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MERGANSER: An Empirical Model To Predict Fish and Loon Mercury in New England Lakes

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Ⓢ *Supporting Information*

ABSTRACT: MERGANSER (MERcury Geo-spatial AssessmeNtS for the New England Region) is an empirical least-squares multiple regression model using mercury (Hg) deposition and readily obtainable lake and watershed features to predict fish (fillet) and common loon (blood) Hg in New England lakes. We modeled lakes larger than 8 ha (4404 lakes), using 3470 fish (12 species) and 253 loon Hg concentrations from 420 lakes. MERGANSER predictor variables included Hg deposition, watershed alkalinity, percent wetlands, percent forest canopy, percent agriculture, drainage area, population density, mean annual air temperature, and watershed slope. The model returns fish or loon Hg for user-entered species and fish length. MERGANSER explained 63% of the variance in fish and loon Hg concentrations. MERGANSER predicted that 32-cm smallmouth bass had a median Hg concentration of 0.53 $\mu g\ g^{-1}$ (root-mean-square error 0.27 $\mu g\ g^{-1}$) and exceeded EPA's recommended fish Hg criterion of 0.3 $\mu g\ g^{-1}$ in 90% of New England lakes. Common loon had a median Hg concentration of 1.07 $\mu g\ g^{-1}$ and was in the moderate or higher risk category of >1 $\mu g\ g^{-1}$ Hg in 58% of New England lakes. MERGANSER can be applied to target fish advisories to specific unmonitored lakes, and for scenario evaluation, such as the effect of changes in Hg deposition, land use, or warmer climate on fish and loon mercury.

■ INTRODUCTION

Global mercury (Hg) contamination via atmospheric deposition poses a risk of neurological damage to humans and wildlife, and fish consumption is a major exposure pathway.[1] All 50 USA states have advisories to limit fish consumption. These may be conservative blanket advisories or may apply to specific water bodies based on known fish Hg concentrations. For example, Massachusetts recommends consumption limits by species on a lake by lake basis (http://db.state.ma.us/dph/fishadvisory/ accessed January 24, 2012). Mercury levels in fish do not

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correspond directly to Hg loads in deposition.[2−4] Processes within the watershed and lake have a strong bearing on the amount of mercury transported by water and the percentage of Hg that is methylated and bioaccumulated in fish and loons.[4−6] Less than 15% of New England lakes have data on fish and loon mercury concentrations. An assessment tool that uses watershed properties to estimate Hg exposure risk in unmonitored lakes would be useful for resource management and human and wildlife health protection.

Typically 80% to 90% of Hg deposited by atmospheric deposition is retained indefinitely by organic matter in soils.[7] The remainder is transported by dissolved organic carbon (DOC) or particulate organic carbon (POC).[8−10] Thus the movement of organic matter is key to the transport and subsequent aquatic biological uptake of Hg.[10] Forest vegetation and the presence of wetlands, especially those in contact with streams and lakes, facilitate DOC export and dissolved Hg export.[11] Steep slopes, organic-rich soils, agriculture, and urban development contribute to erosion that leads to POC and particle Hg export.[12−14] Lakes with large surface area relative to the size of their watersheds receive a correspondingly larger share of atmospheric Hg directly, i.e., without retention in soils.[5,15] The amount of Hg in lake fauna depends in part on the loading of Hg from the watershed (or direct atmospheric deposition of Hg to the lake surface), the extent of conversion to methylmercury (MeHg), and foodweb structure.

Hg in upper trophic level fauna occurs nearly entirely as MeHg, the form that bioaccumulates. Hg methylation may occur within the watershed prior to entering the lake, such as in wetlands.[16,17] Methylation is also known to occur in upland settings, such as anoxic microsites in the unsaturated zone or within groundwater near the water table[18] or within littoral zone sediments or in the water column of lakes. Littoral zone sediments subject to periodic dewatering and reinundation, such as in managed reservoirs, are also prime methylation locations.[19] Requisites for methylation are sufficient Hg and organic matter and an optimum level of sulfur (S).[20] The amount of Hg that is methylated is frequently only a few percent or less, but occasionally exceeds 10%.[21] The competing process of demethylation often limits the amount of MeHg release to surface waters.[22]

