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Mercury levels in herring gulls and fish: 42 years of spatio-temporal trends in the Great Lakes

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HIGHLIGHTS

- We examine the temporal *Hg* trends in herring gull eggs and fish from Great Lakes.
- Mercury levels gradually declined until the mid-1990s.
- Mercury trends reversed in certain locations of the Great Lakes in the 2000s.
- Dynamic linear modelling offers a robust hindcasting tool with a flexible structure.
- Strong herring gull-rainbow smelt interactions exist in Lakes Superior and Ontario.

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ABSTRACT

Total mercury levels in aquatic birds and fish communities have been monitored across the Canadian Great Lakes by Environment and Climate Change Canada (ECCC) for the past 42 years (1974–2015). These data (22 sites) were used to examine spatio-temporal variability of mercury levels in herring gull (*Larus argentatus*) eggs, lake trout (*Salvelinus namaycush*), walleye (*Sander vitreus*), and rainbow smelt (*Osmerus mordax*). Trends were quantified with dynamic linear models, which provided time-variant rates of change of mercury concentrations. Lipid content (in both fish and eggs) and length in fish were used as covariates in all models. For the first three decades, mercury levels in gull eggs and fish declined at all stations. In the 2000s, trends for herring gull eggs reversed at two sites in Lake Erie and two sites in Lake Ontario. Similar trend reversals in the 2000s were observed for lake trout in Lake Superior and at a single station in Lake Ontario. Mercury levels in lake trout continued to slowly decline at all of the remaining stations, except for Lake Huron, where the levels remained stable. A post-hoc Bayesian regression analysis suggests strong trophic interactions between herring gulls and rainbow smelt in Lake Superior and Lake Ontario, but also pinpoints the likelihood of a trophic decoupling in Lake Huron and Lake Erie. Continued monitoring of mercury levels in herring gulls and fish is required to consolidate these trophic shifts and further evaluate their broader implications.

1. Introduction

Mercury is a contaminant of concern worldwide and its effects on the environment have been studied intensively for decades (Braune, 2007; Munthe et al., 2007; Monson, 2009). The main sources of mercury to aquatic ecosystems are direct deposition from the atmosphere and runoff from surrounding catchment zones. In North America, from the 1900s to the 1970s, mercury emissions from human activities such as metal smelting, chlor-alkali and pulp industries, increased rapidly with approximately 30% of the emissions occurring in the Great Lakes Basin (Pirrone et al., 1998). The Great Lakes is the world's largest freshwater ecosystem, serving as a natural and economic resource to over 35 million people (Evers et al., 2011).

The ecological and toxicological effects of mercury are strongly dependent on its form in the environment. Inorganic mercury can be transformed into methylated (organic) species that are highly

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toxic to aquatic organisms and wildlife (Boening, 2000; Ullrich et al., 2010). Methylmercury bioaccumulates in piscivorous birds, such as herring gulls, so that approximately 90% of the total mercury in eggs of piscivorous birds (Scheuhammer et al., 2001) and more than 95% of total mercury in fish (Bloom, 2004; Raymond and Rossmann, 2009) is methylmercury. Using total mercury concentration as a proxy of methylmercury levels in bird eggs and fish tissue has been supported by a number of studies (e.g., Raymond and Rossmann, 2009; Ackerman et al., 2013).

Total mercury levels in Great Lakes fish have been extensively monitored by Environment and Climate Change Canada (ECCC) since the 1970s through the National Aquatic Biological Specimen Bank (NABSB) Program. The Great Lakes Binational Toxic Strategy (1996–1997) was developed jointly by Canada and the United States with the goal of achieving 50–90% reductions in the deliberate use and/or release of mercury from anthropogenic sources by 2006 (IJC, 1978). To further support Canada's efforts in contaminant reductions, the Clean Air Regulatory Agenda (CARA) was established in 2006 to reduce emissions of hazardous air pollutants, such as mercury, through regulation of industrial sectors and monitoring. The Great Lakes Binational Toxic Strategy is currently not active, so the current framework includes CARA and the Great Lakes Water Quality Agreement (GLWQA) with the list of Chemicals of Mutual Concern under Annex 3. Between 1990 and 2005, total mercury emissions to the atmosphere from inventoried anthropogenic sources in the Great Lakes states declined by approximately 50% (NEI, 1990; NATA, 2005). Progress towards program targets has been assessed through monitoring studies, such as the Herring Gull program in the Great Lakes (e.g., Weseloh et al., 2011) and Atlantic Canada (e.g., Burgess et al., 2013).

Studies in the Great Lakes have examined spatial and temporal patterns of mercury in sediments (e.g., Marvin et al., 2004), body burdens in fish (e.g., Bhavsar et al., 2010), and dietary exposure pathways resulting in elevated levels in herring gull eggs (e.g., Weseloh et al., 2011). In particular, rainbow smelt can serve as a primary food item for herring gulls (e.g., Pierotti and Good, 1994) in the Great Lakes and for piscivorous fish (e.g., lake trout, walleye) and, therefore, play an important role in mercury bioaccumulation in upper trophic levels (Scott and Crossman, 1990).

For the past four decades (mid-1970s - 2007), studies showed that total mercury concentrations in fish generally decreased in the Canadian Great Lakes, but inter-lake differences were also identified. For example, mercury levels in Lake Ontario walleye remained relatively stable (1990s - 2007), in contrast to a recent increase in Lake Erie (Bhavsar et al., 2010). Lake-specific differences have also been reported. For example, mercury concentrations in walleye from Lake Erie were lower in the Western Basin compared to the Eastern Basin (Azim et al., 2011). Weseloh et al. (2011) reported declining trends (1974–2009) for total mercury in herring gull eggs across the Great Lakes. This pattern was partially attributed to the gulls shifting their diets from rainbow smelt to terrestrial food sources, which have lower levels of methylmercury. Changes in trophic interactions should be reflected in long-term trends across monitoring stations in the Great Lakes.

Previous Great Lakes studies focused on total mercury concentrations in two tropic levels, e.g., Weseloh et al. (2011) covered herring gulls and rainbow smelt. In this study, we extend our spatio-temporal comparisons to three trophic levels: herring gulls, piscivorous fish (lake trout, walleye) and planktivorous fish (rainbow smelt). Temporal patterns of mercury levels were quantified using dynamic linear models (DLMs) with a Bayesian framework that accounted for uncertainty in model structure and parameter values. Unlike static regression models that have fixed parameters, DLMs have an evolving structure that allows parameters to shift through time (Sadraddini et al., 2011a,b). This "dynamic" feature facilitated our modelling exercise to more accurately depict the levels of mercury in herring gull eggs, planktivorous and piscivorous fish, while accommodating the year-to-year variability of the signature of important covariates (e.g., lipid content, fish length) (Sadraddini et al., 2011a,b). To gain an ecosystem perspective, mercury levels in rainbow smelt and herring gull eggs, after correcting for the role of important covariates, were used to examine the importance of trophic interactions between prey and predator in modulating the transfer of mercury along the food web.

