

Spatial patterns of mercury in macroinvertebrates and fishes from streams of two contrasting forested landscapes in the eastern United States

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Abstract Controls on mercury bioaccumulation in lotic ecosystems are not well understood. During 2007–2009, we studied mercury and stable isotope spatial patterns of macroinvertebrates and fishes from two medium-sized (<80 km²) forested basins in contrasting settings. Samples were collected seasonally from multiple sites across the Fishing Brook basin (FB_{NY}), in New York's Adirondack Mountains, and the McTier Creek basin (MC_{SC}), in South Carolina's Coastal Plain. Mean methylmercury (MeHg) concentrations within macroinvertebrate feeding groups, and mean total mercury (THg) concentrations within most fish feeding groups were similar between the two regions.

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However, mean THg concentrations in game fish and forage fish, overall, were much lower in FB_{NY} (1300 and 590 ng/g dw, respectively) than in MC_{SC} (2300 and 780 ng/g dw, respectively), due to lower trophic positions of these groups from FB_{NY} (means 3.3 and 2.7, respectively) than MC_{SC} (means 3.7 and 3.3, respectively). Much larger spatial variation in topography and water chemistry across FB_{NY} contributed to greater spatial variation in biotic Hg and positive correlations with dissolved MeHg and organic carbon in streamwater. Hydrologic transport distance (HTD) was negatively correlated with biotic Hg across FB_{NY}, and was a better predictor than wetland density. The small range of landscape conditions across MC_{SC} resulted in no consistent spatial patterns, and no discernable correspondence with local-scale environmental factors. This study demonstrates the importance of local-scale environmental factors to mercury bioaccumulation in topographically heterogeneous landscapes, and provides evidence that food-chain length can be an important predictor of broad-scale differences in Hg bioaccumulation among streams.

Keywords Methylmercury bioaccumulation · Wetlands · Macroinvertebrate feeding ecology · Fish trophic position · Stream · Landscape topographic wetness

Introduction

Mercury (Hg) concentrations commonly exceed human and wildlife health guidelines in forested streams of the eastern United States (Driscoll et al. 2007; Scudder et al. 2009), including many that lack Hg point sources (Fitzgerald et al. 1998; Lindberg et al. 2007). In these ecosystems, atmospheric deposition delivers Hg to the landscape

(Hammerschmidt and Fitzgerald 2005) and land cover characteristics, particularly the extent of wetlands, influence its transformation to methylmercury (MeHg) and subsequent delivery of this bioavailable form to streams (St. Louis et al. 1996; Kolka et al. 1999; Wiener et al. 2006; Brigham et al. 2009; Scudder et al. 2009). The contribution of shallow (surface and shallow-subsurface flow systems) geochemical exchange between terrestrial compartments and aquatic habitats in streams makes them particularly sensitive to watershed characteristics that foster MeHg production and transport (Ward et al. 2010). Hg bioaccumulation in streams without Hg point sources has been strongly correlated to forest cover, wetlands extent and connectivity, and hydrologic alteration (St. Louis et al. 1994; Rypel et al. 2008; Scudder et al. 2009; Ward et al. 2010), as well as to aqueous MeHg, dissolved organic carbon (DOC) quantity and aromaticity, and pH (Krabbenhoft et al. 1995; Mason et al. 1995; Driscoll et al. 1998; Grigal 2002; Chasar et al. 2009; Brigham et al. 2009).

Trophic position is an important predictor of MeHg concentrations of consumer organisms within streams (Chasar et al. 2009; Ward et al. 2010), and is an important predictor of Hg in top predator fishes across lakes (Chen and Folt 2005), but has not factored as highly in prediction of fish Hg across streams and rivers (Chasar et al. 2009; Ward et al. 2010). Many stream studies have covered relatively broad geographic, source condition, and (or) hydro-geomorphic extents, which emphasize landscape and associated physiochemical controls [Brumbaugh et al. 2001; Kamman et al. 2005; Chasar et al. 2009; Scudder et al. 2009; U.S. Environmental Protection Agency (USEPA) 2009; Glover et al. 2010]. However, studies addressing a limited range of environmental conditions (spatial coverage, Hg inputs, geochemical characteristics, land cover) are more appropriate for assessing the influence of community structure or trophic relationships on MeHg bioaccumulation (Tsui et al. 2009). These are critical for resource management, yet generally are lacking for small streams (less than 100 km²).

The current study was conducted over 3 years in two medium-sized (<80 km²) basins, Fishing Brook in the central Adirondack Mountain region of NY (FB_{NY}) and McTier Creek in the Sand Hills portion of the inner Coastal Plain of SC (MC_{SC}). Both basins are considered “biological Hg hotspots” (sensu Evers et al. 2007), with (1) relatively high Hg deposition (National Atmospheric Deposition Program Mercury Deposition Network, <http://nadp.sws.uiuc.edu/mdn/>), (2) substantial forests and riparian wetlands within the watershed, (3) seasonally fluctuating water levels, and (4) resident fish and other biota with Hg concentrations that exceed both human and wildlife fish consumption advisory levels (Evers et al. 2007; Driscoll et al. 2007; Glover et al. 2010; NYDOH 2010; SCDHEC

2010). These basins differ from each other with respect to food web, latitude, topography, trophic diversity, climate, hydrology, wetlands type, and geochemistry. Thus, comparison of within-basin spatial patterns of Hg bioaccumulation between these two contrasting basins should provide insight into important environmental controls on Hg bioaccumulation in small stream basins. The objectives of this study were to (1) describe spatial variation in Hg concentrations in selected lower consumer feeding groups within the FB_{NY} and MC_{SC} basins, (2) describe the relationship of biotic Hg with potential biological, chemical, and landscape controls in each basin, and (3) compare patterns and potential drivers between the two study areas.

Methods

Site selection and basin characterization

Sites were selected to include a range of landscape and geomorphic characteristics across each study basin (Fig. 1; Table 1). Eight sites were selected within FB_{NY} and four were selected within MC_{SC}. In keeping with its more topographically diverse setting, the topographic and land cover variation among FB_{NY} sites was much greater than that among MC_{SC} sites (Table 1). For example, wetland variation was more than ten times greater across FB_{NY} than across MC_{SC}, and the amount of open water in FB_{NY} ranged more than tenfold (from 0 to more than 10% of basin area), while open water in MC_{SC} was less than 2% of basin area in all cases. The FB_{NY} sites also covered large ranges of landscape slope, drainage density, and distance to stream channel (Online Resource #1). These characteristics are captured by the hydrologic transport distance (HTD) metric, which was derived from the 10-m National Elevation Dataset (<http://seamless.usgs.gov>, accessed 8 March 2010) as the overland flow distance metric (OFD, System for Automated Geoscientific Analysis; Cimmery 2007). We believe this metric represents shallow subsurface flow paths to the stream network across the FB_{NY} study area. At high flow conditions, some of this flow may occur as overland flow (Schelker et al. 2011), but at the low-to-moderate flow conditions that prevail during much of the growing season, most water moves as shallow subsurface flow. Therefore, we use the term “HTD” in this study to avoid connoting flow over the land surface. FB_{NY} sites covered a much wider range of mean HTD than the MC_{SC} sites (85 vs. 17 m), and included the smallest (146 m) and largest (241 m) mean HTD values among all study sites (both basins).

