

Mercury trends in fish from rivers and lakes in the United States, 1969–2005

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Abstract A national dataset on concentrations of mercury in fish, compiled mainly from state and federal monitoring programs, was used to evaluate trends in mercury (Hg) in fish from US rivers and lakes. Trends were analyzed on data aggregated by site and by state, using samples of the same fish species and tissue type, and using fish

of similar lengths. Site-based trends were evaluated from 1969 to 2005, but focused on a subset of the data from 1969 to 1987. Data aggregated by state were used to evaluate trends in fish Hg concentrations from 1988 to 2005. In addition, the most recent Hg fish data (1996–2005) were compared to wet Hg deposition data from the Mercury Deposition Network (MDN) over the same period. Downward trends in Hg concentrations in fish from data collected during 1969–1987 exceeded upward trends by a ratio of 6 to 1. Declining Hg accumulation rates in sediment and peat cores reported by many studies during the 1970s and 1980s correspond with the period when the most downward trends in fish Hg concentrations occurred. Downward Hg trends in both sediment cores and fish were also consistent with the implementation of stricter regulatory controls of direct releases of Hg to the atmosphere and surface waters during the same period. The southeastern USA had more upward Hg trends in fish than other regions for both site and state aggregated data. Upward Hg trends in fish from the southeastern USA were associated with increases in wet deposition in the region and may be attributed to a greater influence of global atmospheric Hg emissions in the southeastern USA. No significant trends were found in 62% of the fish species from six states from 1996 to 2005. A lack of Hg trends in fish in the more recent data was consistent with the lack of trends in wet Hg deposition at MDN

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sites and with relatively constant global emissions during the same time period. Although few significant trends were observed in the more recent Hg concentrations in fish, it is anticipated that Hg concentrations in fish will respond to changes in atmospheric Hg deposition, however, the magnitude and timing of the response is uncertain.

Keywords Mercury · Fish · Trends · Wet deposition · Atmospheric emissions

Introduction

Mercury (Hg) is a ubiquitous environmental pollutant that accumulates to harmful concentrations in aquatic ecosystems. Atmospheric deposition is the dominant source of inorganic Hg in many aquatic ecosystems (Fitzgerald et al. 1998; Downs et al. 1998; Wiener et al. 2006; Peterson et al. 2007). Industrial and municipal point-sources of mercury to rivers and lakes were very important historically, but have been greatly reduced since the implementation of stricter regulatory controls in the 1970s (Balogh et al. 1999; Krabbenhoft et al. 2005). Widespread Hg contamination of fish in remote areas with low rates of Hg deposition suggests Hg loading in an ecosystem alone does not define Hg concentration in fish (Krabbenhoft et al. 2005; Wiener et al. 2007). Transport of atmospheric Hg to aquatic ecosystems and Hg cycling within aquatic systems are complex and not fully understood. Catchment size, geochemistry, hydrology, climate, soil, land-use, land cover, and topography all influence delivery of Hg to aquatic ecosystems (Balogh et al. 1999). Human activities contribute atmospheric emissions and direct point source Hg contamination, and create land disturbances that enhance Hg delivery to aquatic ecosystems (Balogh et al. 1999).

Methylation of inorganic atmospheric Hg to monomethylmercury (MeHg), which bioaccumulates and biomagnifies in fish, is in turn influenced by a variety of environmental factors such as organic carbon availability, pH, sulfur cycling, biological productivity, and temperature (Wiener et al. 2006). Fish are probably the most widely studied component of Hg transfer and bioaccumulation in ecosystems because fish consump-

tion is the main Hg exposure pathway for humans and wildlife. The Hg concentration in fish, which is generally assumed to be >95% MeHg (Bloom 1992), ultimately depends on the availability of MeHg in their diet and the degree to which the fish can excrete Hg (Downs et al. 1998). Fish Hg concentrations vary by species due to food web position and biochemical differences among species (Wiener et al. 2003). Hg typically accumulates in fish as their age and size increases due to relative exposure times and food web position (Downs et al. 1998; Chen and Folt 2005). Variability in fish Hg concentrations can be reduced by controlling for fish species and fish age or length (Wiener et al. 2007). Many federal, state, and local agencies are monitoring Hg in fish to identify areas with fish Hg concentrations of concern to human health. Elevated Hg concentrations in fish represent a public health hazard and the primary reason for 80% of the 3,080 fish consumption advisories issued by 48 states in 2006 (<http://www.epa.gov/waterscience/fish/advisories/2006/tech.pdf>).

Datasets of Hg concentrations in fish collected by federal, state, and local agencies were compiled by the USGS Geographic Analysis and Monitoring program and the National Institute of Environmental Health Sciences to be used by environmental and health researchers in investigations of Hg in the environment. The result of this effort was the Environmental Mercury Mapping, Modeling, and Analysis (EMMMA) database (<http://emmma.usgs.gov/>, accessed 30 October 2007). As of 2007, the EMMMA dataset included 74,867 records from 7,759 sites collected by 27 agencies from 1969 to 2005. Samples included both rivers (5,034 sites) and lakes (2,725 sites). Fish data were compiled from every state except Hawaii. The EMMMA database contains 241 fish species and 14 types of tissues. The objectives of this paper are (1) to assess whether EMMMA data can provide evidence of changes in Hg concentrations in fish in the USA between 1969 and 2005 and (2) to compare fish Hg trends to trends in Hg wet deposition and Hg atmospheric emissions. Substantial additional data may exist in state, federal, and other databases; however, additional efforts to compile data were beyond the scope of this paper.

Methods

Trend analyses

Trends were analyzed on 31 different fish species and three tissue types. Each trend analysis was limited to one fish species and tissue type. A restricted range of fish lengths were used to normalize data for differences in fish age and size. Data within 10% of the median length for each species and tissue type were retained for trend analyses. Trends were analyzed on data aggregated by site and by state. Sites were selected for trend evaluation if they had at least seven fish samples within the restricted range of fish lengths that were of the same species and tissue type collected for at least different 5 years over more than a 10-year period between 1969 and 2005. Ninety of the original 7,759 river and lake sites met the criteria for trend analyses (Table 1; Supplemental Table A). Site trends focused on a subset of 50 of the 90 sites having data in the period 1969–1987 (Table 2).

Statewide trends were analyzed on an aggregate of all samples of a single fish species and tissue type from a state that met the following criteria: during the period 1988–2005, fish samples of one species and tissue type within the restricted length were collected for at least seven different years for over at least a 10-year period. In addition,

for each fish species and tissue type evaluated in the statewide trend, at least 30 sites had to have been sampled in the state over the 10-year period, with at least three sites having been sampled in both the first and last years of sample collection. Statewide trends were not analyzed prior to 1988 because of insufficient data. Statewide trends were also analyzed from 1996 to 2005 using the same statewide criteria except that the period of record was reduced to at least five different years over at least a 6-year period between 1996 and 2005. The results of the fish Hg trend analyses from 1996–2005 provide a direct comparison with Hg wet deposition data that the National Atmospheric Deposition Program Mercury Deposition Network (NADP MDN) began collecting in 1995.