2. Materials and methods

2.1. Herring gull field methods

The Great Lakes Herring Gull Monitoring Program has been used to monitor contaminants in wildlife since 1974 (Pekarik and Weseloh, 1998; Hebert et al., 1999; Weseloh et al., 2006). Here, we provide a summary of the field methods. Detailed descriptions are presented in Weseloh et al. (2006). Fresh herring gull eggs have been collected annually for up to 15 colonies located throughout the Great Lakes and connecting channels. The locations of these colonies are shown in Fig. 1. Every year, approximately 10–13 eggs were collected from each colony between late-April and early-May, where the eggs were randomly sampled. The eggs were refrigerated at 4 °C and analyzed at ECCC's National Wildlife Research Center (NWRC) within three weeks.

2.2. Fish field methods

Mercury concentrations in rainbow smelt, lake trout and walleye were obtained from ECCC's National Aquatic Biological Specimen Bank (NABSB) Program (McGoldrick et al., 2010). Under this program, lake trout and walleye were collected using bottom set gillnets and rainbow smelt using a bottom trawl. After their capture, fish were immediately frozen on dry ice and transported to the laboratory. In the laboratory, the fish were partially thawed, weighed, measured, and sex determined. Scales, fins, rays and/or otoliths were removed for aging and the remaining portions including internal organs were homogenized five times using a meat grinder. Fish sampling locations are shown in Fig. 1.

2.3. Mercury analyses

Mercury concentrations were measured in herring gull eggs collected across 15 sites from 1974 to 2015, but not all the sites were analyzed consistently, ranging from 30 to 43 years for each colony. Before 1986, individual eggs were analyzed at each site. From 1986 onwards, eggs were pooled for each sampled location. All subsequent statistical analyses were performed on either pooled site data and mean values per site for years 1986 or earlier, resulting in a sample size of 309 site-years. Mercury samples (approximately 0.5 g of each Gull egg homogenate) were derived using different analytical methods and the full details are described in Weseloh et al., 2011. From 1974 to 1989, samples were analyzed by cold vapour atomic absorption spectrophotometry (CVAAS) at the Ontario Research Foundation (ORF; Mississauga, Ontario). The same method was used at the NWRC from 1990 to 2001. After 2001, samples were analyzed at NWRC using an advanced mercury

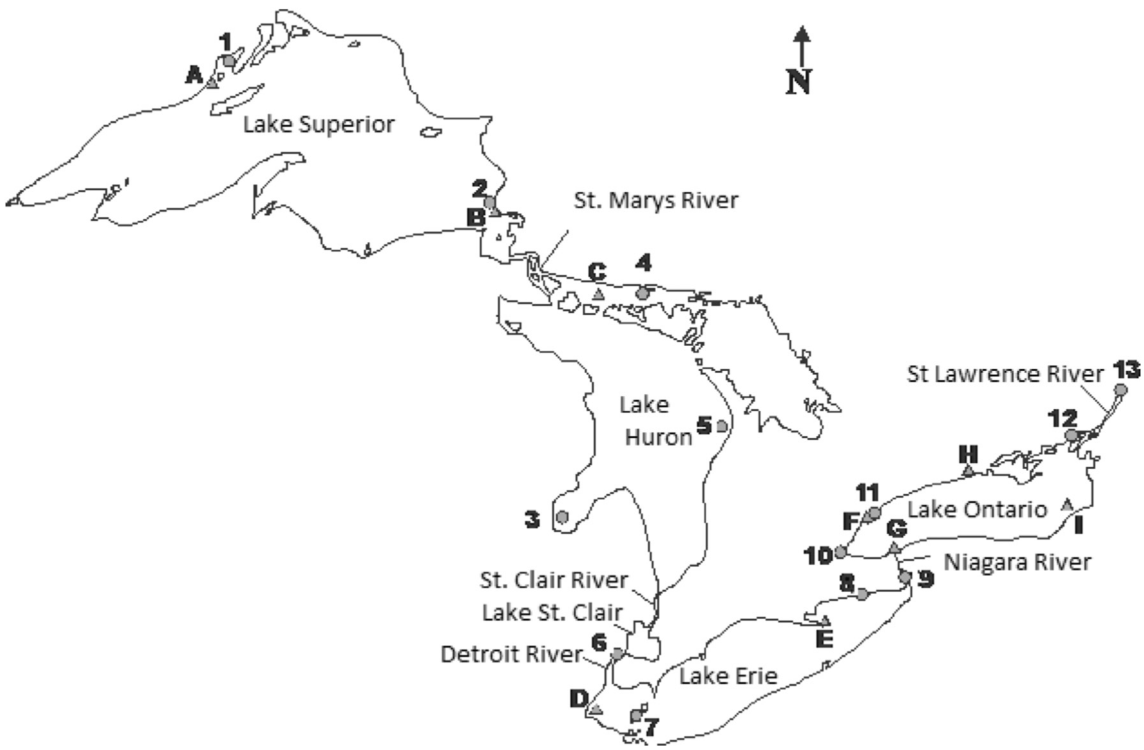

Fig. 1. A map of all the Great Lakes with the monitoring stations indicated for Herring Gulls (1–13) and fish communities (A–H). Herring Gull monitoring sites included two (Granite Island (1) and Agawa Rocks (2)) in Lake Superior, three sites (Channel-Shelter Island (3), Double Island (4), and Chantry Island (5)) in Lake Huron, four sites in Lake Erie (Fighting Island (6), Middle Island (7), Port Colborne (8), and Niagara River (9)), and four sites in Lake Ontario (Hamilton Harbour (10), Toronto Harbour (11), Snake Island (12), and Strachan Island (13)). For the fish communities, Lake Superior had two sites (Thunder Bay Pie-Island (A), and Whitefish Bay (B), one site in Lake Huron (North Channel (C), two sites on Lake Erie (Western Basin (D) and Eastern Basin (E), and four sites in Lake Ontario (Port Credit (F), Cobourg (H), Niagara (G) and Oswego (I).

analyzer (AMA-254), equipped with an ASS-254 auto-sampler for solid samples. To assess accuracy among analytical methods, duplicate herring gull eggs samples and standard reference materials were analyzed where the nominal detection limit for total Hg was 0.05 μg/g (dry weight) for both AMA and CVAAS. There were no significant differences in percent egg moisture for all site-year combinations, but there were some differences in total mercury concentrations between CVAAS and AMA. To allow for direct comparisons, ORF (CVAAS) mercury concentrations were multiplied by 0.934 to be equivalent to NWRC (AMA), while the NWRC (CVAAS) concentrations were multiplied by 1.079 to be equivalent to NWRC (AMA) (Weseloh et al., 2011). Mercury concentrations in whole fish were determined by Environment Canada's National Laboratory for Environmental Testing (NLET) using NLET method 2801. Prior to 2014, this method quantified mercury using CVAAS (for more details see McGoldrick et al., 2010; Bhavsar et al., 2010). In 2014, NLET began quantifying mercury using inductively coupled plasma mass spectrometry (ICP/MS).

2.4. Data consolidation

Herring gull egg time-series (1974–2015) from sites in the Upper and Lower Great Lakes were used for the analyses. In the Upper Great Lakes, Lake Superior had two sites (Agawa Rocks and Granite Island) and Lake Huron had three sites (Channel-Shelter Island, Double Island, and Chantry Island). In the Lower Lakes, Lake Erie had four sites (Fighting Island, Middle Island, Port Colborne, and Niagara River) and Lake Ontario had four sites (Hamilton Harbour, Toronto Harbour, Snake Island, and Strachan Island) (Fig. 1). Fish time series (1977–2013) from the Upper and Lower Great Lakes were used for analyses across seven sites. Samples were collected from two sites in Lake Superior (Thunder Bay-Pie Island and Whitefish Bay), one site in Lake Huron (North Channel), two sites in Lake Erie (Western Basin and Eastern Basin), and four sites in Lake Ontario basin (Port Credit, Cobourg, Niagara and Oswego).