FB_{NY} in-stream habitat varies among sites, from cascading riffles with limited pool or run habitat (site UT) to valley bottom sinuous channels with alternating pools and

Fig. 1 Locations of Fishing Brook and McTier Creek study areas in the eastern United States, and locations of sites from which biota were collected during 2007–2009 for Hg analysis. Site names are given in Table 1

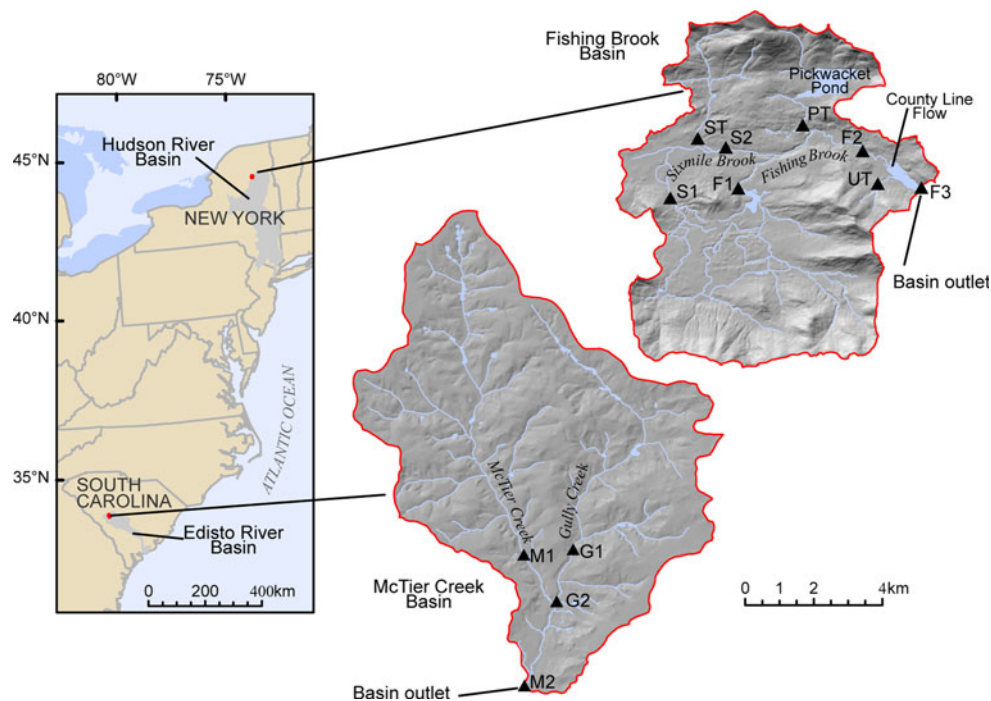


Table 1 Site names, basin-scale land-cover characteristics, pH, and concentrations of FMeHg and DOC

Site abbreviation and name	DA (km ²)	Wet (%)	OW (%)	Dev (%)	HTD (m)	pH	FMeHg (ng/L)	DOC (mg/L)
New York study sites								
UT: Unnamed Trib to County Line Flow near Long Lake	0.96	0.0	0.0	0.00	241	6.5	<0.04	2.63
ST: Sixmile Brook Tributary near Long Lake	1.0	6.8	1.1	0.00	189	7.1	0.34	8.37
S1: Sixmile Brook @ 28 N near Long Lake	4.6	6.8	0.6	0.00	156	6.6	0.35	12.3
PT: Pickwacket Pond Outlet near Long Lake	8.4	4.6	10.5	0.00	233	6.8	0.16	6.92
S2: Sixmile Brook near Long Lake	17.7	13.0	0.6	0.29	175	6.2	0.54	13.4
F1: Fishing Brook @ 28 N near Long Lake	27.1	5.8	1.2	0.10	217	6.4	0.22	8.67
F2: Fishing Brook above County Line Flow near Long Lake	60.6	8.6	2.2	0.21	203	6.5	0.27	8.87
F3: Fishing Brook (County Line Flow) near Newcomb	65.6	8.2	2.7	0.20	205	6.7	0.16	8.30
South Carolina study sites								
G1: Gully Creek Bridge on Old Shoals Rd near Monetta	25.9	5.3	1.6	12.7	176	4.5	0.17	4.92
G2: Gully Creek at McTier Creek	29.9	6.1	1.4	13.1	180	4.4	0.09	4.98
M1: McTier Creek near Monetta	40.5	5.1	1.2	20.9	163	5.4	0.15	5.91
M2: McTier Creek near New Holland	79.4	6.4	1.2	16.8	172	5.1	0.11	7.60

The pH, FMeHg and DOC are means of 5–27 base-flow samples collected during spring, summer, and fall 2007–2009. Sites are listed in order of increasing drainage area within each study area. Wetland land cover for FB_{NY} was derived from the Adirondack Park Agency Wetlands map (<http://www.apa.state.ny.us/Research/uh/uhreporttitle.html>, accessed 7 May 2010). Wetland land cover for MC_{SC} was obtained from NWI wetlands maps produced for the Edisto River Basin Project (Beasley et al. 1996). Developed land cover was derived from National Land Cover Data of 2001 (<http://www.epa.gov/mlcd/nlcd-2001.html>, accessed 1 Mar 2009)

DA drainage area, Wet wetland, OW open water, Dev developed, HTD hydrologic transport distance, FMeHg filtered methylmercury, DOC dissolved organic carbon

runs and limited or no riffle habitat (sites S1, S2, F2). The basin outlet site, F3, differs from all of the other sites in that it is the downstream end of a long, shallow ‘run-of the river’ impoundment, which is a common feature throughout the region. Fish and invertebrate habitat in FB_{NY}

consists of woody debris, rocky substrate, and occasional areas of submerged or emergent macrophytes. MC_{SC} stream sites are generally long runs and pools, with mainly sandy substrate, and habitat structure consisting of largely woody debris, leaf packs, undercut banks, deep pools, and

rootwads along the banks, with few areas of submerged and emergent macrophytes.

Targeted taxa

Macroinvertebrate and fish taxa targeted for collection were those that represented the breadth of primary and secondary consumer functional feeding groups in the study streams, and that could be collected in sufficient numbers and mass from multiple sites and collection periods. Functional feeding group classifications follow Merritt and Cummins (1996) and Thorp and Covich (2001) for macroinvertebrates, and the regional guides of Smith (1985) and Rohde et al. (1994) for fishes of NY and SC, respectively. For macroinvertebrates, the targeted functional feeding groups were shredders (mainly consume living and/or decomposing plant material), scrapers (mainly consume periphyton and associated biofilm), omnivores (consume both plant and animal material), and predators (mainly consume other macroinvertebrates). Invertebrate families within each of the above feeding groups were targeted as follows: shredders—northern case-maker caddisflies (Trichoptera: Limnephilidae); scrapers—flathead mayflies (Ephemeroptera: Heptageniidae); omnivores—crayfish (Decapoda: Cambaridae); and predators—dragonflies (Odonata: Aeshnidae and Libellulidae; kept separate in processing). Because crayfish and dragonflies can cover a large range of sizes (instars, in the case of dragonflies); these were separated into composites of like-sized individuals in field-processing.

