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Mercury and Other Trace Elements in Ohio River Fish Collected Near Coal-Fired Power Plants: Interspecific Patterns and Consideration of Consumption Risks

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ABSTRACT

Many coal-fired electric generating facilities in the United States are discharging higher loads of Hg, Se, and other chemicals to receiving streams due to the installation of flue gas desulfurization (FGD) air pollution control units. There are regulatory concerns about the potential increased uptake of these bioaccumulative trace elements into food webs. We evaluated the concentrations of As, total Hg (THg), methylmercury (MeHg), and Se in Ohio River fish collected proximal to coal-fired power plants, of which 75% operate FGD systems. Fillet samples ($n = 50$) from 6 fish species representing 3 trophic levels were analyzed. Geometric mean fillet concentrations of THg (wet wt), MeHg (wet wt), and Se (dry wt) in 3 species were 0.136, 0.1181, and 3.19 mg/kg (sauger); 0.123, 0.1013, and 1.56 mg/kg (channel catfish); and 0.127, 0.0914, and 3.30 mg/kg (hybrid striped bass). For all species analyzed, only 3 fillet samples (6% of total) had MeHg concentrations that exceeded the US Environmental Protection Agency (USEPA) human health criterion (0.3 mg/kg wet wt); all of these were freshwater drum aged ≥19 y. None of the samples analyzed exceeded the USEPA proposed muscle and whole body Se thresholds for protection against reproductive effects in freshwater fish. All but 8 fillet samples had a total As concentration less than 1.0 mg/kg dry wt. Mean Se health benefit values (HBV_{Se}) for all species were ≥ 4, indicating that potential Hg-related health risks associated with consumption of Ohio River fish are likely to be offset by adequate Se concentrations. Overall, we observed no measurable evidence of enhanced trace element bioaccumulation associated with proximity to power plant FGD facilities, however, some enhanced bioaccumulation could have occurred in the wastewater mixing zones. Furthermore, available evidence indicates that, due to hydraulic and physical factors, the main stem Ohio River appears to have low net Hg methylation potential. *Integr Environ Assess Manag* 2015;11:474–480. © 2015 SETAC

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INTRODUCTION

The Ohio River main stem is 1582 km in length and includes a drainage area of 528 000 km^2. Annually, approximately 300 million tons of cargo are transported along the river; coal and other energy products comprise approximately 70% of total tonnage (ORSANCO 2013). There are 49 power generating facilities along the river main stem. Most of these are coal-fired stations with varying generating capacities and types of coal burned. Many coal-fired facilities have installed flue gas desulfurization (FGD) units to comply with Clean Air Act sulfur dioxide (SO_2) emission reduction requirements. These units typically remove 95% to 99% of flue gas SO_2. FGD systems also capture volatile trace elements and chloride, with a significant mass fraction of these constituents subsequently sequestered in wallboard-grade gypsum or disposed of in solid waste landfills. A portion of the FGD trace element and dissolved solids mass is also transferred to wastewater, which is treated and discharged to receiving streams. As, Hg, and Se are considered the trace elements of most concern for potential aquatic toxicity. Moreover, some federal and state regulators are concerned that wastewater discharges of these bioaccumulative trace elements could result in increased fish tissue concentrations in affected receiving streams (USEPA 2013).

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Chemical analysis of Ohio River fish tissue has a long history, beginning in 1975 with ORSANCO (Ohio River Valley Water Sanitation Commission), but there are relatively few peer-reviewed reports documenting trace element analyses of Ohio River fish. Schmitt (2002) reported levels of As, total Hg (THg), Se, and other trace elements in whole body carp and largemouth bass composite samples collected in 1995 at Ohio River locations and major tributaries. The geometric mean THg concentration in largemouth bass samples for all Ohio River basin sites combined in that study was 0.14 mg/kg (wet wt). Walters et al. (2010) reported concentrations of THg in small and large fish species collected from the Missouri, Ohio, and Mississippi Rivers. These authors estimated that THg concentrations in sampled species exceeded the US Environmental Protection Agency (USEPA) human health methylmercury (MeHg) criterion of 0.3 mg/kg (wet wt; USEPA 2001) for 7% of the entire Ohio River length. That study measured THg (not MeHg) in whole body samples. However, USEPA recommends that fillet samples be analyzed and if the

THg concentration exceeds or approaches the human health criterion, a confirmatory MeHg analysis should be conducted (USEPA 2010). Thus, the prevalence of fish exceeding the human health criterion in the Ohio River, as suggested by Walters et al. (2010), should be considered provisional.