2.5. Dynamic linear modelling framework

A series of DLMs were developed to examine the temporal trends of total mercury in herring gull eggs and fish across the Canadian side of the Great Lakes. In contrast with regression analysis, DLM parameter estimates are dynamic and influenced only by prior and current information, not by subsequent data (Lamon et al., 1998). Thus, an important DLM feature is that the sequence of the time series is maintained. At each time step, the level of the response variable is related to the current and past time steps, unlike in the traditional regression, where the entire time series is used (Pole et al., 1994; Stow et al., 2004). DLMs also minimize the effects of outliers and data gaps and the parameters are related to each other stochastically by virtue of an error term (Stow et al., 2004).

For each herring gull site, DLMs were fitted to ln-transformed total mercury egg concentrations using the program WinBUGS (Spiegelhalter et al., 2003). The main components for the DLM equations were the observation equation followed by the system equations. In the model below, both lengths and lipids (e.g., Visha et al., 2015) were used as covariates in all the fish DLMs while lipids was the only covariate used in the herring gull DLMs.

Observation equation:

$$\ln[THg]_{itk} = level_{tk} + \beta_{tk1}\ \ln[length]_{itk} + \beta_{tk2}\ \ln[lipid]_{itk} + \psi_{itk} \qquad \psi_{itk} \sim N[0, \Psi_{tk}]$$

System equations:

$level_{tk} = level_{t-1,k} + rate_{tk} + \omega_{tk1}$	$\omega_{tk1} \sim N\,[0, \Omega_{tk1}]$
$rate_{tk} = rate_{t-1,k} + \omega_{tk2}$	$\omega_{tk2} \sim N\,[0, \Omega_{tk2}]$
$\beta_{t1,k} = \beta_{t1-1,k} + \omega_{tk3}$	$\omega_{tk3} \sim N\,[0, \Omega_{tk3}]$
$\beta_{t2,k} = \beta_{t2-1,k} + \omega_{tk4}$	$\omega_{tk4} \sim N\,[0, \Omega_{tk4}]$
$1/\Omega^2_{tkj} = \zeta^{t-1} \cdot 1/\Omega^2_{1kj},\ 1/\Psi^2_{tk} = \zeta^{t-1} \cdot 1/\Psi^2_{1k}$	$t > 1$ *and* $j = 1$ *to* 4
$level_1, rate_1, \beta_1 \sim N(0, 10000)$	$t = 1$
$1/\Omega^2_{1jk},\ 1/\Psi^2_{1k} \sim G(0.001, 0.001)$	$K = 1$ *to number of sites*

where $\ln[THg]_{itk}$ is the observed mercury concentration in either the pooled herring gull egg sample or whole-body fish sample i from site k and year t; $level_{tk}$ is the mean mercury concentration at year t with length and lipids as covariates for all the fish models, where $ln[length]_{itk}$ and $ln[lipid]_{itk}$ are the observed (standardized) fish length and lipid content, respectively, at sample i from site k and year t. The rate of change of mercury $level_{tk}$ at a given time is represented as $rate_{tk}$, β_{tk1} and β_{tk2} are the length and lipid (regression) coefficients, respectively, with ψ_{itk} as the error term. The discount factor ζ represents the aging of information with the passage of time, $N(0, 10000)$ is a normal distribution with mean *0* and variance *10000*, and $G(0.001, 0.001)$ is the gamma distribution with shape and scale parameters of 0.*001*. The prior distributions for the parameters of the initial year $level_{1k}$, $rate_{1k}$, β_{1k1}, β_{1k2}, $1/\Omega^2_{1kj}$, and $1/\Psi^2_{1k}$ are considered "non-informative" or vague. This procedure was repeated for each fish (lake trout and rainbow smelt), and site combination where DLMs were fitted to Ln-transformed whole-fish total mercury concentrations. There were a total of 18 models for fish sites across the Canadian Great Lakes: nine for rainbow smelt, eight for lake trout, and one for walleye (only for the Western Basin of Lake Erie). The herring gull egg DLMs were based on the 13 colonies sampled across the Canadian portion of the Great Lakes (Table 1).

The sequential updating of a DLM forecasts for time t based on prior knowledge of the parameters, observed at the time (Lamon et al., 1998). Based on Bayes' Theorem, knowledge about the parameters is updated using the likelihood of the data and the prior information (Congdon, 2003). A discount factor is applied to this new posterior, so that older observations are weighted less than newer ones. The discounted posterior becomes the prior for the next time step and the process repeats. In this analysis, we introduced non-constant and data-driven variances (with respect to time) using a discount factor on the first period prior (West and Harrison, 1989). Discount factors between 0.8 and 1.0 were examined during model specification and 0.95 was selected which provided a balance between model performance (e.g., deviance) and uncertainty of year-specific regression coefficients. This discount factor resulted in the highest model performance, while maintaining lowest possible coefficient of variation corresponding model estimates (Visha et al., 2015).

Using the WinBUGS software, we obtained sequences of realizations from the model posterior distributions with Markov-chain Monte Carlo (MCMC) simulations (Gilks et al., 1998). We used a general normal proposal Metropolis algorithm that is based on a symmetric normal proposal distribution. For each analysis, we used three chain runs of 100,000 iterations, keeping every 20th iteration to minimize serial correlation. Convergence of the MCMC chains was checked using the Brooks–Gelman–Rubin (BGR) scale-reduction factor (Brooks and Gelman, 1998). The BGR factor is the ratio of among chain variability to within chain variability. The chains have converged when the upper limits of the BGR factor are close to one. The accuracy of the posterior parameter values was inspected by assuring that the Monte Carlo error (an estimate of the difference between the mean of the sampled values and the true posterior mean) for all parameters was less than 5% of the sample standard deviation (Spiegelhalter et al., 2003).

2.6. Trophic interactions

It was not possible to compare mercury patterns between herring gulls and fish communities across all of the sampling stations because the spatial and temporal overlap was not consistent. Since rainbow smelt are frequently cited as the preferred prey for herring gull (e.g., Koster et al., 1996), we used this species to evaluate the strength trophic interactions. To ensure temporal overlap across trophic levels, a 31 year time period (1981–2011) was analyzed for Lakes Ontario, a 28 year time period (1981–2008) for Erie, 31 years (1982–2012) for Lake Huron, and 31 years (1983–2013) for Lake

Table 1
Descriptive statistics, mean, standard deviation (SD), minimum, maximum, median, interquartile range (IQR), skewness, and kurtosis for mercury concentrations (μg/g, wet weight) in Herring Gull eggs from 1974–2015.