Targeted fish taxa included both “forage fish” and predatory game fish. Forage fish are typically prey for larger predatory fish, and are smaller, shorter-lived species, with varying degrees of herbivory, detritivory, invertivory, or omnivory. The primary targeted forage fishes were invertivore/herbivores, particularly common shiner (*Luxilus cornutus*) and creek chub (*Semotilus atromaculatus*) in FB_{NY}, and shiners (*Notropis* spp., primarily yellowfin shiner, *N. lutipinnis*) in MC_{SC}. Blacknose dace (*Rhinichthys atratulus*, an invertivore) and northern redbelly dace (*Phoxinus eos*, an herbivore/invertivore) were also collected from FB_{NY}, and tessellated darter (*Etheostoma olmstedii*) and blackbanded darter (*Percina nigrofasciata*), which are both benthic invertivores, were also collected from MC_{SC}. For predatory game fish, we targeted piscivorous, locally important game species that are widely distributed in small streams of each region. Brook trout (*Salvelinus fontinalis*) >180 mm total length (TL) was the predatory game fish targeted in FB_{NY}. The brook trout diet, although predominately invertebrates, does include other fish (Smith 1985), and brook trout are typically the top predator in small Adirondack streams. Small streams in the

MC_{SC} study area can support a larger variety of predatory game fish (Rohde et al. 1994), so multiple species were included, as follows: largemouth bass (*Micropterus salmoides*) >200 mm TL, warmouth (*Chaenobryttus gulosus*) >100 mm TL, chain pickerel (*Esox niger*) >200 mm TL, redbfin pickerel (*E. americanus americanus*) >150 mm TL, American eel (*Anguilla rostrata*), and bullhead species (*Ameiurus* spp.). The full list of macroinvertebrate and fish taxa collected is provided in Online Resource #2.

Sample collection and field processing

Biota and stream water samples were collected seasonally from spring through fall during 2007–2009. Macroinvertebrates were collected from all sites. Fish were collected from a subset of FB_{NY} sites and from all MC_{SC} sites. The basin outlet sites (F3 and M2, Fig. 1) were sampled 8 and 7 times, respectively, during the course of the study; most other sites were sampled 3–5 times. Field measurements of pH and sampling of stream water were conducted within a week of biotic sampling. Water samples were collected with trace-metal clean techniques, and analyzed for filtered-water methylmercury (FMeHg) and DOC as described in Bradley et al. (2011).

Macroinvertebrates were collected by hand-picking, kick-netting, and bank-jabbing from all distinct habitat types (including cobbles, soft surface bed sediment, macrophytes and woody debris) with the goal of collecting three taxon-specific composites of 30 individuals each or at least 1 g wet weight per composite, with a minimum of 15 like-sized individuals per composite. Fish were collected by electrofishing, angling, and passive capture with traps and gill nets.

Specimens were placed in new plastic zip-seal bags with site water, and stored in coolers on wet ice for field processing as soon as possible. The average holding time for all samples was approximately 4 h. Field processing of macroinvertebrates and fish was done in accordance with trace-metal clean techniques (detailed in Scudder et al. 2008). Macroinvertebrates were sorted with pre-cleaned plastic forceps, rinsed in de-ionized water, dried, weighed, and stored on dry ice. Forage fish were processed as whole-body specimens, either individually or as composites of similarly sized individuals. Forage fish were weighed, measured (TL), rinsed in de-ionized water, and double-bagged. Predatory game fish were weighed, measured, and rinsed in de-ionized water. A standard skinless fillet was collected from one side, rinsed, weighed, and double-bagged. All fish samples were immediately placed on dry ice for transport to the laboratory, where they were kept frozen until further processing and analysis.

Hg and stable isotope analysis

Fish tissue was analyzed for total mercury (THg); MeHg (the form of Hg that is accumulated in organisms through diet) is known to comprise >95% of the Hg in fish tissue (Grieb et al. 1990; Bloom 1992). Macroinvertebrates were directly analyzed for MeHg due to the potential for widely varying taxonomic differences in MeHg to THg ratios (Mason et al. 2000). Henceforth, biotic Hg refers to MeHg; either directly measured (macroinvertebrates), or measured as THg and assumed to be primarily MeHg (fish).

Prior to analysis, samples were freeze-dried to constant weight and ground (in their entirety) to a fine powder, with a stainless-steel ball mill (Retsch Model MM200) or an ultracentrifugal mill (Retsch Model ZM200). Macroinvertebrate samples were analyzed for MeHg at the U.S. Geological Survey Wisconsin Hg Research Laboratory, with a dilute nitric acid extraction and cold-vapor atomic fluorescence spectroscopy (Hammerschmidt and Fitzgerald 2005). Laboratory precision for triplicates was 7.6% ($\pm 7.2\%$ standard deviation, s.d.), and accuracies for MeHg concentration in blind submissions of standard reference materials, as mean percentage of certified MeHg value \pm s.d., were as follows: NIST 2976 ($90.9 \pm 27.4\%$); TORT-2 ($93.1 \pm 14.2\%$); NRCC DOLT-3 ($83.5 \pm 9.7\%$). Fish samples were analyzed for THg at the Trace Element Research Laboratory (Texas A&M University, College Station, Texas) with USEPA Method 7473 (combustion and atomic absorption with a Milestone DMA-80 direct Hg analyzer). Accuracies for THg concentration in blind submissions of standard reference materials, as mean percentage of certified THg value \pm s.d., were: NIST 2976 ($90.5 \pm 14.5\%$); TORT-2 ($118.5 \pm 23.5\%$); and NRCC DOLT-3 ($98.2 \pm 10.7\%$).

Macroinvertebrate and fish samples were also analyzed for $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$. These stable isotope analyses were conducted at the Stable Isotope Geochemistry Laboratory at Florida State University's National High Magnetic Field Laboratory. Samples were analyzed with a ThermoQuest NC2500 Elemental Analyzer interfaced with a Finnegan MAT Delta Plus XP isotope ratio mass spectrometer. Isotope ratios were measured relative to reference gases and calibrated to known carbon and nitrogen standards ($\delta^{13}\text{C}_{\text{PDB}}$ and $\delta^{15}\text{N}_{\text{air}}$, ranging from -12.7 to -32.1‰ and -5.3 to 2.5‰ , respectively). Additional QA/QC included blind duplicates and blind standard reference material samples (glutamic acid, USGS-40, NIST-8573) included in sample runs (approximately two for every ten samples). Precision and accuracy for nitrogen and carbon isotopic ratios were $<0.4\text{‰}$, and generally $<0.2\text{‰}$, respectively.