The EMMMA dataset had no information about the sampling or analytical methods used by the 27 agencies that provided data. Changes or differences in sampling or analytical methods in this dataset can result in false trends. Differences in methods used by each agency were minimized by aggregating data by site and by state. Data from most sites and states were collected by only one agency.

Statistical analysis was performed using Version 9.1.3 of the Statistical Analysis System (SAS Institute 2002). Trends were determined through the use of the SAS LIFEREG (linear regression)

Table 1 Summary of trend data aggregated by site and by state

Data aggregation	Site		State	
	1969–2005	1969–1987	1988–2005	1996–2005
Sites (<i>n</i>)	90	50	2,609	2,801
States (<i>n</i>)	32	31	8	6
Major sources of data	State and federal programs	National Contaminant Biomonitoring Program	State programs	State programs
Hg concentrations in fish (µg/g)	<0.01–2.6	<0.01–1.2	<0.001–4.8	<0.001–4.8
Median Hg concentration in fish (µg/g)	0.23	0.10	0.22	0.24
Dominant fish species	Common carp Largemouth bass	Common carp White sucker	Largemouth bass Channel catfish	Largemouth bass Channel catfish
Fish species (<i>n</i>)	26	21	12	14
Upper trophic level species (%)	50	26	73	73
Tissue type	Whole fish, skin-on and skin-off fillets	Whole fish	Skin-on and skin-off fillets	Skin-on and skin-off fillets

Site-specific data analysis focused on a subset of data from 1969 to 1987. Statewide data were analyzed over one shorter period, 1996–2005, for a direct comparison with wet Hg deposition data

Table 2 Trends in Hg concentrations in whole fish from 1969 to 1987 at 50 National Contaminant Biomonitoring Program sites

Site	Site name	State	Species	<i>n</i>	Begin year	End year	Hg (μg/g) median	<i>p</i> value	Percent change
1	Apalachicola River, J Woodruff Dam	FL	SS	13	1969	1986	0.11	0.135	-2.11
2	Arkansas River	AR	CC	9	1969	1984	0.08	0.004	-5.92
3	Chena River	AK	LNS	8	1969	1981	0.08	0.402	2.41
4	Columbia River, Cascade Locks	OR/WA	LSS	10	1969	1984	0.15	0.005	-9.06
5	Connecticut River, Windsor locks	CT	WCF	12	1969	1984	0.15	0.000	-7.22
6	Delaware River	NJ/PA	WS	12	1969	1984	0.06	0.050	-4.59
7	Hudson River	NY	G	10	1969	1980	0.13	0.414	-2.89
8	Illinois River	IL	CC	13	1969	1986	0.06	0.255	-1.49
9	Klamath River	CA	YP	7	1969	1986	0.19	0.000	-3.31
10	Lake Erie	PA	YP	11	1969	1986	0.09	0.000	-8.23
11	Lake Huron	MI	CC	14	1969	1986	0.06	0.206	3.11
11	Lake Huron	MI	YP	10	1969	1986	0.08	0.221	4.68
12	Lake Ontario	NY	RB	7	1969	1981	0.36	0.000	-10.36
13	Lake Superior	WI	BL	10	1969	1986	0.11	0.000	-5.36
13	Lake Superior	WI	LT	7	1969	1986	0.26	0.082	-3.52
14	Missouri River, Garrison Dam	ND	W	7	1969	1986	0.20	0.081	-2.74
15	Red River of the North	MN/ND	S	9	1969	1986	0.45	0.001	-4.86
16	Penobscot River	ME	WS	13	1969	1986	0.25	0.001	-4.71
17	Rio Grande	TX	GS	9	1969	1986	0.05	0.004	-5.69
18	Sacramento River	CA	CC	7	1969	1986	0.10	0.001	-7.13
19	Salmon River	ID	LSS	7	1969	1986	0.19	0.179	-4.88
20	Snake River	ID/WA	LSS	13	1969	1986	0.11	0.679	0.95
21	Utah Lake	UT	CC	10	1969	1986	0.04	0.396	2.65
21	Utah Lake	UT	WB	8	1969	1986	0.06	0.588	1.85
22	White River	AR	BMBU	8	1969	1986	0.24	0.009	7.66
23	Yakima River	WA	LSS	8	1969	1986	0.08	0.362	-3.26
24	Verdigris River, Bartlesville Reservoir	OK	CC	10	1970	1986	0.06	0.765	-0.60
25	Columbia River	WA	CC	8	1970	1980	0.06	0.051	-8.66
26	Cumberland River	TN	CC	9	1970	1986	0.08	0.125	-1.65
27	Colorado River, Imperial Reservoir	AZ/CA	CC	10	1970	1984	0.01	0.001	6.36
28	James River	SD	CC	10	1970	1986	0.09	0.453	-1.09
28	James River	SD	GE	8	1971	1986	0.18	0.677	1.82
29	Kennebec River	ME	WS	7	1971	1984	0.16	0.091	-1.49
29	Kennebec River	ME	YP	10	1970	1986	0.50	0.000	-9.13
30	Lake Michigan	WI	BL	14	1970	1986	0.05	0.003	-5.75
30	Lake Michigan	WI	LT	7	1971	1986	0.14	0.000	-10.26
31	Colorado River, Lake Powell	AZ	CC	13	1971	1986	0.12	0.270	-1.52
31	Colorado River, Lake Powell	AZ	LMB	7	1970	1984	0.18	0.126	-3.34
32	Mississippi River	IA/WI	CC	9	1970	1984	0.10	0.213	-3.82
33	Missouri River	NE/IA	CC	13	1970	1986	0.06	0.596	-1.78
33	Missouri River	NE/IA	GE	7	1970	1981	0.10	0.000	-10.32

Table 2 (continued)

