. Since Cd concentrations in other taxa and sites were comparable in 2010 and 2015, the high concentrations in spiders at EFK 23.4 and EFK 6.3 probably do not represent increased environmental concentrations of that metal.

14. Cadmium concentrations at most locations and most species in 2015 were comparable to concentrations found in 2010.

15. Concentrations of Sb, Be, Li, and Tl were generally not elevated at any site.

16. The highest concentrations of Cr were found in Mitchell Branch at MIK 0.2 at ETTP. This site is downstream of the location were a C^{+6} plume was discovered enter the stream via storm drain 170.

17. Concentrations of As in invertebrates and fish were generally low except in snails from WCK 3.9, MCK 1.4, MIK 0.2, and CRK 32.2.

18. Except for MCK 1.4, Pb concentrations were not elevated at any site. However, concentrations of Pb at MCK 1.4 were only slightly elevated in invertebrates at that site.

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APPENDIX A. DETAILS OF INVERTEBRATE SAMPLE TAXONOMIC COMPOSITION

Table A.1. Details of 2015 sample taxonomic composition

Watershed	Water body	Site[a]	Contaminants of concern	Species collected
East Fork Poplar Creek	East Fork Poplar Creek	EFK 24.4	Hg	Caddisfly larvae (*Cheumatopsyche* and *Hydropsyche*) Central stonerollers (*Campostoma anomalum*)
		EFK 24.4	Metals, PCBs	Megaloptera larvae (*Nigronia* and *Corydalus cornutus*) and Odonata nymphs (dragonflies and damselflies) Central stonerollers (*Campostoma anomalum*)
	East Fork Poplar Creek	EFK 23.4	Hg, metals, PCBs	Spiders (*Dolomedes vittatus* only, males and females) Central stonerollers (*Campostoma anomalum*)
		EFK 23.4	Metals	Odonata nymphs (dragonflies and damselflies)
		EFK 23.4	Hg	Caddisfly larvae (*Cheumatopsyche* and *Hydropsyche*)
		EFK 23.4	PCBs	Megaloptera larvae (*Nigronia* and *Corydalus cornutus*)
	East Fork Poplar Creek	EFK 6.3	Hg, metals, PCBs	Spiders (*Dolomedes scriptus* and *Dolomedes vittatus*, males and females) and Odonata nymphs (dragonflies and damselflies) Central stonerollers (*Campostoma anomalum*)
White Oak Creek	White Oak Creek	WCK 3.9	Metals, PCBs	Snails (*Pleurocera* sp.) Central stonerollers (*Campostoma anomalum*)
		WCK 3.9	Hg	Odonata (dragonfly nymphs) Central stonerollers (*Campostoma anomalum*)
	White Oak Creek	WCK 2.3	Hg, metals, PCBs	Spiders (*Dolomedes scriptus* and *Dolomedes vittatus*, males and females) Central stonerollers (*Campostoma anomalum*)
		WCK 2.3	Metals, PCBs	Megaloptera larvae (*Nigronia* and *Corydalus cornutus*) and Odonata nymphs (dragonflies and damselflies)
		WCK 2.3	Hg	Odonata nymphs (dragonflies and damselflies)
	Melton Branch	MEK 0.2	Metals, PCBs	Megaloptera larvae (*Nigronia* and *Corydalus cornutus*) and Odonata nymphs (dragonflies and damselflies) Central stonerollers (*Campostoma anomalum*)
		MEK 0.2	Hg	Megaloptera larvae (*Nigronia*) Central stonerollers (*Campostoma anomalum*)

Table A.1. Details of 2015 sample taxonomic composition (continued)

Watershed	Water body	Site[a]	Contaminants of concern	Species collected
Bear Creek	Bear Creek	BCK 9.9	Metals	Megaloptera larvae (*Nigronia* and *Sialis*) Central stonerollers (*Campostoma anomalum*)
		BCK 9.9	Hg	Caddisfly larvae (*Pycnopsyche*) Central stonerollers (*Campostoma anomalum*)
		BCK 9.9	PCBs	Snails (*Elimia claviformes*) Central stonerollers (*Campostoma anomalum*)
Chestnut Ridge (McCoy Branch)	McCoy Branch	MCK 1.4	Metals	Caddisfly larvae (*Cheumatopsyche, Hydropsyche, Chimarra*) Blacknose dace (*Rhinichthys atratulus*)
		MCK 1.4	Hg	Megaloptera larvae (*Nigronia*) Blacknose dace (*Rhinichthys atratulus*)
		MCK 1.4	PCBs	Megaloptera larvae (*Nigronia*), Odonata nymphs (dragonflies and damselflies), and Caddisfly larvae (*Cheumatopsyche, Hydropsyche, and Chimarra*) Blacknose dace (*Rhinichthys atratulus*)
ETTP	Mitchell Branch	MIK 0.2	Hg, metals, PCBs	Snails (*Pleurocera* sp.) Central stonerollers (*Campostoma anomalum*)
	K-1007-P1 Pond	P1 Pond	Hg, metals, PCBs	Adult female dragonflies (*Erythemis simplicicollis*) Bluegill (*Lepomis macrochirus*)
	K-901-A Pond	K-901	Hg, metals, PCBs	Adult damselflies (*Argia apicalis*) Gizzard shad (*Dorosoma cepedianum*)
	K-720 Slough	K-720	Hg, metals, PCBs	Adult female dragonflies (*Erythemis simplicicollis*) Gizzard shad (*Dorosoma cepedianum*)
Off-site	Clinch River	CRK 15	Hg, metals, PCBs	Adult mayflies (male subimagos, *Hexagenia bilineata*) Gizzard shad (*Dorosoma cepedianum*)
	Clinch River	CRK 15	Hg, PCBs, gamma	Common snapping turtle (*Chelydra serpentine)*
	Clinch River	CRK 32.2	Hg, metals, PCBs	Snails (*Pleurocera canaliculatum*) Gizzard shad (*Dorosoma cepedianum*)
	Clinch River	CRK 32.2	Hg, PCBs, gamma	Common snapping turtle (*Chelydra serpentine)*
	Poplar Creek	PCK 1.6	Hg, metals, PCBs	Adult damselflies (*Argia apicalis*) Gizzard shad (*Dorosoma cepedianum*)
	Tennessee River	TRM 531[c]	Hg, metals, PCBs	Adult mayflies (male imagos, *Hexagenia bilineata*) Gizzard shad (*Dorosoma cepedianum*)