Data analysis

Preliminary analysis of stable isotopes was done to verify functional feeding group assignment, to ensure the absence of strong seasonal and (or) annual patterns within sites before pooling samples among seasons and years, and to ensure trophic dependence within sites among prey and consumers. A summary of $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ data used in these analyses is provided in Online Resource #2. Normalization of $\delta^{13}\text{C}$ to lipid content was not done because corrections are generally small ($<1\text{‰}$) for epibenthic macroinvertebrates and fish, and non-normalized data are appropriate as a screening tool for diet dependence at the ecosystem scale (Post et al. 2007). Data were pooled across seasons and years after preliminary analyses indicated limited, nonsignificant, or inconsistent temporal variation within sites and taxa in Hg concentration and stable isotopes.

A one-way analysis of variance (ANOVA) followed by Tukey's HSD post-hoc multiple comparison tests was used to examine differences between group means in biotic Hg, $\delta^{15}\text{N}$, and estimated trophic position (i.e., taxon- or group-specific regional differences, differences among sites within regions, and differences among feeding groups). Hg concentrations were log-transformed when necessary to meet assumptions of normal distribution before ANOVA, and Welch's ANOVA was used in cases of failure of Levene's test for homogeneity of variances. Spearman rank correlation was used to examine the relation of group mean Hg concentrations with environmental variables. A Type I error (α) of 0.05 was used as a significance rule for all statistical tests, unless otherwise noted. Chemical data were pooled from base-flow samples collected multiple times (see Online Resource #3) per season during April through November in 2007–2009, and calculated as a growing season mean over spring, summer, and fall periods. Arithmetic means were used where all values were detected. Occasionally, FMeHg was not detected (<0.04 ng/L); in these cases, maximum likelihood means (Helsel 2005) were used (obtained from LIFEREG procedure in SAS). At site UT, all FMeHg samples were <0.04 ng/L, thus mean FMeHg was <0.04 ng/L. Growing-season means were a simple arithmetic mean of spring, summer, and fall means. Concentrations of FMeHg below the detection limit (0.04 ng/L) were treated as the detection limit value for plotting. Statistical analyses were done in SAS software, version 9.1 (SAS Institute, Inc., Cary, NC).

Spatial comparisons of biotic Hg were evaluated by taxon or feeding group (to minimize variability introduced by differences in feeding ecology), as well as (for regional comparison) by the broader 'forage fish' and 'game fish' groupings. Analyses of within-basin spatial patterns within each region focused on taxa and feeding groups collected

from at least five of the eight FB_{NY} sites or three of the four MC_{SC} sites. Data were pooled across sampling periods after preliminary analysis within sites and taxa indicated limited or no significant seasonal or annual bias in Hg and $\delta^{15}\text{N}$. Biotic Hg concentrations were based on dry weight, except where fish Hg was expressed as wet-weight concentration for comparison with human and wildlife consumption guidelines. A moisture content of 80% was applied uniformly for this conversion.

Average trophic position was calculated for consumer groups to account for potential site-to-site differences in $\delta^{15}\text{N}$ of nitrogen source inputs. Trophic position was calculated by taking the group mean of the difference between sample $\delta^{15}\text{N}$ and the mean $\delta^{15}\text{N}$ of the invertebrate feeding group consistently having the lowest $\delta^{15}\text{N}$ across sites (Anderson and Cabana 2007), and applying a fractionation constant of 3.4 (Post 2002).

Results

Shredders were the most widely collected macroinvertebrate feeding group across FB_{NY} (collected from all eight sites), followed by predators (six sites). Predators and omnivores were the most widely collected across MC_{SC} (four sites), but shredders were collected in very small numbers (as expected in blackwater Coastal Plain streams, Smock et al. 1985) from three MC_{SC} sites. Scrapers were collected from five FB_{NY} sites and no MC_{SC} sites. Among forage fish, common shiner and creek chub were each collected from five of the eight FB_{NY} sites and shiners were collected from all four of the MC_{SC} sites. Brook trout large enough to be considered predatory game fish (and classified as invertivore/piscivores) were collected from only two FB_{NY} sites; smaller brook trout (classified as invertivores) were collected from four sites. Benthic invertivores (darter species) and predatory game fishes were each collected from three MC_{SC} sites. These data, as well as those of other taxa and (or) feeding groups that were incidentally collected (and included in analysis of biomagnification patterns) are listed in Online Resource #2.

Regional comparison of biotic Hg and trophic position

Mean Hg within invertebrate feeding groups that were collected from both study areas were similar between the two regions (Fig. 2a). This was also the case for fish feeding groups, with the notable exception of much lower Hg for invertivorous fish from FB_{NY} than from MC_{SC} (ANOVA $F_{1, 204} = 32, P < 0.0001$). When fish were more broadly grouped, mean Hg concentrations were lower for both game fish and forage fish (ANOVA $F_{1, 33} = 7, P 0.015$, and $F_{1, 33} = 583, P < 0.0001$, respectively)

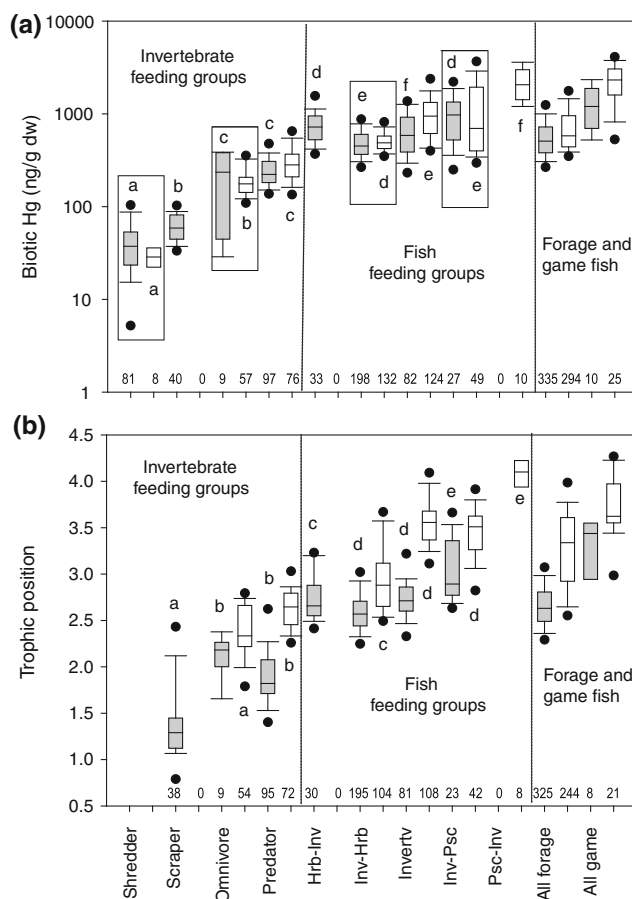


Fig. 2 a Hg concentrations and b trophic positions of macroinvertebrates and fish from streams in the Fishing Brook, NY (gray boxes) and McTier Creek, SC (white boxes) basins. Each box is based on data pooled among sites (eight NY and four SC) and sampling periods (April–Nov. 2007–2009). Lower and upper boundaries of each box represent 25th and 75th percentiles, lower and upper whiskers represent 10th and 90th percentiles, black dots represent 5th and 95th percentiles, and the line within the box represents the median. Abbreviations are as follows: Hrb-Inv herbivore–invertivore, Inv-Hrb invertivore–herbivore, Invtv invertivore, Inv-Psc invertivore–piscivore, Psc-Inv piscivore–invertivore. Groups that are not significantly different (nsd, alpha = 0.05; ANOVA on log-transformed values followed by Tukey’s HSD test) are indicated as follows: rectangle indicates nsd between study areas for particular organism group; the same letter indicates nsd within regions (letters for NY are above the boxes and letters for SC are below). Numbers of samples are above the x-axis

