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Environmental contaminants in freshwater fish and their risk to piscivorous wildlife based on a national monitoring program

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Abstract Organochlorine chemical residues and elemental concentrations were measured in piscivorous and benthivorous fish at 111 sites from large U.S. river basins. Potential contaminant sources such as urban and agricultural runoff, industrial discharges, mine drainage, and irrigation varied among the sampling sites. Our objectives were to provide summary statistics for chemical contaminants and to determine if contaminant concentrations in the fish were a risk to wildlife that forage at these sites. Concentrations of dieldrin, total DDT, total PCBs, toxaphene, TCDD-EQ, cadmium, chromium, mercury, lead, selenium, and zinc exceeded toxicity thresholds to protect fish and piscivorous wildlife in samples from at least one site; most exceedences were for total PCBs, mercury, and zinc. Chemical concentrations in fish from the Mississippi River Basin exceeded the greatest number of toxicity thresholds. Screening level wildlife risk analysis models were developed for bald eagle and mink using no adverse effect levels (NOAELs), which were derived from adult dietary exposure or tissue concentration studies and based primarily on reproductive endpoints. No effect hazard concentrations

(NEHC) were calculated by comparing the NOAEL to the food ingestion rate (dietary-based NOAEL) or biomagnification factor (tissue-based NOAEL) of each receptor. Piscivorous wildlife may be at risk from a contaminant if the measured concentration in fish exceeds the NEHC. Concentrations of most organochlorine residues and elemental contaminants represented no to low risk to bald eagle and mink at most sites. The risk associated with pentachloroanisole, aldrin, Dacthal, methoxychlor, mirex, and toxaphene was unknown because NOAELs for these contaminants were not available for bald eagle or mink. Risk differed among modeled species and sites. Our screening level analysis indicates that the greatest risk to piscivorous wildlife was from total DDT, total PCBs, TCDD-EQ, mercury, and selenium. Bald eagles were at greater risk to total DDT and total PCBs than mink, whereas risks of TCDD-EQ, mercury, and selenium were greater to mink than bald eagle.

Keywords Biomonitoring · Ecological risk assessment · Toxicity · Dietary exposure · Biomagnification

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Introduction

The bioaccumulation of contaminants by fish represents a potentially significant risk to piscivorous wildlife. Persistent, accumulative substances such as DDT (as DDE),

polychlorinated biphenyls (PCBs), mercury, and selenium can accumulate in fish and pose a threat to piscivorous wildlife. Fish often comprise a substantial dietary portion of species such as mink (*Mustela vison*), river otter (*Lutra canadensis*), osprey (*Pandion haliaetus*), and bald eagle (*Haliaeetus leucocephalus*), and therefore represent a significant route of contaminant exposure. In addition, wildlife typically consume most or all of the fish, including internal organs, in which concentrations of many contaminants are generally greater than those in the muscle tissue (fillet) typically used to assess human health risks.

The Large River Monitoring Network (LRMN) of the Biomonitoring of Environmental Status and Trends (BEST) Project was initiated to document spatial trends in accumulative chemical contaminants, health indicators, and reproductive biomarkers. From 1995 through 2004, the BEST-LRMN Project measured concentrations of organochlorine pesticides, PCBs, and elemental contaminants in whole-body composite samples of piscivorous and benthivorous fish from nine U.S. river basins (Schmitt 2002; 2004; Schmitt et al. 2005; Hinck et al. 2006a, b; 2007, 2008). Hinck et al. (2006b) determined that avian and mammalian piscivorous wildlife were at risk to mercury, but not organochlorine pesticides or PCBs, in northern pike (*Esox lucius*), longnose sucker (*Catostomus catostomus*), and burbot (*Lota lota*) from the Yukon River Basin. However, the risk of chemical contaminants to piscivorous wildlife that live and forage at BEST-LRMN sites in the conterminous United States has not been evaluated. Therefore, this paper reports a wildlife risk analysis that was conducted to determine the risk of these contaminants to avian (bald eagle) and mammalian (mink) receptors that consume the large adult fish such as those collected by the BEST-LRMN Project and have a broad geographic range. Unlike many ecological risk assessments that estimate fish contaminant concentrations from sediment or soil contaminant concentrations, contaminant concentrations in whole-body fish samples from the BEST-LRMN Project were used in the analysis to give a more accurate estimate of risk to piscivorous wildlife.

The BEST-LRMN Project was preceded by the National Contaminant Biomonitoring Program (NCBP). The NCBP measured organochlorine residues and elemental contaminants in whole-body fish composite samples from 1976 through 1986 at 107

locations across the United States (Schmitt et al. 1999). The NCBP demonstrated that concentrations of organochlorine pesticides, PCBs, and certain elemental contaminants such as mercury were generally decreasing in fish and provided descriptive statistics for concentration distributions in fish from across the United States. The geometric means and 85th percentile concentrations from the NCBP have been previously used as benchmarks for national trends of contaminant concentrations in whole-body fish (Schmitt et al. 1999). Some of the BEST-LRMN sites were co-located at historical NCBP sites, and trend analysis for contaminant concentrations were included in individual basin reports (Schmitt 2002; 2004; Schmitt et al. 2005; Hinck et al. 2006a; 2007).

This paper provides an example of how contaminant concentrations in fish from a routine monitoring program can be used to screen for risk of contaminant exposure to piscivorous wildlife. This approach follows the procedures developed by the U.S. Environmental Protection Agency (USEPA) to conduct screening-level ecological risk assessments (USEPA 1998). Although these assessments are usually applied locally to assess specific sites, they have been used in regional assessments. For example, Yearley et al. (1998) examined the risk of mercury to wildlife in northeastern U.S. lakes, and Hinck et al. (2006b) determined the risk of contaminant exposure to avian and mammalian receptors in the Yukon River Basin. Our objectives were (1) to provide summary statistics for organochlorine chemical residues and elemental contaminants measured by the BEST-LRMN Project and (2) to screen for contaminants that represent a risk to bald eagle and mink and identify the areas where the risk to these receptors was greatest. The wildlife risk analysis data from Hinck et al. (2006b) were updated in this paper to provide results for all BEST-LRMN samples. These results were then compared to site specific studies to determine if piscivorous wildlife had previously been identified as at risk to contaminant exposure.

Materials and methods

Detailed sampling, field, and laboratory procedures have been previously described (Schmitt 2002; 2004; Schmitt et al. 2005; Hinck et al. 2006a, b; 2007; 2008). A summary is presented here.

Sampling and field procedures

Fish were collected from 111 sites in nine river basins (Table 1; Fig. 1). Sites were located in the Apalachicola River Basin ($n=3$), Colorado River Basin ($n=14$), Columbia River Basin ($n=16$), Mobile River Basin ($n=4$), Mississippi River Basin ($n=48$), Pee Dee River Basin ($n=3$), Rio Grande Basin ($n=10$), Savannah River Basin ($n=3$), and Yukon River Basin ($n=10$; Table 1). Of these, 51 were NCBP sites that had been sampled historically (Schmitt et al. 1999). Each site was sampled once. Sampling sites were located on the mainstem and large tributaries of the river basins with known access points without regard to contaminant sources; however, the sites did represent a range of contaminant sources (e.g., chemical manufacturing, agricultural, and urban areas). Most fish were captured by electrofishing; hook-and-line, gill nets, and fyke nets were used only in the Yukon River Basin. Piscivorous species included black bass (*Micropterus* spp.), white bass (*Morone* spp.), catfish (Ictaluridae), northern pike, northern pikeminnow (*Ptychocheilus oregonensis*), burbot, trout (Salmonidae), pikeperch (*Sander* spp.), and goldeye (*Hiodon alosoides*). Benthivorous species included common carp (*Cyprinus carpio*) and sucker (Catistomidae; Table 2).

Laboratory analyses

Fish were shipped to the laboratory frozen on dry ice and stored at -20°C until prepared for analysis. Individual whole fish were partly thawed, cut into pieces, and ground to a fine texture. The ground sub-samples were then grouped to create a single homogenous composite sample for each site, species, and gender combination. A total of 409 composite samples were included in the chemical analysis.

One sub-sample (10 g) of each composite sample was solvent-extracted and analyzed gravimetrically for lipid content and by high-resolution capillary gas chromatography with electron capture detection for organochlorine pesticide residues and total PCBs (sum of individual congeners) after size exclusion and adsorption column cleanup. Pentachlorobenzene, pentachloroanisole, aldrin, Dacthal, heptachlor, endosulfan I, endosulfan II, endosulfan sulfate, and methoxychlor were not measured in Columbia, Rio Grande, and Mississippi River Basin samples. Quality

assurance (QA) measures for the organochlorine pesticide and PCB analyses included the analysis of blanks, triplicate analyses, and matrix spikes. The detection limit for each compound was calculated by adding the average procedural blank concentration to three times the procedural blank standard deviation (Keith 1991) and varied among study years with detection limits changing by two orders of magnitude for some organochlorine residues (Table 3). Concentrations were converted from ng/g wet weight (ww) to $\mu\text{g/g}$ ww for statistical and wildlife risk analyses.

A second sub-sample of each fish composite sample (100 g) was freeze-dried for elemental analysis. Percent moisture was determined as weight lost during lyophilization. For samples from the Mississippi, Rio Grande, and Columbia River Basins, one portion of the dried material was acid-digested and analyzed by atomic absorption spectroscopy and inductively coupled plasma emission spectroscopy. For all other samples, three sub-samples were analyzed for elemental contaminants. One portion of the dried material was digested in nitric acid and analyzed by inductively coupled plasma mass spectroscopy for cadmium, copper, chromium, nickel, lead, and zinc. A second portion was dry-ashed (magnesium nitrate-nitric acid-HCl) and analyzed by hydride generation atomic absorption spectroscopy for arsenic and selenium. A third portion was analyzed directly for total mercury using thermal combustion, amalgamation, and atomic absorption spectroscopy. QA measures for the elemental analyses included the analysis of reagent blanks, duplicate samples, certified reference materials, and fortified samples. Dry-weight (dw) detection limits were determined individually for each element in each sample. Elemental concentrations (including detection limits) were converted from $\mu\text{g/g}$ dw to $\mu\text{g/g}$ ww for statistical analysis and reporting using the moisture content of each sample (Table 4).

A third sub-sample (10 g) was solvent-extracted and subjected to reactive cleanup for use in the H4IIE bioassay (Tillitt et al. 1991; Whyte et al. 2004). Concentrations of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin equivalents (TCDD-EQ; pg/g ww) were determined by slope ratio assay as modified by Ankley et al. (1991). QA measures for the H4IIE bioassay included analysis of duplicate samples and reference materials. Concentrations were converted to $\mu\text{g/g}$ ww for the wildlife risk analysis.