Table A.1. Details of 2015 sample taxonomic composition (continued)

Watershed	Water body	Site[a]	Contaminants of concern	Species collected
Reference	Emory River	ERM 6.0[c]	Hg, metals, PCBs	Adult mayflies (male imagos, *Hexagenia bilineata*) Gizzard shad (*Dorosoma cepedianum*)
		HCK 20.6	Hg, metals, PCBs	Spiders (*Dolomedes scriptus* and *Dolomedes vittatus*, males and females) Central stonerollers (*Campostoma anomalum*)
	Hinds Creek	HCK 20.6	Metals	Mayfly nymphs (*Hexagenia limbata*) and dragonfly and damselfly nymphs
		HCK 20.6	Hg	Mayfly nymphs (*Hexagenia limbata*)
		HCK 20.6	PCBs	Megaloptera larvae (*Nigronia* and *Corydalus cornutus*) and Odonata nymphs (dragonflies and damselflies)
	Clinch River	CRK 38.4	Hg, PCBs, gamma	Common snapping turtle (*Chelydra serpentine)*
	Freels Bend	Freels Bend	PCBs	Adult damselflies (*Argia apicalis*)

Notes: BCK = Bear Creek kilometer, CRK = Clinch River kilometer, ERM = Emory River mile, EFK = East Fork Poplar Creek kilometer, HCK = Hinds Creek kilometer, MCK = McCoy Branch kilometer, MEK = Melton Branch kilometer, MIK = Mitchell Branch kilometer, PCK = Poplar Creek kilometer, TRM = Tennessee River mile, WCK = White Oak Creek kilometer.