Emery and Spaeth (2011) reported THg concentrations greater than 0.3 mg/kg wet wt in 42% of all composite, skin-on fillet samples (3 fish per composite) of hybrid striped bass (*Morone saxatilis* x *M. chrysops*) collected at several lock and dam tailwater habitats in the Ohio River. Although the analysis of fillet samples is more meaningful for evaluating potential fish consumption risks, the lack of tissue MeHg results makes it impossible to determine the actual prevalence of samples exceeding the human health criterion. Because the fish sampled were the largest (and presumably the oldest) hybrid striped bass at each location, and because this species is piscivorous with rapid growth during the first 2 years (Hodson 1989), elevated THg fillet concentrations might be expected.

Clearly, this discussion reinforces the importance of selecting both a standardized tissue type and a trace element species for analysis that are most meaningful for regulatory use (and human health risk).

In this study, we report concentrations of As, THg, MeHg, and Se in fillet and whole body samples for 2 species of forage fish and 4 other species collected near coal-fired power plants on the Ohio River. The objectives were to: 1) evaluate tissue concentrations of these elements in fish collected near power plants, 2) document associations between trace element concentrations and fish meristic variables (age, length, weight), 3) assess potential human health risks associated with fish consumption by determining the prevalence of fillet MeHg concentrations that exceed the USEPA MeHg human health criterion, and 4) elucidate the potential mitigation of human health risks by evaluating the relative molar ratios of THg and Se in fish tissue.

METHODS

Fish collection and tissue samples

Fish tissue sampling was conducted during August 8–15 and October 18–21, 2010 near 12 coal-fired power plants along the Ohio River main stem. Table 1 indicates the facility name, Ohio River kilometer location, facility total generating capacity, and whether the facility is equipped with an FGD system. The geographic locations of plant sites are presented in Figure 1. Although fish were obtained proximal to the power plants (within 100–200 m), they were not captured within the wastewater discharge mixing zone.

Fish were collected via night electrofishing using a boat-mounted pulsed-DC powered system. Fish species targeted for capture were from trophic level (TL) 3 and TL4, as these species represent the increasing extent of biomagnification in freshwater aquatic systems. TL3 species were predominantly omnivorous, whereas TL4 species were largely piscivorous (based on diet preference data found in the literature). Skin-off fillet samples were collected from channel catfish (*Ictalurus punctatus*) and freshwater drum (*Aplodinotus grunniens*) (TL3) and hybrid striped bass (*Morone saxatilis* x *M. chrysops*) and sauger (*Sander canadense*) (TL4). Composite whole body samples were obtained from channel shiner (*Notropis wickliffi*) and emerald shiner (*Notropis antherinoides*). The shiner forage fish species were collected at only 1 power plant site. The number of fish per forage fish composite sample ranged between 6 and 31. Most fillet samples analyzed were composite samples of 3 or 4 fish, however 12 of the 50 total samples were from individual fishes. When a sufficient number of a species was obtained at a given location, composite samples representing 2 size classes (based on fish total length) were prepared.

Table 1. Location of Ohio River coal-fired power plant sites where fish were collected for trace element content

Facility name	Ohio River location downstream of Pittsburgh (km)	Generating capacity (MW)	FGD system installed?
Sammis	85.3	1260	Yes
Cardinal	123.3	1830	Yes
Mitchell	181.1	1600	Yes
Willow Island	258.4	243	No
Mountaineer	390.4	1300	Yes
Kyger Creek	418.6	1085	Yes
Stuart	653.2	2380	Yes
Beckjord	729.3	1124	No
Tanners Creek	798.0	995	No
Clifty Creek	901.6	1302	Yes
Gallagher	982.1	280	Yes
Cane Run	993.0	563	Yes

Fish length (total length, mm) and weight (g) were measured at the time of capture, and scales (or otoliths for freshwater drum) were obtained for age determination. Trophic transfer functions (TTFs) were calculated using mean forage fish concentrations of THg, MeHg, and Se and fillet concentrations in channel catfish, sauger, and hybrid striped bass.