collected from FB_{NY} (1300 and 560 ng/g, respectively) than MC_{SC} (2300 and 780 ng/g, respectively). The U.S. EPA’s 0.3 $\mu\text{g/g}$ (wet weight basis) MeHg criterion for human health was exceeded in 31% of FB_{NY} game fish specimens and in 80% of MC_{SC} specimens. Similarly, the 0.1 $\mu\text{g/g}$ wildlife guideline was exceeded by a smaller percentage of forage fish specimens collected from FB_{NY} (50%) than MC_{SC} (62%).

Shredders exhibited the lowest $\delta^{15}\text{N}$ among all feeding groups in both study areas (see Online Resource #2) and were the base consumer proxy for trophic position

calculations. The resulting median trophic positions of FB_{NY} game fish (3.3) and forage fish (2.7) were 12% lower and 22% slower, respectively, than those of MC_{SC} game fish (3.7), and forage fish (3.3). The large regional difference in game fish and forage fish trophic positions corresponded with observed biotic Hg differences between the regions. Mean trophic positions of all macroinvertebrate and fish feeding groups tested were also significantly lower in FB_{NY} than in MC_{SC} (ANOVA F from 7 to 401, P from 0.013 to <0.0001), even for feeding groups for which Hg did not differ between regions. The trophic position difference was especially large (33% lower, ANOVA $F_{1, 204} = 32$, $P < 0.0001$) in the case of invertivore fish from FB_{NY} (2.7) than MC_{SC} (3.6), and corresponded with the large regional difference in biotic Hg of that feeding group.

The expected pattern of increasing Hg concentration within regions with increasing carnivory of the feeding group was generally similar between the two regions (Fig. 2a), and differences among feeding groups were highly significant (ANOVA $F_{7, 88} = 292$, $P < 0.0001$ for FB_{NY} ; $F_{6, 54} = 204$, $P < 0.0001$ for MC_{SC}). Mean Hg concentration of predator invertebrates (secondary consumers) was higher than that of shredders and scrapers (both primary consumers), and mean Hg of forage fish feeding groups were higher than that of predatory invertebrates. Among primary consumers in FB_{NY} , Hg was higher in scrapers than in shredders, and among forage fish groups in FB_{NY} , Hg was higher in the herbivore-invertivore group than in the others. A notable difference between the two study areas is the larger overlap among most fish feeding groups in FB_{NY} (including predatory game fish), than in MC_{SC} .

Hg, $\delta^{15}N$ and trophic position spatial patterns

Site-to-site differences in Hg concentrations for macroinvertebrates and for forage fish were relatively large across FB_{NY} (Fig. 3a), and small across MC_{SC} (Fig. 3b). In FB_{NY} , Hg concentrations were generally highest in the Sixmile sites (S2-3), lowest in the high-gradient tributary (UT; shredder macroinvertebrates only), and intermediate in the Fishing Brook sites (F1-3). In addition, biotic Hg was generally lower in the open water section of Fishing Brook (F3), than in F2 (which is well upstream of the open water section, see Fig. 1). Darters and predatory invertebrates from G1, the MC_{SC} tributary site, had significantly lower Hg than those from either mainstem site (Fig. 3b), but there were no discernable patterns for shiners and crayfish.

Whereas spatial variation of $\delta^{15}N$ within feeding groups was generally small across FB_{NY} (Fig. 3a), it was very large across MC_{SC} (Fig. 3b). Shredders collected from the high-gradient, non-wetland FB_{NY} site (site UT) were an

exception to this; mean shredder $\delta^{15}N$ was nearly an order of magnitude lower than any other FB_{NY} site. The MC_{SC} spatial pattern in $\delta^{15}N$ was fairly consistent, with more enriched $\delta^{15}N$ at M1, and least at G1, corresponding with the relative amounts of agricultural development in these sub-basins (Table 1).

Hg spatial patterns in relation to stream chemistry and landscape characteristics

The spatial pattern of mean Hg concentrations for most consumer groups collected from FB_{NY} was positively correlated to mean growing-season FMeHg and DOC (Fig. 4). In contrast, there was little discernable pattern in MC_{SC} , largely due to the limited range of conditions across the MC_{SC} sites (Fig. 4). Mean Hg concentrations within FB_{NY} shredder invertebrates, predator invertebrates, and shiner groups were positively correlated to growing-season mean FMeHg concentration, although the shiner regression somewhat exceeded our significance criterion. DOC concentration was positively correlated to Hg in shredders, which were collected across a wider range of DOC than other groups. High DOC (greater than 12 mg/L) was related to high biotic Hg in shiners and predator invertebrates (although their correlations were not significant). There was no significant or consistent relation between pH and Hg in any of the feeding groups in FB_{NY} due to the very limited pH range.