APPENDIX B. ANALYTICAL RESULTS

Table B.1. PCB concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015

Site	Taxon	N	Aroclor 1248		Aroclor 1254		Aroclor 1260		Total PCBs	
			Mean	SD	Mean	SD	Mean	SD	Mean	SD
East Fork Poplar Creek watershed										
EFK 24.5	Various insect larvae	3	<0.0038	-	0.117	0.023	0.157	0.051	0.274	0.073
	Stonerollers	3	0.816	0.120	0.843	0.150	0.728	0.142	2.386	0.411
EFK 23.4	Spiders	3	<0.0038	-	0.029	0.014	0.147	0.021	0.176	0.018
	Fishfly larvae	3	<0.0056	-	0.120	0.026	0.137	0.023	0.257	0.040
	Stonerollers	3	<0.047	-	0.940	0.060	0.760	0.147	1.700	0.177
EFK 6.3	Spiders	3	<0.0067	-	0.020	0.002	0.036	0.004	0.055	0.005
	Dragonfly and damselfly larvae	3	<0.0059	-	<0.0059	-	0.008	0.001	0.008	0.001
	Stonerollers	3	<0.012	-	<0.012	-	0.120	0.026	0.120	0.026
White Oak Creek watershed										
WCK 3.9	Snails	3	<0.017	-	0.207	0.031	0.543	0.134	0.750	0.156
	Stonerollers	3	0.201	0.034	0.694	0.108	0.852	0.140	1.747	0.242
WCK 2.3	Spiders	3	<0.005	-	0.032	0.012	0.107	0.014	0.139	0.026
	Various insect larvae	3	<0.0059	-	0.017	0.014	0.088	0.028	0.105	0.042
MEK 0.2	Various insect larvae	3	<0.0038	-	<0.0038	-	0.004	-	0.004	-
	Stonerollers	3	<0.012	-	<0.012	-	0.035	0.017	0.035	0.017
Bear Creek watershed										
BCK 9.9	Snails	3	<0.023	-	0.720	0.219	0.750	0.355	1.470	0.572
	Stonerollers	6	<0.11	-	1.500	0.573	2.350	0.550	3.850	1.110
Chestnut Ridge (McCoy Branch)										
MCK 1.4	Various insect larvae	3	<0.004	-	<0.004	-	<0.004	-	<0.004	-
	Blacknose dace	3	<0.012	-	<0.012	-	0.014	0.001	0.014	0.001
East Tennessee Technology Park										
MIK 0.2	Snails	3	<0.077	-	3.700	0.400	0.970	0.044	4.670	0.407
	Stonerollers	3	<0.35	-	5.700	2.343	1.633	0.896	7.333	2.818
K-1007-P1 Pond	Dragonfly adults	3	<0.0026	-	0.005	0.003	0.020	0.006	0.025	0.009
	Bluegill	6	<0.058	-	1.230	0.240	0.762	0.206	1.992	0.436
	Largemouth bass	6	<0.29	-	2.712	1.439	2.522	1.684	5.233	3.105
K-720 Slough	Dragonfly adults	3	<0.0027	-	0.004	0.002	0.009	0.001	0.013	0.003
	Gizzard shad	6	<0.022	-	0.172	0.039	0.207	0.050	0.378	0.088
K-901-A Pond	Damselfly adults	3	<0.005	-	0.010	0.000	0.056	0.002	0.065	0.001
	Gizzard shad	6	<0.36	-	<0.36	-	5.050	0.779	5.050	0.779
Off-site locations										
CRK 32.2	Snails	3	<0.0031	-	0.009	0.001	0.009	0.003	0.018	0.004
	Gizzard shad	3	<0.011	-	<0.011	-	0.054	0.017	0.054	0.017
CRK 15	Mayfly adults	3	<0.0028	-	0.032	0.005	0.021	0.002	0.053	0.007
	Gizzard shad	3	<0.012	-	<0.012	-	0.097	0.012	0.097	0.012
PCK 1.6	Damselfly adults	3	<0.0059	-	0.006	0.001	0.017	0.004	0.021	0.001
	Gizzard shad	3	<0.012	-	<0.012	-	0.280	0.108	0.280	0.108
TRM 531	Mayfly adults	3	<0.0028	-	0.018	0.002	0.008	0.001	0.026	0.002
	Gizzard shad	3	<0.012	-	<0.012	-	0.022	0.008	0.022	0.008
Reference										
HCK 20.6	Spiders	3	<0.004	-	<0.004	-	<0.004	-	<0.004	-
	Various insect larvae	3	<0.0059	-	<0.0059	-	<0.0059	-	<0.0059	-
	Stonerollers	6	<0.012	-	0.016	0.001	<0.012	-	0.016	0.001
Freels Bend	Damselfly adults	3	<0.0034	-	0.004	0.000	0.004	0.001	0.009	0.001

Table B.1. PCB concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015 (continued)

Site	Taxon	N	Aroclor 1248		Aroclor 1254		Aroclor 1260		Total PCBs	
			Mean	SD	Mean	SD	Mean	SD	Mean	SD
ERM 6.0	Gizzard shad	3	<0.012	-	<0.012	-	0.049	0.010	0.049	0.010

Notes: BCK = Bear Creek kilometer, CRK = Clinch River kilometer, ERM = Emory River mile, EFK = East Fork Poplar Creek kilometer, HCK = Hinds Creek kilometer, MCK = McCoy Branch kilometer, MEK = Melton Branch kilometer, MIK = Mitchell Branch kilometer, PCK = Poplar Creek kilometer, SD = standard deviation, TRM = Tennessee River mile, WCK = White Oak Creek kilometer.

Table B.2. Total mercury (HgT) and methylmercury (MeHg) concentrations (µg/g wet mass) and percent %MeHg in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015.
Values are means (Mean) and ±1 standard deviation (SD) in italics. Asterisks indicate locations where MeHg exceeded total mercury in biota.