The Hg burden in fish and loons is the net result of myriad processes in the terrestrial landscape and within lakes. Readily quantifiable watershed and lake metrics, and land use/land cover features often serve as proxies for these underlying processes. Thus prediction of fish and wildlife Hg in lakes where we lack information lends itself to an empirical modeling approach. This approach is embodied in MERGANSER (MERcury Geo-spatial AssessmeNtS for the New England Region), a model for New England lakes that uses a wide array of landscape and lake features to predict fillet Hg concentrations for 12 fish species and blood Hg concentrations in common loon (*Gavia immer*), a prominent high trophic level avian piscivore. We calibrated MERGANSER to available fish and loon Hg concentrations. MERGANSER can guide fish advisories for specific lakes or groups of lakes (e.g., an entire state) and can also test scenarios, e.g., how changes in Hg deposition, land use, and climate will affect fish and loon Hg.

■ MODEL DESCRIPTION

Approach. MERGANSER is an empirical model to predict Hg concentrations in fish fillet and loon blood in New England lakes. So that MERGANSER can be applied to any lake by using existing data, we restricted predictor variables to those obtainable for all lakes from continuous geographic coverages, such as land cover characteristics and estimates of atmospheric Hg deposition. These static MERGANSER predictors represent current conditions and assume a steady state; a land use designation (e.g., agriculture) does not account for short-term perturbations to Hg cycling that conversion to that land use may generate. Use of known strong predictors such as pH, DOC, and lake trophic status was excluded as they were not universally available. When multiple fish or loon Hg analyses existed from a single lake, we treated each case independently in formulating the model.

Lake Identification and Watershed Delineation. To identify the 4404 New England lakes ultimately included in MERGANSER, we used the 1:24 000 National Wetlands Inventory (http://www.fws.gov/wetlands/ accessed June 21 2010) as the base map. The NWI has high-resolution spatial coverage of lakes and categorized wetlands. We merged the NWI lakes database with the 1:100 000 National Hydrography Data set Plus (NHDPlus; http://www.epa.gov/waters/ accessed June 21, 2010) to delineate the watershed for each lake. NHDPlus also provided additional lake watershed characteristics, but its coarser scale excluded some NWI lakes. For these "non-network" lakes, we computed watershed boundaries using standard GIS watershed delineation tools and the NHDPlus digital elevation model (DEM) flow direction grid. We limited consideration to lakes 8 ha or larger, and excluded lakes whose drainage area extended into Canada due to difficulty in reconciling Canada and USA data sources. Despite the complexity and spatial contrasts within large lakes, we retained New England's largest lakes (except Lake Champlain, which has drainage area in Canada) in MERGANSER.

Predictor Variables. We tested 54 predictor variables (Supporting Information (SI), Table S1) capturing the following: (1) physical characteristics (lake and watershed area and their ratio, slope, elevation, precipitation, temperature); (2) atmospheric deposition (wet and dry Hg, total sulfur); (3) land cover categories of the National Land Cover Data set (NLCD); (4) wetland categories classified in the NWI, including open water (representing nested upstream lakes); and (5) miscellaneous (lake and watershed alkalinity, dam presence/absence, human population density, and annual nitrogen and phosphorus fluxes from the lake-watersheds derived from the SPARROW model[23]). Most variables were log-normally distributed, and values were thus natural log-transformed.

Wet and dry Hg deposition were estimated for MERGANSER via a blend of (1) an observation-based model, which interpolated observations and adjusted them for elevation and land cover type,[24] and (2) estimates using the Regulatory Modeling System for Aerosols and Deposition (REMSAD) (http://www.nescaum.org/documents/mercury-modeling-report_2007-1005b_final.pdf/). We took the maximum value of the two estimation methods as the best representation of the likely Hg deposition. The observation-based method produced higher estimates more distant from point sources (where monitoring stations are generally located), while the REMSAD model produced higher estimates closer to point sources (where monitoring stations are generally lacking). Deposition within REMSAD 36 km^2 grid cells was smoothed to avoid discontinuities along cell boundaries, but deposition estimates for specific lakes could be compromised by this coarse grid, particularly near point sources.