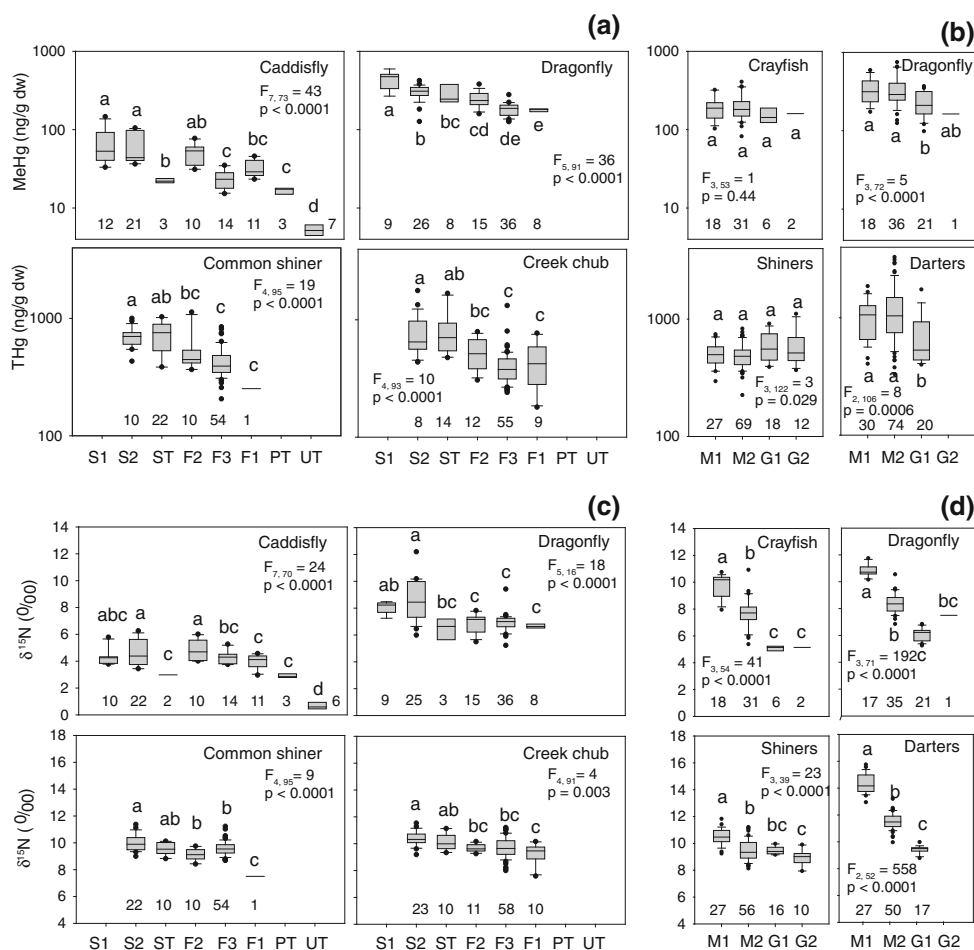
The pattern of consumer Hg across the FB_{NY} study area (Fig. 5) was significantly related to the composition and pattern of the FB_{NY} landscape. Shredder Hg was positively correlated to wetland abundance. There were no significant correlations, however, between wetland abundance and Hg in consumers that were collected across a narrower range of site conditions (i.e., wetland abundance 6–13%). Hg in all three FB_{NY} groups was strongly and inversely correlated to mean HTD (but with an elevated P -value for shiners due to the small number of sites).

Discussion

Inter-basin comparison of biotic Hg

MC_{SC} had much higher Hg in fish, and corresponding increased probability of exceeding human and wildlife advisory levels for forage and game fish (80 and 62%, respectively) compared with FB_{NY} (31 and 50%, respectively). Elevated fish Hg burdens have been widely reported in Coastal Plain streams relative to those in other forested settings across the United States (Scudder et al. 2009) and in other ecoregions within South Carolina (Glover et al. 2010). This pattern corresponded to generally

Fig. 3 Site-to-site variation of Hg and nitrogen stable isotope ratio ($\delta^{15}\text{N}$) in the Fishing Brook, NY (FB_{NY}) and McTier Creek, SC (MC_{SC}) basins. **a** and **b** are Hg in FB_{NY} and MC_{SC} , respectively; **c** and **d** are $\delta^{15}\text{N}$ in FB_{NY} and MC_{SC} , respectively. Lower and upper boundaries of each box represent 25th and 75th percentiles, lower and upper whiskers represent the 10th and 90th percentiles, black dots represent the 5th and 95th percentiles, and the line within the box represents the median. Site names are listed in Table 1. Sites are arranged in order of increasing mean HTD. Groups that are not significantly different (alpha > 0.05; ANOVA on log-transformed values followed by Tukey's HSD test) are indicated with the same letter. Numbers of samples are shown below each box



higher trophic position for secondary consumers in MC_{SC} , where the mean predatory game fish trophic position was 3.7 (s.d. 0.32) compared with 3.3 (s.d. 0.34) in FB_{NY} .

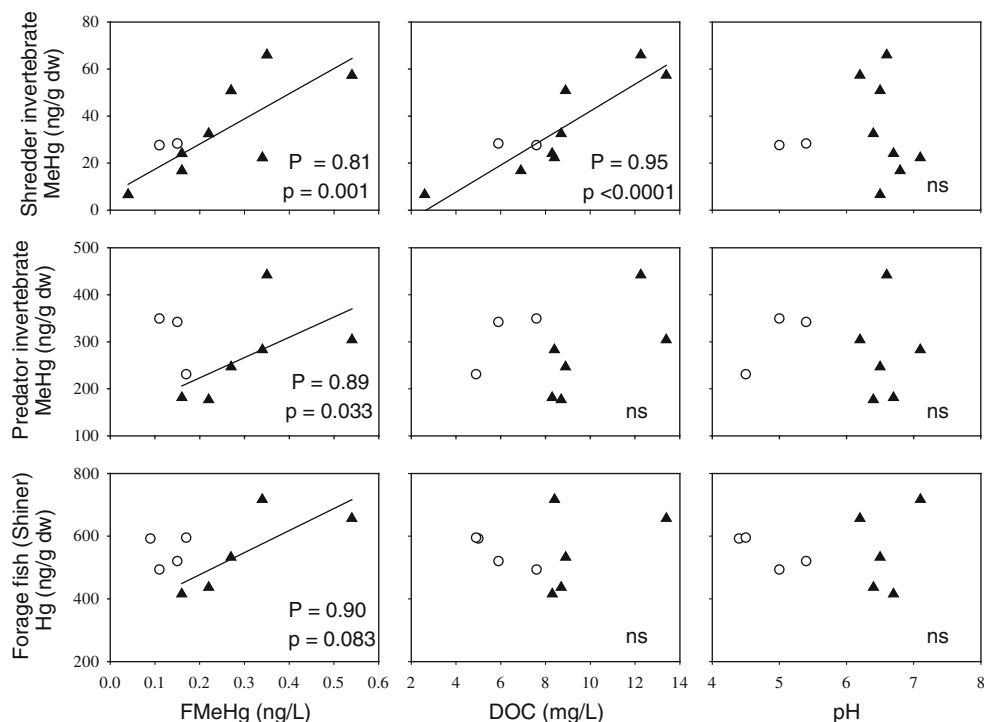
Food chain length, or number of trophic positions in the food chain below a predatory fish, is an important factor influencing fish Hg concentrations (Wiener et al. 2003); however, much of the data supporting this comes from studies of lacustrine ecosystems (reviewed in Wiener et al. 2003). Chasar et al. (2009) observed strong correlations between biotic MeHg and $\delta^{15}\text{N}$, a surrogate of trophic position, in seven of eight streams across the U.S., although relative trophic position of the fish did not appear to be as important as FMeHg at the base of the food web in explaining biotic Hg patterns across a large environmental gradient (in which mean FMeHg varied by several orders of magnitude). Among three Wisconsin streams, with a more limited range in FMeHg, Chasar et al. (2009) did observe an increase in fish Hg with trophic position. Food web and physico-chemical characteristics can be important drivers of consumer Hg burdens, if differences in food chain length are large relative to differences in MeHg concentrations at the base of the food web (Ward et al.