Table 1 Freshwater fish collection sites of the BEST-LRMN Project

Basin site no.	River	Nearest city or feature	Latitude longitude
Apalachicola River Basin			
330	Chattahoochee	Omaha, GA	32°13'19.80" N, 84°55'35.10" W
331	Flint	Albany, GA	31°34'34.86" N, 84°08'49.80" W
332	Apalachicola	Blountstown, FL	30°25'58.20" N, 85°01'17.10" W
Colorado River Basin			
311	Yampa	Lay, CO	40°25'38.00" N, 107°51'24.00" W
312	Green	Ouray NWR, UT	40°08'31.00" N, 109°39'17.00" W
313	Green	San Rafael, UT	38°45'56.00" N, 110°05'16.00" W
314	Gunnison	Delta, CO	38°45'59.58" N, 108°02'30.30" W
315	Colorado	Loma, CO	39°09'39.00" N, 108°48'28.56" W
316	Colorado	Gold Bar Canyon, UT	38°34'02.00" N, 109°38'51.00" W
317	San Juan	Hogback Diversion, NM	36°44'41.00" N, 108°41'28.00" W
319	Colorado	South Cove, AZ	36°05'23.70" N, 114°06'12.30" W
320	Colorado	Willow Beach, AZ	35°52'33.12" N, 114°39'53.10" W
321	Colorado	Needles, CA	34°43'44.64" N, 114°20'12.96" W
322	Colorado	Imperial Dam, AZ	32°54'05.94" N, 114°28'09.48" W
323	Gila	Hayden, AZ	33°01'22.14" N, 110°44'16.32" W
324	Gila	Phoenix, AZ	33°22'33.42" N, 112°18'19.20" W
325	Gila	Arlington, AZ	33°19'06.92" N, 112°40'26.46" W
Columbia River Basin			
41	Snake	Hagerman, ID	42°47'36.21" N, 114°56'18.10" W
42	Salmon	Riggins, ID	45°35'43.42" N, 116°16'55.00" W
43	Snake	Lewiston, ID	46°24'54.28" N, 117°02'03.49" W
44	Yakima	Granger, WA	46°20'49.31" N, 120°12'27.03" W
45	Willamette	Oregon City, OR	45°19'03.47" N, 122°39'57.50" W
46	Columbia	Cascade Locks, OR	45°41'23.11" N, 121°51'00.41" W
96	Snake	Ice Harbor Dam, WA	46°41'51.68" N, 118°53'07.88" W
97	Columbia	Pasco, WA	46°31'49.22" N, 119°16'42.07" W
98	Columbia	Grand Coulee, WA	47°57'44.85" N, 118°58'53.84" W
117	Flathead	Creston, MT	48°09'01.09" N, 114°11'29.71" W
501	Columbia	Beaver Army Terminal, OR	46°10'57.86" N, 123°04'13.87" W
502	Columbia	Warrendale, OR	45°38'00.82" N, 121°58'42.57" W
503	Columbia	Vernita Bridge, WA	46°37'28.40" N, 119°51'31.45" W
504	Columbia	Northport, WA	48°58'21.70" N, 117°38'48.92" W
505	Willamette	Portland, OR	45°33'04.51" N, 122°41'43.74" W
506	Columbia	Vancouver, WA	45°35'44.21" N, 122°32'13.61" W
Mobile River Basin			
326	Tombigbee	Lavaca, AL	32°15'53.60" N, 88°00'44.21" W
327	Coosa	Childersburg, AL	33°19'57.76" N, 86°21'55.87" W
328	Alabama	Eureka Landing, AL	31°23'14.06" N, 87°42'42.19" W
329	Mobile	Bucks, AL	31°03'15.85" N, 87°59'48.07" W
Mississippi River Basin			
15	Mississippi	Luling, LA	29°59'53.2" N, 90°25'31.1" W
23	Kanawha	Winfield, WV	38°29'06.0" N, 81°48'57.6" W
24	Ohio	Marietta, OH	39°24'36.8" N, 81°26'26.3" W
25	Cumberland	Clarksville, OH	36°32'28.6" N, 87°22'04.7" W
26	Illinois	Beardstown, IL	40°07'50.6" N, 90°20'45.6" W
27	Mississippi	Guttenburg, IA	42°43'37.2" N, 91°01'30.0" W
28	Arkansas	Pine Bluff, AR	34°16'27.0" N, 94°57'12.0" W

Table 1 (continued)

Basin site no.	River	Nearest city or feature	Latitude longitude
29	Arkansas	Keystone Res., OK	36°07'54.0" N, 96°20'47.0" W
30	White	Devall's Bluff, AR	34°47'01.0" N, 91°26'28.0" W
31	Missouri	Nebraska City, NE	40°40'15.9" N, 95°49'44.6" W
32	Missouri	Garrison Dam, ND	47°28'27.3" N, 101°26'15.5" W
67	Allegheny	Natrona, PA	40°39'54.0" N, 79°41'24.0" W
68	Wabash	New Harmony, IN	38°11'58.4" N, 87°58'36.0" W
70	Ohio	Metropolis, IL	37°07'40.8" N, 88°39'25.2" W
71	Tennessee	Savannah, TN	35°12'52.0" N, 88°18'36.0" W
72	Wisconsin	Woodman, WI	43°05'42.0" N, 90°48'57.6" W
73	Des Moines	Keosauqua, IA	40°44'52.8" N, 91°59'38.4" W
74	Mississippi	Little Falls, MN	45°58'48.0" N, 94°22'00.0" W
75	Mississippi	Cape Girardeau, MO	37°18'36.0" N, 89°31'01.2" W
76	Mississippi	Memphis, TN	38°08'30.3" N, 90°03'36.6" W
77	Arkansas	John Martin Res., CO	38°03'55.0" N, 102°56'02.0" W
78	Verdigris	Oologah, OK	36°31'16.0" N, 95°33'37.0" W
79	Canadian	Eufaula, OK	35°16'43.0" N, 95°34'39.0" W
80	Yazoo	Redwood, MS	32°24'36.0" N, 90°55'27.0" W
81	Red	Alexandria, LA	31°20'48.0" N, 92°27'37.0" W
82	Red	Lake Texoma, TX/OK	33°52'08.0" N, 96°47'04.0" W
83	Missouri	Hermann, MO	38°42'24.1" N, 91°26'17.5" W
84	Big Horn	Hardin, MT	45°52'12.2" N, 107°34'34.0" W
85	Yellowstone	Sidney, NE	47°34'46.8" N, 104°13'10.7" W
86	James	Olivet, SD	43°13'45.0" N, 97°41'05.0" W
89	Platte	Louisville, NE	40°59'33.1" N, 96°12'30.9" W
90	Kansas	Bonner Springs, KS	39°02'47.0" N, 94°47'05.0" W
111	Mississippi	Lake City, MN	44°22'49.8" N, 92°07'33.0" W
112	Mississippi	Dubuque, IA	42°26'27.6" N, 90°35'06.0" W
201	Big Sunflower	Anguilla, MS	32°58'18.0" N, 90°46'40.0" W
202	Bogue Phalia	Leland, MS	33°24'22.0" N, 90°50'26.0" W
203	Steele Bayou	Rolling Fork, MS	32°54'71.0" N, 90°57'10.0" W
204	Tensas	Tendal, LA	32°25'56.0" N, 91°21'57.0" W
205	S. Skunk	Oskaloosa, IA	41°21'19.0" N, 92°39'31.0" W
206	Iowa	Morengo, IA	41°50'23.0" N, 92°11'54.0" W
207	Cache	Cotton Plant, AR	35°02'32.0" N, 91°19'12.0" W
208	Cache	Egypt, AR	35°51'23.0" N, 90°56'15.0" W
209	S. Fork Iowa	New Providence, IA	42°19'26.0" N, 93°10'10.0" W
210	Iowa	Rowan, IA	42°45'36.0" N, 93°37'23.0" W
211	Cedar	St. Charles City, IA	43°03'45.0" N, 92°40'23.0" W
212	Little R. Ditch	Moorehouse, MO	36°50'03.0" N, 89°43'48.0" W
213	Wolf	LaGrange, TN	35°01'57.0" N, 89°14'48.0" W
400 ^a	Leetown Res.	Kearneysville, WV	39°21'2.15" N, 77°55'32.69" W
Pee Dee River Basin			
336	Pee Dee	Rockingham, NC	34°53'22.14" N, 79°51'24.89" W
337	Pee Dee	Pee Dee, SC	34°21'23.22" N, 79°41'35.19" W
338	Pee Dee	Bucksport, SC	33°42'18.09" N, 79°11'24.00" W
Rio Grande Basin			
16	Rio Grande	Mission, TX	26°09'28.74" N, 98°20'02.82" W
63	Rio Grande	Elephant Butte Res., NM	33°12'48.55" N, 107°13'27.26" W
64	Rio Grande	Alamosa, CO	37°25'06.42" N, 37°23'38.57" W
65	Pecos	Red Bluff Lake, TX	32°00'00.00" N, 104°58'30.00" W

Table 1 (continued)

Basin site no.	River	Nearest city or feature	Latitude longitude
511	Arroyo Colorado	Harlingen, TX	26°11'44.28" N, 97°36'20.52" W
512	Rio Grande	Brownsville, TX	25°52'12.96" N, 97°27'06.30" W
513	Rio Grande	Below Falcon Dam, TX	20°08'06.66" N, 99°08'06.42" W
514	Rio Grande	Below Amistad Res., TX	29°26'49.06" N, 101°03'10.58" W
515	Rio Grande	Foster Ranch, Langtry, TX	29°46'40.91" N, 101°45'13.22" W
516	Rio Grande	El Paso, TX	31°47'55.00" N, 31°48'15.00" W
Savannah River Basin			
333	Savannah	Augusta, GA	33°22'00.18" N, 81°56'46.44" W
334	Savannah	Sylvania, GA	33°01'16.86" N, 81°31'04.50" W
335	Savannah	Port Wentworth, GA	32°13'26.34" N, 81°08'47.04" W
Yukon River Basin			
301	Yukon	Eagle, AK	65°22'28.56" N, 142°30'20.88" W
302	Porcupine	Fish Hook Bend, AK	67°12'50.94" N, 142°09'58.56" W
303	Porcupine	Fort Yukon, AK	66°35'21.06" N, 145°19'59.40" W
304	Ray	Near the Bridge, AK	65°53'01.26" N, 149°48'00.72" W
305	Tanana	Fairbanks, AK	64°45'02.52" N, 148°03'15.84" W
306	Tolovana	Nenana, AK	64°56'56.40" N, 149°44'13.44" W
307	Yukon	Tanana, AK	65°08'09.72" N, 152°24'54.72" W
308	Yukon	Galena, AK	64°41'56.22" N, 156°58'36.30" W
309	Innoko	Innoko NWR, AK	63°38'33.66" N, 158°00'43.56" W
310	Yukon	Kotlik, AK	63°02'12.18" N, 163°34'22.56" W

^aReference site in original study (Schmitt 2002)

Summary statistics including the number of detected concentrations, unweighted geometric means, median, and maximum concentrations were computed to examine distributions of each chemical contaminant for the entire dataset. The 85th percentile also was computed to compare to historical NCBP concentrations (Schmitt et al. 1999). Organochlorine residues and elemental contaminant concentrations were compared to toxicity thresholds based on whole-body fish concentrations and wildlife toxicity values from the scientific literature. Criteria for inclusion of a toxicity threshold in our evaluation were that the threshold was based on a whole-body fish concentration and associated with reproductive performance, growth, or survival. Wildlife toxicity values were used when whole-body toxicity threshold were not available. Censored values (i.e., concentrations less than the detection limit) were replaced by one half the detection limit in all figures.

Screening level wildlife risk analysis

Risk to piscivorous wildlife was evaluated with models based on adult dietary exposure or tissue

concentrations. Dietary-based toxicity reference values (TRVs) based on no observed adverse effect levels (NOAELs) from the scientific literature were used for most contaminants (Linder et al. 1980; Hill and Camardese 1986; Sample et al. 1996; Tillitt et al. 1996; USEPA 1995). An adjustment factor for differences in body size was applied when the mammal NOAEL TRV was not based on a mink study; a scaling factor of 1 was used for interspecies extrapolation among birds (Sample et al. 1996). Consensus TRVs adopted by the USEPA were used for arsenic, cadmium, chromium, copper, nickel, and lead (USEPA 1995; 2005a, b, c, d; 2007a, b). Tissue-based TRVs developed from NOAEL egg concentrations were used for total DDT, total PCB, and TCDD-EQ in the bald eagle model because developing embryos are more sensitive to these contaminants than adult birds (Elliott and Harris 2001). The TRV for TCDD-EQ was based on a toxic equivalent that was developed using consensus toxic equivalent factors (van den Berg et al. 1998; Elliott and Harris 2001). A tissue-based TRV was also used for mercury in the bald eagle model (Giesy et al. 1995) after the use of a dietary-based TRV (0.0064 mg/kg/d; Sample