Site	Taxon	N	Hg_T		MeHg		%
			Mean	SD	Mean	SD	MeHg
East Fork Poplar Creek							
EFK 24.5	Caddisfly larvae	3	2.517	0.470	0.028	0.003	1.09
	Stonerollers	3	0.762	0.050	0.050	0.004	6.61
EFK 23.4	Spiders	3	0.597	0.065	0.431	0.071	72.21
	Caddisfly larvae	3	0.667	0.194	0.043	0.003	6.46
	Stonerollers	3	0.403	0.031	0.078	0.007	19.40
EFK 6.3	Spiders	3	0.310	0.096	0.231	0.023	74.62
	Damselfly/dragonfly larvae	3	0.251	0.038	0.242	0.025	96.29
	Stonerollers	3	0.168	0.012	0.120	0.015	71.63
White Oak Creek							
WCK 3.9	Damselfly/dragonfly larvae	3	0.148	0.034	0.157	0.038	100*
	Stonerollers	3	0.090	0.015	ND	ND	N/A
WCK 2.3	Spiders	3	0.087	0.009	0.083	0.005	95.85
	Damselfly/dragonfly larvae	3	0.053	0.014	0.064	0.016	100*
MEK 0.2	Fishfly larvae	3	0.028	0.002	0.028	0.005	98.10
	Stonerollers	3	0.028	0.001	0.024	0.001	84.69
Bear Creek							
BCK 9.9	Fishfly larvae	3	0.056	0.009	0.053	0.007	95.23
	Stonerollers	6	0.077	0.010	0.067	0.010	86.86
Chestnut Ridge (McCoy Branch)							
MCK 1.4	Fishfly larvae	3	0.028	0.005	0.027	0.001	93.56
	Blacknose dace	3	0.085	0.013	0.102	0.025	100*
ETTP							
MIK 0.2	Snails	3	0.264	0.026	0.094	0.015	35.70
	Stonerollers	3	0.061	0.007	0.043	0.010	69.96
K-1007-P1 Pond	Dragonfly adults	3	0.067	0.009	0.079	0.008	100*
	Bluegill	6	0.078	0.0141	0.067	0.008	67.41
K-720 Slough	Dragonfly adults	3	0.033	0.010	0.037	0.010	100*
	Gizzard shad	6	0.074	0.016	0.021	0.003	28.02
K-901-A Pond	Damselfly adults	3	0.037	0.006	0.045	0.010	100*
	Gizzard shad	6	0.051	0.003	0.013	0.003	25.54
Off-site							
CRK 32.2	Snails	3	0.018	0.006	0.006	0.001	32.37
	Gizzard shad	3	0.027	0.004	0.011	0.004	39.31
CRK 15	Mayfly adults	3	0.027	0.002	0.022	0.001	80.74
	Gizzard shad	3	0.053	0.009	0.026	0.009	48.70
PCK 1.6	Damselfly adults	3	0.139	0.008	0.181	0.012	100*
	Gizzard shad	3	0.227	0.050	0.038	0.013	16.64
TRM 531	Mayfly adults	3	0.021	0.003	0.014	0.001	68.69
	Gizzard shad	3	0.025	0.001	0.013	0.002	50.40
Reference							
HCK 20.6	Spiders	3	0.025	0.002	0.021	0.005	80.95
	Mayfly larvae	3	0.011	0.000	0.007	0.001	65.26
	Stonerollers	6	0.031	0.004	0.028	0.006	91.58
ERM 6.0	Mayfly adults	3	0.021	0.000	0.015	0.002	70.47
	Gizzard shad	3	0.030	0.000	0.015	0.002	50.17

Table B.3. Total metal concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015.
Values are means and ±1 standard deviation (below each mean in italics). Asterisks indicate locations where lithium was not measured in fish.