Site	Site name	State	Species	<i>n</i>	Begin year	End year	Hg (µg/g) median	<i>p</i> value	Percent change
34	North Platte River, Lake McConaughy	NE	CC	9	1970	1986	0.10	0.087	-3.21
34	North Platte River, Lake McConaughy	NE	W	8	1970	1986	0.08	0.011	-7.37
35	Nueces River	TX	GS	10	1970	1986	0.02	0.043	2.47
36	Red River, Lake Texoma	OK/TX	CC	9	1970	1986	0.09	0.962	0.19
37	<i>Rogue River, Goldray Dam</i>	OR	<i>BB</i>	<i>10</i>	<i>1970</i>	<i>1986</i>	<i>0.12</i>	<i>0.000</i>	<i>-11.98</i>
38	Wisconsin River	WI	CC	10	1970	1986	0.17	0.003	-4.78
39	Yazoo River	MS	SMBU	8	1970	1986	0.17	0.703	0.49
40	Green River	UT	CC	10	1971	1986	0.11	0.082	2.53
41	Mississippi River	MN	WS	9	1971	1986	0.23	0.760	-1.29
42	Ohio River	IL/KY	CC	8	1971	1984	0.14	0.000	-5.64
43	Ohio River	OH/KY	CC	9	1971	1987	0.06	0.006	-3.56
44	Raritan River	NJ	WS	9	1971	1986	0.18	0.032	3.46
45	Rio Grande	CO	WS	7	1971	1986	0.03	0.000	-6.31
46	St. Lawrence River	NY	WS	7	1971	1987	0.11	0.770	-1.02
47	Susquehanna River, Conowingo Dam	MD	CC	7	1971	1986	0.08	0.000	-5.37
48	Wabash River	IN/IL	CC	8	1971	1986	0.15	0.002	-4.59
49	Des moines River	IA	CC	11	1972	1986	0.07	0.041	6.34
50	Cooper River, Lake Moultrie	SC	CCF	8	1976	1986	0.04	0.208	3.14

Significant trends ($p < 0.05$) are shaded. Downward trends starting above and ending below EPA advisory guideline (0.3 µg/g MeHg) are in italics. All other trends are below EPA guideline
BMBU bigmouth buffalo, *BL* bloater, *BB* brown bullhead, *CCF* channel catfish, *CC* common carp, *GS* gizzard shad, *GE* goldeye, *G* goldfish, *LT* lake trout, *LMB* largemouth bass, *LSS* largescale sucker, *LNS* longnose sucker, *RB* rock bass, *S* sauger, *SMBU* smallmouth buffalo, *SS* spotted sucker, *W* walleye, *WB* white bass, *WCF* white catfish, *WS* white sucker, *YP* yellow perch

procedure, which yields estimates of parametric regression models for censored data using the method of maximum likelihood (Allison 1995). Hg concentrations in fish were natural log transformed to remove much of the heterogeneity of residuals. Linear changes in log Hg concentrations were considered significant if the p value was less than 0.05. Three diagnostic statistics, Cook’s D, leverage, and standardized residual statistics were used to identify data values that substantially influenced the fit of the regression line (Helsel and Hirsch 1992). Data values that diagnostic statistics identified as outliers were removed resulting in changes in trends at two sites. The annual percent change in fish Hg concentrations was calculated using the equation, $(e^B - 1) \times 100$, where B is the slope of the regression line (Hirsch et al. 1991).

Ancillary data

Ancillary data were obtained for the 90 sites that met the trends criteria. Land use, land cover, and watershed size were compiled for each of the trend site watersheds from the National Hydrography Dataset Plus (NHDPlus; USEPA 2006; U. S. Geological Survey 2000). NHDPlus is a compilation of geospatial data that includes features from the National Hydrography Dataset, the National Elevation Dataset, and the National Watershed Boundary Dataset.

Dataset compositions

The 1969–2005 trends data aggregated by site consists of 80 river sites and 10 lake sites. However,

13 of the river sites were at impoundments, where conditions were more similar to a reservoir or lake than a river (Supplemental Table A). Fourteen rivers had more than one site and 14 sites had more than one species of fish that met the trend analyses criteria. The average length of record at the sites was 17 years, and the maximum 29 years (Supplemental Table A). The 90 sites were located in 32 states; however, 20 of the 90 sites were in South Carolina. Two data sources provided 82% of the data—the National Contaminant Biomonitoring Program (NCBP; 54 sites), and the South Carolina Department of Health and Environmental Conservation (ScDHEC; 20 sites).

The NCBP analyzed Hg concentrations in whole fish at 117 sites on large rivers and lakes

across the USA from 1969 through 1987 (May and McKinney 1981; Lowe et al. 1985; Schmitt and Brumbaugh 1990; Schmitt et al. 1999). The Biomonitoring of Environmental Status and Trends (BEST) program, an extension and revision of the NCBP, collected additional samples at selected NCBP sites in the Mississippi River and Columbia River basins during 1995 and 1997, respectively (Schmitt et al. 2005; Hinck et al. 2006a). Of the original 117 NCBP sites, 54 met our criteria for trends analyses from 1969 to 2005, and 50 of the sites met criteria for trends analysis from 1969 to 1987. More than half the fish collected by the NCBP at the 54 sites were from the lower trophic level, mainly common carp (*Cyprinus carpio*, 34%) and sucker species (*Catostomus* sp., 23%).

Table 3 Trends in Hg concentrations based on fish data aggregated by state from 1988 to 2005

State	Sites	<i>n</i>	Species	Begin year	End year	Median Hg (µg/g)	<i>p</i> value	Percent change	Trend
Southeastern USA									
<u>GA</u>	<u>112</u>	<u>266</u>	<u>LMB</u>	<u>1991</u>	<u>2001</u>	<u>0.24</u>	<u><0.001</u>	<u>7.43</u>	<u>UP</u>
GA	56	105	CCF	1991	2001	0.10	0.063	6.76	NONE
<u>LA</u>	<u>324</u>	<u>1,049</u>	<u>LMB</u>	<u>1994</u>	<u>2005</u>	<u>0.39</u>	<u><0.001</u>	<u>3.78</u>	<u>UP</u>
LA	113	168	CCF	1994	2004	0.10	0.007	11.2	UP
<u>LA</u>	<u>178</u>	<u>328</u>	<u>FD</u>	<u>1994</u>	<u>2004</u>	<u>0.37</u>	<u>0.013</u>	<u>6.23</u>	<u>UP</u>
LA	72	125	RS	1994	2004	0.16	0.075	11.4	NONE
LA	158	383	WC	1994	2004	0.21	0.601	1.14	NONE
LA	178	378	BC	1995	2005	0.24	0.686	0.83	NONE
LA	169	444	B	1994	2005	0.52	0.019	-3.26	DOWN
<i>LA</i>	<i>42</i>	<i>66</i>	<i>BMBU</i>	<i>1995</i>	<i>2004</i>	<i>0.28</i>	<i>0.045</i>	<i>-6.66</i>	<i>DOWN</i>
NC	37	61	BG	1989	1999	0.10	0.771	0.76	NONE
SC	129	963	B	1993	2004	0.80	0.003	3.02	UP
SC	70	194	CCF	1994	2004	<0.25	0.277	-2.29	NONE
SC	188	1,556	LMB	1993	2004	0.38	<0.001	-3.13	DOWN
Midwestern USA									
IA	31	34	LMB	1994	2005	0.13	0.947	0.25	NONE
IA	44	60	CC	1993	2005	0.10	0.005	-6.11	DOWN
IA	87	142	CCF	1988	2005	0.09	<0.001	-6.14	DOWN
IN	194	285	CC	1988	2004	0.17	0.856	-0.11	NONE
IN	56	74	CCF	1988	2004	0.14	0.316	-1.04	NONE
IN	75	91	LMB	1991	2004	0.19	0.999	0.00	NONE
MI	55	158	LMB	1988	1997	0.31	0.632	-0.75	NONE
MN	43	81	CCF	1990	2000	0.21	0.364	1.80	NONE
MN	78	157	CC	1990	2001	0.13	0.085	-2.16	NONE
<i>MN</i>	<i>53</i>	<i>142</i>	<i>NP</i>	<i>1988</i>	<i>2001</i>	<i>0.27</i>	<i>0.001</i>	<i>-4.89</i>	<i>DOWN</i>
MN	75	202	W	1989	2001	0.23	0.036	-3.16	DOWN

