Distribution and trends of mercury in aquatic and terrestrial biota of New York, USA: a synthesis of 50 years of research and monitoring

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Abstract

Mercury (Hg) inputs have particularly impacted the northeastern United States due to its proximity to anthropogenic emissions sources and abundant habitats that efficiently convert inorganic Hg into methylmercury. Intensive research and monitoring efforts over the past 50 years in New York State, USA, have informed the assessment of the extent and impacts of Hg exposure on fishes and wildlife. By synthesizing Hg data statewide, this study quantified temporal trends of Hg exposure, spatiotemporal patterns of risk, the role that habitat and Hg deposition play in producing spatial patterns of Hg exposure in fish and other wildlife, and the effectiveness of current monitoring approaches in describing Hg trends. Most temporal trends were stable, but we found significant declines in Hg exposure over time in some long-sampled fish. The Adirondack Mountains and Long Island showed the greatest number of aquatic and terrestrial species with elevated Hg concentrations, reflecting an unequal distribution of exposure risk to fauna across the state. Persistent hotspots were detected for aquatic species in central New York and the Adirondack Mountains. Elevated Hg concentrations were associated with open water, forests, and rural, developed habitats for aquatic species, and open water and forested habitats for terrestrial species. Areas of consistently elevated Hg were found in areas driven by atmospheric and local Hg inputs, and habitat played a significant role in translating those inputs into biotic exposure. Continued long-term monitoring will be important in evaluating how these patterns continue to change in the face of changing land cover, climate, and Hg emissions.

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Introduction

Mercury (Hg) pollution has impacted fish, wildlife, and human health in the northeastern United States, and New York specifically, for over 50 years. Emissions from natural and anthropogenic sources are transported via atmospheric circulation, resulting in deposition onto the landscape far from the point of origin (Driscoll et al. 2013). Point sources of Hg can augment atmospheric Hg influx and have created highly contaminated sites (Becker and Bigham 1995; Wang and Driscoll 1995). In response to concerns about human exposure to Hg and its effects on human health, monitoring efforts began in New York State in 1969. Since then, New York State has been a focal point of Hg research and monitoring, including emissions and deposition (Mao et al. 2017a, b; Ye et al. 2019), habitat sensitivity to deposition (Driscoll et al. 2007; Simonin et al. 2009; Riva-Murray et al. 2011; Burns and Riva-Murray 2018), spatiotemporal patterns (Levinton and Pochron 2008; Dittman and Driscoll 2009; Evers et al. 2011b; Razavi et al. 2019; Buxton et al. 2020; Dzielski et al. 2020; Richter and Skinner 2020; Millard et al. 2020; Riva-Murray et al. 2020, Razavi et al. 2020), effects on wildlife and humans (Baldigo et al. 2006; McKelvey et al. 2007; Gillet and Seewagen 2014; Schoch et al. 2014; Webster et al. 2021), invasive species (Brown et al. 2022), and the effects of meteorology and changing climate on Hg dynamics (Blackwell et al. 2014; Mao et al. 2017b; Adams et al. 2020). Given this wealth of information, this paper synthesizes 50 years of Hg research and monitoring efforts to assess the spatiotemporal patterns and current status of Hg exposure in biota across New York State, including the role of land cover and atmospheric deposition in statewide spatial patterns of Hg exposure in biota.

Synthesizing Hg data across species is challenged by complex transmission pathways (Scheuhammer et al., 2007; Sandheinrich and Wiener, 2011; Scheuhammer et al., 2015; Evers (2018)) and methylation processes in aquatic and terrestrial habitats (Gabriel and Williamson 2004; St. Louis et al. 2004; Cristol et al. 2008; Kim et al. 2008), after which methylmercury (MeHg) bioaccumulates and biomagnifies in food webs (Boening 2000; Ullrich et al. 2001). Wetland soils and aquatic sediment are important drivers of the methylation leading to variation in MeHg exposure across aquatic and terrestrial ecosystems (Ullrich et al. 2001, Cristol et al. 2008). Indicator species are often used to describe spatiotemporal patterns in exposure due to biomagnification and the sensitivity of higher trophic animals (Lane et al. 2011; Zananski et al. 2011; Weseloh et al. 2011; Yu et al. 2011; Schoch et al. 2014; Stenhouse et al. 2018), with some tissues, such as blood for birds and muscle for fish, being better indicators for these species (Jackson et al. 2015; Ackerman et al. 2016; Eagles-Smith et al. 2016).

Examining spatial patterns and temporal trends in Hg concentrations in organisms to identify locations of temporally persistent biotic Hg exposure that exceed adverse effects thresholds for populations or communities ('biological Hg hotspots') can be used to identify geographic areas of concern. Biological hotspots can be assessed by combining information over space in species likely to bioaccumulate Hg to determine areas with elevated tissue concentrations or where high percentages of the sampled population have Hg concentrations that exceed adverse effects thresholds (Evers et al. 2007; Ackerman et al. 2016; Eagles-Smith et al. 2016). Regions often lack data to properly assess spatial or temporal patterns, with such efforts only possible in well-studied areas (Weseloh et al. 2011; Eagles-Smith et al. 2016) or species (Yang et al. 2020). With Hg concentrations observed to exceed critical thresholds in New York State, an abundance of Hg data are available for synthesis. For wildlife, much of the data lies in piscivorous birds (Evers et al. 2008; Schoch et al. 2011; DeSorbo et al. 2020; Sauer et al. 2020) and invertivorous birds (Lane et al. 2011, 2020; Sauer et al. 2020), whereas, fish tissue Hg concentrations have been monitored for human health purposes for decades (Boulton, Hetling (1972), Kamman et al. 2005; Simonin et al. 2009; Yu et al. 2011).

This study synthesizes Hg exposure data across aquatic and terrestrial biota in New York State, combining over 20 databases with over 37,000 samples collected over the past 50 years. Using this comprehensive dataset, we evaluate the past and current patterns of Hg exposure in aquatic and terrestrial biota and the landscape-scale factors that influence these exposure patterns. The objectives of this analysis are to use these aggregated data sets to evaluate: (1) annual trends in total mercury (THg) in biota across ecoregions in New York State; (2) the decadal changes in the spatial distribution of effects hotspots for aquatic biota; and (3) the role of land cover and atmospheric Hg deposition on the spatial distribution of aquatic and terrestrial THg concentrations. For the third objective, we examine how land cover differentially affects aquatic and terrestrial species by comparing the relative contributions of habitat and atmospheric drivers of Hg exposure. We hypothesized that surrounding terrestrial habitats influenced Hg exposure in aquatic biota, while wetland habitats were an important driver of terrestrial Hg exposure.

Methods

Data compilation

Mercury data from biotic and abiotic endpoints were aggregated across New York State across multiple projects

Table 1 Tissue standardization equations for all tissue types converted for birds, mammals, and fish

Taxa	Tissue Convert to	Mercury value conversion Equations	Source
Bird			
Songbird	Blood	(tail feather – 0.64)/3.38 = blood	Jackson et al. 2011
Raptor	Blood	(body feather - 0.85)/2.14 = blood	Jackson et al. 2011
Non-raptor piscivore	Blood	0.673* ln(feather) - 1.673 = ln(blood)	Eagles-Smith et al. 2008
	Blood	0.673* ln(feather) - 1.673 = ln(blood)	Eagles-Smith et al. 2008
Aquatic non-piscivore[a]		(1.5544* egg) + 0.2238 = blood	Eagles-Smith et al. 2008
		0.673 * ln(feather) - 1.673 = ln(blood)	Eagles-Smith et al. 2008
		(ln(egg) + 0.75914)/0.9316 = ln(blood)	Eagles-Smith et al. 2008
		0.970* ln(liver) - 1.929 = ln(blood)	Evers et al. 2005
		1.003* ln(kidney) - 2.008 = ln(blood)	Evers et al. 2003
		1.080* ln(muscle) - 1.024 = ln(blood)	Evers et al. 2005
Mammal			
Bat	Fur	(274.27* blood) - 0.82 = fur	Wada et al. 2010
Mink	Blood	liver/0.46 = fur	Eccles et al. 2017
River Otter	Blood	kidney/0.64 = fur	Eccles et al. 2017
		brain/0.13 = fur	Eccles et al. 2017
		liver/0.80 = fur	Eccles et al. 2017
		kidney/0.62 = fur	Eccles et al. 2017
		brain/0.15 = fur	Eccles et al. 2017
Fish			
All Fish	Whole	0.74 * muscle = whole	Eagles-Smith et al. 2016

All tissues were measured in parts per million wet weight (ppm ww) except feathers and fur, which were measured in ppm fresh weight (fw), and eggs measured in ppm fresh wet weight (fww). The original tissue type used for invertebrate, amphibian, and reptile tissues is listed in the equations

[a]This group includes ducks, geese, waders, shorebirds, and seabirds. For full list of species, see Appendix B

(Appendix A, Table A1). Much of the data were gathered from public databases, including the New York Department of Environmental Conservation (NYSDEC) Fish Contaminants Database (https://www.dec.ny.gov/chemical/8437.html) and the U. S. Geological Survey National Water Information System (https://doi.org/10.5066/F7P55KJN). The remaining data were obtained from private data sources, including individual researchers and data compilations by the Biodiversity Research Institute. Data collection dates ranged from 1969 to 2017, with 63% collected from 1998 onward.