Site	Taxon	N	Metal (µg/g wet mass)														
			Ag	As	Be	Cd	Cr	Cu	Li	Mo	Ni	Pb	Sb	Se	Tl	U	Zn
East Fork Poplar Creek																	
EFK 24.5	Various insect larvae	3	0.036	0.21	<0.013	0.136	0.153	6.2	0.13	0.783	0.69	0.377	0.015	0.72	0.003	0.119	55.3
			0.013	*0.04*	-	*0.042*	*0.045*	*1.2*	*0.069*	*0.46*	*0.249*	*0.124*	*0.012*	*0.3*	*0.001*	*0.047*	*17.1*
	Stonerollers	3	0.067	0.10	0.005	0.022	0.183	3.3	*	0.063	0.357	0.19	0.02	0.6	0.004	0.131	43.7
			0.017	*0.02*	*0.001*	*0.004*	*0.067*	*0.61*		*0.008*	*0.122*	*0.07*	*0.004*	*0.046*	*0*	*0.052*	*4.6*
EFK 23.4	Spiders	3	0.112	0.08	<0.007	2.56	0.556	27.3	0.14	0.204	0.547	0.025	<0.017	0.55	0.007	0.026	88.3
			0.122	*0.06*	-	*2.643*	*0.904*	*15.3*	*0.113*	*0.3*	*0.739*	*0.008*	-	*0.161*	*0.009*	*0.025*	*19.6*
	Dragonfly and damselfly larvae	3	0.019	0.2	<0.013	0.104	0.07	4.0	<0.18	0.109	0.513	0.19	<0.016	0.48	0.005	0.047	40.0
			0.005	*0.03*	-	*0.02*	*0.031*	*0.3*	-	*0.012*	*0.086*	*0.04*	-	*0.075*	*0*	*0.011*	*6.1*
	Stonerollers	3	0.075	0.13	<0.012	0.029	0.383	1.1	0.19	0.095	0.463	0.25	0.011	0.57	0.009	0.107	51.3
			0.037	*0.02*	-	*0.009*	*0.11*	*0.3*	*0.017*	*0.032*	*0.133*	*0.095*	*0.005*	*0.19*	*0*	*0.006*	*4.5*
EFK 6.3	Spiders	3	0.26	0.08	<0.013	3.263	0.063	47.7	<0.18	0.052	<0.25	0.038	<0.017	0.88	0.003	0.017	94.3
			0.217	*0.04*	-	*3.463*	*0.05*	*36.6*	-	*0.032*	-	*0.012*	-	*0.214*	*0.001*	*0.01*	*41.8*
	Dragonfly and damselfly larvae	3	0.04	0.31	<0.013	0.125	0.105	4.2	<0.18	<0.041	0.397	0.343	0.018	0.48	<0.004	0.033	29.0
			0.01	*0.12*	-	*0.033*	*0.062*	*0.61*	-	-	*0.091*	*0.085*	*0.011*	*0.151*	-	*0.005*	*2.0*
	Stonerollers	3	0.024	0.06	<0.013	0.019	1.333	0.6	0.17	0.07	0.94	0.133	<0.016	0.49	0.007	0.044	31.3
			0.008	*0.02*	-	*0.002*	*0.321*	*0.19*	*0.085*	*0.035*	*0.226*	*0.056*	-	*0.129*	*0.002*	*0.009*	*2.3*
White Oak Creek																	
WCK 3.9	Snails	3	0.29	1.67	<0.013	0.337	0.78	44.7	0.34	0.177	0.877	0.847	0.011	0.59	0.009	0.123	37.0
			0.044	*0.31*	-	*0.045*	*0.193*	*8.6*	*0.087*	*0.05*	*0.155*	*0.2*	*0.005*	*0.15*	*0.002*	*0.021*	*4.6*
	Stonerollers	3	0.061	0.19	0.014	0.039	1.473	20.7	*	0.109	0.83	0.757	0.033	0.47	0.005	0.025	44.7
			0.007	*0.03*	*0.003*	*0.004*	*1.062*	*2.1*		*0.018*	*0.327*	*0.127*	*0.003*	*0.026*	*0.001*	*0.006*	*1.2*
WCK 2.3	Spiders	3	0.19	0.11	<0.012	0.753	0.044	25.3	<0.16	0.083	<0.23	0.011	0.02	0.97	<0.0036	0.005	95.0
			0.03	*0.05*	-	*0.273*	*0.026*	*4.0*	-	*0.023*	-	*0.004*	*0.014*	*0.677*	-	*0.003*	*16.7*
	Various insect larvae	3	0.039	0.33	<0.013	0.045	0.183	8.1	<0.18	0.061	0.453	0.3	0.03	0.47	<0.004	0.011	34.7
			0.014	*0.05*	-	*0.012*	*0.055*	*1.1*	-	*0.037*	*0.038*	*0.046*	*0.031*	*0.068*	-	*0.002*	*6.7*
MEK 0.2	Various insect larvae	3	0.009	0.21	<0.013	0.024	0.513	4.4	<0.17	0.05	<0.24	0.133	0.023	0.59	<0.0038	0.007	33.7
			0.004	*0.06*	-	*0.013*	*0.595*	*0.7*	-	*0.009*	-	*0.035*	*0.005*	*0.221*	-	*0.002*	*2.9*
	Stonerollers	3	0.005	0.05	<0.013	<0.016	0.263	0.6	0.307	0.037	0.443	0.081	0.034	0.48	0.004	0.006	32.0
			0.003	*0.01*	-	-	*0.095*	*0.18*	*0.074*	*0.014*	*0.114*	*0.01*	*0.001*	*0.145*	*0.003*	*0.001*	*2.0*

Table B.3. Total metal concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. (continued)