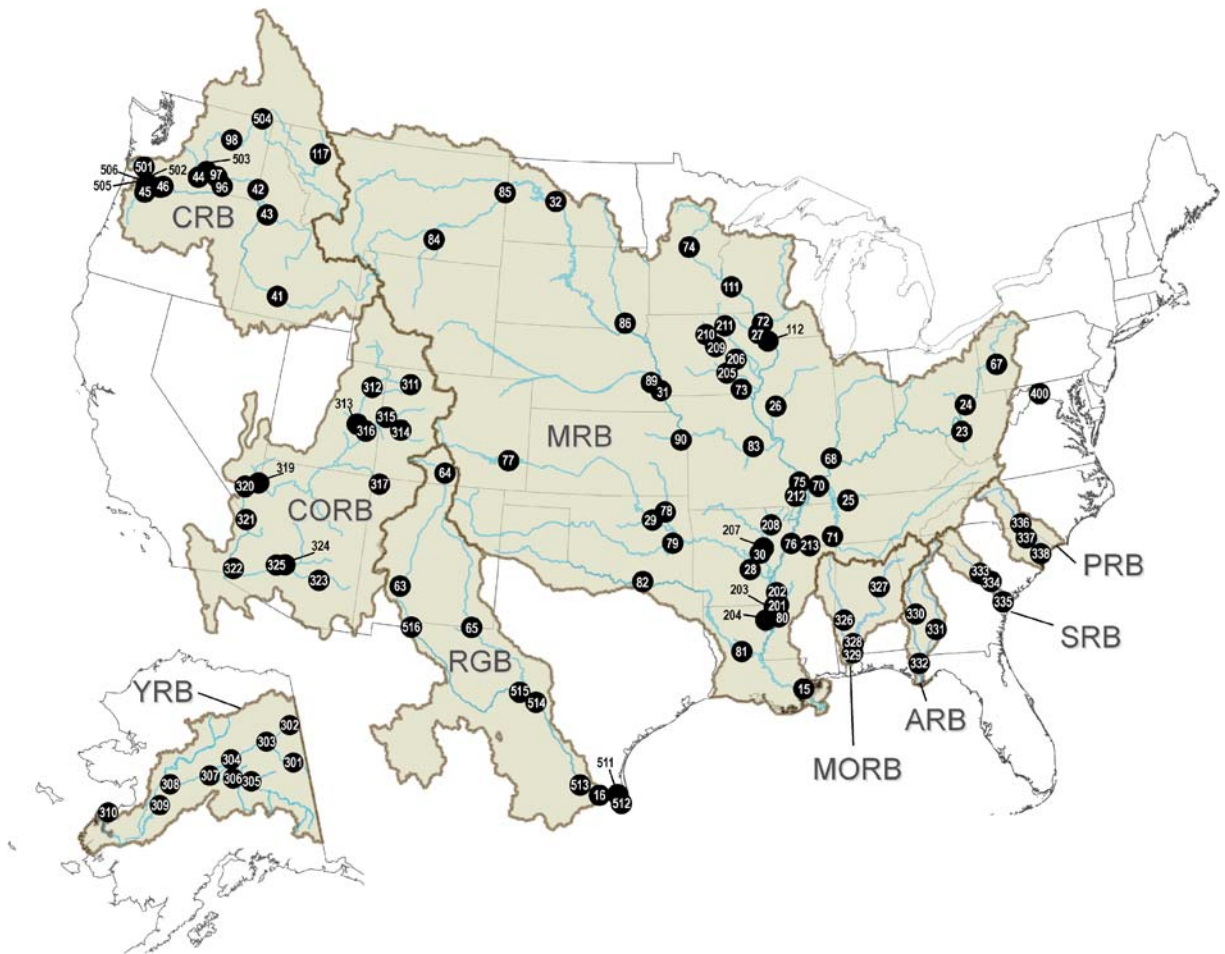


Fig. 1 Freshwater fish collection sites in the Yukon River Basin (YRB), Columbia River Basin (CRB), Colorado River Basin (CORB), Rio Grande Basin (RGB), Mississippi River Basin (MRB), Mobile

River Basin (MORB), Apalachicola River Basin (ARB), Savannah River Basin (SRB), and Pee Dee River Basin (PRB). See Table 1 for specific site information

Table 2 Benthivorous and piscivorous species collected from each basin

River Basin	Benthivores	Piscivores
Yukon	Longnose sucker	Northern pike, burbot
Columbia	Common carp, largescale sucker	Largemouth bass, smallmouth bass, northern pikeminnow, walleye, rainbow trout
Colorado	Common carp, white sucker	Largemouth bass, smallmouth bass, channel catfish, brown trout
Rio Grande	Common carp	Largemouth bass, smallmouth bass, spotted bass, white bass, northern pike, channel catfish
Mississippi	Common carp, white sucker, smallmouth buffalo	Largemouth bass, smallmouth bass, spotted bass, striped bass, white bass, northern pike, goldeye, sauger, brown trout
Mobile	Common carp	Largemouth bass
Apalachicola	Common carp	Largemouth bass
Savannah	Common carp, redhorse sucker	Largemouth bass
Pee Dee	Common carp	Largemouth bass

Table 3 Detection limits ($\mu\text{g/g}$ wet weight unless otherwise noted) for organochlorine residues

Analyte	Mississippi River Basin (1995)	Rio Grande and Columbia River Basins (1997)	Yukon River Basin (2002)	Colorado River Basin (2003)	Mobile, Apalachicola, Savannah, and Pee Dee River Basins (2004)
Pentachlorobenzene	NM	NM	0.00005–0.00006	0.00007	0.00007
Hexachlorobenzene	0.01	0.01	0.0021–0.0025	0.00026	NA
Pentachloroanisole	NM	NM	0.00023–0.00025	0.00013	0.00010
α -HCH	0.01	0.01	0.00018–0.00020	0.00008	0.00010
β -HCH	0.01	0.01	0.00017–0.00019	0.00019	0.00030
γ -HCH	0.01	0.01	0.00039–0.00044	0.00051	0.00010
δ -HCH	0.01	0.01	0.00010–0.00011	0.00005	0.00010
Aldrin	NM	NM	NA	0.00027	0.00009
Dieldrin	0.01	0.01	0.00016	0.00015	0.00008
Endrin	0.01	0.01	0.00011–0.00012	0.00010	0.00025
Dacthal	NM	NM	0.00029–0.00032	0.00050	0.00013
Heptachlor	NM	NM	0.00019–0.00021	0.00019	0.00010
Heptachlor epoxide	0.01	0.01	0.00019–0.00021	0.00001	0.00010
Oxychlordane	0.01	0.01	0.00008–0.00009	0.00008	0.00010
<i>cis</i> -chlordane	0.01	0.01	0.00019–0.00022	0.00004	0.00028
<i>trans</i> -chlordane	0.01	0.01	0.00031–0.00035	0.00023	NA
<i>cis</i> -nonachlor	0.01	0.01	0.00008–0.00009	0.00010	NA
<i>trans</i> -nonachlor	0.01	0.01	0.00021–0.00023	NA	NA
<i>o,p'</i> -DDE	0.01	0.01	0.00015–0.00016	0.00008	0.00081
<i>o,p'</i> -DDD	0.01	0.01	0.00061–0.00070	0.00055	0.00010
<i>o,p'</i> -DDT	0.01	0.01	0.00012–0.00013	0.00053	0.00010
<i>p,p'</i> -DDE	0.01	0.01	0.00039–0.00042	NA	NA
<i>p,p'</i> -DDD	0.01	0.01	0.00038–0.00043	0.00067	NA
<i>p,p'</i> -DDT	0.01	0.01	0.0014–0.0016	0.0014	0.00047
Endosulfan I	NM	NM	0.00034–0.00039	0.00021	0.00010
Endosulfan II	NM	NM	0.00014–0.00019	0.00017	0.00010
Endosulfan sulfate	NM	NM	0.00079–0.00090	0.00005	0.00014
Methoxychlor	NM	NM	0.0017–0.0019	0.00035	0.00010
Mirex	0.01	0.01	0.00001	0.00005	0.00010
PCB	0.05	0.03	0.02	0.048	NA
Toxaphene	0.05	0.03	0.011	0.024	0.010
TCDD-EQ (pg/g)	0.2–1.0	0.4–1.3	1.7	0.1–0.5	1.0–2.6

Study year is shown in parentheses

HCH Hexachlorocyclohexane, *PCB* polychlorinated biphenyl, *NM* analyte not measured, *NA* all concentrations were greater than the detection limit

et al. 1996) was found to be too conservative, resulting in risk at most sites ($n=105$). Models assuming a diet of 100% fish were used to screen for risk. Site-specific data for other exposure pathways such as water and sediment were not available for all sites and were not considered in this risk evaluation; exposure from these other pathways was expected to be low and therefore would contribute little to the overall risk. A no effects hazard concentration (NEHC) was calculated for each spe-

cies modeled using the equation $\text{NEHC } (\mu\text{g/g}) = [\text{dietary-based NOAEL TRV (mg/kg body weight/day)}] / \text{food ingestion rate (kg food/kg body weight/day)}$ or $[\text{tissue-based NOAEL TRV (mg/kg egg)}] / \text{biomagnification factor (BMF)}_{\text{fish} \rightarrow \text{egg}}$. The bald eagle-specific BMFs were 28 for total PCBs, 19 for TCDD-EQ, 22 for total DDT, and one for mercury (Giesy et al. 1995). Piscivorous wildlife may be at risk from a contaminant if the measured concentration in the fish composite sample exceeds the NEHC. Risk

Table 4 Detection limits (µg/g wet weight) for elemental contaminants

Analyte	Mississippi River Basin (1995)	Rio Grande and Columbia River Basins (1997)	Yukon River Basin (2002)	Colorado River Basin (2003)	Mobile, Apalachicola, Savannah, and Pee Dee River Basins (2004)
Arsenic	0.11–0.27	0.03–0.32	NA	NA	0.01
Cadmium	0.02–0.05	0.02–0.06	0.02–0.03	0.02–0.03	0.01
Chromium	0.16	NA	0.22–0.27	0.24–0.35	NA
Copper	NA	NA	NA	NA	NA
Mercury	0.04–0.05	0.02–0.06	NA	NA	NA
Nickel	0.16–0.26	0.22–0.33	0.21–0.27	0.24–0.35	0.01
Lead	0.01–0.04	0.03–0.14	0.21–0.27	0.24–0.35	0.01
Selenium	0.12–0.18	0.22–0.27	NA	NA	NA
Zinc	NA	NA	NA	NA	NA

Study year is shown in parentheses

NA All concentrations were greater than the detection limit

was categorized as none (<NEHC), low (1–10× NEHC), or high (>10× NEHC). Model species considered in the risk analysis included the bald eagle as an avian receptor and mink as a mammalian receptor. Both of these species have distributions that span most of the United States and consume the large, adult fish such as those collected by the BEST-LRMN Project. Some contaminants we evaluated have been associated with the death of bald eagles and mink. For example, reproductive impairment of mink has been directly related to PCBs in Great Lakes fish (Aulerich and Ringer 1977; Heaton et al. 1995), and mercury has been implicated in increased mink mortality (Wren et al. 1987). Bald eagle survival and reproduction have been negatively affected by dieldrin, *p,p'*-DDE, and PCBs in the environment (Reichel et al. 1984; Wiemeyer et al. 1984). Benthivorous and piscivorous fish were examined separately because fish in higher trophic levels (piscivorous fish) generally have higher concentrations of accumulative contaminants such as total PCBs and mercury, but benthivorous fish species are the preferred prey in some bald eagle populations (Haywood and Ohmart 1986).

Results

Environmental contaminant concentrations

Organochlorine concentrations were less than the detection limits in most samples (Table 5). *Trans*-nonachlor, *p,p'*-DDE, *p,p'*-DDD, total PCBs, and TCDD-EQ were the most frequently detected com-

pounds. Higher detection limits for the Mississippi, Rio Grande, and Columbia River Basins than for those other basins sampled influenced mean and median concentrations of many organochlorine residues (Table 5). Mean concentrations ranged from 0.00007 to 0.08089 µg/g, and median concentration ranged from 0.00007 to 0.0723 µg/g (most <0.01 µg/g). Mean and median concentrations were greatest for total chlordanes, *p,p'*-DDE, total DDT, total PCBs, and toxaphene (Table 5). The 85th percentile ranged from 0.00021 µg/g for pentachlorobenzene and heptachlor to 0.450 µg/g for total PCBs and were >0.02 µg/g for *trans*-nonachlor, total chlordanes, *p,p'*-DDE, *p,p'*-DDD, total DDT, total PCBs, and toxaphene (Table 5). Maximum concentrations ranged from 0.00111 µg/g for aldrin to 11.37 µg/g for total DDT but were <0.08 µg/g for most organochlorine residues (Table 5). The median TCDD-EQ concentration was 0.0000017 µg/g (Table 5). Mean concentrations of select organochlorine pesticides and total PCBs were less than those reported in fish from the historical NCBP (Table 5). Mean concentrations of Dacthal and heptachlor were tenfold lower than those measured in NCBP samples, and mean concentrations of chlordane components (oxychlordane, *cis*- and *trans*-chlordanes and nonachlors) also were lower in BEST-LRMN samples (0.002–0.005 µg/g) than NCBP samples (0.007–0.011 µg/g; Table 5). Mean *p,p'*-homolog concentrations (0.003–0.045 µg/g) were lower than those reported in NCBP samples (0.010–0.070 µg/g; Table 5). Concentrations of total PCBs and toxaphene also were lower in BEST-LRMN samples compared to those in NCBP samples.