Laboratory methods of Hg determination varied across sampling endpoints and studies, but methods for THg typically followed U.S. Environmental Protection Agency (USEPA) Method 245.6 (1991) until succeeded by SW-846 Method 7473 (1998). Notably, New York State Department of Environmental Conservation (NYSDEC) did not switch until 2014 (Richter and Skinner 2020). Both THg determination methods have high intra-method precision and use different standards, though there was consistent variation in standard recovery (Lowery et al. 2007). NYSDEC found high consistency when changing methods concordance correlation coefficient between the two methods $\rho_c = 0.987$

(95% CI = 0.975–0.993). Samples were analyzed with these methods when THg concentrations closely approximated tissue MeHg (Wagemann et al. 1997; Rimmer et al. 2005). MeHg concentrations were quantified (USEPA Method 1631) when MeHg was less than the dominant fraction in tissue types (e.g., whole invertebrates; see Appendix A, Table A2). When quality assurance/quality control information on Hg determination was lacking from a study, those data were excluded from the analysis.

Data validation and standardization

A representative tissue type was selected for each taxonomic group to simplify analysis (Appendix A, Table A2). All data within these taxonomic groups were converted to this single tissue type using past tissue comparisons (Table 1). Data were converted to standard units (parts per million; ppm) for tissue type by normalization with the generally accepted form of weight (i.e., wet (ww), dry (dw), or fresh (fw) weight). When individual sampling events—individuals sampled at the same time and place—involved multiple tissue samples, the tissue that accurately represented Hg from that location and was most frequently used was

Table 2 Correlation of proportional land cover type for New York State (National Land Cover Database, 2011) with the top four principal components that represent 92% of the total variation in the data

Land Cover Type	Major Principal Components			
	PC1	PC2	PC3	PC4
Open Water	−0.71	−0.59	−0.22	0.16
Developed, Open Space	−0.03	0.10	0.07	−0.42
Developed, Low Intensity	−0.05	0.07	0.09	−0.39
Developed, Medium Intensity	−0.05	0.05	0.10	−0.37
Developed, High Intensity	−0.03	0.03	0.07	−0.21
Bare Rock/Sand/Clay	−0.01	0.00	0.00	−0.01
Deciduous Forest	0.68	−0.57	−0.37	−0.03
Evergreen Forest	0.12	−0.14	0.69	0.43
Mixed Forest	0.10	−0.03	0.26	0.26
Shrub/Scrub	0.01	0.04	−0.02	0.07
Grasslands/Herbaceous	0.00	0.01	−0.01	0.01
Pasture/Hay	0.00	0.53	−0.49	0.45
Woody Wetlands	0.02	0.11	0.03	−0.07
Emergent Wetlands	−0.02	0.01	−0.01	−0.02

Correlation coefficients greater than 0.5 represent a strong relationship between the land cover and the component

chosen for analysis (see Appendix A for prioritization). Individuals sampled on numerous occasions over time (e.g., across seasons or years) were included for analysis. Blood and muscle tissues were given the highest priority, while feathers and organs were given lower priority due to lower concentrations of MeHg, and high inter-tissue variance (Low et al., 2020). Samples that do not represent MeHg from New York State (e.g., feathers from bird species that developed in areas outside of the state) or blood samples that were impacted by feather molt (e.g., songbird nestlings; Ackerman et al., 2011) were not considered for analysis.

Whole-body and muscle Hg samples from fish were length-standardized to control for variation in Hg exposure due to size and age. Length correction of tissue-converted Hg concentrations was conducted using a general linear mixed model (Eagles-Smith et al. 2016). The statistical model quantified the species-specific effect of total length on tissue Hg concentration using this form:

$$\log([Hg]) \sim Intercept + Total\,Length + \textbf{Species} + \textbf{Species}/\textbf{Length}$$

Bolded terms are random effects, with the interaction estimating a random slope. Model fit was excellent (conditional $R^2 = 0.93$). Each Hg measurement was adjusted to the mean total length of the sampled species by predicting from the model.

Habitat and foraging guild designations were also categorized for each species. Taxa were divided into two main groups, aquatic and terrestrial, based on the primary food web that each species utilized. While all fish were considered aquatic, and most birds and mammals considered terrestrial, the exceptions were piscivorous birds (e.g., Common Loons) and mammals (e.g., North American river otter; see Appendix B for classifications of each species as well as scientific names). Species were then categorized into foraging guilds (piscivore, carnivore, invertivore-piscivore, invertivore, omnivore, herbivore, and planktivore) based on diet information (Frimpong and Angermeier 2013; Wilman et al. 2014; Froese and Pauly 2018; Myers et al. 2019) using the following criteria: (1) if the diet of a species consisted of 80% or greater of an item (e.g., fish or invertebrates), the species was categorized within that foraging guild (e.g., piscivores or invertivores), (2) invertivore-piscivores were species whose diet consisted of both aquatic invertebrates (e.g., insects, mollusks, crustaceans) and small fish, (3) carnivores were species whose diet consisted primarily of terrestrial vertebrates, (4) granivores were included within the herbivore category, (5) omnivores were defined as species that consumed a variety of prey items including plant material (e.g., algae, plants, detritus), and animals (e.g., invertebrates, fish), and (6) planktivores included species that primarily feed on zooplankton and phytoplankton.

Adverse effects thresholds from past laboratory and field studies were used to assess the negative impacts of Hg exposure (Appendix A, Table A3). A range of Hg synthesis efforts were used for this assessment, and the effects of Hg on population demography were well-represented. However, information was lacking for mammals on these endpoints. As a result, we selected the most robustly supported physiological effect. Adverse effects thresholds were generalized within broad taxonomic groups and foraging guilds when more specific information was unavailable (e.g., using Carolina Wren effects thresholds for all songbirds).

Spatial and environmental covariate data

A grid cell system (1/8° resolution) was created to standardize and summarize sampling efforts across New York State. EPA Level III Ecoregions (hereafter 'ecoregions'), areas distinguished by habitat, topography, and soil structure (https://www.epa.gov/eco-research/level-iii-and-iv-ecoregions-state), were used to summarize data in an ecologically-relevant spatial framework. Only data where researchers provided the latitude/longitude of the sampling effort were included in the analysis. Atmospheric Hg deposition and land cover data were summarized across New York State using the grid system to examine the impact of atmospheric deposition and habitat on spatial patterns of Hg exposure. Dry and wet Hg deposition rates were simulated throughout the northeastern United States over the period of March – November 2010 by Ye et al.

(2018) using a three-dimensional chemical transport model, the Community Multiscale Air Quality (CMAQ)-newHg-Br. Ye et al. (2018) used the Lambert Conformal projection, so deposition values (μg/m^2) were transferred between grid types based on the nearest grid cell centroids. Land cover data were acquired from the National Land Cover Database (NLCD 2011; Homer et al., 2015). Principal component analysis created independent variables that combine information across land cover types (Table 2). Methodological details are found in Appendix C, but briefly, four principal components (PCs) were selected for use as analytical covariates and describe land cover variation from open water to deciduous forest (PC1), open water and deciduous forest to agricultural habitats (PC2), agricultural habitats to evergreen forests (PC3), and rural to urban habitats (PC4).