Analytical procedures

Frozen fillet and whole-body samples were shipped to Brooks Rand Laboratories. Samples were homogenized (Standard Methods 2540 G) (APHA 2005) and analyzed for total As and Se (USEPA Method 1638, modified DRC) (USEPA 1996), THg (USEPA Method 1631, Appendix E) (USEPA 2002), and MeHg (USEPA Method 1630, modified) (USEPA 1998). Quality assurance-quality control analyses conducted on each shipped batch included analysis of homogenization and laboratory-fortified blanks, method blanks, certified reference materials (CRM; DORM-3 and IAEA 407), duplicates, matrix spikes, and matrix spike duplicates. A detectable concentration was that measured at or above the method detection limit.

Data analysis

For each sample, we calculated the percent of THg as MeHg, as well as the molar ratio of Se to THg (μmol/μg). For tissue samples from TL3 and TL4 species, we used Pearson correlation analysis to evaluate associations between trace element concentrations and fish length, weight, and age. To assess aquatic life health, we evaluated concentrations of tissue

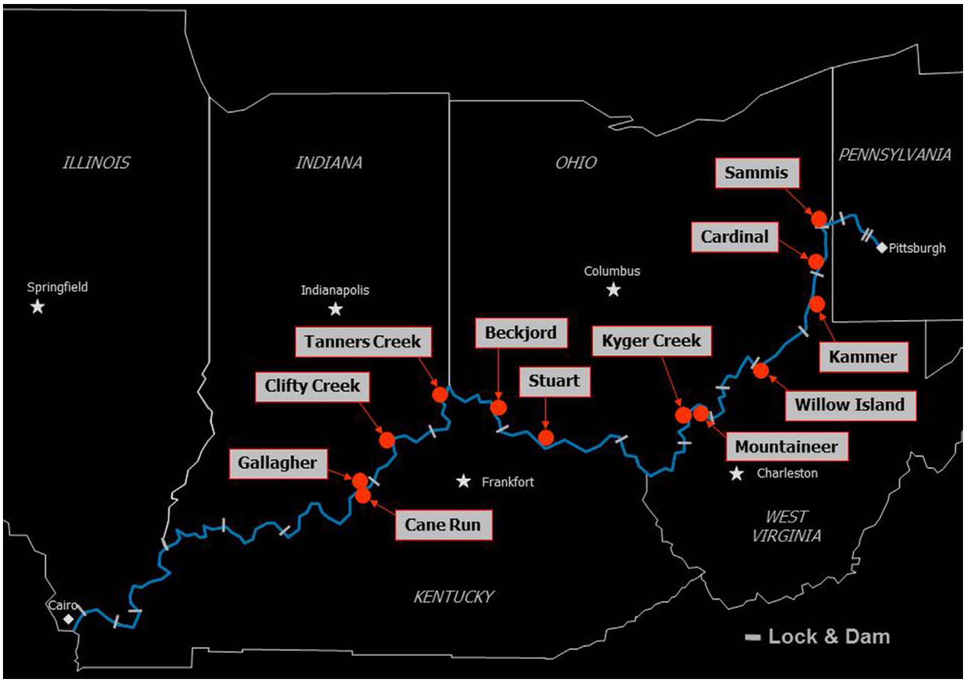

Figure 1. Location of 12 coal-fired power plants on the Ohio River, where fish were collected for tissue analysis.

Se relative to USEPA's proposed muscle and whole body tissue thresholds for protection against adverse reproductive effects (USEPA 2014). There is some evidence that potential human health risks from consumption of fish containing Hg may be mitigated by the Se content in fish tissue (Zhang et al. 2014). Thus, we calculated a Se health benefit value (HBV_{Se}) (Ralston and Raymond 2013) for all tissue samples analyzed using the following equation:

$$HBV_{Se} = \frac{\left(Se_{\mu M} - Hg_{\mu M}\right)}{Se_{\mu M} \times \left(Se_{\mu M} + Hg_{\mu M}\right)},$$

where $Se_{\mu M}$ and $Hg_{\mu M}$ are molar concentrations of Se and THg, as defined on a wet wt basis.