Table 5 Organochlorine chemical residue concentrations ($\mu\text{g/g}$ wet weight) in whole-body fish composite samples from 1995 to 2004

Analyte	1995–2004					NCBP
	No. >DL/n	Mean	Median	85th	Maximum	Mean
Lipid (%)	409/409	4.89	4.96	8.99	18.10	N/A
Pentachlorobenzene	56/135	0.00007	0.00007	0.00021	0.00119	N/A
Hexachlorobenzene	110/409	0.00277	0.01	0.01	0.068	<0.01
Pentachloroanisole	113/135	0.00056	0.00057	0.00258	0.021	<0.01
α -HCH	62/409	0.00146	0.01	0.01	0.01	0.005
β -HCH	55/409	0.00192	0.01	0.01	0.01	N/A
γ -HCH	40/409	0.00178	0.01	0.01	0.01	<0.01
δ -HCH	54/409	0.00134	0.01	0.01	0.01	N/A
Aldrin	38/135	0.00012	0.00019	0.00027	0.00111	N/A
Dieldrin	198/409	0.00411	0.01	0.017	0.250	0.008
Endrin	76/409	0.00215	0.01	0.01	0.71	0.005
Dacthal	46/135	0.00025	0.00032	0.00070	0.00928	0.007
Heptachlor	14/135	0.00009	0.00019	0.00021	0.00382	0.007
Heptachlor epoxide	115/409	0.00189	0.01	0.01	0.075	N/A
Oxychlorodane	116/409	0.00229	0.01	0.01	0.028	0.007
<i>cis</i> -chlordane	175/409	0.00374	0.01	0.013	0.120	0.011
<i>trans</i> -chlordane	150/409	0.00307	0.01	0.01	0.35	0.008
<i>cis</i> -nonachlor	172/409	0.00351	0.01	0.01	0.047	0.009
<i>trans</i> -nonachlor	229/409	0.00548	0.01	0.02	0.31	0.013
Σ Chlordane	N/A	0.0236	0.03	0.057	0.529	N/A
<i>o,p'</i> -DDE	89/409	0.00238	0.01	0.01	0.022	N/A
<i>o,p'</i> -DDD	99/409	0.00283	0.01	0.01	0.34	N/A
<i>o,p'</i> -DDT	67/409	0.00189	0.01	0.01	0.24	N/A
<i>p,p'</i> -DDE	382/409	0.0447	0.05	0.29	8.30	0.070
<i>p,p'</i> -DDD	250/409	0.00907	0.01	0.04	2.80	0.019
<i>p,p'</i> -DDT	88/409	0.00332	0.01	0.01	0.31	0.010
Σ DDT	N/A	0.0809	0.07	0.38	11.37	0.110 ^a
Endosulfan I	13/135	0.00011	0.00021	0.00037	0.00390	N/A
Endosulfan II	47/135	0.00015	0.00017	0.00050	0.05400	N/A
Endosulfan sulfate	80/135	0.00054	0.00079	0.00220	0.07900	N/A
Methoxychlor	15/135	0.00021	0.00035	0.00180	0.00960	N/A
Mirex	118/409	0.00183	0.01	0.01	0.08	0.006
PCB	256/409	0.084	0.067	0.450	3.30	0.335
Toxaphene	83/409	0.03	0.05	0.05	0.83	0.066
TCDD-EQ	255/409	0.0000015	0.0000017	0.000007	0.0000680	N/A

Number of samples with a concentration greater than the detection limit (DL), geometric mean, median, and 85th percentile concentrations are presented. The geometric mean from 1986 National Contaminant Biomonitoring Program (NCBP) samples also is provided for comparison (Schmitt et al. 1999). Σ Chlordane is the sum of heptachlor epoxide, oxychlorodane, *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, and *trans*-nonachlor, and Σ DDT is the sum of *o,p'*- and *p,p'*-DDE, DDD, and DDT. Censored values were represented by one half the detection limit in the summed values

N/A not applicable

Elemental contaminant concentrations exceeded detection limits in most samples (Table 6). Mean concentrations ranged from 0.03 $\mu\text{g/g}$ for cadmium to 35.2 $\mu\text{g/g}$ for zinc, and median concentrations ranged from 0.04 to 36.0 $\mu\text{g/g}$ (Table 6). The 85th percentile was <1.0 $\mu\text{g/g}$ for arsenic, cadmium, mercury, nickel,

lead, and selenium and >1.0 $\mu\text{g/g}$ for chromium, copper, and zinc. Maximum concentrations ranged from 0.51 $\mu\text{g/g}$ for cadmium to 150 $\mu\text{g/g}$ for zinc but were <10 $\mu\text{g/g}$ for most elemental contaminants (Table 6). Mean concentrations of arsenic, cadmium, copper, mercury, lead, selenium, and zinc were greater

Table 6 Elemental contaminant concentrations ($\mu\text{g/g}$ wet weight) in whole-body fish composite samples from 1995 to 2004

Analyte	1995–2004					NCBP	
	No.>DL/ <i>n</i>	Mean	Median	85th	Maximum	Mean	85th
Moisture (%)	409/409	72.2	72.6	75.2	79.9	N/A	N/A
Arsenic	233/409	0.11	0.19	0.27	1.95	0.08	0.24
Cadmium	180/409	0.03	0.04	0.09	0.51	0.01	0.04
Chromium	393/409	0.76	0.60	2.38	71.8	N/A	N/A
Copper	409/409	0.80	0.86	1.26	3.92	0.79	1.7
Mercury	409/409	0.13	0.13	0.27	0.78	0.09	0.18
Nickel	274/409	0.30	0.30	0.73	5.59	N/A	N/A
Lead	233/409	0.07	0.10	0.27	9.29	0.06	0.21
Selenium	398/409	0.59	0.53	0.98	4.66	0.42	0.66
Zinc	409/409	35.2	36.0	76.0	150	21.2	31.7

Number of samples with a concentration greater than the detection limit (DL), geometric mean, median, and 85th percentile concentrations are presented. The geometric mean and 85th percentiles from 1986 NCBP samples are provided for comparison (Schmitt et al. 1999)

N/A Not applicable

than those reported in fish from the historical NCBP (Table 6). Most differences in mean concentrations between BEST-LRMN samples and NCBP samples were $<0.04 \mu\text{g/g}$, but mean differences for selenium ($0.17 \mu\text{g/g}$) and zinc ($14.0 \mu\text{g/g}$) were greater (Table 6). The 85th percentile concentrations of arsenic, cadmium, mercury, lead, selenium, and zinc also were greater than those reported in fish from the NCBP (Table 6). Differences in mean concentrations of arsenic, cadmium, mercury, and lead were $<0.10 \mu\text{g/g}$, but differences in copper ($0.44 \mu\text{g/g}$), selenium ($0.32 \mu\text{g/g}$), and zinc ($54.8 \mu\text{g/g}$) were greater (Table 6).

Toxicity thresholds

Concentrations in whole-body fish samples exceeded literature-based toxicity thresholds for dieldrin, total chlordane, total DDT, total PCBs, toxaphene, TCDD-EQ, cadmium, chromium, mercury, lead, selenium, and zinc (Table 7). Concentrations of hexachlorobenzene, arsenic, and copper did not exceed toxicity thresholds for fish and wildlife at any sampling location. Toxicity thresholds meeting our criteria were not found for pentachlorobenzene, pentachloroanisole, α -hexachlorocyclohexane (HCH), β -HCH, δ -HCH, aldrin, endrin, Dacthal, endosulfans, methoxychlor, mirex, or nickel (Table 7). Concentrations of some contaminants differed between trophic levels. Concentrations of dieldrin, endrin, total chlordane, total DDT, total PCBs, toxaphene, TCDD-EQ, chromium, and selenium generally were similar in benthivorous and piscivorous

fish (Fig. 2). Concentrations of endrin, total DDT, and toxaphene in Mississippi River Basin samples were greater in benthivores compared to piscivores (Fig. 2). Concentrations of cadmium, lead, and zinc were greater in benthivores than in piscivores, but mercury was greater in piscivorous fish than in benthivorous fish (Fig. 2).

The greatest concentrations and therefore the greatest exceedences of toxicity thresholds were found in Mississippi River Basin samples. Toxicity thresholds for dieldrin and total chlordane were exceeded by only a few samples from the Mississippi River Basin (Fig. 2); carp samples from Sites 76 and 206 exceeded thresholds for both of these contaminants. Total DDT concentrations exceeded toxicity thresholds in 126 samples representing all basins except the Yukon, Savannah, and Pee Dee River Basins and were greatest in samples from the Columbia, Colorado, Rio Grande, and Mississippi River Basins (Fig. 2). A total of 172 samples representing all basins except the Rio Grande and Yukon River Basins exceeded protective thresholds for total PCBs. Total PCB concentrations were greatest in samples from the Columbia, Colorado, Mississippi, Mobile, and Apalachicola River Basins (Fig. 2). Toxaphene concentrations exceeded toxicity thresholds in 42 samples representing all basins except the Savannah River Basin but were generally greatest in samples from the Columbia, Rio Grande, and Mississippi River Basins (Fig. 2). TCDD-EQ concentrations exceeded toxicity thresholds for fish

Table 7 Toxicity thresholds for fish and wildlife and the number of samples and sites that exceed these concentrations^a

Contaminant	Toxicity threshold		No. of samples ^b	No. of sites ^b
	Conc. (µg/g)	Reference		
Hexachlorobenzene	0.33	Newell et al. 1987	0	0
γ-HCH	0.1	Newell et al. 1987	0	0
Dieldrin	0.12–5.65	Newell et al. 1987; Shubat & Curtis 1986; Peakall 1996	5	2
ΣChlordane	0.30	Eisler 1990	5	2
ΣDDT	0.15–3.0	Anderson et al. 1975; Blus 1996; Beckvar et al. 2005	126	42
PCB	0.11–0.48	Hornshaw et al. 1983; Newell et al. 1987	172	54
Toxaphene	0.035–0.90	Mayer et al. 1975	42	18
TCDD-EQ	0.0000044–0.00003	Nosek et al. 1992; Heaton et al. 1995; Tillitt et al. 1996; Walker et al. 1996	108	49
Arsenic	2.2–11.6	Gilderhus 1966; McGeachy and Dixon 1992	0	0
Cadmium	0.23–15.6	Spehar 1976; Eisler 1985; Hansen et al. 2002	17	12
Chromium	1.0	Eisler 1986	121	59
Copper	11.1–42.0	Stouthart et al. 1996	0	0
Mercury	0.1–0.3	Barr 1986; Yeardeley et al. 1998; Beckvar et al. 2005;	278	99
Lead	0.4–8.8	Holcombe et al. 1976	12	9
Selenium	0.75–1.0	Lemly 1996; Hamilton 2004	101	43
Zinc	40–64	Spehar 1976	198	97

Toxicity thresholds for dieldrin, toxaphene, arsenic, cadmium, lead, selenium, and zinc were based on whole-body fish concentrations. Toxicity thresholds for hexachlorobenzene, γ-HCH, ΣChlordane, ΣDDT, PCB, TCDD-EQ, and chromium were based on wildlife toxicity values. Toxicity thresholds were not available for pentachlorobenzene, pentachloroanisole, α-HCH, β-HCH, δ-HCH, aldrin, endrin, Dacthal, endosulfans, methoxychlor, mirex, and nickel. ΣChlordane is the sum of heptachlor epoxide, oxychlordane, *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, and *trans*-nonachlor, and ΣDDT is the sum of *o,p'*- and *p,p'*-DDE, DDD, and DDT

^aNumber of samples=409 for all contaminants except for TCDD-EQ ($n=408$). Number of sites=111

^bThe number of samples or sites to exceed lowest toxicity threshold available for a contaminant

and wildlife in 108 samples representing all basins except the Yukon and Savannah River Basins and were greatest in samples from the Columbia, Mississippi, and Mobile River Basins (Fig. 2). Cadmium concentrations were generally greater in carp and other benthivores than in piscivores and exceeded toxicity thresholds in 17 samples representing the Columbia, Colorado, and Mississippi River Basins (Fig. 2). Conversely, mercury concentrations were consistently greater in piscivores and exceeded toxicity thresholds in 278 samples representing all basins (Fig. 2). Protective toxicity thresholds for lead were exceeded by 12 samples representing the Columbia, Rio Grande, Mississippi, and Apalachicola River Basins, and concentrations were greatest in benthivore samples from the Columbia River Basin (Fig. 2). Selenium concentrations exceeded toxicity thresholds in 101 samples representing all basins except the Savannah and Pee Dee River Basins and were

greatest in samples from the Colorado, Rio Grande, and Mississippi River Basins (Fig. 2). Zinc concentrations were generally greater in carp and other benthivores than piscivores, but concentrations in northern pike were greater than longnose sucker from the Yukon River Basin. Zinc exceeded toxicity thresholds in 198 samples representing all basins (Fig. 2).