Analysis framework for describing spatial and temporal patterns in Biotic Hg exposure

Variation in species-specific THg concentrations over time and space was quantified using general linear mixed models. Models were fit in a Bayesian framework using packages *rstan* and *rstanarm* (Carpenter et al., 2017; Goodrich et al., 2018; Stan Development Team, 2018) within the R statistical modeling environment (R Core Team 2021). Three general linear mixed models were used to describe spatiotemporal patterns in aquatic and terrestrial biota. Aquatic species tissue Hg concentrations were estimated for the whole-body endpoint, and terrestrial species were estimated for whole blood. WAIC (Widely Applicable Information Criterion) was used to determine the most effective formulations for each model (Watanabe, 2013; further details on analytical methods are found in Appendix C).

In Model 1, temporal trends in tissue THg concentrations of species sampled more than 10 years were assessed with more than 400 samples. The model was formulated as:

$$\log([Hg]) \sim Intercept + \textbf{\textit{Year}} : \textbf{\textit{Species}} : \textbf{\textit{Ecoregion}} \\ + \textbf{\textit{Grid Cell}} + \textbf{\textit{Site Index}}$$

As above, bolded effects were specified as a Gaussian random effect with nesting used for year, species, and ecoregion. Site index is the unique combination of latitude and longitude of the sampling event, whereas grid cell is the grouping of sampling events within the larger 1/8° grid. Species for this effort included Bluegill, Brown Bullhead, Lake Trout, Largemouth Bass, Northern Pike, Smallmouth Bass, Pumpkinseed, Walleye, White Perch, White Sucker, Yellow Perch, Common Loon, Red-eyed Vireo, Saltmarsh Sparrow, and Seaside Sparrow; see Appendix B for scientific names.

For Model 2, the grid cells where species exceed adverse effects thresholds were estimated for terrestrial and aquatic species. A log-Gaussian linear model was run for each species group and specified as:

$$\log([Hg]) \sim Intercept + \textbf{\textit{Year}} + \textbf{\textit{Grid Cell}} : \textbf{\textit{Species}} \\ + \textbf{\textit{Site Index}} + Foraging\ Guild \\ + Standardized\ Tissue\ Type$$

Here, foraging guild and standard tissue type were included to account for differences in foraging ecology and sampling strategy (see Appendix C). Estimates from Model 2 were used to determine biological Hg hotspots at the grid cell scale based on the number of species with estimated values above effect thresholds. A secondary analysis was conducted in five periods for aquatic species (1969–1979, 1980–1989, 1990–1999, 2000–2009, 2010–2017; hereafter 'decades') to examine temporal changes in aquatic biological Hg hotspots.

Finally, Model 3 estimated spatial variation in tissue Hg concentrations. Model 3 is the same as Model 2, but tissue Hg concentration was estimated for each sampling location rather than by grid cell. Predictions made from Model 3 were used as data for a spatial modeling framework to determine the environmental drivers of spatial patterns in Hg concentrations. Using a preferential sampling model (Diggle et al. 2010), the drivers of spatial patterns of Hg concentrations were examined while minimizing bias associated with spatially non-random data collection. Using a non-homogenous Poisson process implemented using a stochastic partial differential equation framework (SPDE; Lindgren et al., 2011) estimated in a Bayesian framework using integrated nested Laplace approximation (INLA; Rue et al., 2009), and implemented in R (R Core Team 2021) using package *INLA* (Rue et al. 2017). This model estimates the spatial sampling distribution with the spatial pattern of THg exposure using this formulation:

$$\log([Hg]) \sim Intercept + S(\textbf{\textit{SPDE}}) + LC_{PC1} \\ + LC_{PC2} + LC_{PC3} + LC_{PC4} \\ + Hg\ Deposition_{Dry} + Hg\ Deposition_{Wet}$$

Where $S(SPDE)$ is a spatial random effect estimated using Matérn covariance, LC are the four principal components of land cover covariates, and Hg is the wet and dry deposition rates estimated from the CMAQ-newHg-Br model. Sampling effort is estimated via a log-Gaussian Cox process model fit jointly with S(SPDE) to account for potential biases. Each model was assessed using predictive assessment (see Appendix C for more detailed methods), and ΔWAIC was used to determine the relative predictive power of this model to a base model. A power analysis of
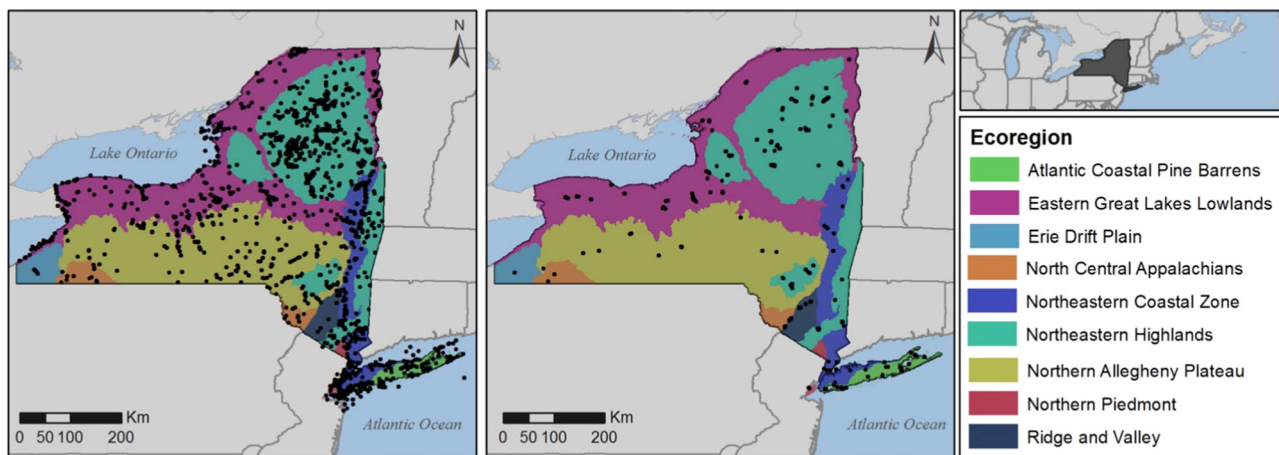
Fig. 1 Distribution of mercury sampling locations for aquatic (left) and terrestrial (right) food webs across New York State from 1969–2017 in relation to USEPA Level 3 ecoregions. Aquatic sampling includes 32,294 data points from 1878 unique sampling locations. Terrestrial sampling includes 5197 data points from 411 unique sampling locations

spatial patterns and temporal trends was also conducted for select species (Appendix D).

Results

Aquatic and terrestrial species were sampled across nine ecoregions in New York State (Fig. 1). Samples for aquatic species were collected from 197 taxa on 32,294 occasions from 1969 – 2017 across 545 grid cells. The arithmetic mean tissue- and length-standardized THg concentration from these data was 0.53 ppm ww whole body (range: non-detect – 203 ppm ww). Terrestrial species were sampled on 5,197 occasions over 140 grid cells from 1970 – 2017 across 206 taxa. The arithmetic mean tissue-converted THg concentrations for these species was 0.25 ppm ww blood (range: non-detect – 4.4 ppm ww). Further information on species- and tissue-specific THg or MeHg concentrations can be found for all data in Appendix B.

Bayesian R^2 values were excellent and ranged from 0.80–0.85 for Models 1-3, except for the pre-1980 spatial hotspot model ($R^2 = 0.65$). Small sample size is a likely cause of this poorer model fit, but the result was still considered adequate for comparison. Posterior predictive checks provided no indication that model assumptions were violated.

Temporal trends in Hg concentrations

The temporal trends indicator species data set included 22,705 Hg samples from 15 species, including 11 species of fish ($n = 20,908$ samples) and four species of birds ($n = 1797$). This data set included samples from 430 grid cells and all nine ecoregions: Northeastern Highlands ($n = 7546$), Eastern Great Lakes Lowlands ($n = 8801$),

Northeastern Coastal Zone ($n = 2213$), Northern Allegheny Plateau ($n = 2570$), Atlantic Coastal Pine Barrens ($n = 1119$), North Central Appalachians ($n = 176$), Ridge and Valley ($n = 101$), Northern Piedmont ($n = 94$), and Erie Drift Plain ($n = 55$).