2010). In this study, the relative difference in food chain length between MC_{SC} and FB_{NY} (i.e., 12% longer in MC_{SC}) was sufficiently large to overcome any potential influence of the lower mean growing season FMeHg in MC_{SC} than FB_{NY} (relative difference 120%), lower DOC (relative difference 48%), lower wetland coverage (relative difference 18%) and the higher HTD (relative difference 17%).

Trophic position and consumer Hg

As expected, trophic position was an important within-site predictor of consumer Hg in both study areas. The increasing biotic Hg with increasing carnivory and piscivory in the diet corresponded with literature-based feeding group classifications in most, but not all, cases. In FB_{NY} , Hg in scrapers was at least twice that in shredders, both for all sites pooled and within sites and sampling dates, although both are considered primary consumers. Tsui et al. (2009) reported higher Hg in scrapers than shredders, related to a higher Hg content of periphyton as compared with detritus. Algal uptake may be an important MeHg

Fig. 4 Hg in shredder invertebrates, predator invertebrates, and invertivore–herbivore fishes (shiners, Cyprinidae: *Notropis* spp. or *Luxilus cornutus*) in relation to growing season mean FMeHg, DOC, and pH for Fishing Brook, NY (solid triangles) and McTier Creek, SC (open circles) stream sites. Biotic Hg concentrations are arithmetic means of MeHg (invertebrates) or THg (fish) collected during April–November 2007–2009. Chemical concentrations are arithmetic means from base flow samples collected during the same time period. Regression lines and Spearman rank correlations (P) are shown for NY sites



entry point for stream food webs, and primary consumption by scrapers, such as Heptageniidae, may be an important pathway of Hg transfer to higher trophic levels (Mason et al. 2000; Castro et al. 2007; Cremona et al. 2009; Ward et al. 2010). In our case, the higher Hg in scrapers may be attributed at least in part to their higher trophic position in relation to shredders, partly a function of the macroinvertebrate family targeted for this feeding group in our study. Higher trophic position estimates for Heptageniidae than other scraper families (and Limnephilidae caddisfly larvae) were also documented by Anderson and Cabana (2007), who suggest this is due to a greater tendency toward omnivory by Heptageniidae than most other scraper families. The high Hg of herbivore-invertivore fish in FB_{NY} compared with other forage fish feeding groups in FB_{NY} also did not agree with what would be expected for this classification, but did correspond with their higher trophic position relative to invertivore–herbivores, which suggests that the northern redbelly dace are consuming a relatively large proportion of invertebrates, and highlights the benefits of incorporating stable isotope data with literature classifications in contaminant studies.

Spatial patterns in biotic Hg

The relative spatial homogeneity of biotic Hg in MC_{SC} corresponds to a general lack of spatial variability in topography or stream geochemistry among study sites. Relatively homogeneous geochemistry for MC_{SC} was also

reported by Bradley et al. (2011). Strong relationships between fish Hg burdens, wetland percentage and water chemistry have been reported for South Carolina (Guentzel 2009; Glover et al. 2010), among sites spanning a greater range of ecoregions, waterbody types and sizes, and wetlands coverage (from less than 1% to more than 30%). Local reach and habitat characteristics that were not evaluated in the current study may be important sources of spatial variation in MC_{SC}, potentially explaining the significantly lower biotic Hg in darters and dragonfly larvae from the Gully Creek tributary site. Bradley et al. (2011) report significantly lower aqueous MeHg downstream of a small pond in the MC_{SC}, but biota were not collected from that site for the present study.

Spatial variation in consumer Hg was large across FB_{NY}. This large variation in biotic Hg on a relatively small spatial scale has been reported in Adirondack lakes (Driscoll et al. 1994; Simonin et al. 2008), and across streams within a somewhat larger (150 km²) basin in northern California (Tsui et al. 2009), but has not previously been reported for small Adirondack streams. Bradley et al. (2011) also report large spatial variation in geochemistry across the FB_{NY} study area.

Factors influencing biotic Hg spatial variation in FB_{NY}

The spatial pattern of Hg in FB_{NY} consumers corresponded closely with that of aqueous MeHg and, for some consumer groups, with DOC. A significant relationship between

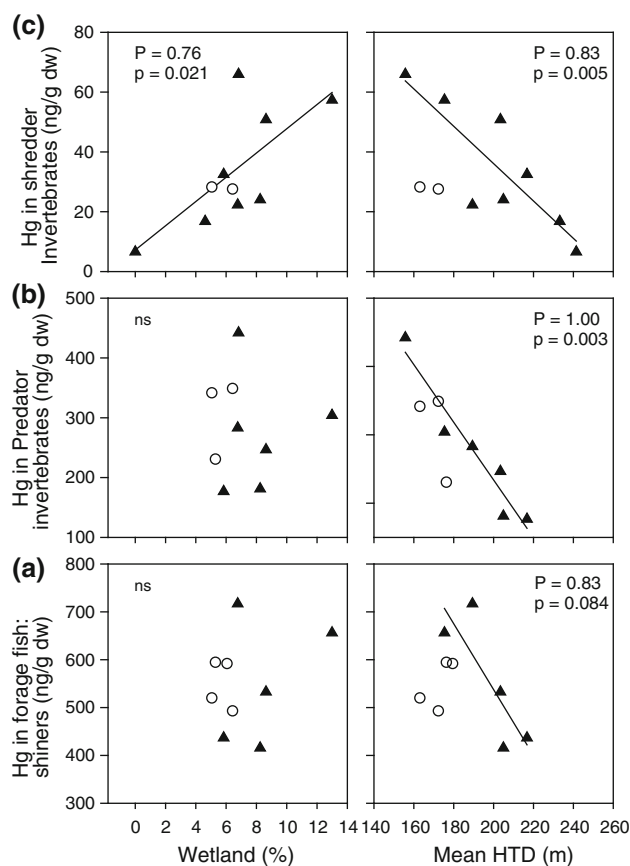


Fig. 5 Hg in: **a** shiners, **b** predatory invertebrates (dragonfly larvae), and **c** shredder invertebrates (caddisfly larvae) in relation to wetland abundance and mean HTD for NY (triangles) and SC (open circles). Biotic Hg concentrations are arithmetic means of MeHg (invertebrates) or THg (fishes) collected during April–November 2007–2009. Regression lines and Spearman rank correlations (P) are shown for NY sites

biotic Hg, dissolved MeHg, and DOC has been reported previously over broader geographic scales and across much larger geochemical gradients (e.g., Chasar et al. 2009; Scudder et al. 2009). Some studies report inhibitory effects of DOC on algal uptake and hence, bioaccumulation (Driscoll et al. 1994; Kamman et al. 2005; Gorski et al. 2008); however, DOC concentrations in our study were consistently lower than were those previously reported to inhibit algal uptake of MeHg.