Site	Taxon	N	Metal (µg/g wet mass)														
			Ag	As	Be	Cd	Cr	Cu	Li	Mo	Ni	Pb	Sb	Se	Tl	U	Zn
Bear Creek																	
BCK 9.9	Caddisfly larvae	3	0.008	0.2	<0.013	0.35	0.16	3.9	0.22	0.42	0.51	0.37	<0.016	0.22	0.003	0.75	63.0
			0.007	*0.021*	-	*0.10*	*0.02*	*0.17*	*0.04*	*0*	*0.06*	*0.02*	-	*0.04*	*0.002*	*0.11*	*1.0*
	Stonerollers	6	0.005	0.15	<0.013	0.21	0.20	0.6	0.35	0.027	0.30	0.16	0.01	0.43	0.006	0.28	30.2
			0.002	*0.07*	-	*0.06*	*0.08*	*0.37*	*0.14*	*0.01*	*0.12*	*0.07*	*0.006*	*0.13*	*0.001*	*0.039*	*2.317*
Chestnut Ridge (McCoy Branch)																	
MCK 1.4	Caddisfly larvae	3	0.008	2.43	0.033	0.08	0.57	5.8	0.80	0.627	0.65	1.1	0.026	1.4	0.021	0.03	36.0
			0.009	*0.38*	*0.005*	*0.01*	*0.11*	*0.38*	*0.05*	*0.021*	*0.14*	*0*	*0.004*	*0.17*	*0.001*	*0.003*	*2.0*
	Western blacknose dace	3	0.009	0.21	<0.013	0.03	0.61	0.4	<0.17	0.041	1.02	0.05	<0.016	1.43	0.007	<0.0052	45. 7
			0.003	*0.06*	-	*0.02*	*0.19*	*0.43*	-	*0.002*	*0.85*	*0.001*	-	*0.23*	*0.003*	-	*4.7*
ETTP																	
MIK 0.2	Snails	3	0.092	3.4	<0.013	0.50	2.3	29.3	<0.18	0.19	18.0	0.8	0.03	0.68	0.033	0.97	30. 7
			0.043	*0.62*	-	*0.14*	*0.35*	*6.81*	-	*0.036*	*1.732*	*0.22*	*0.011*	*0.03*	*0.004*	*0.20*	*1.2*
	Stonerollers	3	0.004	0.26	<0.013	0.03	0.22	2.3	0.35	0.027	0.87	0.17	<0.016	0.37	0.008	0.03	31.0
			0.002	*0.08*	-	*0.00*	*0.03*	*0.15*	*0.05*	*0.013*	*0.22*	*0.04*	-	*0.03*	*0.001*	*0.007*	*1.0*
K-1007-P1 Pond	Dragonfly adults	3	0.217	0.05	<0.013	0.09	<0.071	11.5	<0.18	0.064	<0.25	<0.019	0.011	0.23	0.005	<0.0053	34.7
			0.129	*0.02*	-	*0.07*	-	*1.86*	-	*0.02*	-	-	*0.006*	*0.11*	*0.003*	-	*3.9*
	Bluegill	6	0.005	0.05	<0.013	<0.017	0.07	<0.24	<0.18	0.069	<0.25	0.04	<0.017	0.25	0.003	<0.0054	29.5
			0.004	*0.04*	-	-	*0.04*	-	-	*0.018*	-	*0.02*	-	*0.09*	*0.002*	-	*3.271*
K-720 Slough	Dragonfly adults	3	0.03	0.12	<0.012	0.17	<0.066	12.7	<0.17	0.187	<0.23	0.04	0.015	0.25	0.036	<0.005	46.7
			0.002	*0.05*	-	*0.07*	-	*1.53*	-	*0.021*	-	*0.03*	*0.013*	*0.11*	*0.025*	-	*7.0*
	Gizzard shad	6	0.005	0.35	0.011	0.02	0.84	0.8	0.16	0.048	0.53	0.47	<0.016	0.34	0.017	0.06	15.0
			0.002	*0.02*	*0.005*	*0.01*	*0.43*	*0.09*	*0.06*	*0.006*	*0.20*	*0.05*	-	*0.09*	*0.002*	*0.01*	*0.9*
K-901-A Pond	Damselfly adults	3	0.023	0.05	<0.013	0.10	0.33	8.6	<0.18	0.064	<0.25	0.03	0.015	0.41	0.003	<0.0053	43.3
			0.005	*0.04*	-	*0.05*	*0.08*	*0.4*	-	*0.011*	-	*0.02*	*0.006*	*0.08*	*0.002*	-	*2.5*
	Gizzard shad	6	<0.0049	0.28	<0.013	<0.016	7.37	0.8	0.1	0.055	0.32	0.22	0.009	0.39	0.004	0.03	22.8
			-	*0.03*	-	-	*1.13*	*0.08*	*0.04*	*0.011*	*0.16*	*0.03*	*0.003*	*0.08*	*0.002*	*0.003*	*0.98*
Offsite																	
CRK 32.2	Snails	3	0.367	2.77	0.016	0.24	0.88	22.7	0.5	0.173	5.1	0.55	0.01	0.84	0.028	0.12	26.0
			0.191	*0.42*	*0.009*	*0.03*	*0.21*	*8.96*	*0.14*	*0.015*	*0.46*	*0.14*	*0.005*	*0.05*	*0.004*	*0.04*	*4.4*
	Gizzard shad	3	<0.0027	0.08	<0.0096	<0.019	0.1	0.8	<0.13	<0.035	0.17	0.12	<0.016	0.48	0.006	0.03	16.0
			-	*0.01*	-	-	*0.06*	*0.07*	-	-	*0.13*	*0.02*	-	*0.03*	*0*	*0.01*	*1.7*
CRK 15	Mayfly adults	3	0.007	0.02	<0.013	0.03	<0.069	4.2	<0.17	0.088	<0.24	<0.018	<0.015	0.53	<0.0039	<0.0052	22.7
			0.004	*0.01*	-	*0.00*	-	*0.32*	-	*0.006*	-	-	-	*0.13*	-	-	*1.2*
	Gizzard shad	3	0.004	0.16	<0.01	<0.02	0.35	1.1	0.09	<0.036	0.30	0.22	<0.017	0.51	0.008	0.03	16.0
			0.003	*0.01*	-	-	*0.03*	*0.177*	*0.04*	-	*0.06*	*0.05*	-	*0.04*	*0.001*	*0.003*	*1.0*

Table B.3. Total metal concentrations (µg/g wet mass) in biota collected from the Oak Ridge Reservation and off-site and reference locations, 2015. (continued)