There were 68 species/ecoregion combinations with three or more years of sampling, most of which did not show a significant trend in biota Hg concentrations (Fig. 2, see Appendix B for parameter estimates and 95% credible intervals). Increases in tissue Hg concentrations were found in only Northern Pike (piscivore) in the Northeastern Highlands ($\beta = 0.03[0.01, 0.05]$). Decreases were found in 7 ecoregion/species combinations, including three of the 11 fish species sampled in the Eastern Great Lakes Lowlands. Specifically, annual declines in THg were found in omnivorous fish, Pumpkinseed ($\beta = -0.02[-0.03, -0.01]$) and White Sucker ($\beta = -0.02[-0.03, -0.01]$); and a single invertivorous piscivore, Yellow Perch ($\beta = -0.01[-0.2, -0.0]$) in the Eastern Great Lakes Lowlands. Yellow Perch declined in the Northeastern Highlands ($\beta = -0.02[-0.03, 0.00]$). The Northeastern Coastal Zone had two species showing a declining trend: White Perch ($\beta = -0.04[-0.06, -0.02]$) and Brown Bullhead ($\beta = -0.02[-0.04, -0.01]$). The North Central Appalachians had a single declining species, Yellow Perch ($\beta = -0.06[-0.13, -0.0]$). The Erie Drift Plain, Atlantic Coastal Pine Barrens, Northern Piedmont, and Ridge and Valley ecoregions had only 3–4 species represented and exhibited no significant trends in biotic Hg concentrations. The Northern Allegheny Plateau had 10 fish species with sufficient sampling, but no trends were detected.

Overall, declining trends were documented in omnivorous or invertivorous/piscivorous fish from four ecoregions, with Yellow Perch being the only species to decline in multiple ecoregions. Piscivores across all ecoregions did not

Fig. 2 Significance of annual trend in tissue THg concentration derived from a general linear mixed model in each of nine USEPA Level III Ecoregions in New York. Colored tiles indicate that the 95% Credible Interval of the trend did not overlap zero, blue indicates a negative trend, and red a positive trend. Trends were estimated for whole organism ppm, ww, for fish, and blood ppm, ww, for birds
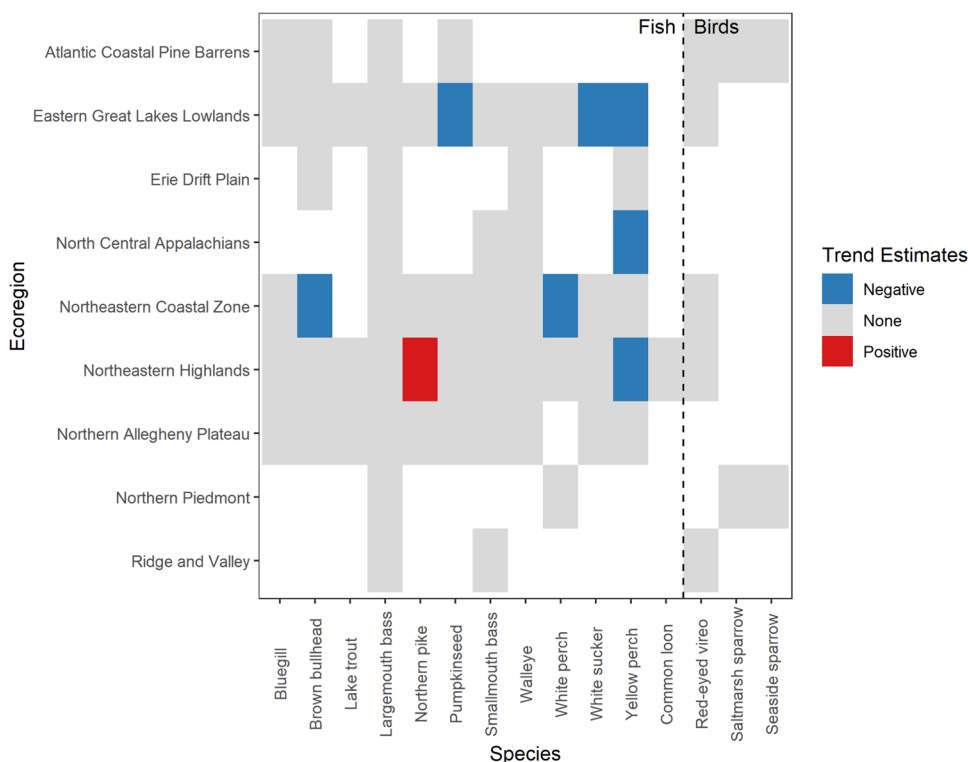
show declines, and one species (Northern Pike) showed an increase in the Northeastern Highlands.

Mercury biological hotspot identification

In general, spatiotemporal coverage was much greater for the aquatic food web than the terrestrial food web with 31,537 aquatic observations (Appendix A, Table A1) in 510 grid cells over 50 years, resulting in 2554 grid cell/species combinations. The median whole-organism equivalent THg ranged from 0.012 to 1.93 ppm, with an arithmetic mean value of 0.20 ppm. The terrestrial food web assessment included 4612 observations in 111 grid cells over 21 years, resulting in 935 grid cell/species combinations. The median blood-equivalent THg ranged from 0.004 to 1.09 ppm ww with an arithmetic mean value of 0.12 ppm ww

Foraging guild was an important predictor of species-level differences in Hg exposure within aquatic and terrestrial ecosystems. Using Model 2, we found that aquatic piscivores had the highest predicted values mean THg concentration $\mu = 0.29[0.04, 1.91]$; followed by invertivore-piscivores ($\mu = 0.17[0.03, 1.14]$), omnivores ($\mu = 0.10[0.01, 0.63]$) and invertivores ($\mu = 0.08[0.01, 0.52]$). Herbivores ($\mu = 0.04[0.01, 0.3]$) and planktivores ($\mu = 0.06[0.01, 0.43]$) exhibited similarly low THg concentrations. Analysis of terrestrial species indicated that carnivores had the highest THg values ($\mu = 0.34[0.05, 2.62]$) followed by invertivores ($\mu = 0.1[0.02, 0.7]$), then omnivores ($\mu = 0.08[0.01, 0.56]$), and finally

herbivores with the lowest predicted values ($\mu = 0.03[0.004, 0.0.2]$).

In aquatic food webs, Hg hotspots were identified in the Northeastern Highlands, the Northern Allegheny Plateau regions, and a small area in the Eastern Great Lakes region (Fig. 3d). Patterns were primarily driven by piscivorous birds (Appendix B; Table B8), including Common Loons (32 grid cells above effects thresholds) and Bald Eagles (eight grid cells above effects thresholds) and by fish, including Walleye (56 grid cells above effects levels), Largemouth Bass (16 grid cells above effects thresholds), and Smallmouth Bass (26 grid cells above adverse effects thresholds). These species also tended to have some of the highest spatial coverage by the sampling effort. Exceptions occurred for Roseate Terns and North American River Otters, with only one and two grid cell(s) sampled, respectively.