Site	n	Mean	SD	Min.	Max.	Median	IQR	Skewness	Kurtosis
Upper Lakes									
Lake Superior									
Granite Island	25	0.213	0.105	0.096	0.475	0.168	0.066	1.337	0.600
Agawa Rocks	25	0.215	0.105	0.122	0.585	0.180	0.075	2.216	5.703
Lake Huron									
Channel-Shelter Island	25	0.158	0.045	0.089	0.264	0.147	0.050	0.824	0.224
Double Island	22	0.175	0.077	0.084	0.385	0.159	0.071	1.456	1.904
Chantry Island	27	0.122	0.054	0.060	0.254	0.110	0.079	0.886	0.133
Lower Lakes									
Lake Erie									
Fighting Island	18	0.172	0.096	0.085	0.542	0.161	0.021	3.630	14.634
Middle Island	25	0.165	0.045	0.099	0.292	0.156	0.047	0.981	1.382
Niagara River	23	0.149	0.051	0.065	0.316	0.139	0.035	1.744	4.605
Port Colborne	25	0.175	0.091	0.072	0.520	0.149	0.078	2.446	8.177
Lake Ontario									
Hamilton Harbour	24	0.167	0.096	0.081	0.435	0.135	0.077	1.900	2.868
Toronto Harbour	25	0.234	0.172	0.096	0.775	0.170	0.103	1.867	3.110
Snake Island	23	0.261	0.148	0.105	0.829	0.214	0.101	2.769	9.706
Strachan Island	27	0.208	0.044	0.150	0.356	0.200	0.045	1.427	3.784

Superior. We developed post-hoc simple regression models with mercury mean levels in herring gull eggs as the response and levels in rainbow smelt as predictors, after correcting for their covariance with lipids in eggs and both fish length and lipid content, respectively:

$$level\left[Hg_{\text{Herring Gulls}}\right]_{tk} = \alpha_1 * level[Hg_{\text{Rainbow Smelt}}]_{tk} + \alpha_2 + \varepsilon_t \; \varepsilon_t \sim N\left[0, \sigma^2\right]$$

where $level[Hg_{\text{Herring Gulls}}]_{tk}$ is the mean mercury concentration in pooled herring gull egg samples at year t and site k, $level[Hg_{\text{Rainbow Smelt}}]_{tk}$ is the mean mercury concentrations at time t and site k in the prey item, after partialling out the effects of length and lipid content; α_1 is the slope coefficient reflecting the change in mean annual mercury levels in herring gull eggs for a unit change of mercury in rainbow smelt. The baseline mercury concentration in herring gull eggs is indicated by the intercept coefficient α_2, and ε_t is an error term, based on a draw from a normal distribution with a mean equal to zero and variance equal to the model error σ^2. This regression analysis allows us to estimate the relationship of mercury concentration between herring gulls and their prey item.

3. Results-discussion

3.1. Descriptive statistics

Herring gull eggs from Lake Ontario had the highest mercury concentrations across all sites between 1974 and 2015, followed by Lake Superior, Lake Erie, and Lake Huron (Table 1). There were also site-specific differences. Eggs from Snake Island (Lake Ontario) had the highest mean mercury concentrations (0.261 μg/g wet weight or ww), while eggs from Chantry Island (Lake Huron) had the lowest levels (0.122 μg/g ww). High standard deviations and asymmetric distributions indicate strong site-specific variation in egg mercury levels. Weseloh et al. (2011) found that egg mercury levels had a similar range across the Great Lakes with significant differences among sampling sites.

Mercury levels in fish were also highly variable (Table 2). Lake trout collected in the vicinity of Pie Island (Lake Superior) had the highest average mercury levels (0.194 μg/g ww), whereas the lowest levels were observed in the Eastern Basin of Lake Erie (0.077 μg/g ww). Overall, fish from Lake Superior had on average (1977–2013) the highest mercury levels compared to fish from the Lower Lakes. This is consistent with other studies (e.g., Monson et al., 2011) showing that mercury impacts are exacerbated in northern lakes because these landscapes are relatively more abundant in forests, where mercury deposition through litterfall is enhanced, and wetlands, where methylmercury production is elevated (Brigham et al., 2009). Higher mercury concentrations were also detected in lake trout in the most eastern stations in Lake Superior and Lake Ontario. Similar geographic patterns in average mercury levels have been detected in walleye and largemouth bass (*Micropterus salmoides*) (e.g., Monson et al., 2011). There are many different drivers contributing to these spatial differences, such as landscape features, trophic interactions, and proximity to current and legacy point sources of mercury (Evers et al., 2011).

Differences in mercury levels were also observed across trophic levels. Higher levels of mercury (on average about 30%) were observed in herring gull eggs compared to lake trout and walleye (only available from the Western Basin of Lake Erie). Reflecting their lower trophic position, rainbow smelt had the lowest mercury levels, which were approximately 67% lower than piscivorous fish and 76% lower than herring gull eggs. Other studies have reported similar increases in mercury levels with trophic position in fish and wildlife, stressing the potentially adverse impacts on their health and reproduction (Evers et al., 2011). Moreover, methylmercury is commonly produced under low oxygen levels in the sediments of the Great Lakes (Evers et al., 2011). Methylmercury accumulates in fish tissues, especially older piscivorous fish through prolonged dietary exposure compared to planktivorous fish (Visha et al., 2015). Round goby (*Neogobius melanostomus*) and zebra mussel (*Dreissena polymorpha*) invasions have altered the food web, potentially contributing to the recent increase in mercury trends across multiple fish and wildlife species (Azim et al., 2011).

Table 2
Descriptive statistics, mean, standard deviation (SD), minimum, maximum, median, interquartile range (IQR), skewness, and kurtosis for mercury concentrations (μg/g, wet weight) in Lake Trout (LT), Walleye (W) and Rainbow Smelt (RS), 1977–2013.

Site	n	Mean	SD	Min.	Max.	Median	IQR	Skewness	Kurtosis
Upper Lakes									
Lake Superior									
Pie Island/LT	333	0.194	0.102	0.050	0.710	0.160	0.138	1.335	2.320
Pie Island/RS	78	0.046	0.023	0.005	0.150	0.043	0.026	1.268	4.370
Whitefish Bay/LT	324	0.168	0.086	0.040	0.790	0.150	0.090	2.281	10.208
Whitefish Bay/RS	72	0.059	0.028	0.009	0.140	0.059	0.030	0.753	0.659
Lake Huron									
North Channel/LT	218	0.111	0.055	0.021	0.300	0.096	0.023	1.219	0.968
North Channel/RS	64	0.043	0.015	0.014	0.080	0.040	0.018	0.158	−0.367
Lower Lakes									
Lake Erie									
Eastern Basin//LT	233	0.077	0.032	0.001	0.189	0.080	0.037	0.289	0.588
Eastern/RS	178	0.028	0.021	0.005	0.110	0.020	0.025	1.958	4.424
Western Basin/RS	113	0.037	0.019	0.005	0.110	0.040	0.030	0.658	0.588
Western Basin/W	661	0.132	0.078	0.015	0.610	0.115	0.035	2.286	8.244
Lake Ontario									
Port Credit/LT	691	0.178	0.067	0.042	0.480	0.180	0.102	0.373	0.222
Port Credit/RS	188	0.057	0.041	0.010	0.230	0.040	0.050	1.263	1.166
Niagara/LT	480	0.155	0.057	0.015	0.347	0.147	0.077	0.497	0.083
Niagara/RS	139	0.047	0.032	0.010	0.210	0.038	0.028	1.837	4.852
Cobourg/LT	342	0.140	0.059	0.005	0.640	0.130	0.067	2.469	15.242
Cobourg/RS	148	0.053	0.039	0.010	0.190	0.037	0.051	1.279	0.858
Oswego/LT	721	0.142	0.058	0.019	0.420	0.135	0.068	0.785	1.483
Oswego/RS	240	0.041	0.027	0.008	0.180	0.033	0.013	2.642	8.791