Significant trends (*p* value < 0.05) are shaded. Trends above EPA advisory guideline (0.3 µg/g MeHg) are in bold text. Upward trends starting below and ending above EPA guideline are underlined. Downward trends starting above and ending below EPA guideline are in italics. Trends below EPA guideline are in regular text

BMBU bigmouth buffalo, *BC* black crappie, *BG* bluegill, *B* bowfin, *CCF* channel catfish, *CC* common carp, *FD* freshwater drum, *LMB* largemouth bass, *NP* northern pike, *RS* redear sunfish, *W* walleye, *WC* white crappie

The latter included white (*Catostomus commersonii*), largescale (*Catostomus macrocheilus*), and longnose (*Catostomus catostomus*) suckers.

The ScDHEC collected fish samples at 201 sites from 1992 to 2005. Of the 20 ScDHEC sites that met our criteria for trend analyses, more than 90% of the fish were upper trophic level species, predominantly largemouth bass (*Micropterus salmoides*; 63%) and bowfin (*Amia calva*; 22%). Although median Hg concentrations in fish from ScDHEC sites were five times higher than at NCBP sites, 24% of samples were below the ScDHEC Hg detection level of 0.25 µg/g.

In contrast, only 2% of NCBP samples were below detection levels, which ranged from 0.05 to 0.01 µg/g. The watersheds of the ScDHEC sites were roughly one-tenth the size of NCBP watersheds, but the median percent wetlands were 10 times higher in the ScDHEC watersheds than in the NCBP watersheds (Supplemental Table B). Biases in the data may result from over-representation of sites in South Carolina because of the higher percentage of wetlands in their watersheds and their relatively high Hg concentration when compared to other trend sites. High amounts of wetlands in South Carolina wa-

Table 4 Trends in Hg concentrations based on fish data aggregated by state between 1996 and 2005

State	Sites	<i>n</i>	Species	Begin year	End year	Median Hg (µg/g)	<i>p</i> value	Percent change	Trend
Southeastern USA									
GA	41	53	CCF	1996	2001	0.11	0.303	11.7	NONE
GA	88	174	LMB	1996	2001	0.30	0.399	-3.28	NONE
LA	102	151	CCF	1996	2004	0.10	0.002	14.9	UP
<i>LA</i>	<i>53</i>	<i>74</i>	<i>FCF</i>	<i>1996</i>	<i>2004</i>	<i>0.32</i>	<i>0.005</i>	<i>15.1</i>	<i>UP</i>
LA	316	988	LMB	1996	2005	0.39	0.003	3.10	UP
LA	62	97	RS	1996	2004	0.16	0.057	15.0	NONE
LA	112	193	BCF	1996	2004	0.14	0.056	7.30	NONE
LA	175	318	FD	1996	2004	0.37	0.223	3.03	NONE
LA	173	373	BC	1996	2004	0.24	0.485	1.47	NONE
LA	153	367	WC	1996	2004	0.21	0.571	-1.27	NONE
LA	42	63	BMBU	1996	2004	0.28	0.085	-6.49	NONE
LA	169	434	B	1996	2005	0.52	0.011	-3.79	DOWN
SC	128	922	B	1996	2004	0.79	<0.001	5.50	UP
SC	51	179	BCF	1996	2004	<0.25	0.001	9.06	UP
SC	143	963	RS	1996	2004	<0.25	0.006	3.38	UP
SC	142	677	BG	1996	2004	<0.25	0.008	4.28	UP
SC	182	1,408	LMB	1996	2004	0.37	0.186	-1.46	NONE
SC	68	186	CCF	1996	2004	<0.25	0.100	-3.88	NONE
Midwestern USA									
IA	37	46	CC	1996	2005	0.09	0.445	-2.48	NONE
IA	44	57	CCF	1996	2005	0.06	0.384	-4.31	NONE
IN	158	185	CC	1996	2004	0.16	<0.001	8.87	UP
IN	39	45	CCF	1997	2004	0.13	0.302	3.19	NONE
IN	71	84	LMB	1996	2004	0.18	0.980	-0.06	NONE
MN	45	110	CC	1996	2001	0.13	0.285	3.19	NONE
MN	44	115	W	1996	2001	0.20	0.088	6.98	NONE
<i>MN</i>	<i>33</i>	<i>82</i>	<i>NP</i>	<i>1996</i>	<i>2001</i>	<i>0.24</i>	<i>0.002</i>	<i>-12.8</i>	<i>DOWN</i>

Significant trends (*p* value < 0.05) are shaded. Trends above EPA advisory guideline (0.3 µg/g MeHg) are in bold text. Upward trends starting below and ending above EPA guideline are underlined. Downward trends starting above and ending below EPA guideline are in italics. Trends below EPA guideline are in regular text

BMBU bigmouth buffalo, *BC* black crappie, *BCF* blue catfish, *BG* bluegill, *B* bowfin, *CCF* channel catfish, *CC* common carp, *FCF* flathead catfish, *FD* freshwater drum, *LMB* largemouth bass, *NP* northern pike, *RS* redear sunfish, *W* walleye, *WC* white crappie

tersheds should promote transformation of inorganic Hg to MeHg. In addition, the smaller South Carolina watersheds would be expected to respond more rapidly to changes in Hg emissions (Grigal 2002). To minimize potential problems resulting from differences in site types, collection protocols, and analytical procedures between the NCBP and the SCDHEC, site-specific trends focused on a subset of 50 NCBP sites sampled from 1969 to 1987 (Table 2) before SCDHEC implemented a monitoring program.

Statewide data were used for the evaluation of trends from 1988 to 2005. The statewide datasets were composed of 14 fish species and two tissue types (skin-on and skin-off fillets). Of the fish species collected, 73% were from the upper trophic level (Table 1). Data for eight states (four midwest and four southeast, Table 3) met the 1988–2005 criteria. The midwestern states implemented their sampling programs earlier than the southeastern states. The average length of record for the eight states was 12 years. Only

Iowa had data that spanned the full period from 1988 to 2005. Statewide trends in fish Hg were analyzed from 1996 to 2005 for comparison with wet Hg deposition during the same period. Data for six states (three midwest and three southeast, Table 4) met the 1996–2005 criteria.