Screening level wildlife risk analysis

Concentrations of dieldrin, endrin, total DDT, total PCBs, and TCDD-EQ in whole-body fish samples represented a risk to piscivorous wildlife at one or more sites. No risk of dieldrin and endrin exposure to bald eagle and mink was found at most sites. Dieldrin concentrations at two Mississippi River Basin sites (Sites 76 and 206; 0.15–0.25 µg/g) exceeded the NEHC for mink (Figs. 2 and 3), and endrin concentrations in samples from Site 76 (0.22–0.71 µg/g) also

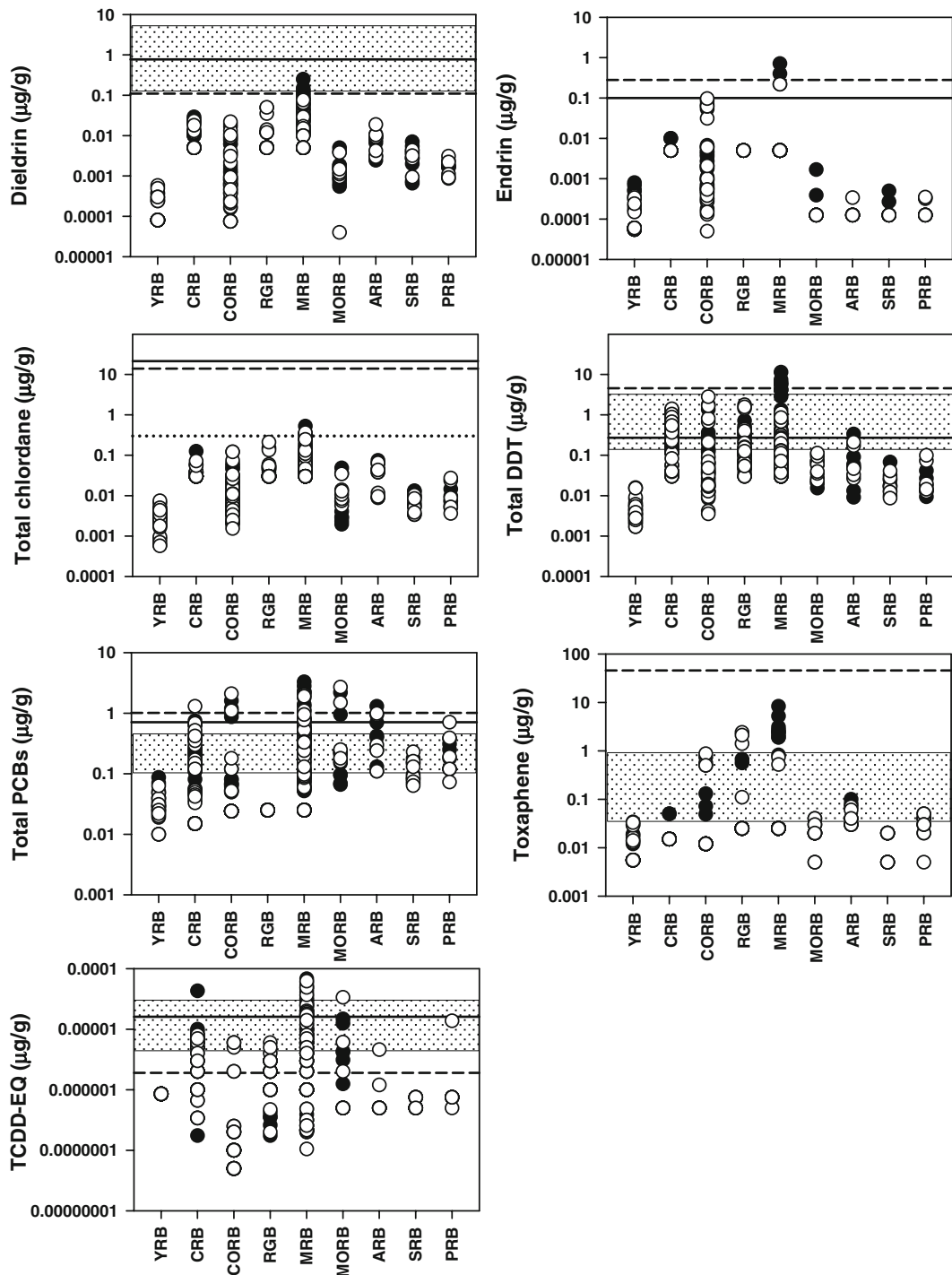


Fig. 2 Organochlorine residue and elemental concentrations (all $\mu\text{g/g}$ wet-weight) in whole-body composite samples of benthivorous (*black circles*) and piscivorous (*white circles*) fish from the Yukon River Basin (*YRB*), Columbia River Basin (*CRB*), Colorado River Basin (*CORB*), Rio Grande Basin (*RGB*), Mississippi River Basin (*MRB*), Mobile River Basin (*MORB*), Apalachicola River Basin (*ARB*), Savannah River

Basin (*SRB*), and Pee Dee River Basin (*PRB*). Literature-based toxicity thresholds are represented by *dotted lines* or *boxes* (see Table 7 for reference). No effect hazard concentrations (NEHCs) for bald eagle (*solid line*) and mink (*dashed line*) are presented for each contaminant (see Table 8 for references and text for calculation)

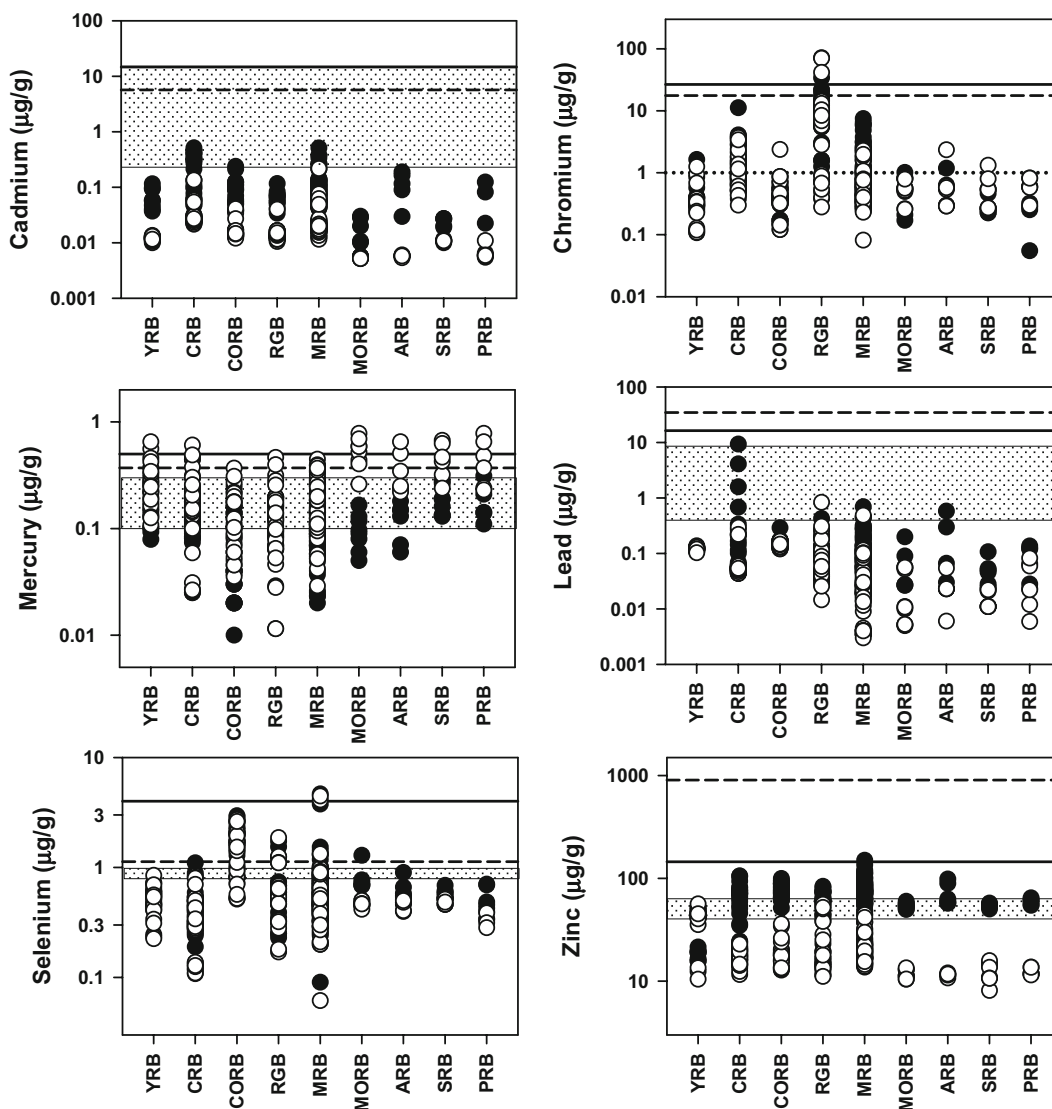


Fig. 2 (continued)

exceeded the NEHC for bald eagle and mink (Fig. 2). Total DDT concentrations did not represent a risk to bald eagle at sites from the Yukon, Mobile, Savannah, and Pee Dee River Basins (Fig. 3). Total DDT concentrations exceeded the NEHC for bald eagle in samples from the Columbia River Basin (34 samples from 12 sites; 0.27–1.41 $\mu\text{g/g}$), Colorado River Basin (nine samples from three sites; 0.28–2.80 $\mu\text{g/g}$), Rio Grande Basin (12 samples from four sites; 0.30–1.78 $\mu\text{g/g}$), Mississippi River Basin (36 samples from 16 sites; 0.27–11.37 $\mu\text{g/g}$), and Apalachicola River Basin (one sample from Site 330; 0.33 $\mu\text{g/g}$; Figs. 2 and 3). The risk of total DDT to mink was lower with few NEHC exceedences. Carp from Sites