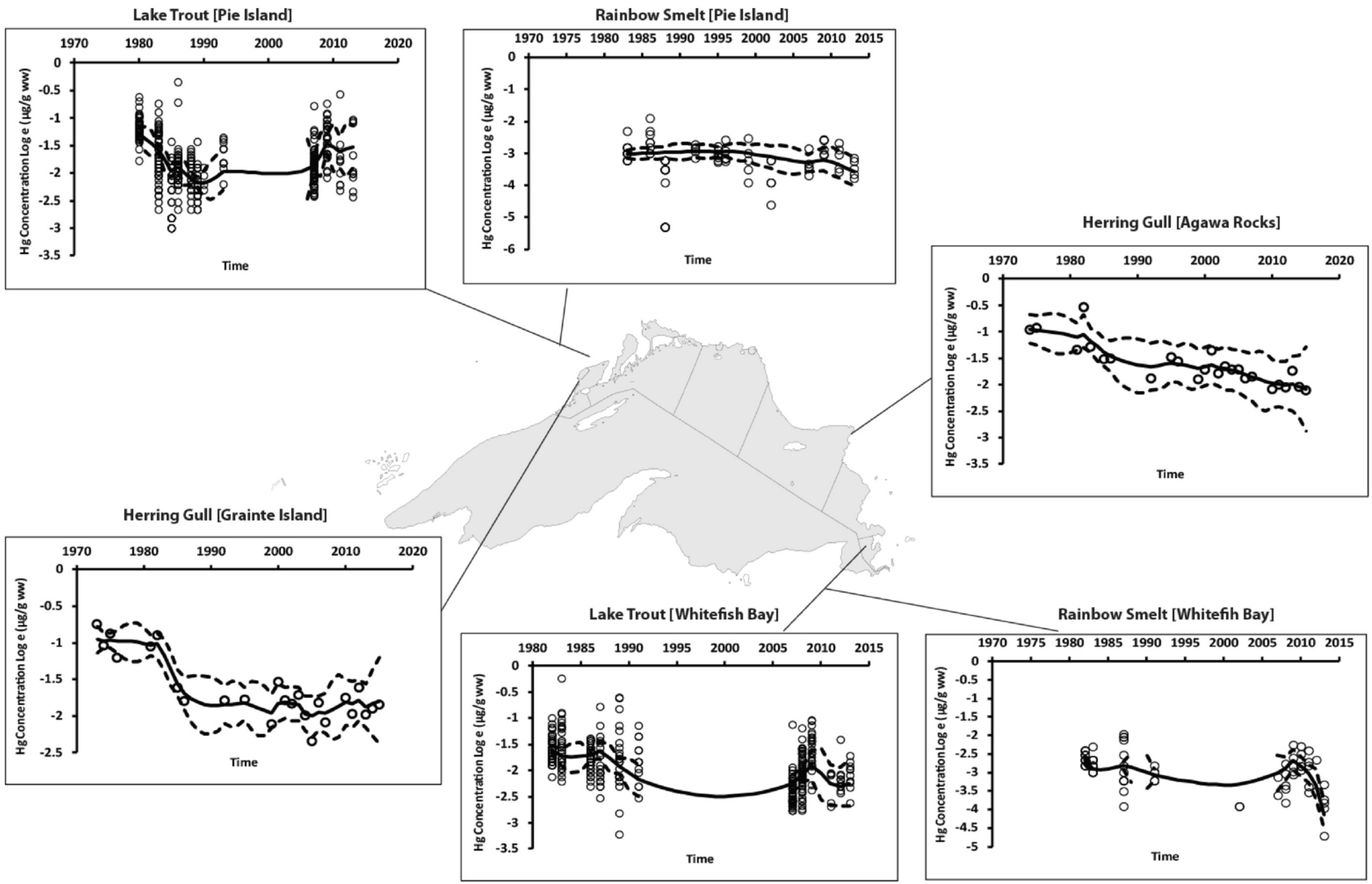

Fig. 2. Predicted (level) concentrations (Ln-transformed in µg/g wet weight) for all sites in Lake Superior for Herring Gulls, Lake Trout and Rainbow Smelt. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted mercury concentrations.

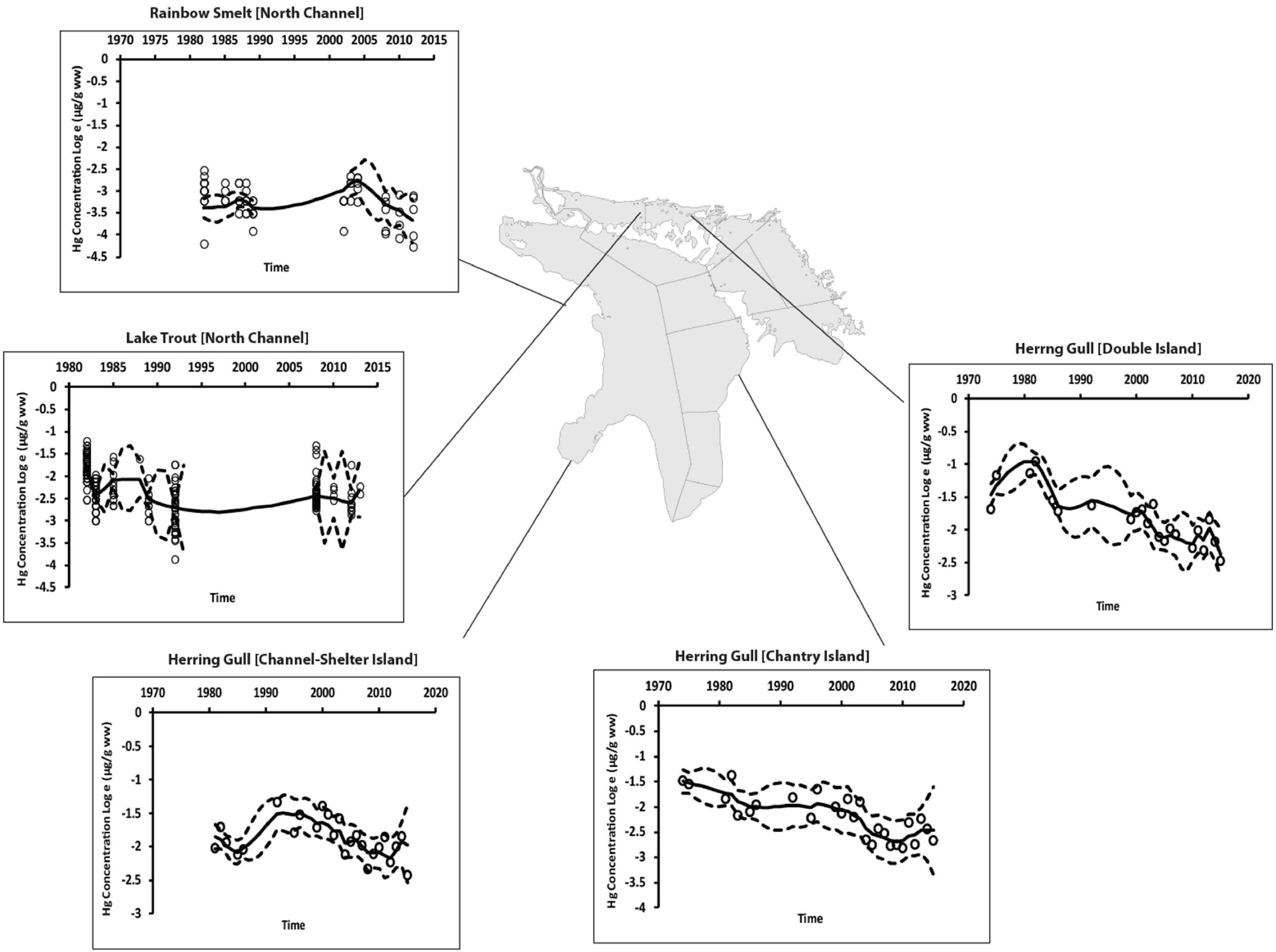

Fig. 3. Predicted (level) concentrations (Ln-transformed in µg/g wet weight) for all sites in Lake Huron for Herring Gulls, Lake Trout and Rainbow Smelt. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted mercury concentrations.