Results

Site trends, 1969–2005

Hg concentrations in fish between 1969 and 2005 from 90 river and lake sites across the USA had no temporal trends at 57% of the sites, downwards trends at 32% of the sites, and upward trends at 11% of the sites (Fig. 1; Supplemental Table A). The direction of trends was similar at river and lake or impounded sites. Fifty percent of the lake and impounded sites and 41% of the river sites had trends, the ratio of downward to upward trends being 9 to 2 for lakes and impoundments

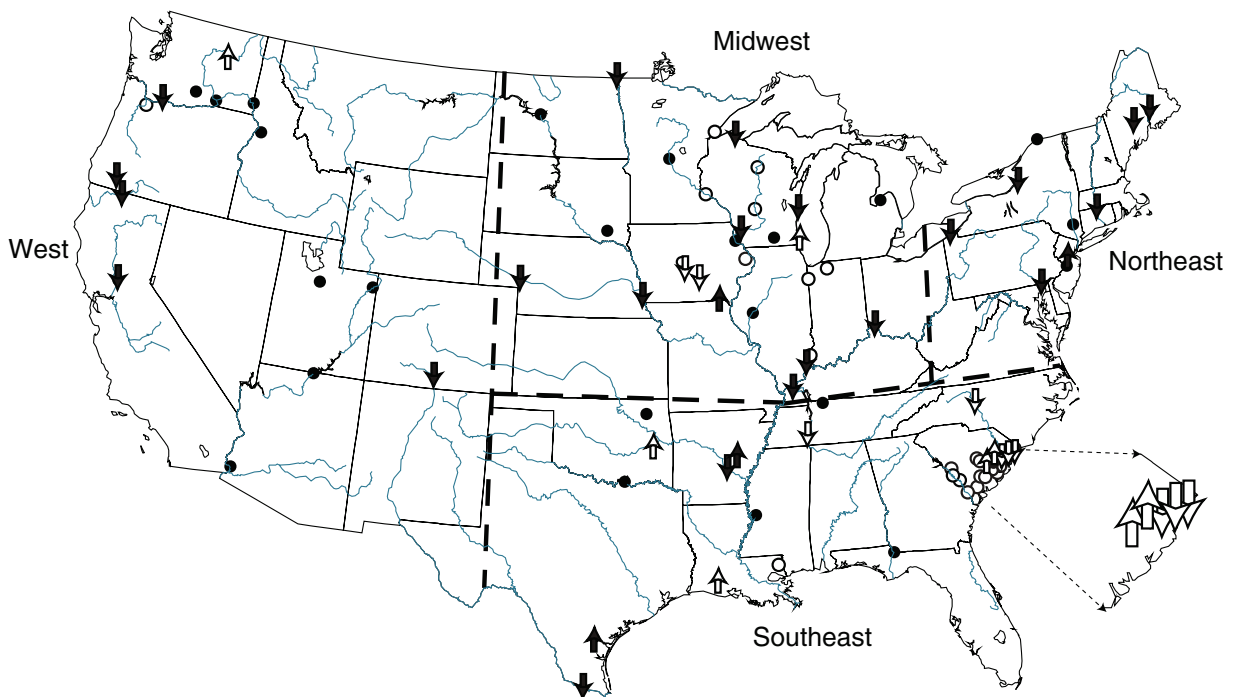


Fig. 1 Map showing Hg trends in fish tissue at 90 sites in the USA, 1969–2005. One site in Alaska (no trend) is not shown. *Arrows* sites with significant trends ($p < 0.05$). *Circles* sites without significant trends. *Black symbols* 50

sites sampled 1969–1987. *White symbols* 40 sites sampled mostly 1988–2005. Trends in South Carolina are shown in larger view. *Heavy dashed lines* mark four regions of the country

and 5 to 2 for rivers. The trophic level of fish species, as defined by their food habits, did not appear to affect Hg trends. Downward trends were found in 26% of fish species in the lower trophic level and in 28% of fish in the upper trophic level. Twice as many downward trends as upward trends were found in fish species from both lower and upper trophic levels. The overall ratio of downward and upward trends in fish Hg concentrations were relatively even at sites across most of the country: the ratio of downward to upward trends was about 6 to 1 except in the southeast, where it was 4 to 3 (Fig. 2).

Twelve of the 39 sites with temporal Hg trends in fish had Hg concentrations in fish tissue greater than 0.3 µg/g, the EPA advisory level for protection of human health (Supplemental Table A). Of the 12 sites, six sites in the northern USA had fish Hg concentrations starting above EPA guidelines and ending below the guidelines, three sites in the southeastern USA had fish Hg concentrations starting below EPA guidelines and ending above EPA guidelines, and three sites in South Carolina with downward trends had fish Hg concentrations that remained above EPA guidelines.

Site trends, 1969–1987

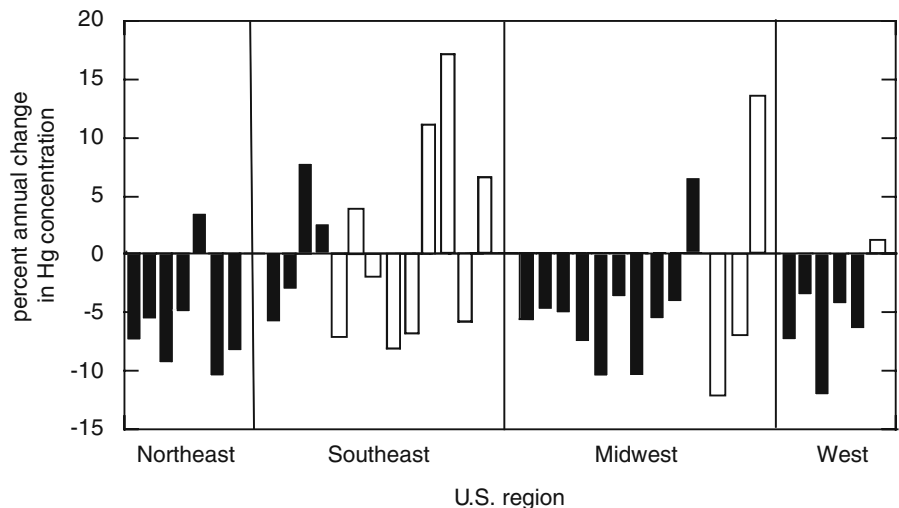
Site trends were focused on a subset of 50 NCBP sites from 1969 to 1987 (Fig. 1; Table 2). These trends in Hg concentrations in fish were down-

ward for 22 sites, upward for four sites, and no trends for 24 sites. More downward Hg trends were found at sites in the northeast, midwest, and west than at sites in the southeast (Fig. 2). Thirteen of the 22 sites with downward trends had an inflection point around 1980, from an initial rapid decrease in fish Hg concentrations in the 1970s to a more gradual decrease, or to no decrease in fish Hg concentration in the 1980s (Fig. 3). May and McKinney (1981), Lowe et al. (1985), Schmitt and Brumbaugh (1990), and Schmitt et al. (1999) reported similar changes in fish Hg concentrations from NCBP data. May and McKinney (1981) found the only significant decrease in national geometric mean fish Hg concentrations in the NCBP data was from 1972 to 1977. No significant changes in national geometric mean Hg concentration were found in the NCBP data from 1978 to 1981 (Lowe et al. 1985) or from 1976 to 1984 (Schmitt and Brumbaugh 1990).