In terrestrial food webs, examining the number of species above effect level thresholds revealed Hg hotspots in the Eastern Great Lakes Lowlands region and the Atlantic Coastal Pine Barrens and Northeastern Coastal Zone on Long Island (Fig. 3a). On Long Island, these patterns were primarily driven by Seaside and Saltmarsh Sparrow Hg concentrations (three and six grid cells above effects thresholds, respectively), whereas patterns in the Eastern Great Lakes Lowlands region were primarily driven by multiple bat species (Appendix B; Table B8). Areas in central New York also had high numbers of
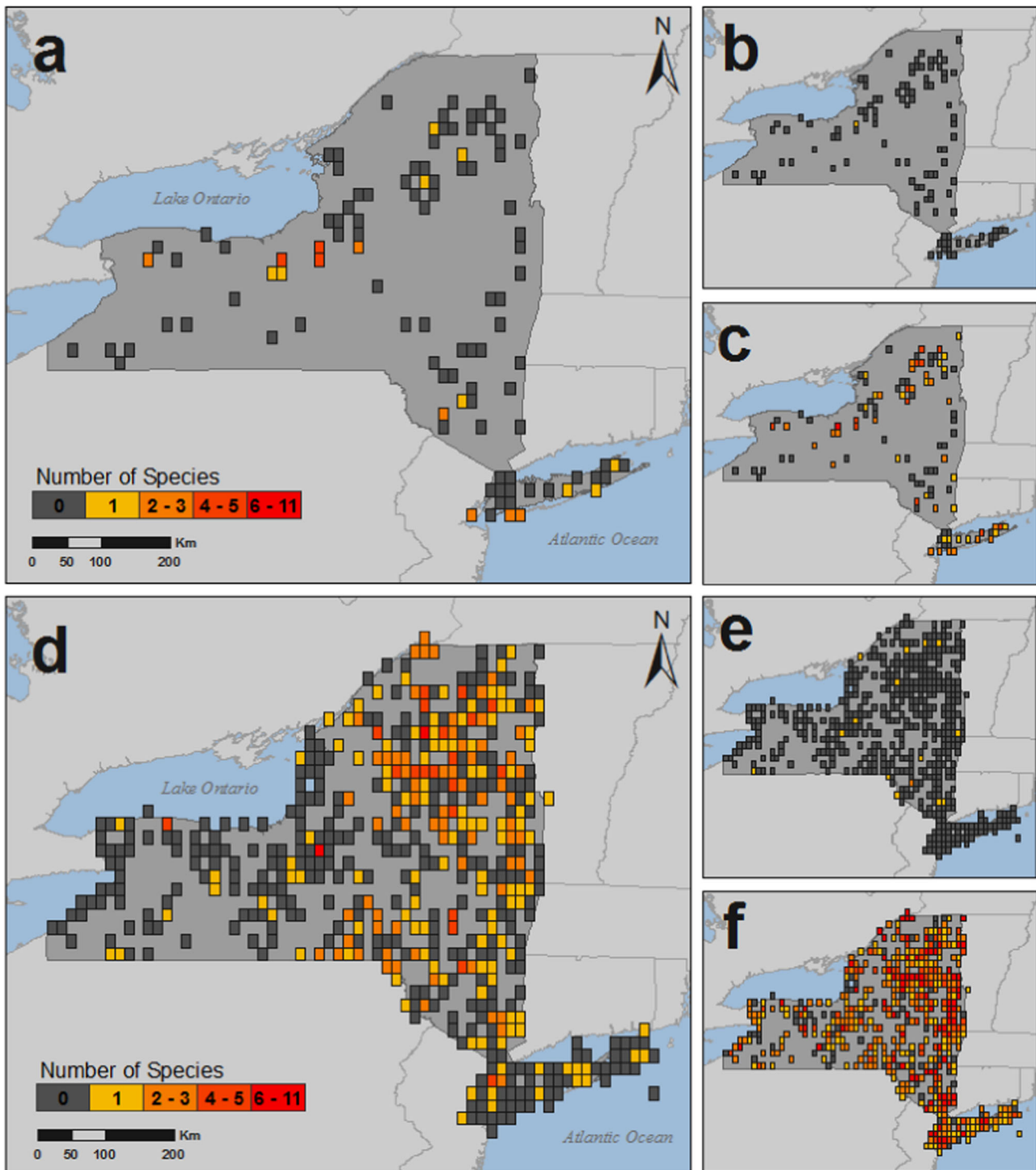
Fig. 3 The distribution and number of species above effects thresholds for terrestrial (**a**–**c**) and aquatic (**d**–**f**) food webs in New York State from 1969–2017. Species counts are based on whether the median (**a**, **d**), full 95% credible interval (**b**, **e**), or any of the 95% credible interval (**c**, **f**) falls above the effects level threshold for that species. High risk scenarios (**c**, **f**) describe areas that have little chance of being a Hg hotspot even with lax criteria and low risk scenarios (**b**, **e**) show the opposite. Estimates are based on general linear mixed model run in a Bayesian modeling framework, and thresholds are based on low effect level literature values

species affected, though spatial sampling effort was low in this area, and the spatial extent of this pattern is unclear (Fig. 3b–f).

Spatiotemporal hotspot analysis revealed changes in Hg hotspots and the sampling distribution over time for aquatic biota (Fig. 4a–e). One hotspot in the Eastern Great Lakes
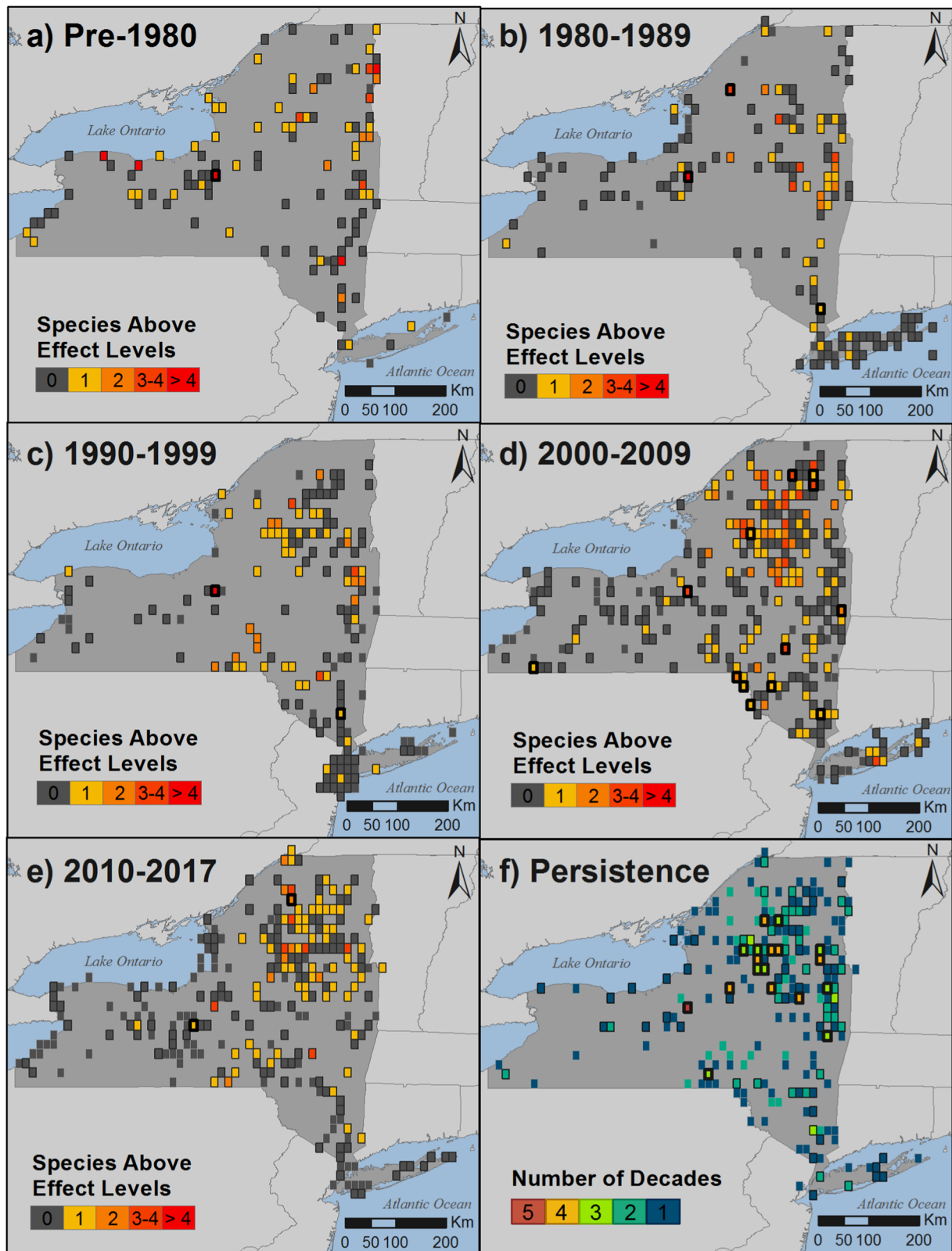
Fig. 4 Changes in the number of aquatic species above mercury effects thresholds over time in the State of New York. **a–e** Approximate decadal changes in the number of species above effects levels for each grid cell sampled. Colors represent the number of species with median THg above effects levels. Grid cells with gray outlines represent areas where the 95% credible interval of at least one species intersects the effect threshold. Grid cells with black bolded outlines represent areas where the 95% credible interval of at least one species is above the effect threshold. **f** Hotspot persistence over time with colors representing by the number of approximate decades with 1+ species above effect thresholds. Outlined grid cells represent those sampled in >2 decades. Black bolded grid cells represent those with >2 decades above effect thresholds, including the most recent time period (2010–2017)

Lowlands ecoregion has persisted throughout the sampling period (Fig. 4f), though the number of species above effects levels peaked in the 1990s at 13. Additional areas in the Northeastern Highlands, the Northeastern Coastal Zone, and the Northern Allegheny Plateau showed hotspot persistence over 2-4 decades. However, inconsistencies in sampling effort over time also play a role in these patterns (Appendix B; Table B9).