3.2. Herring gull eggs: spatio-temporal patterns

Overall, across all stations in the Upper Lakes (Superior and Huron), mercury levels in herring gull eggs decreased for the first three decades (1970 to 2000), with the exception of a slight peak in the mid-1990s in Shelter Bay (Lake Huron), and then continued to decline through the 2000s (Figs. 2–3 and Figs. 1 SI–2 SI). Variability in the rates of change in mercury levels was quantified with odds ratios, as in Visha et al. (2015), and here we report these odds as percentages. Rates varied across sites and the steepest declines in mercury concentrations, of about 3% per year, were observed in the 1980s at the two Lake Superior sites, Granite Island (89% chance of a decline) and Agawa Rocks (82% chance of a decline). During the 1990s, mercury concentrations remained relatively stable (Fig. 1 SI). Then, in the 2000s, mercury levels resumed their decline at the two Lake Superior sites, but this time the rates slowed to about 2% per year, with the chance of a decline falling to about 56% (Fig. 1 SI). Similarly, in Lake Huron throughout the 2000s, mercury declines of between 2 and 4% per year were observed (Fig. 2 SI) with the odds of a decline in randomly selected fish individuals ranging from 60% to 65%.

Similarly, mercury concentrations in herring gull eggs declined for the first three decades across all sites of the Lower Lakes (Erie and Ontario) (Figs. 4–5 and Figs. 3 SI–4 SI). Three sites continued to show declines through the 2000s, but trend reversals were detected at the other five sites. The steepest declines of about 6% per year (1970s–2000s) were detected at three Lake Ontario sites (Toronto Island, Snake Island, and Hamilton Harbour) (Fig. 5), followed by a 4% decline at two sites in Lake Erie (Fighting Island and Niagara River) (Fig. 4), while the remaining locations (Port Colborne in Lake Eire and Strahan Island in Lake Ontario) had declines of about 2% (Figs. 4 and 5). During the 2000s, the rates increased by 1% at all the Lake Erie sites with the highest chance of mercury increase in gull eggs from Fighting Island (75%) and the lowest chance (52%) of an increase in eggs from Niagara (Fig. 4). In Lake Ontario, a slight increase was only observed in Toronto Harbour with a chance of an increase slightly above 50%, while the rates decreased at all of the remaining sites (Fig. 5). Other studies in North America found that total mercury levels have also recently increased in common loons (*Gavia immer*) from northern Wisconsin (Meyer et al., 2011) and in bald eagles (*Haliaeetus leucocephalus*) from Voyageur National Park (Pittman et al., 2011).

Variability in spatio-temporal patterns in herring gull egg mercury levels was also observed by Weseloh et al. (2011), which used a slightly shorter time series (1974–2009). Similar to the change-point regression analysis of the latter study, we were able to detect trend reversals at all sites in Lake Erie and at one site in Lake Ontario. These additional insights were obtained because our DLM framework has a dynamic structure offering year-specific rates of change of the mercury concentrations, based on data from the present and recent past. In contrast, change-point regression provides fixed slopes over two (or more) periods, which are estimated by every single data-point within the identified time spans.

3.3. Fish: spatio-temporal patterns

In the Upper Lakes, mercury concentrations in lake trout declined on average by about 5 to 6% from the mid-1980s to 2000 (Figs. 2–3 and 1 SI–2 SI). In the 2000s, these trends reversed and mercury concentrations increased at the Lake Superior stations between 2% (Whitefish Bay, 60% chance of an increase) and 3% (Pie Island, 59% chance of an increase). Similarly, mercury concentrations increased by 2% in lake trout from Lake Huron (North Channel, 54% odds of an increase). In contrast, mercury levels in rainbow smelt continued to decrease in the Upper Lakes with the chance of a decrease consistently above 50% across all stations.

In Lake Erie, mercury concentrations in fish declined across all stations from the 1970s to 1999 and continued to decline only in rainbow smelt at the Western Basin station in Lake Erie for the subsequent period (Figs. 4 and 3 SI). At all the other stations, the trends reversed in the 2000s, by about 2% in walleye from the Western Basin (66% chance of an increase), 1% in lake trout from the Eastern Basin (53% chance of an increase), and 1% in rainbow smelt (53% chance of an increase) from the Eastern Basin. In contrast, at four Lake Ontario stations (Niagara, Port Credit, Coburg, and Oswego), mercury levels in lake trout declined sharply from the 1970s to 1999 between 3 and 5% (Figs. 5 and 4 SI). During the 2000s, mercury levels in lake trout continued to decline at two stations (Niagara and Port Credit), but at a slower rate of about 1% with a chance of decline ranging from 52 to 60%. Trend reversals of about 1% were observed in lake trout at the two remaining stations with the odds of an increase ranging from 52 to 55%. Mercury concentrations in rainbow smelt also declined sharply (between 3 and 5%) across all stations through the first three decades and continued to decrease but with slower rates (1%–3%).

Similar total mercury trend reversals have been reported in wildlife from the entire Great Lakes Region (Monson et al., 2011). Bhavsar et al. (2010) used Mann-Kendall's trend analysis to show that total mercury levels declined from the 1970s to 2007 in lake trout and walleye across the Canadian Great Lakes, except in Lake Erie walleye, where the trends have reversed since 2005 (Zananski et al., 2011). Our study had slightly longer time series (1970s - 2015) and the evolving structure of DLMs allowed to accurately capturing recent shifts in mercury levels in fish communities. Another recent study using historical data (1970–2012) from lakes across Ontario showed that trends in mercury levels in piscivorous fish, walleye, northern pike (*Esox lucius*), and lake trout, switched from declining to increasing starting from the late 1990s and continued throughout the 2000s (Gandhi et al., 2014).

3.4. Long term trends

Mercury levels in the Great Lakes environment have declined over the last four decades concurrent with decreased air emissions from regional sources. Sediment cores from the Lower Lakes (e.g., Lake Ontario) suggest that local and regional sources of atmospheric mercury emissions are an important source of loadings into the Great Lakes compared to global sources (Drevnick et al., 2011). Substantial efforts have been made to control point-source discharge and this is supported by sediment cores which show a decline after the mid-1980s in the Great Lakes region (Drevnick et al., 2011). Long term mercury trends in herring gull eggs and fish (e.g., this study; Bhavsar et al., 2010) show a regional decline from the 1970s to the 2000s, consistent with declines in regional atmospheric emissions and sediment accumulation rates.