201, 202, 203, and 204 in the Mississippi River Basin (5.49–11.37 $\mu\text{g/g}$) were the only samples with total DDT concentrations that exceeded the NEHC for mink (Figs. 2 and 3). Total PCB concentrations did not represent a risk to piscivorous wildlife at sites in the Yukon, Rio Grande, Savannah, and Pee Dee River Basins (Fig. 3). Total PCB concentrations exceeded the NEHC for bald eagle in samples from the Columbia River Basin (two samples from two sites; 0.75–1.3 $\mu\text{g/g}$), Colorado River Basin (six sample from two sites; 0.87–2.1 $\mu\text{g/g}$), Mississippi River Basin (20 samples from nine sites; 0.75–3.3 $\mu\text{g/g}$), Mobile River Basin (four samples from Site 327; 0.95–2.7 $\mu\text{g/g}$), and Apalachicola River Basin (three

samples from Site 330; 0.94–1.3 $\mu\text{g/g}$; Figs. 2 and 3). The risk of total PCB exposure was lower to mink than bald eagle (Fig. 3). Concentrations of total PCBs exceeded the NEHC for mink in samples from the Columbia River Basin (one sample from Site 503; 1.3 $\mu\text{g/g}$), Colorado River Basin (four samples from two sites; 1.1–2.1 $\mu\text{g/g}$), Mississippi River Basin (12 samples from five sites; 1.1–3.3 $\mu\text{g/g}$), Mobile River Basin (three samples from Site 327; 1.5–2.7 $\mu\text{g/g}$), and Apalachicola River Basin (one sample from Site 330; 1.3 $\mu\text{g/g}$; Figs. 2 and 3). No risk of TCDD-EQ exposure to bald eagle and mink was found at sites in the Yukon or Savannah River Basins (Fig. 3). TCDD-EQ concentrations exceeded the NEHC for bald eagle in samples from the Columbia River Basin (one sample from Site 96; 0.000043 $\mu\text{g/g}$), Mississippi River Basin (29 samples from 18 sites; 0.000017–0.000068 $\mu\text{g/g}$), and Mobile River Basin (one sample from Site 327; 0.000034 $\mu\text{g/g}$; Figs. 2 and 3). The risk of TCDD-EQ exposure was greater to mink than bald eagle (Fig. 3). TCDD-EQ concentrations exceeded the NEHC for mink in samples from the Columbia River Basin (38 samples from 15 sites; 0.000002–0.000043 $\mu\text{g/g}$), Colorado River Basin (six samples from three sites; 0.000002–0.000006 $\mu\text{g/g}$), Rio Grande Basin (25 samples from 10 sites; 0.000002–0.000006 $\mu\text{g/g}$), Mississippi River Basin (106 samples from 42 sites; 0.000002–0.000068 $\mu\text{g/g}$), Mobile River Basin (eight samples from four sites; 0.000002–0.000034 $\mu\text{g/g}$), Apalachicola River Basin (one sample from Site 330; 0.0000046 $\mu\text{g/g}$), and Pee Dee River Basin (one sample from Site 337; 0.000014 $\mu\text{g/g}$; Figs. 2 and 3). A NOAEL TRV was not available for pentachlorobenzene, pentachloroanisole, aldrin, Dacthal, methoxychlor, mirex, or toxaphene in bald eagle and hexachlorobenzene, pentachloroanisole, Dacthal, and mirex in mink (Table 8); therefore, the risk of these contaminants is unknown.

Chromium, mercury, selenium, and zinc concentrations in whole-body fish samples represented a risk to piscivorous wildlife at one or more sites. Chromium represented a risk to piscivorous wildlife only in the Rio Grande Basin, where concentrations were comparatively high; they exceeded the NEHC for bald eagle, mink, or both in Rio Grande samples from Sites 63 (71.8 $\mu\text{g/g}$), 511 (34.0 $\mu\text{g/g}$), 512 (18.3 $\mu\text{g/g}$), 513 (21.1–70.2 $\mu\text{g/g}$), and 515 (21.8 $\mu\text{g/g}$; Figs. 2 and 4). No risk of mercury exposure to bald eagle was associated with samples from the Colorado, Rio

Grande, or Mississippi River Basins (Fig. 2). Mercury concentrations exceeded the NEHC for bald eagle in the Yukon River Basin (two samples from two sites; 0.56–0.65 $\mu\text{g/g}$), Columbia River Basin (one sample from Site 43; 0.61 $\mu\text{g/g}$), Mobile River Basin (five samples from three sites; 0.52–0.78 $\mu\text{g/g}$), and Apalachicola-Flint-Chattahoochee River Basin (two samples from Site 332; 0.51–0.65 $\mu\text{g/g}$), Savannah River Basin (two samples from Site 334; 0.63–0.67 $\mu\text{g/g}$), and Pee Dee River Basin (two samples from Site 338; 0.65–0.78 $\mu\text{g/g}$; Figs. 2 and 4). For mink, samples from the Yukon River Basin (five samples from three sites; 0.37–0.65 $\mu\text{g/g}$), Columbia River Basin (six samples from four sites; 0.38–0.61 $\mu\text{g/g}$), Rio Grande Basin (two samples from two sites; 0.40–0.46 $\mu\text{g/g}$), Mississippi River Basin (four samples from three sites; 0.39–0.45 $\mu\text{g/g}$), Mobile River Basin (seven samples from four sites; 0.40–0.78 $\mu\text{g/g}$), Apalachicola River Basin (two samples from Site 332; 0.51–0.65 $\mu\text{g/g}$), Savannah River Basin (four samples from three sites; 0.43–0.67 $\mu\text{g/g}$), and Pee Dee River Basin (four samples from two sites; 0.37–0.78 $\mu\text{g/g}$) exceeded the NEHC for mercury (Figs. 2 and 4). Selenium did not represent a risk to piscivorous wildlife at any site in the Yukon, Columbia, Apalachicola, Savannah, or Pee Dee River Basins (Figs. 2 and 4). Selenium concentrations exceeded the NEHC for bald eagle at one site in the Mississippi River Basin (Site 77; 4.11–4.66 $\mu\text{g/g}$) and for mink in samples from the Colorado River Basin (34 samples from 12 sites; 1.29–2.95 $\mu\text{g/g}$), Rio Grande Basin (six samples from three sites; 1.24–1.87 $\mu\text{g/g}$), Mississippi River Basin (ten samples from three sites; 1.18–4.66 $\mu\text{g/g}$), and the Mobile River Basin (one sample from Site 329; 1.29 $\mu\text{g/g}$; Figs. 2 and 4). No risk of zinc exposure to piscivorous wildlife was found at most sites; zinc concentrations in one Mississippi River Basin carp sample (Site 79; 150 $\mu\text{g/g}$) represented a risk to bald eagle (Fig. 2). Concentrations of arsenic, cadmium, copper, nickel, and lead did not represent a risk to wildlife at any site.

Discussion

Overall, mean concentrations of organochlorine residues were less than historical NCBP concentrations; concentrations of arsenic, cadmium, copper, mercury,

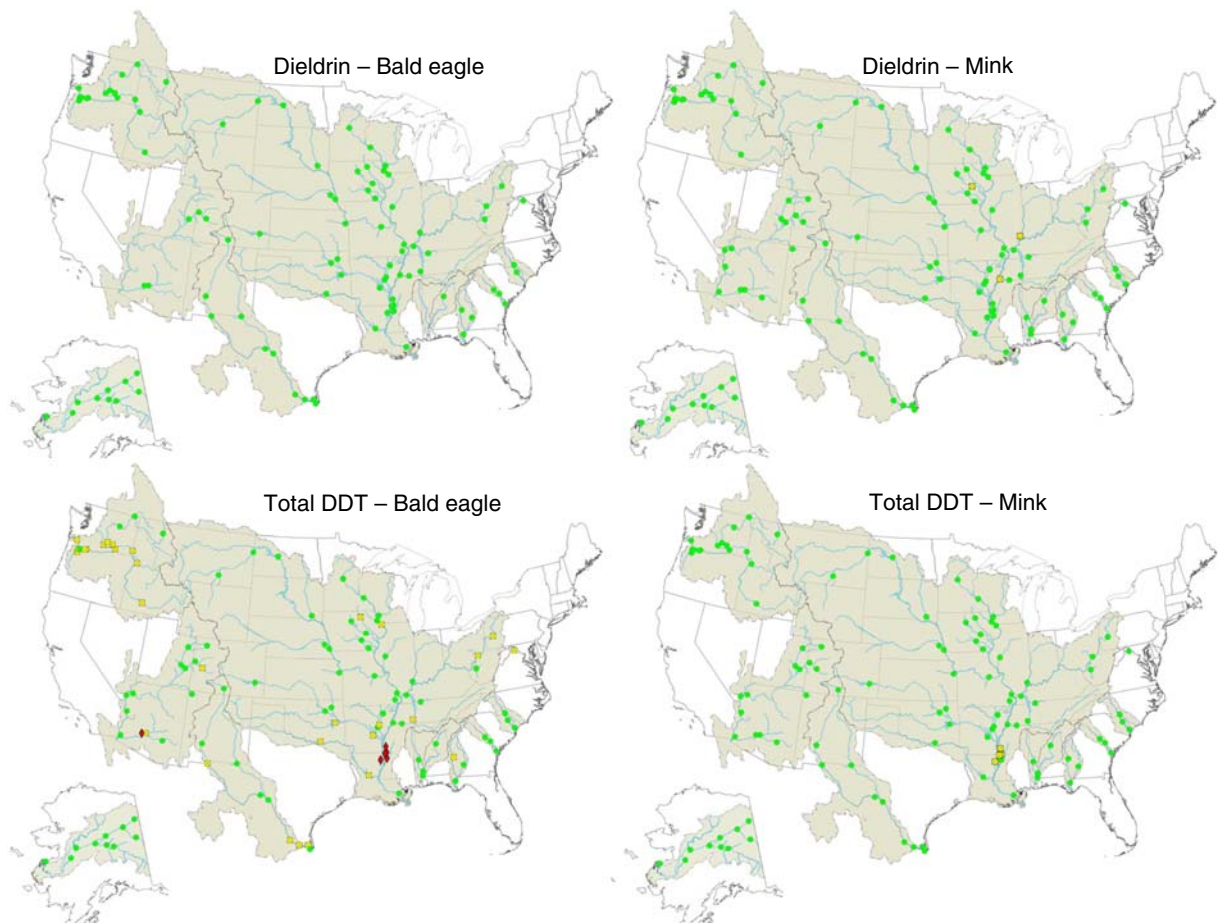


Fig. 3 Sites that exceed the no effects hazard concentration (NEHC) for selected organochlorine residues. See Fig. 1 for specific site locations and Table 8 for NEHC concentrations

lead, selenium, and zinc were greater than NCBP concentrations. Direct comparison of these datasets is precluded by collection location and species composition differences, which can bias mean concentrations. Concentrations of organochlorine pesticides and total PCBs were expected to decline because their use has been banned or regulated in the United States. However, high concentrations of organochlorine chemical residues remain evident in fish near point sources such as former manufacturing facilities and areas of high historical use. Elemental contaminants emanating from point sources have also been regulated, as have many pesticides containing potentially toxic trace elements. However, elemental contaminants also are natural constituents of the earth's crust; some are essential trace elements for terrestrial and aquatic organisms. In spite of regulation, elemental

concentrations in fish remain elevated in some areas due to irrigated agriculture (Hamilton 2004), mining (Schmitt et al. 2006), and the combustion of fossil fuels (Baumann and Gillespie 1986; Yearley et al. 1998), and both organochlorine and elemental contaminants are distributed globally by atmospheric transport. Considered independently, concentrations of most organochlorine residues and elemental contaminants we evaluated represented no or low risk to bald eagle and mink. Among the contaminants evaluated, the greatest risk to bald eagle and mink was associated with total DDT, total PCBs, TCDD-EQ, mercury, and selenium. The risk of pentachloroanisole, aldrin, Dacthal, methoxychlor, mirex, or toxaphene to bald eagle or mink was unknown; NEHCs could not be calculated because NOAELS were not available for these compounds.