Statewide trends, 1988–2005

Fish data, aggregated by state and by species, were used for evaluation of trends in fish Hg concentrations from 1988 to 2005 (Table 3). No trends in fish Hg concentrations were found in 52% of fish species from eight states. The only upward statewide trends in fish Hg concentrations were found in southeastern states (Supplemental Fig. A). Upward statewide trends in Hg concentrations

Fig. 2 Direction and magnitude of significant trends (*p* value < 0.05) in Hg concentration in fish tissue between 1969 and 2005. *Black bars* trends at 26 sites sampled 1969–1987. *White bars* trends at 13 sites sampled mostly 1988–2005



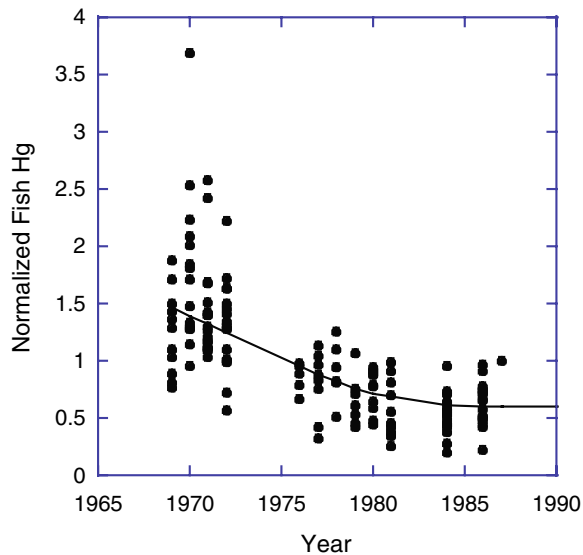


Fig. 3 Normalized Hg concentrations in whole fish collected by the National Contaminant Biomonitoring Program from 22 sites with significant ($p < 0.05$) downward trends, 1969–1987. Fish Hg concentrations were normalized by expressing the Hg concentrations of each fish as a percentage of the mean Hg concentration for that fish species and site. The curve is fitted using the locally weighted least squared error method. Trend analysis results for these sites are in Table 2

in fish were detected in Louisiana largemouth bass, channel catfish (*Ictalurus punctatus*), and freshwater drum (*Aplodinotus grunniens*); in South Carolina bowfin (*A. calva*); and in Georgia largemouth bass. Upward trends were above EPA advisory level for protection of human health guidelines in South Carolina bowfin. Upward trends in Louisiana largemouth bass and freshwater drum and Georgia largemouth bass started below EPA guidelines in the early 1990s and ended above the guidelines in the early to mid-2000s. Downward trends were detected in Louisiana bowfin and bigmouth buffalo (*Ictiobus cyrinellus*) and in South Carolina largemouth bass. Downward trends in Louisiana bowfin and South Carolina largemouth bass remained above the EPA advisory level into the mid-2000s (Supplemental Fig. A). States in the midwest had fewer trends than states in the southeast. No trends were detected in eight out of 12 fish species from states in the midwest (Table 3). All the statewide trends in fish Hg concentrations

were downward in midwestern states (Table 3). Downward trends were detected in Iowa common carp (*C. carpio*) and channel catfish and in Minnesota northern pike (*Esox lucius*) and walleye (*Sander vitreus*) (Supplemental Fig. A). Downward Hg trends in Iowa common carp and channel catfish were below EPA guidelines from the late 1980s and early 1990s to 2005. Downward trends in Minnesota northern pike and walleye started at or above EPA guidelines in the late 1980s and fell below the guidelines by 2001. Downward trends in midwestern states have been observed in other studies. Downward trends in walleye from northern Wisconsin lakes were also reported by Madsen and Stern (2007) between 1982 and 2005. Rasmussen et al. (2007) found trends between 1982 and 2005 in Wisconsin walleye varied with latitude; northern lakes had downward trends, middle latitude had no trends, and southern lakes had upward trends.

Some of the differences in fish Hg trends between states in the midwest and states in the southeast may reflect the period in which the samples were collected. Data from most states did not span the entire period from 1988 to 2005. In the midwest, most of the samples were collected during the late 1980s and 1990s, whereas most of the samples in the southeast were collected during the late 1990s and early 2000s. Monson (2009) reported downward trends for Hg concentrations in northern pike and walleye from Minnesota lakes before the mid-1990s, followed by upward trends after the mid-1990s.

Statewide trends, 1996–2005

Fish Hg data, aggregated by state and by species, were also evaluated from 1996 to 2005 for comparison with wet Hg deposition during the same period (Table 4). No Hg trends were found in 62% of fish species from six states. Fish Hg trends from states in the southeast were mostly upward. Upward trends were found in four South Carolina fish species [bowfin, blue catfish (*I. furcatus*), redear sunfish (*Lepomis microlophus*), and bluegill (*Lepomis macrochirus*)]. In Louisiana, upward trends in Hg concentrations were found in three species [flathead catfish (*Pylodictis olivaris*), channel catfish, and largemouth bass], and a downward

trend in one species (bowfin). Trends in Hg concentrations in the midwest were mixed; upward trends in Indiana channel catfish and downward trends in Minnesota northern pike (Table 4).

Discussion

Comparison to atmospheric emissions

An important question is whether trends in fish Hg concentration mirror trends in Hg emissions or trends in Hg inputs to aquatic ecosystems, or both. Major sources of anthropogenic Hg include mining, emissions from fossil fuel combustion, industrial processes (metal smelting and refining, chemical production, cement, and pulp and paper), and waste incineration (USEPA 1997). From about 1940 to 1970, a period of rapid industrial growth and inadequate wastewater treatment, local point-sources were the dominant source of Hg in many urbanized areas (Pirrone et al. 1998; Balogh et al. 1999). Improved wastewater treatment, stricter regulation of discharges to air and surface waters (USEPA Clean Water Act, Clean Air Act, Safe Drinking Water Act, and Toxic Substance Control Act), and reductions in industrial use of Hg since the 1960s have eliminated most Hg point-sources in urban areas (Balogh et al. 1999). The rapid decrease in Hg concentrations in fish collected by the NCBP in the 1970s likely results from this overall reduction of Hg in direct releases to the atmosphere and surface waters (Fig. 3; May and McKinney 1981; Krabbenhoft et al. 2005). Regulations since 1990 have targeted air emissions (Clean Air Amendments; USEPA 2004; USEPA 2007). Estimated anthropogenic Hg emissions from the USA have decreased by almost 50% in the northeast and over 30% in the southeast and midwest between the mid-1990s and 2002 (Supplemental Fig. B). Over a similar period from the mid-1980s through the early 2000s, the BEST program reported that temporal trends in fish Hg concentrations were not evident in nine major river basins (Mississippi, Columbia, Rio Grande, Yukon, Colorado, Mobile, Apalachicola, Savannah, and PeeDee; Schmitt 2002; Hinck et al. 2006a; Schmitt et al. 2005; Hinck et al. 2007; Hinck et al. 2006b;

Hinck et al. 2004). We also found few downward trends in Hg concentrations in fish during this period (Tables 3 and 4).