Table 1. MERGANSER Predictors and Coefficient Values, with Standard Error, *t*- and *p*-Values, and Variance Inflation Factors (Overall Model r^2 Was 0.63)

predictor	units	coeff	standard error	*t*-value	*p*-value	variance inflation factor	source[a]
spatial variables							
intercept		−11.1270	0.463	−24.03	<0.0001	0.00	
ln (total Hg deposition)	μg m^{-2} yr^{-1}	0.2773	0.046	6.00	<0.0001	4.06	Miller et al. (2005); this paper
ln (watershed area)	km^2	0.0354	0.006	5.90	<0.0001	2.24	NHDPlus
ln (% forest canopy area)	%	0.2400	0.085	2.83	0.0047	6.49	NLCD
ln (% wetland area)	%	0.0666	0.012	5.60	<0.0001	2.01	NWI
fourth root of population density, 2000 Census	km^{-2}	0.0514	0.008	6.16	<0.0001	2.33	NHDPlus
ln (slope)	unitless: y/x	−0.1400	0.032	−4.32	<0.0001	3.15	NHDPlus
ln (mean annual temp, 1971−2000)	°C	−0.4446	0.057	−7.83	<0.0001	5.51	NHDPlus
ln (% agricultural land)	%	0.0198	0.007	2.86	0.0042	1.92	NLCD
weighted watershed alkalinity	unitless[c]	−0.1207	0.009	−13.17	<0.0001	1.68	EPA
interaction term: % shrubland and ln (total Hg deposition)		0.0092	0.001	9.26	<0.0001	2.05	NLCD, Miller et al. (2005)
interaction term: % forest canopy and ln (total Hg deposition)		0.0035	0.000	7.46	<0.0001	9.32	NLCD, Miller et al. (2005)
interaction term: ln (total Hg deposition) and watershed alkalinity[b]		0.1928	0.027	7.06	<0.0001	1.93	Miller et al. (2005), EPA
user-input variables							
loon	binary: 1/0	9.4821	0.202	46.83	<0.0001	37.36[d]	user specified
ln (length)	mm	1.5310	0.036	42.24	<0.0001	39.95[d]	user specified
ln (length): brook trout	mm	−0.1337	0.010	−13.78	<0.0001	1.63	user specified
ln (length): brown bullhead	mm	−0.1846	0.007	−24.97	<0.0001	1.14	user specified
ln (length): eastern chain pickerel	mm	−0.0720	0.008	−8.99	<0.0001	1.25	user specified
ln (length): lake trout	mm	−0.1716	0.008	−21.14	<0.0001	1.94	user specified
ln (length): land locked salmon	mm	−0.1600	0.010	−15.76	<0.0001	1.26	user specified
ln (length): largemouth bass	mm	−0.0150	0.005	−3.15	0.0016	1.87	user specified
ln (length): pumpkinseed	mm	−0.0362	0.012	−2.90	0.0037	1.10	user specified
ln (length): white perch	mm	0.0457	0.006	8.07	<0.0001	1.30	user specified
ln (length): white sucker	mm	−0.2292	0.013	−17.76	<0.0001	1.15	user specified

[a] For full description of sources see Table S1 in the Supporting Information. [b] This term was optimized as: (Watershed weighted alkalinity −2.48208){ln (total Hg deposition) − 3.38803}. [c] From EPA scale (units in μequiv L^{-1}): 1 (<50); 2 (50−100); 3 (100−200); 4 (200−400); 5 (>400); weighted by areal extent. [d] The high values of the variance inflation factors for "loon" and "ln(length)" result from an artifact of the model formulation; because the loon Hg variable is standardized, no length variable is needed, yet the model requires a value. When "loon" is set to 1, length is set to 1 so ln(length) = 0. Conversely, when loon = 0, meaning the user is requesting fish Hg, "length" will have a value. The resulting strong anticorrelation has no effect on the model because it has no effect on any other coefficients including the coefficient for ln(length); MERGANSER effectively uses only one of these variables at a time.