RESULTS

A total of 123 quality assurance-quality control samples were analyzed. The distribution of these were as follows: laboratory-fortified blanks (4), DORM-3 CRM (4), IAEA 407 CRM (22), duplicates (31), matrix spikes (27), matrix spike duplicates (31), and method blanks (4). All of the analyses (100%) achieved the applicable precision, accuracy, and/or percent difference criteria.

A total of 50 tissue samples were analyzed for trace element content (21 samples in August 2010; 29 samples in October). Data on sample sizes, species length ranges, species age-class ranges, and geometric mean trace element concentration (±1 SD) are presented in Table 2.

For As, 84% of tissue samples (42 of 50 samples) had concentrations less than 1.0 mg/kg dry wt. The 8 samples with As concentrations greater than 1.0 mg/kg included 3 freshwater drum, 3 hybrid striped bass, 1 channel catfish, and 1 channel shiner. Excluding freshwater drum and forage species, species-specific geometric mean As concentrations ranged between 0.240 mg/kg (channel catfish) and 0.723 mg/kg dry wt (hybrid striped bass). For comparison, EPRI (2008) summarized As bioaccumulation data for both freshwater and marine fish species. For fish designated as TL4 species, EPRI reported a median whole body As concentration for freshwater species (not from contaminated sites) of 0.20 mg/kg wet wt; assuming a moisture content of 75%, the corresponding dry weight value is 0.80 mg/kg. Thus, our data indicate that As levels in Ohio River fish are comparable to those from other studies.

Like As, fillet Se concentrations in our study were generally low (Table 2). The highest concentration measured among all samples was 6.43 mg/kg dry wt. (sauger) which, like the geometric mean concentrations for all species, was well below the USEPA muscle tissue threshold of 11.8 mg/kg dry wt. For whole-body samples, geometric mean whole body Se concentrations for emerald shiner (3.06 mg/kg dry wt) and channel shiner (3.25 mg/kg dry wt) were also well below the USEPA whole body threshold of 8.1 mg/kg dry wt.

Sunfish (i.e., genus *Lepomis* and *Micropterus*) are Se-sensitive warmwater species (USEPA 2014). Although tissue Se concentrations for these species were not evaluated in this study, bluegill sunfish (*Lepomis machrochirus*) were collected at river kilometer 390 (at Mountaineer Plant) during 2010–2011 for an unrelated study. The geometric mean fillet Se concentration (total of 5 samples representing individual fish or composite samples) was 5.52 mg/kg dry wt, approximately 50% of the USEPA fillet threshold criterion.

The geometric mean concentrations of THg and MeHg were similar for channel catfish, hybrid striped bass, and sauger (Table 2). THg and MeHg concentrations in freshwater drum samples, however, were significantly greater than these or the forage species, with the geometric mean MeHg concentration equaling the USEPA MeHg human health criterion (0.30 mg/kg wet wt). Because the 3 freshwater drum analyzed in this study were larger (>540 mm total length) and older

Table 2. Sample size, range of fish length and age, and geometric mean tissue trace element concentration for 6 Ohio River fish species

				Geometric mean concentration (± SD)			
Species	*n*	Total length range (mm)	Fish age (range)	As (mg/kg dry wt)	THg (mg/kg wet wt)	MeHg (mg/kg wet wt)	Se (mg/kg dry wt)
Emerald shiner[a]	2	47–92	N/A	0.485 (0.159)	0.043 (0.003)	0.0351 (<0.0001)	3.06 (0.63)
Channel shiner[a]	1	37–49	N/A	1.50	0.026	0.0215	3.25
Channel catfish	13	393–629	4–7	0.240 (0.244)	0.123 (0.090)	0.1013 (0.0717)	1.56 (0.74)
Hybrid striped bass	7	224–491	1–4	0.723 (0.481)	0.127 (0.052)	0.0914 (0.0440)	3.30 (1.43)
Sauger	24	220–382	1–3	0.324 (0.116)	0.136 (0.051)	0.1181 (0.0467)	3.19 (0.89)
Freshwater drum	3	428–584	5–21	1.289 (0.384)	0.412 (0.092)	0.3050 (0.0580)	2.19 (0.55)

[a]Whole body composite samples.