However, trend reversals on mercury levels have been recently observed by collaborations between the U.S. EPA, ECCC, and Ontario Ministry of the Environment and Climate Change (OMECC) (e.g., Monson et al., 2011; Zananski et al., 2011) in top fish predators, such as walleye and lake trout. Increases on mercury levels in herring gulls and fish communities have been attributed to increased temperatures, which can accelerate methylation across the Great Lakes (Monson et al., 2011; Austin and Colman, 2008). Similar effects could have been induced by shifts in trophodynamics resulting from invasive species, e.g., zebra mussels (*Dreissena polymorpha*) and round gobies (*Neogobius melanostomus*), which tend to accumulate contaminants and subsequently transfer them to upper trophic levels (Azim et al., 2011; Monson et al., 2011). Other factors include lower water levels, greater exposed shoreline

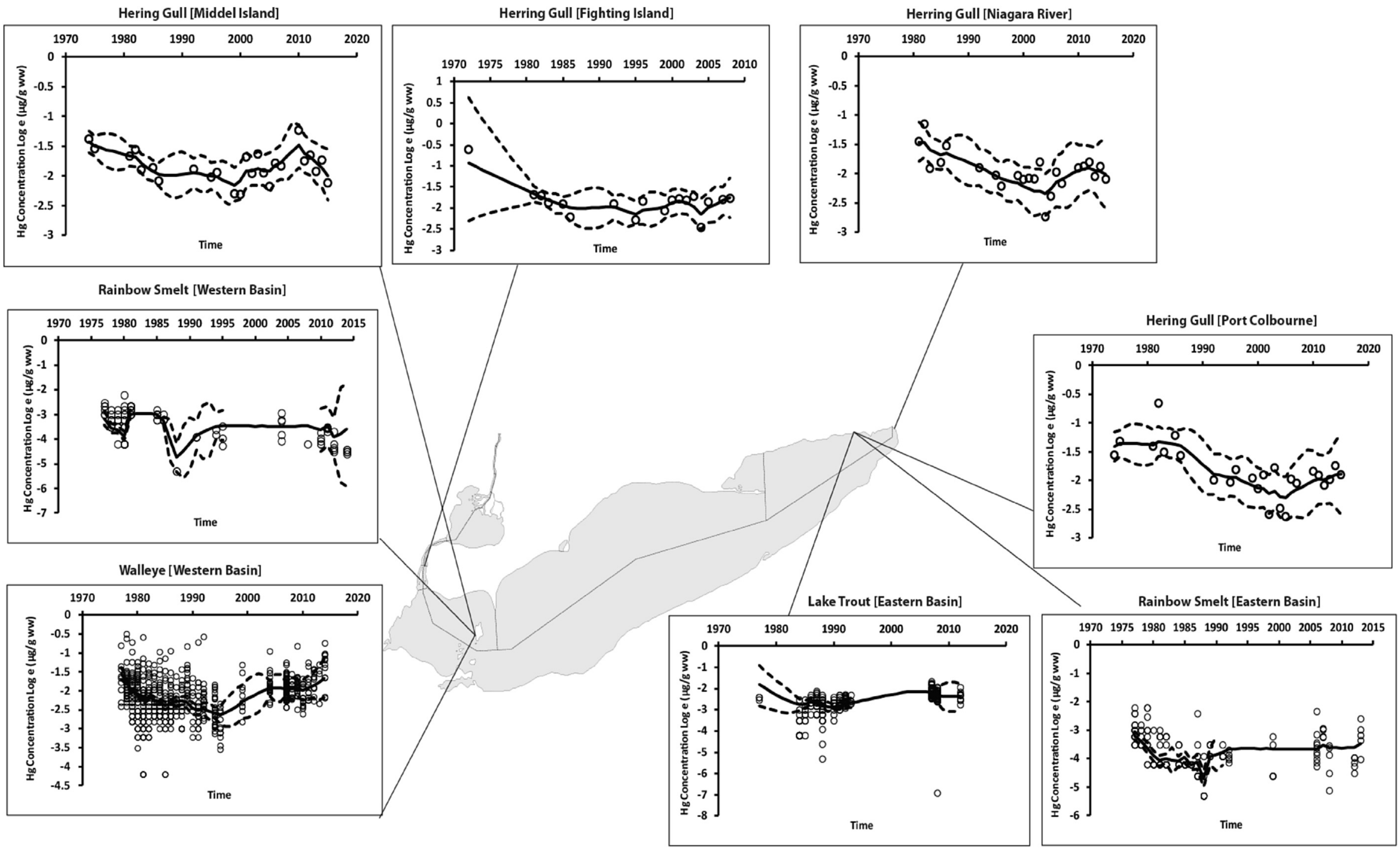

Fig. 4. Predicted (level) concentrations (Ln-transformed in µg/g wet weight) for all sites in Lake Erie for Herring Gulls, Lake Trout, Rainbow Smelt and Walleye. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted mercury concentrations.