Response Variable: Fish Fillet Hg. Fish fillet Hg concentrations were extracted from the U.S. Environmental Protection Agency (EPA) publicly accessible Wildlife database (http://oaspub.epa.gov/aed/wildlife.search), which stores results from EPA's Regional Environmental Monitoring and Assessment Program (REMAP) and State monitoring programs. Of the 4404 modeled MERGANSER lakes, 351 lakes had fish tissue Hg values with 3470 individual values from 12 species. Species with fewer than 50 cases were excluded. Fish Hg concentrations were available from 1988 to 2006, with the majority from 1996 to 2006, thus limiting the effects of temporal trends in fish Hg due to changing deposition and land use. Distributions of fish species and size are given in Figure S1 in the SI.

Response Variable: Loon Blood Hg. Loon Hg data were generated by the BioDiversity Research Institute (BRI) in Maine. BRI normalizes loon Hg concentrations for gender, age, and tissue type (blood or egg) to an adult female loon blood Hg equivalent, or Female Loon Unit (FLU).[4] FLU values are linearly correlated with yellow perch fish tissue Hg, as yellow perch are the primary loon diet during the breeding season.[25] As loon Hg varied widely among multiple loon territories on the same lakes, we restricted consideration to single-territory lakes, large enough to reliably support a breeding pair but too small for a second pair (25 to 80 ha). This resulted in 253 FLU values from 87 lakes, 18 of which overlapped with the fish data set.

Model Formulation. We tested a nonlinear (multiplicative) model, but achieved greater prediction accuracy with a linear least-squares approach for MERGANSER. We evaluated individual predictor variables (usually natural log-transformed) and expected interaction terms by iterative stepwise addition and subtraction of terms, guarding against overfitting the model by accepting only terms that were scientifically defensible and significant at $p < 0.05$. Minimal covariance among predictors was confirmed by inflation factors <10 for all predictors (Table 1). To test whether sample lakes were representative of the general population, we evaluated the distributions of predictor values in both data sets and found no strong biases (Figure S2, SI). Finally, we evaluated various stratification schemes such as dam/no dam, "first-order" vs nested lakes, and lake size classes. Stratification was not adopted because model performance gains were insufficient to warrant the additional complexity that stratification would pose to the end-user.

For the response variable, previous efforts have standardized Hg concentrations from various fish to a common length and species (e.g., 12-cm Yellow Perch Equivalents[3]). MERGANSER achieved better results using unadjusted fish Hg as the response variable, while directly incorporating fish species and length as predictor variables. Unlike the static predictors derived from spatial coverages, species and length are dynamic predictors that are user-specified for any lake. MERGANSER users are cautioned to constrain Hg predictions to the 12 fish species (and loon) modeled and to the known geographic species ranges.

To economize on the number of variables, fish length (without specifying species) sufficed for three species whose Hg concentrations fell on the dominant Hg:length relation. For the remaining nine species, a species binary term effectively adjusted Hg concentrations up or down accordingly. Because the loon Hg response variable FLU was standardized, no length term was needed; toggling the loon binary variable directed MERGANSER to return an FLU value based solely on the parameters for a specific lake. Fish tissue Hg concentrations and FLU values were log-normally distributed, so ln(fish Hg) and ln(FLU) were set as the model response variables.

Model Validation. MERGANSER was validated by using an additional 1577 fish and 54 loon cases, distributed broadly across New England and with distribution of predictor values similar to the calibration cases (Figure S2, SI).

■ RESULTS

Predictor Variables. Of the 54 spatial predictors tested (Table S1, SI), MERGANSER accepted 10: total Hg deposition, watershed area, percent forest canopy area, percent wetland area, percent agricultural land, percent scrub-shrub land (in an interaction term), human population density, slope, mean annual air temperature, and watershed alkalinity (Table 1). The resulting model explained 63% of the variance in fish and loon Hg. Example predictions for 32-cm smallmouth bass (*Micropterus dolomieu*) are presented as a probability of exceeding the EPA recommended Hg concentration criterion of 0.3 μg g^{-1}, wet weight (Figure 1).

Total Hg deposition was a stronger predictor than wet or dry Hg deposition separately, so it was retained in the final model. Total Hg deposition was also a component in each of the three interaction terms, with watershed alkalinity, scrub-shrub land, and forest land, all with positive coefficients.