(≥19 y) compared to all other fish, these elevated MeHg concentrations likely are the result of lifetime bioaccumulation of Hg. For TL3 and TL4 species, the distribution of fillet MeHg concentrations by fish collection location is presented in Figure 2. These results indicate no discernible longitudinal pattern (i.e., increasing or decreasing concentration relative to river kilometer) for species-specific tissue MeHg concentrations.

We observed a highly significant ($p < 0.001$) correlation between sauger weight and sauger tissue THg concentration (Figure 3). Both THg and MeHg concentrations for sauger were positively correlated with fish age ($p < 0.001$). Such significant THg and MeHg correlations were not observed for other species. Trophic transfer factors of THg and MeHg between the shiner species and TL4 species (hybrid striped bass and sauger) ranged from 3.2 to 4.2. Our examination of a limited number of sauger stomach content samples confirmed that shiners (and to a lesser extent, small gizzard shad) were a primary dietary source for this species.

As concentrations in the shiner species were greater than those in both TL3 and TL4 species, resulting in negative TTF values (although there is a limited sample size). For Se, the geometric mean tissue concentrations for shiner and TL4 species were within a narrow range (between 3.06 and 3.25 mg/kg dry wt), indicating no food web bioaccumulation of Se in this study.

The median percentage of THg as MeHg in fillet samples ranged from 73% (freshwater drum and hybrid striped bass) to 85% (sauger) (Figure 4). For all species, the 75th percentile % MeHg was less than 90%. Thus, for samples in this study, the data do not confirm a common regulatory assumption that MeHg constitutes greater than 90% of THg in most fish species (USEPA 2010).

Geometric mean molar ratio and HBV_{Se} values for all TL3 and TL4 species are provided in Table 3. For all species evaluated, the geometric mean molar ratio of Se to THg values ranged from 2.5 (freshwater drum) to 14.8 (hybrid striped bass). Geometric mean HBV_{Se} values for all species were greater than 1.0, and ranged from 4.0 (channel catfish) to 9.1 (hybrid striped bass).

DISCUSSION

Previous studies have reported inconsistent results for evidence of trace element enrichment in abiotic and biotic samples collected near coal-fired power plants. Hg-enriched sediment cores were reported by Furl and Meredith (2011) whereas Sackett et al. (2010) found that sunfish collected proximal to coal-fired power plants had elevated tissue Se concentrations. In contrast, both Pinkney et al. (1997) and Weir et al. (2010) observed no evidence of increased THg

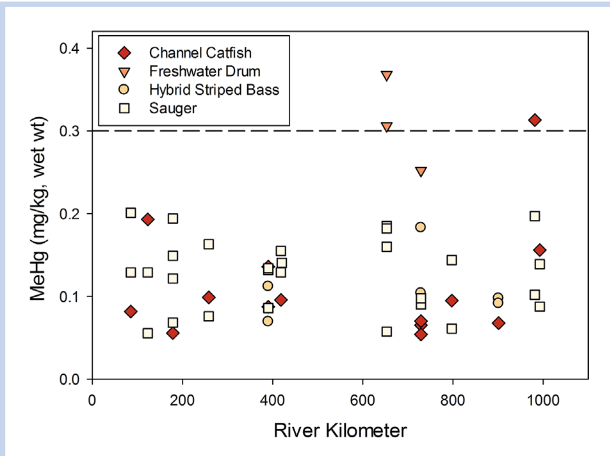

Figure 2. Skin-off fillet concentrations of methylmercury (mg/kg wet wt) in 4 Ohio River fish species, 2010. The dashed line represents USEPA's methylmercury human health fish tissue criterion.

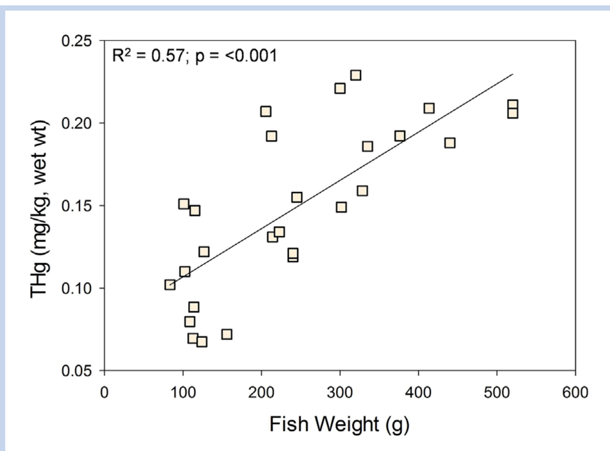

Figure 3. Plot of sauger wet weight versus total Hg fillet concentration, all Ohio River locations, 2010.