Site	Taxon	N	Metal (µg/g wet mass)														
			Ag	As	Be	Cd	Cr	Cu	Li	Mo	Ni	Pb	Sb	Se	Tl	U	Zn
Offsite (continued)																	
PCK 1.6	Damselfly adults	3	0.033	0.03	<0.013	0.076	0.072	7.1	<0.17	0.035	<0.25	0.029	<0.016	0.27	0.007	<0.0053	31.7
			0.008	*0.02*	-	*0.022*	*0.067*	*0.31*	-	*0.014*	-	*0.017*	-	*0.08*	*0*	-	*2.1*
	Gizzard shad	3	0.009	0.23	0.014	0.022	0.857	1.3	0.28	0.026	0.71	0.363	<0.016	0.37	0.011	0.068	19.0
			0.002	*0.01*	*0.002*	*0.011*	*0.214*	*0.12*	*0.072*	*0.015*	*0.04*	*0.015*	-	*0.06*	*0.002*	*0.007*	*1.0*
TRM 531	Mayfly adults	3	0.007	<0.032	<0.013	0.029	<0.07	6.9	<0.17	0.076	<0.25	<0.018	<0.016	0.58	<0.0039	<0.0053	21.3
			0.001	-	-	*0.009*	-	*0.38*	-	*0.014*	-	-	-	*0.11*	-		*1.2*
	Gizzard shad	3	0.003	0.13	0.005	0.015	0.343	0.9	0.12	0.018	0.53	0.183	0.008	0.49	0.01	0.041	15.3
			0.002	*0.02*	*0*	*0.009*	*0.067*	*0.21*	*0.046*	*0*	*0.24*	*0.031*	*0*	*0.12*	*0.002*	*0.008*	*1.5*
Reference																	
HCK 20.6	Spiders	3	0.037	0.07	<0.012	0.297	<0.062	26.7	<0.15	0.055	<0.22	0.023	<0.014	0.81	<0.0035	<0.0047	75.3
			0.017	*0.03*	-	*0.133*	-	*11.8*	-	*0.034*	-	*0.015*	-	*0.17*	-	-	*15.4*
	Various insect larvae	3	0.006	0.41	0.012	0.01	0.313	2.9	0.25	0.117	0.47	0.593	0.016	0.69	0.008	0.012	42.3
			0.003	*0.09*	*0.005*	*0.005*	*0.081*	*0.25*	*0.067*	*0.015*	*0.06*	*0.116*	*0.007*	*0.17*	*0.004*	*0.001*	*8.4*
	Stonerollers	6	0.003	0.30	0.014	<0.019	0.553	0.9	0.44	0.03	0.56	0.347	0.013	0.36	0.008	0.012	35.2
			0.001	*0.08*	*0.006*	-	*0.213*	*0.34*	*0.096*	*0.015*	*0.14*	*0.099*	*0.006*	*0.12*	*0.002*	*0.002*	*2.1*
ERM 6.0	Mayfly adults	3	0.014	<0.032	<0.013	0.057	<0.07	6.8	<0.18	0.063	<0.25	<0.018	<0.017	0.72	<0.004	<0.0053	22.3
			0.002	-	-	*0.009*	-	*0.31*	-	*0.009*	-	-	-	*0.04*	-	-	*2.1*
	Gizzard shad	3	0.01	0.28	0.021	0.025	1.967	0.8	0.38	0.07	1.03	0.603	0.031	0.47	0.008	0.071	14.7
			0.009	*0.04*	*0.005*	*0.003*	*0.306*	*0.09*	*0.036*	*0.016*	*0.21*	*0.108*	*0.008*	*0.13*	*0.001*	*0.009*	*1.2*

Table B.4. Mean (±1 standard deviation) concentrations of dioxins and furans in fish collected from WCK 1.5, MEK 0.2, and the ERM 6.0 reference site, pg/g, 2015. For gizzard shad, n = 6, for largemouth bass, n = 6, for stonerollers, n = 3. Whole body composites are composed of 10 fish each. Toxic equivalents (TEQ) were calculated as described in Van den Berg et al. (1998).