Of the land-cover terms, percent wetland area within a watershed had the strongest positive effect on fish Hg. Knowing the importance of wetlands as methylation sites, we tested the effect of various wetland types from the NWI, and the area of wetland contiguous to the lake and its inlet streams as model inputs, but none of these predictors performed as well as overall percent wetland. Percent forest canopy and percent agriculture each had smaller, but also positive effects.

Watershed alkalinity was an area-weighted average of the EPA alkalinity class designation (scale 1 to 5), which in turn was based on spatially interpolated values from regionwide field measurements in lakes and streams (<ftp://ftp.epa.gov/wed/ecoregions/1_Latest_Updates/alk_rev_09.Zip). The watershed-based alkalinity index was a slightly better predictor than a lake-based alkalinity. Fish Hg was negatively related to alkalinity. Because alkalinity can change with rock type over short spatial scales, some lakes may be mischaracterized in this low-resolution coverage, yet alkalinity was one of the strongest watershed predictors (Table 1).

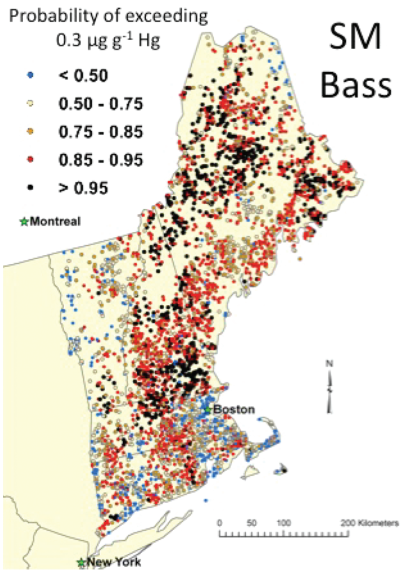

Figure 1. Map of New England states, USA, showing MERGANSER predictions for probability of fish tissue Hg concentration exceeding EPA'S recommended criterion of 0.3 μg g^{-1}, wet weight, in 32-cm smallmouth bass in lakes larger than 8 ha.

Of the remaining predictors, human population density (fourth root) and lake watershed drainage area had positive effects on fish Hg, whereas mean annual temperature and watershed slope had negative effects. Surprisingly, S deposition was not a significant predictor.

To test for geographic anomalies in fish Hg unrelated to any of our established predictors, such as ecological factors, we used a coverage of EPA-designated ecoregions. In contrast to the model of Sackett et al.[26] for North Carolina, ecoregion terms were not significant in MERGANSER ($p > 0.05$). This result was consistent with the lack of a spatial pattern in model residuals.

For loons, MERGANSER returns the standardized FLU when the "loon" binary variable is toggled on. This term had the highest *t*-value of all predictor variables (Table 1), and its inclusion increased model variance explained (r^2) from 61% to 63%. MERGANSER probabilities for exceedance of the severe risk threshold for loon Hg (as FLU) are presented (Figure 2).

Error and Probability. The root-mean-square error for MERGANSER was 0.51. For a predicted fish Hg value of 0.2 μg g^{-1}, there is 95% confidence that the true value falls between 0.07 and 0.55 μg g^{-1}. From predicted Hg concentrations and assuming a log-normal error distribution, we could compute the likelihood of a species exceeding a target Hg concentration in a given lake. This calculation underlies the exceedence probability distribution maps by species and length (Figures 1 and 2).

Model residual distribution was near log-normal and heteroscedastic (SI, Figure S3), suggesting appropriate model parametrization. There were no grossly anomalous high or low outliers.

Model Validation. For the validation data set ($n = 1631$), predictions correlated linearly on a 1:1 line with observations

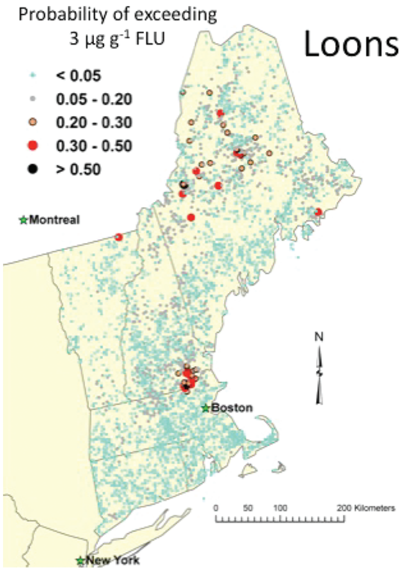

Figure 2. Map of MERGANSER predictions for probability of loon Hg (as FLU) exceeding 3 μg g^{-1} in New England lakes. Loons with FLU > 3 μg g^{-1} are likely to exhibit behavioral deviations and reduced reproductive success.

with an r^2 of 0.52 (SI, Figure S4). Nearly all observations fell within the 95% confidence band of the predictions.