Spatial effects of habitat and Hg deposition on biota Hg exposure

Mean tissue THg was estimated at 1732 unique sites for aquatic species and 411 sites for terrestrial species in the preferential sampling analysis. Relative model fit for aquatic and terrestrial SPDE models suggested that the covariate models were better than the base models (ΔWAIC > 100 for each), and absolute goodness-of-fit was adequate for both models. The models were better at predicting Hg concentrations in sampled areas, but both were good, and we had no evidence of breaking model assumptions.

Sampling intensity differed considerably between the aquatic and terrestrial food webs, with aquatic sampling having around three times the number of observations and, therefore, a significantly higher sampling intensity intercept than the terrestrial model ($\mu = 3.8[3.3, 4.3]$ to $\mu = 0.8[0.1, 1.5]$). However, higher sampling efforts for both food webs occurred in the same general areas: Long Island, the Catskills, and the Adirondack Mountains (hereafter, the Adirondacks). Sampling intensity was lower in western New York for both species groups and more scattered overall for terrestrial species. After accounting for sampling intensity, estimates of spatial autocorrelation in Hg exposure varied between the aquatic and terrestrial food webs. Aquatic species showed correlation over greater distances than terrestrial species, as indicated by a higher range in the spatial random effect than terrestrial species ($0.84[0.64, 1.1]$ vs. $0.5[0.38, 0.66]$).

THg in aquatic species was high in the Northeastern Highlands—specifically the Adirondacks—with other pockets of elevated concentrations in the Atlantic Coastal Pine Barrens and Northeastern Coastal Zone (Long Island and the Catskills), and in the Eastern Great Lakes Lowlands (central and western New York; Fig. 5, left). For terrestrial species, the highest THg concentrations occurred in coastal Long Island (Fig. 5, right), but there were sporadic grid cells of elevated concentrations in the Catskills, Adirondacks, and the Finger Lakes. Differential relationships with land cover drive the dissimilarities in spatial distributions of THg among species groups. Aquatic species THg concentrations were correlated with upland forest land cover rather than open water (PC1, $\beta = 0.76[0.61, 0.92]$), undeveloped rather than developed habitats (PC4, $\beta = 1.23[0.82, 1.65]$), and a

forest/open water combination rather than agricultural land cover types (PC2, $\beta = -0.51[-0.78, -0.24]$) (Fig. 6, top). Total Hg concentrations in terrestrial species showed a different pattern. Higher tissue THg concentrations were associated with open water over upland forests (PC1, $\beta = -1.2[-1.44, -0.88]$) and had no strong correlations with the other land cover variables (Fig. 6, bottom). For aquatic and terrestrial species, dry and wet Hg deposition were not important predictors of tissue THg concentrations in the study area (Fig. 6). Western New York exhibited high uncertainty for both aquatic and terrestrial THg estimates, particularly along the southern border of the state, while Long Island, the Catskills, and the Adirondacks exhibited low uncertainty (Fig. 5).

Discussion

After assessing 50 years of Hg data from New York State, we found mostly stable trends with a few significant declines in some well-sampled fish species and variable Hg exposure risk across the state. Land cover was a significant driver of these spatial patterns, but affected aquatic and terrestrial biota differently. Temporal declines in Hg were observed in fish with long sampling histories extending from the 1970s in the Eastern Great Lakes Lowlands and the Northeastern Highlands. Tissue Hg concentrations increased in only one species (Northern Pike in the Northern Highlands). The communities with the highest risk for adverse effects are found in the Adirondacks for aquatic species and Long Island for terrestrial species. Biological Hg hotspots shifted over the past 50 years for aquatic species, but high-risk areas persisted in the Northeastern Highlands. Spatial patterns in aquatic and terrestrial Hg exposure were correlated with land cover features, but not with 2010 estimates of statewide atmospheric deposition of Hg. Mercury exposure in aquatic species was positively correlated with forest cover, rural land cover types, and landscapes with a mixture of forest and water cover types. In contrast, Hg exposure in terrestrial species was positively related to water surface area (both fresh and marine). These findings emphasize the modular role of land cover in Hg availability for aquatic and terrestrial food webs and the connections between aquatic and terrestrial environments (Cristol et al. 2008; Tsui et al. 2018).

Annual trends of Hg in aquatic and terrestrial biota

Most well-studied species sampled in New York State showed no significant annual trends in Hg concentration for ecoregions over 50 years of study. The relatively coarse spatial scale could obscure finer scale patterns (e.g., Schoch et al. 2020; Millard et al. 2020), but when changes were
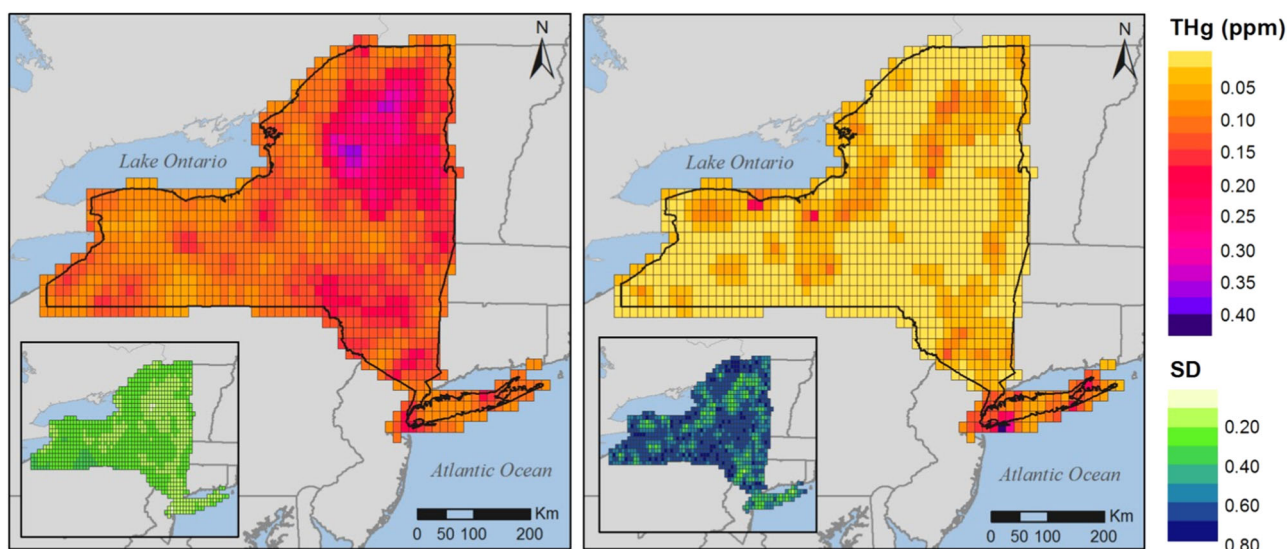
Fig. 5 Spatial distributions of average tissue THg concentrations in aquatic (left) and terrestrial (right) biota estimated from a spatially explicit linear model that corrects for bias in preferential sampling across New York State from 1969–2017. Aquatic THg is estimated for the whole-body tissue type, and terrestrial is estimated for the blood. Inset maps are standard deviations (SD) of the estimates

detected, like declines in fish Hg concentrations, other studies corroborated our results. An analysis of the New York portions of Lake Erie and Lake Ontario showed lower concentrations of Hg in fish in the 1980s through the 2000s than in 1970, contrasting with the relative stability in recent decades (Richter and Skinner 2020). Similarly, Weseloh et al. (2011) found decreasing Hg concentrations in Herring Gull eggs in the Great Lakes from the 1970s to 1980s but no significant trends in the 2000s. This pattern of declining Hg concentrations in fish and wildlife has also been observed in other parts of the northeastern United States and parts of the Great Lakes (Scheider et al. 1998; Kinghorn et al. 2007; Bhavsar et al. 2010). In sum, this study and others indicate a decline in biotic Hg concentrations in the Great Lakes in the 1970s and early 1980s with relative stability since, and changes in atmospheric deposition are a likely driver (Drevnick et al. 2012).