		WCK 1.5		MEK 0.2	ERM 6.0
FURANS	TEF	Gizzard shad *(whole body)*	Largemouth bass (*fillets)*	Stoneroller minnow *(whole body)*	Gizzard shad *(whole body)*
Octachlorodibenzofuran	0.0001	56.37 ± 15.53	0.98 ± 1.31	2.15 ± 1.12	0.41 ± 0.12
1,2,3,4,6,7,8-Heptachlorodibenzofuran	0.01	34.90 ± 10.47	0.47 ± 0.21	1.10 ± 0.51	0.50 ± 0.20
1,2,3,4,7,8,9-Heptachlorodibenzofuran	0.01	2.69 ± 0.96	<0.62 ± 0.01	<0.62 ± 0.00	<0.62 ± 0.01
1,2,3,4,7,8-Hexachlorodibenzofuran	0.1	5.32 ± 1.33	0.38 ± 0.21	<0.62 ± 0.00	<0.62 ± 0.01
1,2,3,6,7,8-Hexachlorodibenzofuran	0.1	6.26 ± 1.85	0.53 ± 0.16	<0.62 ± 0.00	<0.62 ± 0.01
1,2,3,7,8,9-Hexachlorodibenzofuran	0.1	0.59 ± 0.06	<0.62 ± 0.01	<0.62 ± 0.00	<0.62 ± 0.01
2,3,4,6,7,8-Hexachlorodibenzofuran	0.1	1.60 ± 1.01	0.53 ± 0.23	<0.62 ± 0.00	<0.62 ± 0.01
1,2,3,7,8-Pentachlorodibenzofuran	0.05	4.87 ± 1.43	0.61 ± 0.20	<0.62 ± 0.00	<0.62 ± 0.01
2,3,4,7,8-Pentachlorodibenzofuran	0.5	3.75 ± 1.20	0.69 ± 0.24	0.55 ± 0.12	<0.62 ± 0.01
2,3,7,8-Tetrachlorodibenzofuran	0.1	22.97 ± 6.75	2.77 ± 1.22	1.16 ± 0.05	0.65 ± 0.04
Total Heptafurans		103.77 ± 29.91		2.58 ± 2.21	
Total Hexafurans		74.17 ± 18.02		1.52 ± 0.72	
Total Pentafurans		44.37 ± 10.19		1.89 ± 0.57	
Total Tetrafurans		40.03 ± 6.04	2.82 ± 1.27	1.33 ± 0.34	0.65 ± 0.04
DIOXINS					
Octachloro-dibenzo[b,e][1,4]dioxin	0.0001	1064.00 ± 320.36	7.04 ± 1.88	31.10 ± 9.64	23.77 ± 1.64
1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin	0.01	218.00 ± 61.51	4.24 ± 3.13	6.17 ± 2.02	1.25 ± 0.05
1,2,3,4,7,8-Hexachlorodibenzo-p-dioxin	0.1	2.31 ± 0.44	0.37 ± 0.19	<0.62 ± 0.00	<0.62 ± 0.01
1,2,3,6,7,8-Hexachlorodibenzo-p-dioxin	0.1	18.17 ± 4.76	1.90 ± 0.74	0.49 ± 0.04	<0.62 ± 0.01
1,2,3,7,8,9-Hexachlorodibenzo-p-dioxin	0.1	5.57 ± 1.74	0.51 ± 0.18	<0.62 ± 0.00	<0.62 ± 0.01
1,2,3,7,8-Pentachlorodibenzo-p-dioxin	1	2.40 ± 1.57	0.67 ± 0.12	<0.62 ± 0.00	<0.62 ± 0.01
2,3,7,8-Tetrachlorodibenzo-p-dioxin	1	2.40 ± 0.49	1.27 ± 0.26	<0.25 ± 0.00	<0.25 ± 0.005
Total Heptadioxins		382.00 ± 112.05	4.63 ± 3.41	9.45 ± 3.33	2.91 ± 0.19
Total Hexadioxins		78.57 ± 15.36	2.21 ± 0.95		
Total Pentadioxins		4.62 ± 2.41	0.36 ± 0.41		
Total Tetradioxins		4.59 ± 2.16	1.27 ± 0.26		
Toxic Equivalents (TEQ)		15.87	3.13	1.79	1.73

Table B.5. Contaminant concentrations and % lipids in muscle, liver, and fat of common snapping turtles (*Chelydra serpentine*) collected in 2010 and 2015 from off-site locations. Values represent equal aliquots of tissue composited from three turtles from each site (except CRK15 from 2010, n = 1).

Tissue	Location	Hg (µg/g)		MeHg (µg/g)		PCB (Aroclor-1260) (µg/g)		% Lipids		Cs-137 (pCi/g)	
		2015	2010	2015	2010	2015	2010	2015	2010	2015	2010
Muscle	CRK 15	0.112	0.197	0.099	0.099	0.08	0.04	0.47	0.72	0.321*	0
	CRK 32.2	0.068	0.103	0.088	0.045	0.10	0.05	0.45	0.52	0.257*	0
	CRK 38.4	0.066	0.129	0.080	0.050	0.08	0.04	0.44	4.7	0.329*	0.152*
Liver	CRK 15	1.870	3.407	0.147	0.084	3.27	4.40	15	11	0.277*	0.11*
	CRK 32.2	0.471	2.481	0.112	0.109	1.80	1.90	8	5.3	0.323*	0.119*
	CRK 38.4	0.530	0.916	0.114	0.067	0.77	4.70	9.5	23	0.353*	0
Fat	CRK 15	0.019	0.026	0.009	0.001	21.70	47.00	75	81	0.344*	0.336*
	CRK 32.2	0.012	0.019	0.006	0.005	17.70	28.00	77	73	0.263*	0
	CRK 38.4	0.010	0.015	0.007	0.007	5.45	17.00	68	75	0.355*	0

* = concentration estimated at a level below detection limit
CRK = Clinch River kilometer
MeHg = methylmercury
PCB = polychlorinated biphenyls (only Aroclor 1260 was detected in all samples)
Cs = Cesium
pCi = picocurie