Scenario Testing. We tested MERGANSER for three scenarios depicting likely future trends. In the first scenario, we lowered Hg deposition regionwide to the current minimum value (10.0 μg m^{-2} yr^{-1}). The rationale here is that global background deposition has a floor that is not likely to decrease, while areas impacted by anthropogenic point sources may experience significant reductions. The second scenario addressed growing demand on the biomass resource of New England forests and assessed the effect of converting 20% of each watershed from forest to open land. The third scenario considered a 2 °C mean annual air temperature increase.

All three scenarios reduced predicted median Hg concentrations and the probability of exceeding EPA's recommended Hg criterion. For a 32-cm smallmouth bass, median Hg (exceedance probabilities) decreased from present values of 0.54 μg g^{-1} (87.2%) to 0.31 μg g^{-1} (51.7%) for the reduced deposition, 0.43 μg g^{-1} (75.6%) for reduced forest, and 0.47 μg g^{-1} (81.1%) for increased temperature (SI, Figure S5). All scenarios considered a future stable condition that ignores ecosystem adjustments and lags, e.g. short-term increases in fish Hg due to land disturbance.

These results give an indication of the expected direction of change, but each considers a single variable in isolation, holding other predictors constant, and does not account for interactions and feedback processes. Projected changes in fish tissue Hg might be either counteracted or magnified by changes in other driving variables.

DISCUSSION

Interpretation of Predictor Variables. MERGANSER's primary objective was to predict fish and loon Hg in unmeasured lakes. A secondary objective was to examine the predictors in the model to interpret first-order controls on fish and loon Hg. The physical and biological factors that cause methylmercury to form, persist, and enter the food web are complex, and empirical predictors typically serve as proxies for a process or suite of processes representing the true controls. However, in-lake processes were difficult to represent in MERGANSER because of limited available information on lake characteristics for the target (unmeasured) lakes.

MERGANSER indicated that fish and loon Hg concentrations decrease in response to the decreasing gradient of Hg deposition across New England from southwest to northeast. This result was consistent with a nationwide study,[27] but a Vermont/New Hampshire study found no relation between fish Hg and Hg deposition,[28] possibly due to the much smaller range of Hg deposition than in the present study. MERGANSER predictions were more accurate when using total Hg deposition rather than wet Hg deposition, as wet deposition is usually less than half of total deposition.[29] Dry deposition is enhanced by the presence of forest canopy,[24] as demonstrated by the significant positive interaction terms of Hg deposition with shrub-scrub land and with forest canopy. Consistent with other studies,[5,11] watershed features were also strong predictors of fish and loon Hg. The link between Hg deposition and watershed features in driving Hg bioaccumulation is demonstrated by the significance of watershed alkalinity in MERGANSER. This result is consistent with evidence that fish Hg is higher in more acidic lakes.[30,31] The significance of the Hg deposition−alkalinity interaction term demonstrates positive feedback.

The significance of land cover in MERGANSER agreed with many other studies. Wetland cover is consistently a strong predictor because wetlands are primary sites of methylation and DOC production.[2,16] In contrast to Kramar et al.,[11] we found overall wetland area coverage to be more important than wetland area proximal to the lake. The significance of percent forest canopy was consistent with preferential scavenging of atmospheric Hg by the canopy, high DOC generated on the forest landscape, and increasing evidence that methylation occurs at the water table or in anoxic microsites in unsaturated forest soils.[16] Agricultural land was a weak positive predictor, probably because of the tendency for Hg loss from agricultural lands with eroding soil particles or past use of Hg as a fungicide on crop seeds; the agricultural landscape releases primarily total Hg that is subsequently methylated downgradient. In contrast, Chen et al.[32] found a negative coefficient for percent agriculture in northeastern USA lakes.