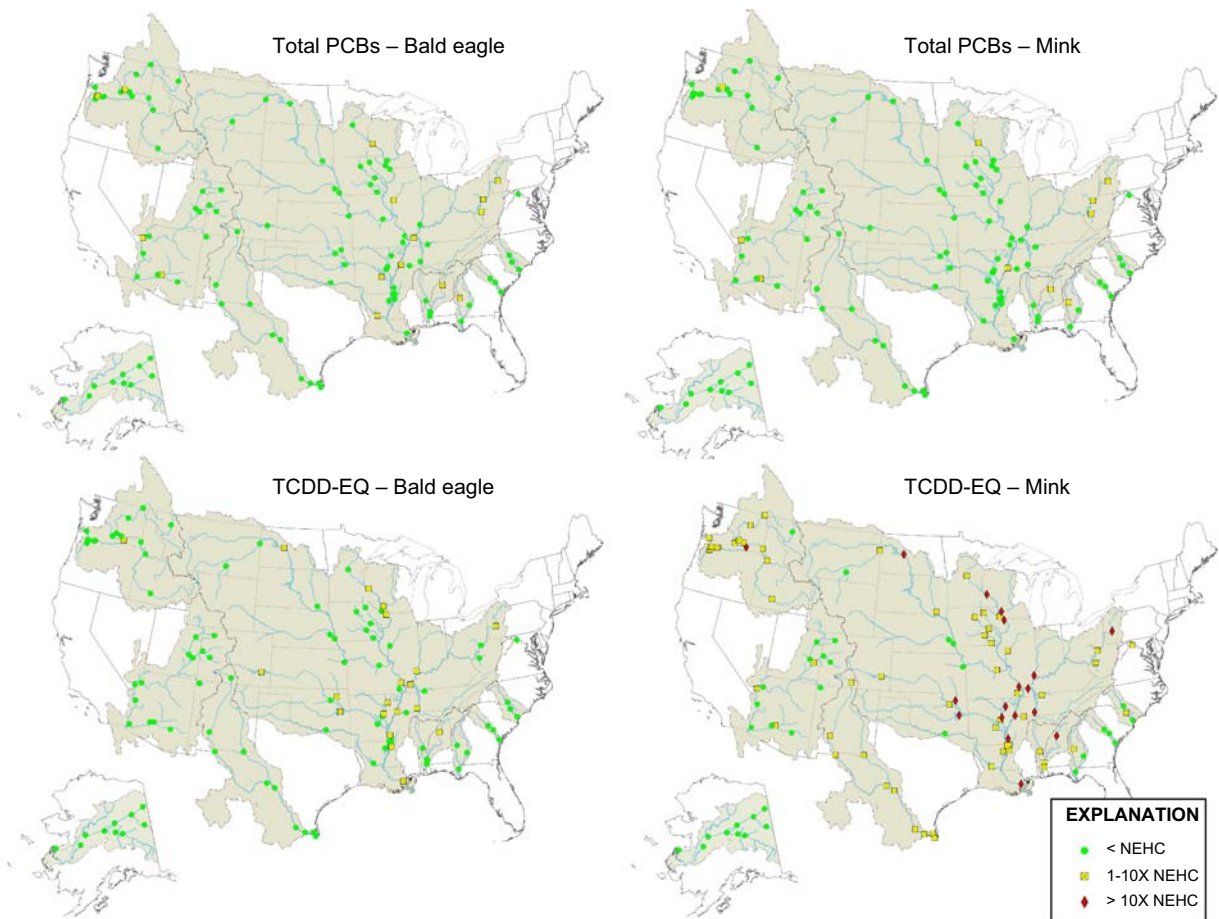


Fig. 3 (continued)

Environmental residues of DDT and its degradation products persist in many areas of historic use, primarily in agricultural areas and near manufacturing and formulation sites, even though the insecticide has been banned in the United States since 1972. Total DDT concentrations remained high in whole-body fish samples from multiple sampling sites in our study and represented a greater risk to bald eagle than mink. Total DDT concentrations exceeded the NEHC for bald eagle in at least one sample from all basins except the Yukon and Savannah River Basins, and the risk was greatest at sites in agricultural areas of the lower Columbia, Mississippi, Rio Grande and Colorado River Basins. Other studies have reported that DDT concentrations in these regions continue to represent a risk to avian wildlife (Grubb et al. 1990; Henny et al. 2004; Buck et al. 2005). Concentrations of *p,p'*-DDE were

associated with reproductive effects including egg-shell thinning and lower productivity in osprey and bald eagle populations in the lower Columbia River Basin (Henny et al. 2004; Buck et al. 2005). Conversely, DDT and other contaminants did not affect bald eagle populations nesting along the Salt and Verde Rivers (Grubb et al. 1990), upstream of where our study indicated high risk of total DDT exposure to bald eagles in the Colorado River Basin. Other studies of bald eagles from our study area were not found in the scientific literature. Our screening level risk analysis indicated that bald eagles were at greatest risk to total DDT in winter nesting areas in the lower Mississippi River Basin (i.e., Arkansas and Mississippi). Mink were also at risk to total DDT exposure in the lower Mississippi River Valley at sites in watersheds farmed for cotton. We are not aware of specific studies evaluating total DDT

Table 8 Toxicity reference value (TRV) for bald eagle and mink based on the no observed adverse effect level from dietary or tissue studies^a, no effects hazard concentration (NEHC)^b, and the number of samples and sites that exceed the NEHC^c

Contaminant	Bald eagle				Mink			
	TRV (mg/kg/d)	NEHC (µg/g)	No. of samples	No. of sites	TRV (mg/kg/d)	NEHC (µg/g)	No. of samples	No. of sites
PentaCB	N/A	N/A	N/A	N/A	7.25 ^d	53.3	0	0
HCB	0.225 ^e	2.25	0	0	N/A	N/A	N/A	N/A
α-HCH	0.56 ^f	5.6	0	0	0.014 ^f	0.10	0	0
β-HCH	0.56 ^f	5.6	0	0	0.31 ^f	2.28	0	0
γ-HCH	2 ^f	20	0	0	6.15 ^f	45.2	0	0
δ-HCH	0.56 ^f	5.6	0	0	0.014 ^f	0.10	0	0
Aldrin	N/A	N/A	N/A	N/A	0.154 ^f	1.13	0	0
Dieldrin	0.077 ^f	0.77	0	0	0.015 ^f	0.11	6	4
Endrin	0.010 ^f	0.10	5	2	0.038 ^f	0.28	2	1
ΣChlordane	2.14 ^f	21.4	0	0	1.9 ^f	14.0	0	0
ΣDDT	6 ^g	0.27	92	36	0.62 ^f	4.56	5	4
Endosulfans	10 ^f	100	0	0	0.12 ^f	0.88	0	0
PCB	20 ^g	0.71	35	15	0.137 ^f	1.01	21	10
Methoxychlor	N/A	N/A	N/A	N/A	3.1 ^f	22.8	0	0
Toxaphene	N/A	N/A	N/A	N/A	6.2 ^f	45.6	0	0
TCDD-EQ	0.000303 ^g	0.000016	31	20	0.00000027 ^h	0.0000019	185	76
Arsenic	2.24 ⁱ	22.4	0	0	1.04 ⁱ	7.65	0	0
Cadmium	1.47 ^j	14.7	0	0	0.77 ^j	5.66	0	0
Chromium	2.66 ^k	26.6	4	3	2.40 ^k	17.7	7	5
Copper	4.05 ^l	40.5	0	0	5.60 ^l	41.2	0	0
Mercury	0.5 ^m	0.5	14	9	0.05 ⁿ	0.37	34	22
Nickel	6.71 ^o	67.1	0	0	1.70 ^o	12.5	0	0
Lead	1.63 ^p	16.3	0	0	4.7 ^p	34.6	0	0
Selenium	0.4 ^f	4.0	3	1	0.154 ^f	1.13	51	19
Zinc	14.49 ^f	145	1	1	123 ^f	904	0	0

TRVs were not available for pentachloroanisole, Dacthal, and mirex for either bald eagle or mink. ΣChlordane is the sum of heptachlor epoxide, oxychlordane, *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, and *trans*-nonachlor, and ΣDDT is the sum of *o,p'*- and *p,p'*-DDE, DDD, and DDT

N/A Not available, *pentaCB* pentachlorobenzene, *HCB* hexachlorobenzene

^a Most TRVs were derived from adult dietary exposures. TRVs for ΣDDT, PCB, TCDD-EQ, and mercury in bald eagle were based on egg concentrations

^b NEHC=dietary-based no adverse effects level TRV (mg/kg body weight/day)/food ingestion rate (kg food/kg body weight/day). Food ingestion rate=0.100 kg food/kg body weight/day for bald eagle and 0.136 kg food/kg body weight/day for mink (USEPA 1993). NEHC=tissue-based no adverse effects level TRV (mg/kg egg)/biomagnification factor (BMF). The bald eagle BMFs were 28 for PCB, 19 for TCDD-EQ, 22 for ΣDDT, and 1 for mercury (Giesy et al. 1995)

^c Number of samples=409 for all contaminants except for TCDD-EQ (*n*=408) and pentachlorobenzene, aldrin, endosulfans, and methoxychlor (*n*=135). Number of sites=111 except for pentachlorobenzene, aldrin, endosulfans, and methoxychlor (*n*=37)

^d Linder et al. 1980

^e Hill and Camardese 1986

^f Sample et al. 1996

^g Elliott and Harris 2001

^h Tillitt et al. 1996

ⁱ USEPA 2005a

^j USEPA 2005b

^k USEPA 2005c

^l USEPA 2007a

^m Giesy et al. 1995

ⁿ USEPA 1995

^o USEPA 2007b

^p USEPA 2005d

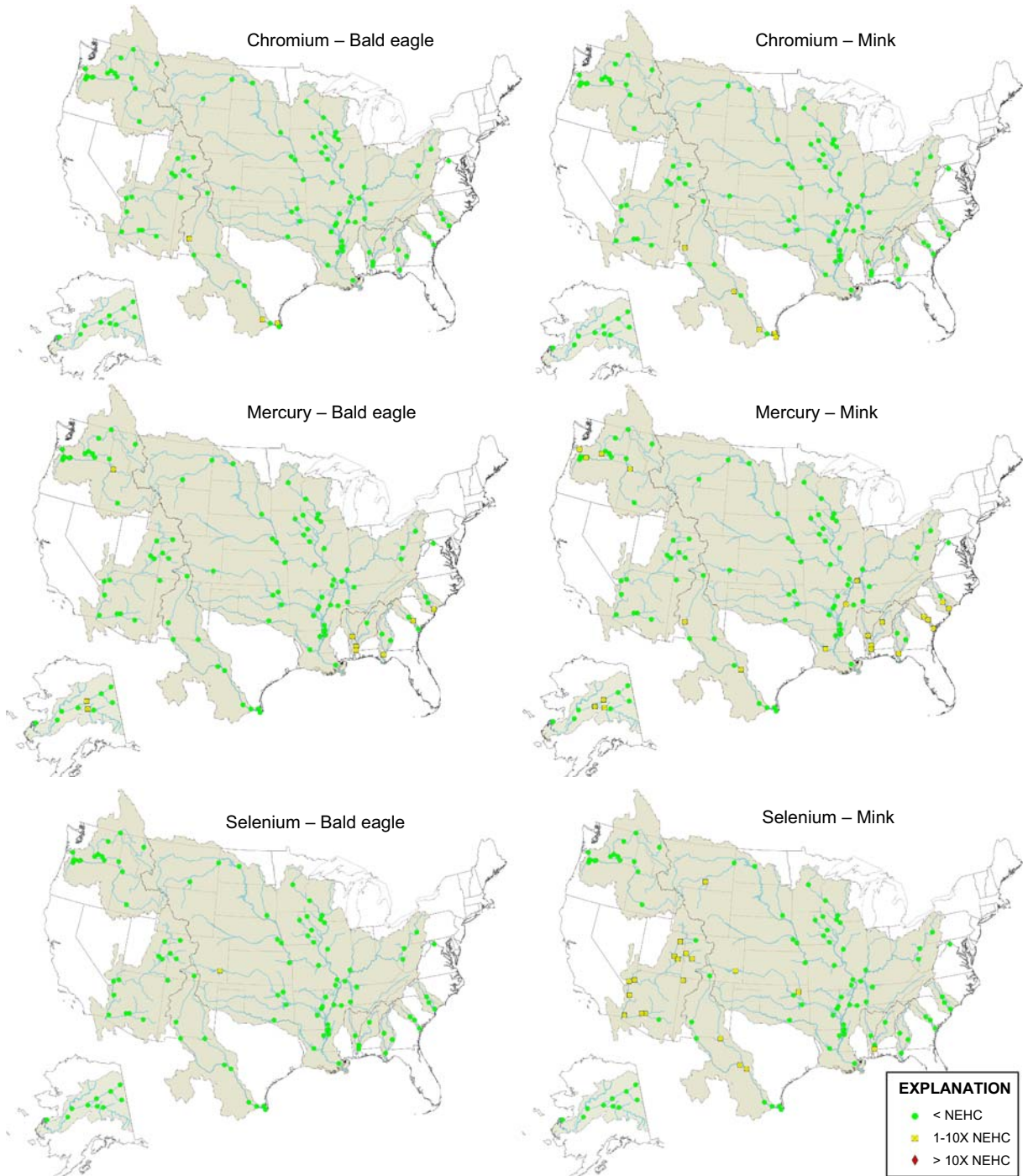


Fig. 4 Sites that exceed the no effects hazard concentration (NEHC) for selected elemental contaminants. See Fig. 1 for specific site locations and Table 8 for NEHC concentrations

concentrations in piscivorous mammal populations from this region.