Several factors could eliminate or delay any response in fish Hg concentrations to changes in anthropogenic Hg emissions. About half of the anthropogenic emissions contributing to Hg deposition in the USA originate from other countries (Seigneur et al. 2004). Although US emissions are decreasing, emissions from Asia and Africa are increasing (Pacyna et al. 2006), with the result that the global anthropogenic emission inventory has remained almost constant since the 1990s (Slemr et al. 2003). Estimated global anthropogenic emissions were between 1,270 and 2,140 tons per year in 1990, 1,900 tons per year in 1995 (Pacyna and Pacyna 2002), and 2,200 tons per year in 2000 (Pacyna et al. 2006). A lag between actual reductions in anthropogenic Hg emissions and decreases in atmospheric Hg occurs because re-emitted, natural, and anthropogenic emission sources each contribute roughly equal amounts of Hg to the atmosphere (Seigneur et al. 2004). Thus, measures of Hg deposition are better indicators of Hg loading than anthropogenic emissions.

Comparison to sediment cores

Although few records of atmospheric Hg deposition exist before 1995 (<http://nadp.sws.uiuc.edu/mdn/>), Hg deposition back to pre-industrial times can be estimated from sediment, peat, and ice cores (Schuster et al. 2002). Dated depth profiles of Hg in sediment cores show changes in Hg accumulation rates over time that correlate well with documented Hg utilization and environmental releases and have been shown to be an accurate record of changes in external loading (Krabbenhoft et al. 2007; Lockhart et al. 2000; Lamborg et al. 2002; Pirrone et al. 1998; Engstrom and Swain 1997). Declining Hg accumulation rates in lake sediments and bog peat have been reported for many areas of the USA from 1970 through 1990 (Norton et al. 1997; Engstrom and Swain 1997; Balogh et al. 1999; Lorey and Driscoll 1999; Kamman and Engstrom 2002; Van Metre et al. 2004; Perry et al. 2005; Mahler et al. 2006). These declining Hg accumulation rates have been at least partially attributed to improved wastewa-

ter treatment, stricter regulatory controls on discharges to air and surface waters, and reductions in industrial use of Hg during the 1960s and 1970s.

Fish Hg trends corresponded with the declining Hg accumulation rates found in many sediment cores and the implementation of stricter regulatory controls on direct releases to the atmosphere and surface waters in the 1970s. Most downward trends in fish Hg occurred between 1969 and 1987 (Fig. 2), with the most rapid decreases in Hg concentrations during the 1970s (Fig. 3). In particular, the direction of fish Hg trends at the 50 NCBP sites from 1969 to 1987 was similar to that in trends found in the USGS National Water Quality Assessment program sediment cores from 35 lake and reservoir sites characterizing Hg from 1970 to 2001 (Mahler et al. 2006). No trends in Hg concentrations were found in about half of the sediment core and fish tissue sites. In addition, there were more downward Hg trends in sediment cores and fish (35% and 44%, respectively) than upward trends (17% and 8%, respectively).

Comparison to wet deposition

Hg deposition occurs in two forms: wet deposition (associated with precipitation) and dry deposition (direct deposition of gaseous Hg(0) and Hg(II), and deposition of particles containing Hg; Lindberg et al. 2007). Many studies have demonstrated that dry deposition processes may contribute amounts of mercury to the landscape equal to or greater than wet deposition (Miller et al. 2005; Seigneur et al. 2004; Grigal 2002; Cohen et al. 2004; Driscoll et al. 2007) however, insufficient dry deposition data are available at this time to quantify trends. Wet deposition of atmospheric Hg in the USA has been measured by the Mercury Deposition Network since the mid-1990s. Forty-nine MDN sites were tested for trends using a Seasonal Kendall approach (Prestbo and Gay 2009). These were primarily rural sites in the eastern USA and Canada. No trends in annual Hg wet deposition rate were found in 76% of the sites (Prestbo and Gay 2009). Downward trends in wet Hg deposition were found at 18% of the sites and upward trends were found in 6% of the sites. Trends in wet Hg deposition were complicated by trends in amounts

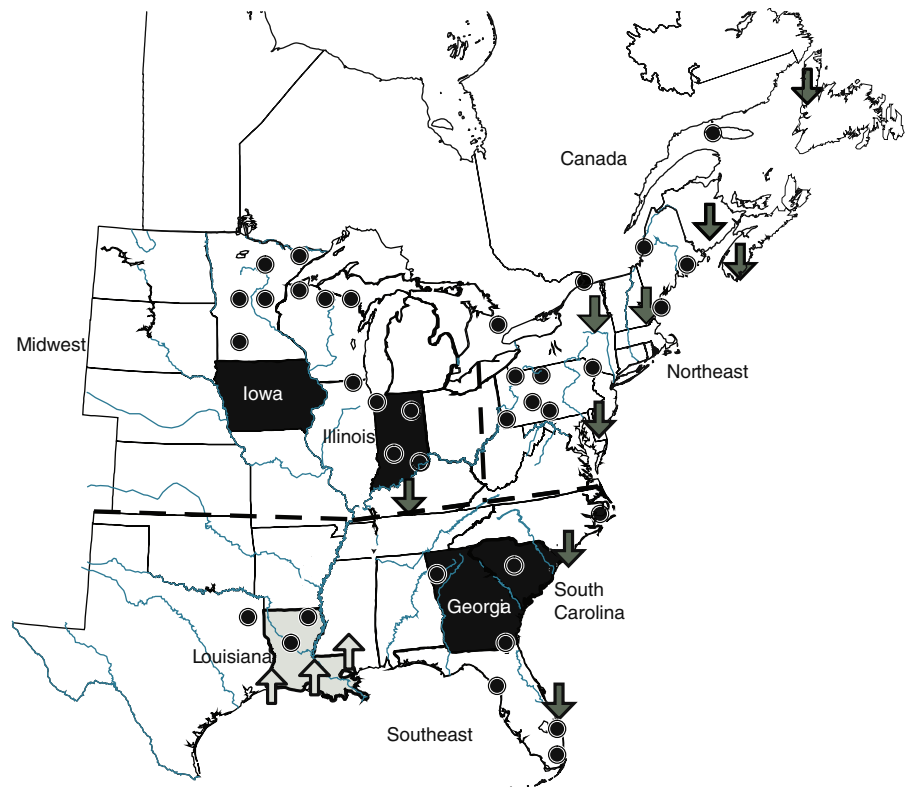
of precipitation. Of the nine sites with downward trends in wet Hg deposition, three sites also had decreasing amounts of precipitation that could account for the decrease in wet deposition. Similarly, two of the three sites (in Louisiana) with upward trends in wet deposition also had increasing amounts of precipitation, possibly explaining the increase in wet deposition over that period. The use of volume-weighted concentrations avoids the effect of variations in precipitation on deposition, but is less relevant when comparing to fish. What affects Hg concentrations in fish is not so much the concentration of Hg in wet deposition, but the amount of Hg entering the ecosystem, in other words, deposition.