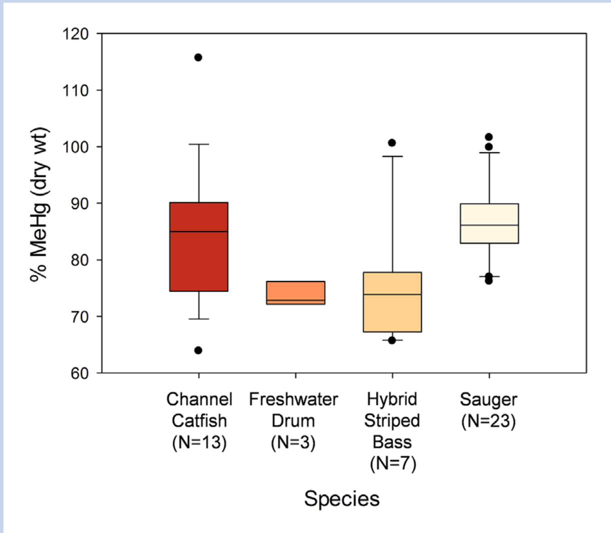

Figure 4. Boxplot of the percent methylmercury as total Hg in 4 Ohio River fish species, 2010.

tissue concentrations in fish and frog samples, respectively, in samples collected adjacent to coal power plants. The results from this study demonstrated no evidence of elevated concentrations of As, THg, MeHg, or Se in fish sampled proximal to coal-fired Ohio River power plants. However, because fish were not collected in the immediate wastewater mixing zones, we could not discard the potential that some of these fish may have had elevated tissue levels of bioaccumulative trace elements.

Except for 3 19+-year-old freshwater drum (6% of total) with fillet MeHg concentrations greater than the USEPA human health criterion, median concentrations of the measured trace elements in fillet and whole body composite samples were all relatively low. For comparison purposes, geometric mean tissue THg, MeHg, and Se concentration data from this study as well as from the 2010 ORSANCO fish tissue monitoring assessment (ORSANCO 2015) are presented in Table 4. For the ORSANCO study, fish were collected near lock and dam tailwater reaches at a moderate to considerable distance from the power plant sites listed in Table 1. Although sample sizes varied between the 2 studies, concentrations of the 3 trace elements in channel catfish and sauger were comparable between the 2 data sets. In contrast, significant differences were observed in geometric mean concentrations of THg and MeHg for freshwater drum and hybrid striped bass samples. For freshwater drum, the mean tissue concentrations of both THg and MeHg presented in this study were greater than USEPA's MeHg criterion, whereas mean THg and MeHg tissue concentrations for fish collected by ORSANCO were below the criterion. For hybrid striped bass the reverse relationship was apparent, with mean THg and MeHg tissue concentrations less than the criterion for data presented in this study, and greater than the criterion for the ORSANCO samples. One weakness in comparing the 2 data sets is that length-normalized Hg tissue concentration data were not available. Such a comparison would be more technically robust than comparing values provided in Table 4.

Table 3. Mean tissue Se–THg molar ratio and HBV_{Se} values for 4 Ohio River fish species

	Geometric mean value	
Species	Se–Hg molar ratio (μmol/μg)	HBV_{Se}
Channel catfish	8.1	4.0
Hybrid striped bass	14.8	9.1
Sauger	12.4	8.4
Freshwater drum	2.5	5.8

Overall, several factors may explain why THg and MeHg concentrations in Ohio River fish are generally low. One factor is an apparently low potential for net Hg methylation. Chan et al. (2012) developed a predictive Hg fate model that was used to assess Hg methylation and bioaccumulation potential for fish from several case study watersheds in the Ohio River basin. Although the authors noted that the accuracy of model-predicted tissue concentrations was low for some watersheds, principal factors associated with Hg bioaccumulation in most watersheds studied were land use (specifically, the percent of forested area), proximity to and coverage of wetlands, and watershed nutrient levels.