Decreasing Hg trends in biota, however, are not universal; previous studies have found increasing trends in Hg in other parts of the Great Lakes (Zananski et al. 2011; Gandhi et al. 2014; Zhou et al. 2017). Differential methylation rates across habitats or shifts in diet are confounding factors in linking Hg deposition to Hg concentrations in biota (Weseloh et al. 2011; Burgess et al. 2013; Gerson and Driscoll 2016). It may be surprising that we observed limited trends in Hg in biota in recent years given the large decreases in Hg emissions and atmospheric deposition over the past decade associated with the Mercury and Air Toxics Standard and shifts in energy sources of generation, particularly in the eastern U.S. (Olson et al. 2020). This lack of recent declines in Hg in biota, given marked declines in emissions, may be due to the effects of legacy Hg, lags in

processing and methylation of Hg in the landscape, or enhanced processing of Hg due to climate impacts (increases in temperature, precipitation, enhanced wet-dry cycles, enhanced mobilization of dissolved organic matter, enhanced thermal stratification of lakes). Changing Hg determination protocols in the 1990's could also lead to higher observations when comparing the two (Lowery et al. 2007), but changes over the past 20 years have been relatively static.

Only Northern Pike in the Northeastern highlands showed an increasing temporal trend in THg concentrations. Pike are primarily piscivores and consume higher trophic-level prey than the other fish in the region. Bioaccumulation in long-lived predators is also a potential cause for exposure increases over time (Orihel et al. 2008), and emphasizes the issues of legacy Hg for top predators.

Spatial Hg hotspots: trends across communities

Analysis of Hg spatial hotspots revealed differing patterns in aquatic and terrestrial food webs, highlighting the importance of species, foraging guild, and sampling resolution. Sampling effort and the size of the sampled communities also clearly played a role in determining the number of species detected with risk of adverse effects. Grid cells are large and include variable habitat composition, which causes community exposure estimates to be coarser than more focused studies. As such, this analysis should be considered descriptive of the number of species currently known to surpass effects thresholds and not necessarily a tool for comparing relative community exposure risk.

Fig. 6 Effect of environmental covariates on aquatic (top) and terrestrial (bottom) tissue THg concentrations in New York State from a spatially explicit linear model that corrects for bias in preferential sampling across space. Habitat variables were created using principal components analysis based on land cover data. Principal Component 1 (PC1) was negatively correlated with open water and positively correlated with deciduous forest, PC2 was negatively correlated with open water and deciduous forest and positively correlated with agricultural habitat, PC3 was negatively correlated with agricultural habitat and positively correlated with forest, and PC4 was negatively correlated with developed habitats and positively correlated with forest and agriculture land cover. Data from New York State biota sampled from 1969–2017. Error bars represent 95% credible intervals of the slope parameter estimate
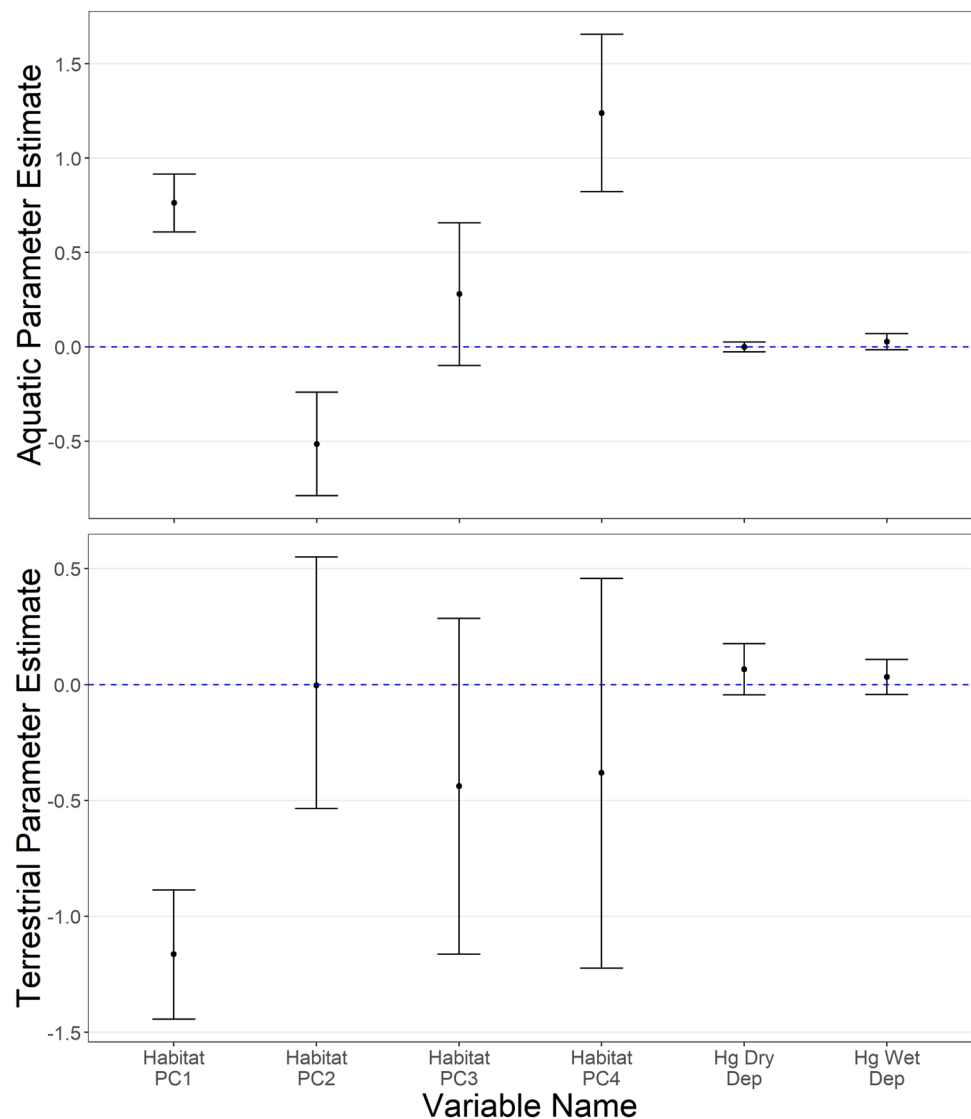
Aquatic species at risk of adverse effects of Hg exposure were primarily located in the Adirondacks, extending to Long Island and the Catskills regions. The Adirondacks have been identified as a biological Hg hotspot in previous regional syntheses of aquatic species, including Common Loons (Evers et al. 2007, 2011c) and multiple fish species (Kamman et al. 2005; Driscoll et al. 2007; Evers et al. 2007). The Catskills region has been identified as an area of concern for fish (Evers et al. 2007) and Bald Eagles (DeSorbo et al. 2020).

At-risk terrestrial species are primarily located in Long Island and central New York State, including invertivorous bats, Saltmarsh Sparrows, and Seaside Sparrows. Elevated THg concentrations have been found in bat fur throughout eastern North America (Yates et al., (2014)), and New York is similar. *Myotis* bat species have shown positive associations with water (Furlonger et al. 2009), and a diet of emergent aquatic insects may play a role in Hg

concentrations in these bats. Previous studies in the western (Ackerman et al. 2016) and eastern U.S. (Jackson et al. 2015; Adams et al. 2020) found that saltmarsh and wetland habitats correlate with higher Hg concentrations in songbirds. In particular, Saltmarsh Sparrows are an obligate year-round tidal marsh species with elevated concentrations of THg on Long Island (Warner et al. 2010; Lane et al. 2011).