Site-specific and statewide trend analyses consistently showed more upward trends in Hg concentrations in fish in southeastern states than other regions of the country (Figs. 1 and 2; Tables 3 and 4; Supplemental Table A). More upward Hg trends in fish in the southeast corresponded with the increased wet deposition in the region. Louisiana was the only state in which upward trends in Hg wet deposition were found (Fig. 4). Butler et al. (2008) also reported an upward trend in Hg wet deposition in the southeastern USA from 1998 to 2005, however, the upward trend was at a MDN site in Florida not Louisiana. The southeastern states may be more heavily influenced by long-range global transport resulting from large convective summer storms that scavenge Hg from the middle and upper troposphere (Butler et al. 2008). Increasing Asian Hg emissions may therefore have a greater impact in the southeastern USA than elsewhere in North America.

Fewer Hg trends in fish and wet deposition from 1996 to 2005 were found in the midwest than in the southeast. Only one downward trend in Hg wet deposition was detected among 14 MDN sites in the midwest (Fig. 4). Of eight midwest fish species analyzed for statewide Hg trends from 1996 to 2005, only two trends were detected: one upward (channel catfish in Indiana) and one downward (northern pike in Minnesota; Table 4). Butler et al. (2008) also reported downward Hg trends in wet deposition at two sites in Minnesota.

Although there were insufficient fish data from the northeastern USA for statewide trend analy-

Fig. 4 Comparison of Hg trends in channel catfish fillets to Hg trends in wet deposition at Mercury Deposition Network sites in the eastern USA and Canada, 1996–2005. Trends in channel catfish were analyzed in five states: Iowa, Illinois, Louisiana, South Carolina, and Georgia. *Grey states* upward trends in channel catfish fillets. *Black states* no trends in channel catfish fillets. *Arrows* direction of wet deposition trends at MDN sites. *Circles* MDN sites without wet deposition trends. *Dashed lines* mark regions of the USA



ses, site-specific data (1969–1987) indicate downward trends at six out of 10 sites in the northeastern USA (Fig. 1; Table 2). More recently, Simonin et al. (2009) reported an average decrease of 14% in Hg concentrations in yellow perch from lakes in New York from 1987 to 2005. These results correspond with those for MDN sites in the northeast, including Nova Scotia and Newfoundland, where downward trends in Hg wet deposition were found at six out of 15 sites (Fig. 4). In contrast, Butler et al. (2008) reported no trends in Hg wet deposition at 12 MDN sites in the northeast from 1998 to 2005. Overall, no trends were found in 62% of statewide fish Hg data and in 80% of MDN wet deposition sites in the eastern USA from 1996 to 2005 (Table 4; Fig. 4). By comparison, Butler et al. (2008) reported no trends in wet deposition at 88% of MDN sites in the eastern USA from 1998 to 2005. Differences between Hg wet deposition trends reported by Prestbo and Gay (2009) and Butler et al. (2008) most likely were due to the different ways of data aggregation (weekly data, versus annual and warm season,

respectively), and lengths of record (1996–2005 versus 1998–2005, respectively).

Response time

Although few Hg trends were detected in either fish tissue or atmospheric wet deposition from 1996 to 2005 in the eastern USA, it is anticipated that Hg concentrations in fish will respond to changes in atmospheric Hg loading. Recent studies have linked levels of atmospheric Hg loading with Hg concentrations in fish (Hrabik and Watras 2002; Atkeson et al. 2003; Hammerschmidt and Fitzgerald 2006), loons (Evers et al. 2007), and mosquitoes (Hammerschmidt and Fitzgerald 2005), and directly with MeHg production (Orihel et al. 2006; Hammerschmidt et al. 2006). Hammerschmidt and Fitzgerald (2006) reported that Hg wet deposition between 1992 and 2004 accounted for about two-thirds of Hg variation in largemouth bass between 1990 and 1995. Hammerschmidt

and Fitzgerald (2006) suggested that reductions in anthropogenic Hg emissions could result in proportionally lower levels of Hg concentrations in fish, but that the magnitude and timing of the response are unclear. The response to reductions in Hg emissions near sources (mostly inorganic Hg) is expected to be more rapid than at remote locations dominated by input from the global Hg pool (mostly elemental Hg; Lindberg et al. 2007). Slower response times are expected in systems with large watershed to water surface ratios due to retention of Hg in the watershed (Grigal 2002). The storage of previously deposited Hg within the watershed coupled with gradual delivery downstream can create a time lag between implementation of Hg emission controls and reductions in Hg accumulation in lake sediments and aquatic biota (Kamman and Engstrom 2002). Therefore, the watershed could remain a major source of Hg to lakes and rivers for many years and delay the response of aquatic ecosystems to changes in emissions (Yang et al. 2002). However, recent Hg emissions appear to be more readily methylated, suggesting at least an initial rapid decrease in fish Hg concentration after reductions in Hg emissions (Harris et al. 2007; Hintelmann et al. 2002). As a result of these complexities and uncertainties, Hg concentrations in fish could respond over months, years, or decades following changes in atmospheric Hg emissions. If the reduction in Hg emissions is relatively small compared to the inventories of Hg in the ecosystem, the response of fish Hg concentrations is expected to occur more slowly and be smaller in magnitude (Krabbenhoft et al. 2007).

Conclusions

A national compilation of fish Hg data from state and federal monitoring programs was used to analyze trends in Hg concentrations in fish. Most downward trends in fish Hg occurred between 1969 and 1987, with the most rapid decreases in Hg concentrations during the 1970s. Fish Hg trends corresponded with the declining Hg accumulation rates found in many sediment cores and the implementation of stricter regulatory controls on direct releases to the atmosphere and surface

waters in the 1970s. The southeastern USA had more upward Hg trends in fish than other regions for both site and state aggregated data. Upward Hg trends in fish from the southeastern USA were associated with increases in wet deposition in the region. The southeastern states may be more heavily influenced by long-range global transport and increases in distant Hg emissions. No Hg trends were found in 62% of fish data, and 76% of wet deposition sites from 1996 to 2005. Although few trends were detected in Hg data in fish or wet atmospheric deposition from 1996 to 2005, it is anticipated that Hg concentrations in fish will decrease in response to decreases in atmospheric Hg loading; however, the magnitude and the timing of the response are uncertain.

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