PCBs were used historically as dielectric, hydraulic, and heat transfer fluids; lubricants; and in carbonless copy paper until the U.S. ban in 1979. Total PCB concentrations exceeded NEHCs for bald eagle and mink at multiple sites. Mink are among the most sensitive organisms to PCBs with their survival and reproduction affected by small dietary PCB doses (Leonards et al. 1995). Risk associated with total PCBs to mink generally was greatest in industrialized watersheds and near historical production and formulation facilities. NEHCs for both mink and bald eagle were exceeded at Sites 23, 24, 67, 76, 111, 320, 324, 327, and 330. Elevated PCB concentrations have been previously documented in water and biota near Site 327 (USFWS 1996; Schmitt et al. 1999; Zappia 2002), and PCBs were identified as a potential cause of reproductive dysfunction in mink from the coastal plain of Georgia, South Carolina, and North Carolina (Osowski et al. 1995). We are not aware of contaminant studies evaluating the risk of total PCBs to piscivorous wildlife in the Mississippi River Basin downstream from Memphis (Site 76) or the Ohio River Valley (Sites 23, 24, and 67). Mink typically do not occur in the southwestern United States, but other piscivorous mammals [i.e., raccoon (*Procyon lotor*)] may be at risk to total PCBs near Sites 320 and 324. The Gila River at Site 324 has low or no flow for part of the year and is recharged by wastewater treatment plant effluent and urban runoff from the Phoenix area. The Gila River supports contaminated prey items including fish with elevated concentrations of organochlorine pesticides and total PCBs (Hinck et al. 2007). Therefore, contaminant exposure to piscivorous wildlife may be increased by limited water and food resources in this arid region, where riparian wildlife concentrate. Moreover, bald eagles breeding in southern Arizona habitually forage in the same areas of free-flowing water, where fish are the most commonly consumed prey item (Haywood and Ohmart 1986; Grubb 1995).

PCBs were formulated as complex mixtures comprising as many as 209 structurally similar compounds (congeners) that vary widely in toxicity and persistence. The toxicity of PCBs to mink, bald eagles, and other organisms is largely attributable to relatively few congeners, specifically the highly toxic non-*ortho*- and mono-*ortho*-chloro substituted com-

pounds that can assume a planar molecular configuration (Aulerich and Ringer 1977; Peterson et al. 1993; Tillitt et al. 1996). These compounds are structurally similar to 2,3,7,8-tetrachloro-*p*-dioxin (TCDD) and are among the compounds collectively known as planar halogenated hydrocarbons (PHHs). Concentrations of PHHs in fish can vary over orders of magnitude depending upon the original composition of the mixture released to the environment, weathering, and toxicokinetic factors, and they largely determine the risk assessment of PCBs in the environment (Safe 1990; van den Berg et al. 2006). It has been widely accepted that concentrations of individual congeners, specifically the planar congeners, rather than total PCB concentrations are necessary to assess the risk of PCBs to piscivorous wildlife. However, recent studies indicate that total PCBs measurements can be used to assess the toxicological risks associated with dioxin-like PCB concentrations in whole-body benthivorous and piscivorous fish (Bhavsar et al. 2007a, b). High-resolution analyses required for PHHs were beyond the scope of our studies; therefore, we determined dioxin-like activity (i.e., TCDD-EQ) with the H4IIE bioassay. As measured in our samples, TCDD-EQ includes the contributions of all the PHHs including the planar PCB congeners. Dioxin-like activity (as TCDD-EQ) was elevated at many of the sites where total PCB concentrations were also comparatively high, indicating that much of the dioxin-like activity was attributable to PCBs. However, the overall risk of TCDD-EQ to bald eagle and mink was more widespread than risk of PCBs, indicating some contributions from other PHHs at sites with high TCDD-EQ concentrations but low total PCB concentrations. TCDD-EQ-related risk was greatest in the Columbia, Rio Grande, Mobile, and Mississippi River Basins, but was minimal in the Yukon, Colorado, Apalachicola, Savannah, and Pee Dee River Basins.

The risk of mercury to piscivorous wildlife was greatest at sites in the southeastern United States (Mobile, Apalachicola, Savannah, and Pee Dee River Basins) and Yukon River Basin. Sources of Hg in these basins include historical gold and cinnabar mines, releases from chemical manufacturing and coal-fired power plants, atmospheric deposition, and wetlands (Rudd 1995; Brumbaugh et al. 2001; Eisler 2004; Paller et al. 2004; Warner et al. 2005). Orihel et al. (2007) demonstrated that organic mercury deposited

directly to aquatic ecosystem was readily converted to methylmercury, which is available to biota. This relationship may explain the high fish concentrations and associated risk of mercury in the southeastern United States and Yukon River Basin, where mercury methylation rates are known to be high (Brumbaugh et al. 2001). Other studies also have reported that mercury concentrations in these regions represent a risk to avian and mammalian wildlife (Halbrook et al. 1994; Osowski et al. 1995; Ambrose et al. 2000; Adair et al. 2003). Mercury has been suspected to cause reproductive effects, organ toxicity, and mortality in prothonotary warbler (*Protonotaria citrea*), mink, and river otter throughout the southeastern United States (Halbrook et al. 1994; Osowski et al. 1995; Adair et al. 2003). Ambrose et al. (2000) reported that mercury concentrations in peregrine falcon (*Falco peregrinus anatum*) breeding in the Yukon River Basin were associated with lower nest success. Conversely, mercury concentrations in bald eagle eggs from the lower Columbia River Basin were not associated with lower productivity (Anthony et al. 1993; Buck et al. 2005).

Sources of excess selenium include natural weathering of seleniferous shales, irrigation practices, uranium ore and coal extraction, and coal-fired power plants (Sorenson 1991). The risk of selenium to bald eagle was lower than to mink. Among basins, mink in the Colorado River Basin were at greatest risk to selenium exposure. Irrigation and mining throughout these areas accelerate the rates of the processes controlling the release and distribution of this naturally occurring element. Selenium exposure to wildlife is most commonly associated with toxicity to birds (Lemly 1996; Hamilton 2004). Historically, avian wildlife was impacted by selenium contamination in the Colorado River Basin near Site 312 (Stephens et al. 1992), but we are not aware of studies evaluating selenium toxicity to piscivorous mammals in this basin.

The wildlife risk analysis presented here is a screening level or Tier 1 assessment. As such, it was based on the most conservative (i.e., lowest) TRVs available to identify all potential contaminants of concern (USEPA 1998). The conservative TRVs we used included values extrapolated from receptors other than bald eagle and mink. As noted by Sample and Suter (1999), such an approach indicates only whether harmful effects are possible, not whether they are probable. Consequently, the use of the most

conservative TRVs may overestimate the actual risk of some contaminants to wildlife. For example, we initially used a TRV for mercury in bald eagle (0.0064 mg/kg/d) derived from a multi-generational study of mallards (*Anas platyrhynchos*; Heinz 1979; Sample et al. 1996). Based on this TRV, Yearley et al. (1998) estimated that mercury concentrations in fish from 98% of the lakes in the northeastern United States represented a risk to piscivorous birds. Our NEHC calculated from this TRV (0.064 g/g) similarly indicated that bald eagles were at risk from mercury at most of our sites (105 of 109 sites), which might be interpreted as indicative of widespread mercury poisoning of bald eagles in the conterminous United States. This conclusion would contradict current data indicating the recovery of U.S. bald eagle populations and their recent removal from the Endangered Species List (U.S. Fish and Wildlife Service 2007), and would indicate that an alternative TRV should be considered. Therefore, we selected a NEHC of 0.5 µg/g developed specifically to estimate mercury risk to bald eagles (Giesy et al. 1995), which resulted in considerably lower estimates of risk to bald eagles at our sites; only nine sites had mercury concentrations >0.5 µg/g. However, there is evidence that piscivorous birds are being negatively affected by mercury in fish elsewhere (e.g., Scheuhammer and Blancher 1994; Scheuhammer et al. 1998; Nocera and Taylor 1998). One might therefore argue that bald eagle populations would be increasing more rapidly if not for mercury, which would support the use of the lower TRVs. This example highlights the limitations of conservative TRVs and the importance of qualifying their use in the screening level phase of wildlife risk analysis. Regardless of the TRV employed, sites at risk would need to be evaluated further to fully characterize the exposure and ecological effects of specific contaminants to bald eagles or mink (e.g., Tier 2 risk assessment).

There are other assumptions and uncertainties associated with the approach we used to evaluate risk to piscivorous wildlife. Some of the NOAELs used here were derived from studies conducted several decades ago, and other NOAELs have been proposed and used elsewhere. Consensus NOAELs based on whole-body fish concentrations, such as those adopted for arsenic, cadmium, chromium, copper, nickel, and lead, are not available for many contaminants. The use of other NOAELs would result in different

NEHCs and could change the perceived risk to piscivorous wildlife. Other wildlife food chain analyses incorporate water and sediment/soil concentrations to estimate contaminant concentrations in fish and evaluate the risk of contaminants to piscivorous wildlife because fish concentrations are not available. While more recent toxicity thresholds may be available for water, soil, and sediment, there are additional uncertainties associated with estimating exposure to piscivorous wildlife from these endpoints. Factors such as organic carbon content, sulfides, hardness, and pH can influence the bioavailability and toxicity of contaminants (e.g., Björnberg et al. 1988; Chapman et al. 1998; Heekyoung Choi et al. 1998). These points emphasize that updated consensus toxicity thresholds based on whole-body fish concentrations associated with reproductive performance, growth, and survival are needed, especially for contaminants such as pentachloroanisole, Dacthal, and mirex for which toxicity thresholds are not currently available. However, studies determining toxicity thresholds in whole-body fish are not likely to be conducted for some of these contaminants because of time and expense requirements for multi-generational studies with birds and mammals and the current focus of research on new or emerging contaminants.

The selection of appropriate receptors in the analysis is important. Bald eagles and mink consume large, adult fish such as those collected and analyzed in this study and are therefore reasonable receptors. The risk of contaminants to other piscivorous wildlife such as belted kingfisher (*Ceryle alcyon*), Caspian tern (*Sterna caspia*), or common loons (*Gavia immer*) may be overestimated if these species are used in the model, because they consume smaller fish that are presumed to have lower contaminant concentrations than larger fish because of their shorter lifespans, higher growth rates, and higher metabolism (Scheuhammer et al. 1998; Rose et al. 1999). It is also unlikely that the diet of adult bald eagles or mink would be entirely composed of fish from one site, which is tacitly assumed in this approach. Nevertheless, this assumption may be valid for nestling birds or cubs fed locally procured prey items. Moreover, the risk of accumulative contaminants to nestlings/cubs and juveniles may be greater because of their small body size and correspondingly higher food ingestion rates compared to adult wildlife. Inexperienced juveniles also may have greater exposure to chemical contaminants

because of their initial small home range or when they scavenge on dead or dying fish associated with natural or anthropogenic events.

As a final consideration, this assessment assumes that the contaminants act independently. Many of the contaminants co-occur at the sites, and their effects are not necessarily independent or additive (e.g., Wren et al. 1987). Effects also may vary depending on other factors such as the disease, nutritional, and reproductive status of the organism. Nevertheless, the NOAELs and subsequent NEHCs represent conservative, screening-level criteria for the assessment of potential adverse effects in wildlife from chemical contaminants in the fish that they consume.

Overall, concentrations of many persistent, bioaccumulative organochlorine chemical residues have declined over the past two decades. Our results indicate that organochlorine residues, in particular *p,p'*-DDE and total PCBs, remain a risk to piscivorous wildlife at some sites. Concentrations of mercury and selenium, which have not declined, also represent a risk at a number of sites. In addition, mercury and selenium concentrations in aquatic systems may rise because of increasing global coal combustion and subsequent atmospheric deposition (USEPA 1997; Mason et al. 1999). The full geographic extent of the risk associated with contaminants in fish cannot be determined from our data because some of the sites were not selected in a truly unbiased manner and only screening level analyses were conducted. Regardless, our findings indicate that environmental contaminants are a risk to piscivorous wildlife at some sites. Characterization of exposure and ecological effects for contaminants at specific sites would be the next step in refining the risks determined in our screening level wildlife risk analysis.

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