Both DOC and MeHg are produced in riparian wetlands (Driscoll et al. 1994; St. Louis et al. 1994; Krabbenhoft et al. 1995), and DOC export from wetlands to streams facilitates MeHg transport downslope and downstream (Driscoll et al. 1995; Grigal 2002, 2003; Hall et al. 2008). The importance of riparian wetlands as sources of Hg within FB_{NY} was confirmed by Schelker et al. (2011). In the current study, wetland abundance was significantly correlated with MeHg only in shredder macroinvertebrates, the only consumer collected from the full range of wetland

abundances, including the site lacking a wetland. Remaining functional groups were collected across a narrower range of wetland coverages—too narrow for wetlands abundance to be a useful predictor of biotic Hg. Our results are similar to those of Ward et al. (2010), who found a 5- to 13-fold range of Hg in stream-dwelling juvenile Atlantic salmon (*Salmo salar*) across a narrow range of wetland abundance, but no strong relation between biotic Hg and wetland abundance.

The lack of strong and consistent correlations between biotic Hg and wetland abundance in this study (despite a large range in biotic Hg across the FB_{NY} study area) may be due to several factors. Imagery used to produce maps can significantly underestimate wetlands area (Tiner et al. 2002), especially where there is high forest cover (Lindsay et al. 2004). However, in our case we used wetland data from high-quality photogrammetric imagery for the FB_{NY} study area (http://www.apa.state.ny.us/Research/uh/uhre_porttile.html, accessed 7 May 2010). It is more likely that wetland area metrics do not sufficiently quantify small-scale landscape variations that are highly relevant to MeHg production and movement. For example, topographic depressions that are not classified as wetland (i.e., are too small, or too ephemeral for wetland classification based on vegetation) can produce significant amounts of aqueous MeHg and DOC (Grigal 2002; Lindsay et al. 2004). Additionally, the potential production of MeHg and DOC in wetlands and subsequent delivery to streams is strongly influenced by wetland characteristics that are not quantified by wetland abundance, including distance to the stream channel (Grigal 2002), hydrologic connection (St. Louis et al. 1996; Bradley et al. 2010), and wetland type and plant composition (St. Louis et al. 1996).

HTD emerged as a robust predictor of Hg in consumers within the FB_{NY} basin. HTD integrates slope, drainage density, and riparian area. These three basin characteristics affect Hg methylation and transport to the stream. HTD is positively related to slope and negatively related to riparian area and drainage density. Lower mean basin slope and greater riparian area favor greater formation of MeHg, while higher drainage density favors efficient MeHg transport to the stream in low-slope areas. Most of the transport of MeHg to streams in the FB_{NY} basins is thought to be via shallow groundwater flow from flat-lying riparian areas where wetlands are common (Schelker et al. 2011). Thus, decreasing HTD across the sub-basins in FB_{NY} reflects both an increase in factors associated with MeHg and DOC sources as well as a decrease in the transport distance of these constituents to the stream habitat. Because the life histories and behaviors of some organisms are timed to take advantage of the habitat and high-quality food sources available in seasonally flooded riparian wetlands (Resh et al. 1988), lower HTD may also indicate

greater opportunity for mobile macroinvertebrates, such as Limnephilidae caddisflies, to feed in wetland habitats.

The superior predictive ability of a topographic-based metric over simple wetland abundance has been noted elsewhere for aqueous MeHg and for DOC, but not for Hg in stream biota. Dennis et al. (2005) used terrain analysis of digital elevation data to determine an “soil drainage index” for predicting base-flow MeHg in Adirondack lakes. Lindsay et al. (2004) found “bottomland wetland area” (a Digital Elevation Map—derived metric that integrates extent, position, and connectivity of wetlands to a stream) to be an important control on runoff dynamics for forested catchments on the Canadian Shield. Canham et al. (2004) modeled DOC concentrations in Adirondack lakes with flow-path distances for particular land covers; and Frost et al. (2006) found that drainage density, slope, and stream length each were negatively related to DOC concentration and, together, accounted for 44% of the variation in DOC across a northwestern Wisconsin watershed.

Other local reach characteristics may contribute to spatial variability in Hg in surface waters and biota. We found consistently lower Hg in consumers collected from the shallow, open water portion of FB_{NY} (F3, County Line Flow) than from the site immediately upstream. This pattern corresponds with documented lower concentrations of aqueous MeHg and DOC in the County Line Flow (Bradley et al. 2011; Schelker et al. 2011). Aqueous MeHg loss in this reach may be due to sedimentation of particle-bound MeHg when going from channelized flow to shallow open water, and/or to demethylation by photoreduction (Bradley et al. 2011). The open water bodies in the FB_{NY} basin include natural lakes such as Pickwacket Pond (upstream of P2) and a shallow ‘run-of-the-river’ impoundment, County Line Flow (F3), which has a high flushing rate and was constructed nearly a century ago. Other types of impoundments, such as younger reservoirs (e.g., Hecky et al. 1991) and beaver impoundments, which are common throughout the Adirondacks and Coastal Plain, may affect Hg cycling and bioaccumulation differently at the reach scale (Driscoll et al. 1995, 1998; Selvendiran et al. 2008; Roy et al. 2009).

Conclusions

The spatial patterns of Hg in consumers in the two contrasting basins in this study can be attributed to food web and physico-chemical factors. The relative importance of each factor differs, however, depending on the geographic scale of interest. At the regional scale, comparing Hg bioaccumulation in consumers of two widely separated areas with very different landscapes, we found that food web processes, particularly food chain length, are the main

predictors of the regional differences in consumer Hg. On a local scale, we found that physico-chemical and landscape processes were the main predictors of biotic Hg in the topographically heterogeneous setting of the Adirondacks. The large within-region variability in Hg among different macroinvertebrate feeding groups and among different forage fish taxa emphasized the importance of considering detailed feeding ecologies, and the benefits of incorporating stable isotope data, for spatial comparisons at local scales.

Landscape metrics can predict Hg bioaccumulation and its spatial variation across small watersheds, as they do across larger geographic extents. Our results demonstrate that local-scale patterns of Hg concentrations in stream biota are strongly associated with hydro-geomorphology and landscape characteristics that influence MeHg production and delivery to streams. Important characteristics are short flow paths from topographic depressions to the stream channel, closely associated riparian wetlands, and elevated concentrations of aqueous MeHg and DOC. Within the topographically heterogeneous Adirondack region of NY, biotic Hg concentrations vary significantly over a relatively small spatial scale. In contrast, within the more homogeneous, flat Coastal Plain of SC, with less variable terrain and water chemistry, Hg concentrations in aquatic biota did not demonstrate any apparent relation with landscape characteristics at a similar spatial scale. Our results highlight the importance of considering local-scale spatial variability when assessing sensitivity of lotic ecosystems to Hg deposition in the geographically extensive Adirondack region of NY and the Coastal Plain region of the southeastern US. These findings have implications for the development of monitoring plans for Hg in water or biota, the ranking of Hg-sensitive areas, and the use of readily available landscape data. Hydro-geomorphic metrics such as HTD can be robust predictors of Hg in streams and stream biota within the heterogeneous Adirondack landscape and similar regions, and may be a more sensitive indicator than wetland density in topographically heterogeneous settings across limited ranges of wetland cover.

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Program. The use of trade, product, or firm names in this article is for descriptive purposes only and does not imply endorsement by the U.S. government.

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