Land use in the Ohio River basin varies geographically (ORSANCO 2014). In northern bordering states (Ohio, Indiana, Illinois) row crop agriculture is predominant. The dominant land use in Pennsylvania and West Virginia is deciduous forest, whereas both of these land uses are roughly in equal proportions in Kentucky and Tennessee. Thus, the percent of forested land use within the entire basin is likely not more than 50%. Because the Ohio River main stem is comprised of a flow-regulated series of navigation pools, the shoreline is characterized by minimal perennial wetland habitat and thus likely has low Hg methylation potential. Ohio River tributary backwater reaches likely represent the most functionally comparable habitat to perennial wetlands. Regarding nutrient levels, median total P concentrations at 18 Ohio River lock and dam sample locations were less than 0.2 mg/L during 2006–2011; many locations had a median concentration less than 0.1 mg/L (ORSANCO 2012). Lewis et al. (2011) cited an average total P concentration of 0.1 mg/L for unpolluted reference sites. Thus, although the Ohio River is clearly not oligotrophic, excessive nutrient levels are not characteristic throughout its length.

EPRI (2013) recently applied a revised version of the Dynamic Mercury Cycling Model (D-MCM) to examine Hg cycling in the Robert C. Byrd navigation pool of the Ohio River. This navigation pool is located between river kilometers 382 and 449, and a major tributary (Kanawha River) enters the pool at about the midpoint. The model used measured THg and MeHg data from water (main stem and tributary), sediments, atmospheric deposition, forage fish tissue, and point-source discharges (including 4 coal-fired power plants) to predict Hg cycling. Based on measured concentrations of THg and MeHg in adjacent upstream and downstream navigation pools, the model predicted negligible methylation of inorganic Hg in either the water column or sediments. Model accuracy was verified by reasonable correspondence between measured and predicted MeHg concentrations in fish species including channel catfish, hybrid striped bass, and sauger. For the Ohio River, variables associated with low predicted methylation rates likely include short hydraulic

Table 4. Geometric mean trace element concentrations in 4 Ohio River fish species based on 2 studies

Species	Trace element	Geomentric mean concentration (mg/kg)[a]			
		This study	*n*	ORSANCO	*n*
Channel catfish	THg	0.123	13	0.104	23
	MeHg	0.1013		0.11	
	Se	1.56		1.14	
Hybrid striped bass	THg	0.127	7	0.416	35
	MeHg	0.0914		0.383	
	Se	3.3		Not analyzed	
Sauger	THg	0.136	24	0.123	5
	MeHg	0.1181		Not analyzed	
	Se	3.19		2.14	
Freshwater drum	THg	0.412	3	0.3	24
	MeHg	0.305		0.254	
	Se	2.19		2.58	

[a]THg and MeHg values in wet wt. Se values in dry wt. ORSANCO results given in wet wt; Se dry wt conversion for all samples assumed 75% moisture content.

residence time for Hg (particularly during high flow events), and the presence of well-oxygenated sediments that have high erosional potential and low total organic carbon.

For fish analyzed in this study, not only are tissue THg and MeHg concentrations relatively low, but Se to THg molar ratios in these samples were somewhat elevated (≥2.5). Likewise, all calculated species-specific calculated HBV_{Se} values exceeded 1.0, with the highest value (9.1) observed for hybrid striped bass. Molar index values greater than 1 are considered protective of potential risks associated with consumption of fish having moderate to high THg concentrations (Zhang et al. 2014). Based on these findings, a reasonable risk management conclusion is that Se concentrations in Ohio River fish tissue may offset any potential human health effects associated with consuming fish with high body burdens of Hg. Peterson et al. (2009) reached a similar conclusion, noting that 98% of fish samples collected in Western US states had Se to Hg molar ratios greater than 1. Traditional human health risk assessments, as well as policies on the implementation of regulatory action in response to elevated fish Hg concentrations (Chan and Jacobs 2012), may therefore warrant revision as more scientific evidence of the protective effects of Se appears in the primary literature. We propose that ORSANCO, as well as states bordering the Ohio River, consider indices such as the HBV_{Se} in their issuance of fish consumption advisories.

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