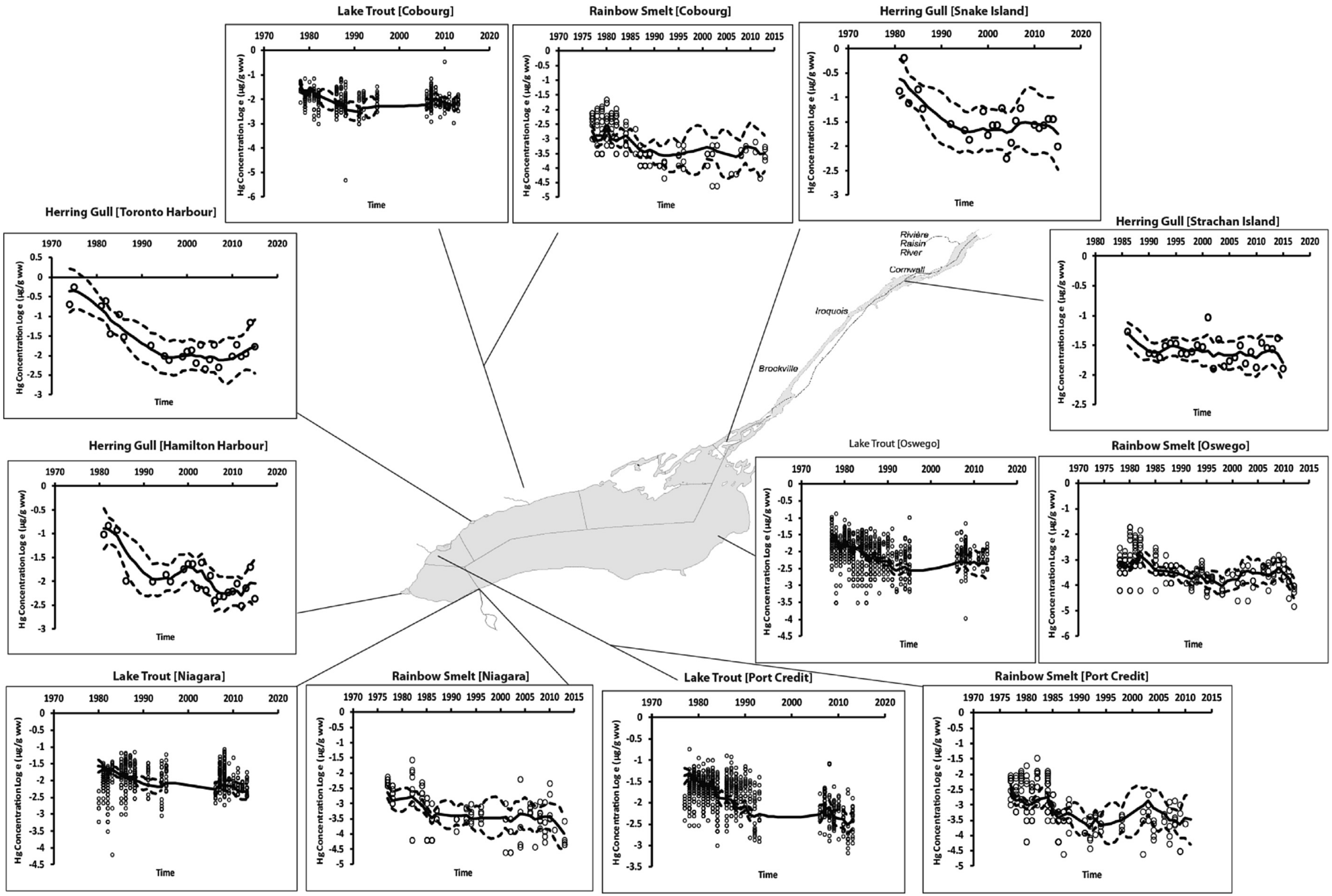

Fig. 5. Predicted (level) concentrations (Ln-transformed in µg/g wet weight) for all sites in Lake Ontario for Herring Gulls, Lake Trout and Rainbow Smelt. The solid and dashed lines correspond to the median and the 95% credible intervals of the predicted mercury concentrations.

associated with drought (Meyer et al., 2011), and reversal of the biodilution effect through decreases in nutrient loading (Zananski et al., 2011). However, the direct causes of the recent increase in mercury levels need further monitoring and research to determine whether these are short-term oscillations or long-term trends. Increased mercury levels in fish have also been reported in the Canadian Artic and Greenland (Carrie et al., 2010; Riget et al., 2010; Wyn et al., 2010) and these studies have attributed the increase to a warming climate. Continued monitoring of mercury levels in herring gulls and fish is required to further quantify the trophic implications from these recent patterns.

3.5. Trophic interactions

Trophic interactions between herring gulls and rainbow smelt were examined with a Bayesian linear regression approach, using a subset of DLM level predictions from the early-1980s to the late 2000s, to optimize spatial and temporal overlap. In Lake Ontario, there was a strong positive relationship (high and well-identified slope α_1) between mercury concentrations in rainbow smelt and herring gull eggs, indicative of strong trophic interactions (Table 3). In Lake Superior, this relationship was also positive, although somewhat weaker, suggesting that bioaccumulation rates in herring gulls are similar to rainbow smelt, their preferred prey item (e.g., Fox et al., 1990). Other studies found similar trophic interactions, for example, yellow perch (*Perca flavescens*) are a common prey item for loons (*Gavia immer*) and mercury levels in loons are closely related to levels in perch (Evers et al., 2011). In contrast, negative relationships were detected in Lake Huron and Lake Erie, indicating a trophic decoupling (prey switching) between herring gulls and rainbow smelt (Table 3).

Herring gulls are primarily exposed to mercury through their diet (Scheuhammer et al., 2007) and thus egg chemical composition will reflect the gull diet over several weeks during the period of egg formation (Hobson et al., 1997). Using the entire time series (1974 to 2009), Weseloh et al. (2011) found that there were no differences between the slopes of mercury levels in herring gulls eggs and rainbow smelt in each of the Canadian Great Lakes; however, they also noted that the rates of mercury decline in gull and smelt may be diverging, possibly due to recent dietary changes. Diet composition of aquatic birds is sensitive to food web structure. In the Great Lakes, herring gulls feed mainly nearshore on small fishes, primarily on rainbow smelt and alewife (*Alosa pseudoharengus*) (Pierotti and Good, 1994). Stable isotope analyses performed on herring gull eggs from 1974 to 1995 by Hebert et al. (1999) showed significant terrestrial enrichment in Lake Erie. In a more recent retrospective study (1981–2005), using both stable isotopes and fatty acid profiles, Hebert et al. (2009) concluded that there was a temporal shift in the herring gull diet from Lake Huron with an increasing reliance on terrestrial foods through time. A study conducted by Ewins et al. (1994) found that herring gulls from eastern Lake Huron (Chantry Island) fed extensively on deer mice in early spring.

Gulls are opportunistic scavengers typically relying on abundant food items, such as mussels and garbage, and dietary shifts can have profound consequences for their exposure to contaminants (Pierotti and Annett, 1991). For example, rainbow smelt are trophically elevated and tend to accumulate greater body burdens of mercury relative to native forage fish species, which in turn represents a greater risk of contaminant exposure for herring gulls that have greater reliance on them (Swanson et al., 2003). Ewins et al. (1994) concluded that reductions in available fish prey items, such as alewife, are responsible for the recent dietary shifts. Hebert et al. (2008) also concluded from their retrospective (25 years) analysis across all the Great Lakes that herring gull diets tracked decreases in pelagic prey fish abundance. All of these studies are consistent with our finding of trophic decoupling in Lakes Huron and Erie.

Table 3
Bayesian regression models used to predict mercury levels in Herring Gull eggs from concentrations in rainbow smelt tissues. The slope coefficient (α_1) reflecting the change in mean annual mercury levels in Herring Gull eggs for a unit change of mercury in Rainbow Smelt, the average baseline mercury concentration in Herring Gulls (α_2), and the model error (σ) are shown.

Lake	Mean (α_1)	Mean (α_2)	SD (α_1)	SD (α_2)	Mean Error(σ)
Lake Superior	0.081	−1.745	0.029	0.028	0.157
Lake Huron	−0.038	−1.913	0.045	0.045	0.246
Lake Erie	−0.026	−1.919	0.035	0.035	0.182
Lake Ontario	0.306	−1.655	0.036	0.036	0.197

4. Conclusions

We presented a unique spatio-temporal comparison of mercury trends in three trophic levels: herring gulls, piscivorous fish (lake trout, walleye) and planktivorous fish (rainbow smelt). Mercury patterns were quantified using dynamic linear modelling which has an evolving structure that allowed parameters (slopes, intercepts) to vary through time compared with the rigid structure of conventional regression models. Our trend analysis illustrated that past (BTS) and current (CARA, GLWQA-Annex 3) programs have been relatively successful in controlling mercury levels in biotic communities. However, mercury concentrations varied considerably in space and time, and across trophic levels indicating that predator-prey dynamics may have modulated the reported trends together with the reductions from exogenous sources. Recent increases in mercury levels at some of the sites in the Lower Lakes have been attributed to various drivers, such as climate change, shifts in trophodynamics resulting from invasive species, and fluctuating water levels. Our Bayesian regression analysis suggests strong trophic interactions between herring gulls and rainbow smelt in Lakes Superior and Ontario and a trophic decoupling in Lakes Huron and Erie. Continued monitoring of mercury levels in herring gulls and fish is required to consolidate this trophic pattern and further evaluate its broader implications.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2016.12.148.

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