Many factors contribute to differences in Hg exposure among species, including trophic position (Lavoie et al. 2010; Yu et al. 2011), foraging habitat (Eagles-Smith et al. 2008), and bioenergetics (Lepak et al. 2012). Our results indicate a greater number of aquatic species at or above Hg adverse effects thresholds than terrestrial species. There is generally a higher potential for Hg methylation in aquatic ecosystems and often adjacent wetlands, leading to greater bioaccumulation in aquatic species (Ullrich et al. 2001). This pattern also links to food web structure of the species

sampled in both systems. Aquatic species above adverse effect thresholds tended to be upper trophic level piscivores and invertivore-piscivores. In contrast, species identified in the terrestrial food web were primarily invertivores, thus lower in the food web, with few upper trophic level species sampled. There are well-established links between trophic level and Hg bioaccumulation (Kidd et al. 1995; Lavoie et al. 2010; Coelho et al. 2013), and previous studies have found higher Hg concentrations in piscivores than lower trophic-level foraging guilds (Eagles-Smith et al. 2016).

Landscape factors influencing Hg exposure in aquatic and terrestrial biota

New York State land cover influenced aquatic and terrestrial tissue Hg exposure in strikingly different ways. Aquatic tissue Hg concentrations were positively correlated with forest cover, while terrestrial tissue Hg concentrations were positively correlated with open water. These patterns suggest the importance of connectivity between habitat types in driving elevated Hg in biota. A significant proportion of MeHg in terrestrial biota is derived from aquatic sources (Cristol et al. 2008; Jackson et al. 2015; Tsui et al. 2018; Sauer et al. 2020), and our results emphasize these linkages among ecosystems. Water body size has variable effects on aquatic bioaccumulations. Larger water bodies often have more complex food chains that facilitate Hg biomagnification, but there is evidence that demethylation rates are higher in larger water bodies (Burns et al. 2012). In contrast, terrestrial species can be generalists that forage opportunistically on aquatic resources (Jackson et al. 2015; Tsui et al. 2018; Twining et al. 2021), and animals using habitats adjacent to water or wetlands are more likely to be exposed to elevated MeHg. The amount of wetland habitat can be a significant predictor of Hg exposure in songbirds (Edmonds et al. 2012; Jackson et al. 2015; Adams et al. 2020; Sauer et al. 2020), and terrestrial wetland specialists, such as Palm Warblers and Saltmarsh Sparrows (Jackson et al. 2015), could have higher Hg exposure due to their restricted habitat use. Rural agricultural landscapes in New York State have also been linked to elevated MeHg exposure in songbirds and are important for determining exposure risk in conjunction with aquatic diet (Twining et al. 2021).

Elevated THg concentrations in aquatic species were also more common in habitats that were undeveloped and mixtures of forest and open water. Many studies have found positive relationships between rural or forested habitats and Hg exposure in aquatic species (Evers et al. 2007, 2011a; Chumchal et al. 2008; Simonin et al. 2008; Yu et al. 2011; Riva-Murray et al. 2011; Riva-Murray et al. 2020), but our study suggests that these smaller-scale relationships can aggregate to influence statewide distributions of Hg exposure.

Both dry and wet Hg deposition were not important for predicting spatial distributions of aquatic or terrestrial Hg exposure, though this lack of relationship could be due to correlations between deposition and habitat, and mismatched spatiotemporal scales. Land cover influences atmospheric Hg deposition, particularly dry deposition (Yu et al. 2013; Ye et al. 2018), and is higher in urban areas (Ye et al. 2018). Urban landscapes have low methylating potential; thus, inputs often do not translate into biotic Hg exposure (Chalmers et al. 2014). While we do not find evidence of its importance in this study, there is strong evidence that Hg deposition can be a strong predictor of biotic Hg in other contexts. Past studies have found correlations between fish Hg concentrations and atmospheric Hg deposition in freshwater and marine ecosystems (Hammerschmidt and Fitzgerald 2006; Lee et al. 2016; Riva-Murray et al. 2020). Newly deposited Hg is considered more bioavailable (e.g., more easily methylated, more likely to enter food chains) than legacy Hg (Harris et al. 2007). We recommend future research use temporally dynamic estimates of Hg deposition to better compare with biotic sampling and add knowledge of local Hg sources (Suchanek et al. 2008), to understand better the relative importance of landscape-scale drivers of Hg exposure.

Monitoring tissue Hg in biota: present and future

Long-term and continuous is required for effective monitoring programs, as greater than ten years of data with 10–15 sites within each year have been considered optimal across a range of contaminant monitoring programs (Nicholson et al. 1997; Rigét et al. 2010; Gewurtz et al. 2011). Generally, efforts should be allocated more in time (number of years sampled) than space (sites sampled within a year). Based on our power analysis (Appendix D; Yang et al., 2020), if the sampling duration was extended to 20 years, only 5–10 sites would be needed per year. Most species sampled in New York State have not yet reached those measures except for a few species tied to long-term public or environmental health (e.g., sport fish, Richter and Skinner, 2020; and Common Loons, Schoch et al., 2020). Increasing the number of sites sampled per year does not always help detect changes over short sampling durations, as we found sampling for less than 10 years does not help detect trends even with 25 sites sampled within a year. No practical number of sites can substitute for monitoring time; thus, long-term sampling efforts must be prioritized to evaluate temporal trends in Hg exposure.

For future monitoring of Hg in biota, stratifying sampling locations by ecoregion and exposure risk could help improve study design. Our study revealed effective sampling rates varied by ecoregion. These differences are likely related to site-scale characteristics and environmental

processes such as surface water acidity, habitat type, or food web dynamics (Razavi et al. 2020; Yang et al. 2020). Another stratifying parameter could be the potential for adverse effects of Hg. Less sampling effort may be allocated for ecoregions with Hg concentrations far below threshold levels. For example, Yellow Perch in the Northern Coastal Zone exhibited low Hg concentrations (0.13 ppm ww whole body; 43% of the effects threshold limit), and reduced sampling effort could be acceptable as the value of high precision estimates is lower.

Other considerations for future Hg monitoring include: (1) establishing an online platform for scientific organizations and universities involved in monitoring to share sampling plans and results to avoid duplicated efforts in time and money; (2) standardizing tissue types to be sampled for each species for better monitoring of Hg or other contaminants (e.g., polychlorinated biphenyls) to reduce uncertainty based on tissue conversion; and (3) choosing the appropriate bioindicators for the monitoring objective(s). Some tissue types (e.g., adult feathers) have more sampling variability and may not be appropriate for specific monitoring questions (Low et al. 2020). If the goal is to monitor temporal changes in Hg loadings, lower trophic level organisms or younger generations of the higher trophic level organisms are more sensitive to recent changes. Long-lived, high trophic level species bioaccumulate Hg, creating a temporal lag in the monitored response (Harris et al. 2007). If the goal is to monitor wildlife health, the monitoring scheme should align with other key metrics such as reproductive success and diet. Finally, power analyses are a powerful tool and should be repeated every 5–10 years as new data become available to improve our understanding of our ability to detect trends (Levine et al. 2014) and provide guidance on the sampling intensity required.

Conclusions

After 50 years of research on Hg, we have identified consistent patterns of Hg contamination in biota across New York State. Most well-studied species showed no significant changes in tissue THg concentrations, though the species that did decrease were primarily fish from the Great Lakes sampled since the 1970s. The Adirondacks and Long Island remain areas of persistent Hg exposure that likely leads to adverse effects in many species, but elevated Hg in biota is not restricted to those regions. Land cover is a significant factor driving these patterns, particularly forests, water, and rural habitat types, and can modulate Hg exposure in habitats with local or remote Hg sources. While particularly useful in the northeastern United States, these results are generalizable to other temperate, forested regions and can inform Hg monitoring and management decisions in many contexts.

While there is an understanding of the factors that created the present distribution of Hg exposure, it is difficult to forecast the future of Hg in biota in New York State's biota. Many of the factors affecting atmospheric Hg deposition and Hg methylation are predicted to be affected by policy or climate change (Schindler 2001; Ye et al. 2019). As land cover, climate, Hg deposition, and emissions change over time, the spatial and trophic distributions of MeHg exposure are expected to change. Moving forward, continued research and monitoring efforts in New York State and other locations with well-documented Hg exposure risk to biota are warranted to quantify the effects of these changes on Hg exposure risk in fish and wildlife. Spatially robust and temporally consistent monitoring efforts on critical species or communities will be crucial to quantifying changes in these ecosystems and determining the impacts of environmental changes on MeHg distribution and abundance in aquatic and terrestrial ecosystems.

Compliance with ethical standards

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