The significant positive influence of basin size and population density in MERGANSER may reflect enhanced Hg mobilization from landscape disturbance.[33] These results are seemingly at odds with a synthesis by Grigal,[34] who showed a decrease in stream Hg export per unit area with increasing basin size, primarily for undisturbed forested watersheds without lakes. Large New England lake basins tend to be relatively developed, possibly masking the relation found by Grigal.[34] Some large basins also contain reservoirs, where fluctuating water levels enhance methylation.[35]

The negative coefficient on mean annual air temperature in MERGANSER likely results from the growth dilution effect,[36] whereby higher fish growth rates in warmer waters effectively outpace the Hg accumulation rate, resulting in lower fish Hg. The negative coefficient on watershed slope arises because steep average slopes enhance landscape runoff efficiency and minimize landscape features that promote methylation (i.e., poor drainage, carbon accumulation, and anoxia, conditions prevalent in wetlands).

Comparison to Other Empirical Hg Models. Because of the expense of mercury analysis, and the lack of information on many lakes, a model that predicts fish Hg from easily derived spatial variables is useful for risk assessment. Like MERGANSER, many studies have followed an empirical approach to relate fish Hg to watershed features, and/or lake and soil chemistry, but the latter data are not always readily available. For example, Gabriel et al.[37] achieved a comparable r^2 to MERGANSER with only one variable: O-horizon soil total Hg concentration. But this was done for only 10 lakes on a single fish species and age (young-of-year yellow perch) within a small region in boreal Minnesota. In a different year, an even better model resulted with two variables: watershed area and lake water dissolved Fe. Also working in the boreal zone, Roué-LeGall et al.[38] found that watershed-to-lake-area ratio, percent wetland, and Hg deposition were sufficient to predict fish Hg. Simonin et al.[39] found that fish length, lake chemistry (water column Hg, pH, specific conductance, and chlorophyll *a*), presence of an outlet dam, and wetlands contiguous to the lake were the most important factors controlling fish Hg in New York lakes. Chen et al.,[32] analyzing four separate multiple lake studies from the northeastern USA, found that highest fish Hg generally occurred in low pH, low productivity lakes in forested landscapes. Kamman et al.[28] used a multivariate approach to model the likelihood that standardized yellow perch (*Perca flavescens*) would violate the EPA Hg criterion using alkalinity, pH, conductivity, and lake flushing rate, with a maximum misclassification error rate of 13%.

In an effort similar to MERGANSER for North Carolina, Sackett et al.[26] used a large database to predict fish Hg and found that fish species and lake trophic status, ecoregion, percent agricultural land, and water body type were the important drivers of fish Hg. Where they had water quality data, they expanded the model for those water bodies and obtained the highest r^2 (0.81) by the addition of pH only. Qian et al.[40] modeled fish Hg in four southeastern USA states and found that fish species and size, pH, and an unexplained "spatial variable" explained much of the variance in fish Hg.

Wente[41] modeled a USA data set of more than 30 000 fish Hg concentrations, with the objective of differentiating spatial/temporal variation from sample characteristic variation (e.g., fillets vs whole fish samples) in fish Hg concentrations. This model formulation acknowledges the inherent spatial/temporal variation but unlike MERGANSER, does not attempt to predict it. Krabbenhoft et al.[42] used watershed attributes, including topographic indices, to predict MeHg in the water column in USA-wide lakes and streams. A topographic approach also successfully explained streamwater Hg variations at a 65-km^2 catchment in the Adirondacks, NY.[43]

Model Transferability. We view MERGANSER as a predictive tool that is broadly applicable across the New England region. In contrast to other empirical Hg models,[26,37] we avoided use of water quality indices (except modeled alkalinity) that may improve predictive power but restrict applicability. As formulated, MERGANSER predicts length-specific Hg concentration for common fish species in New England lakes to within about a factor of 2. Also, the inclusion of fish-eating wildlife is unique among these models, and allows the user to encompass wildlife health impacts in addition to human health impacts from Hg exposure. Future improvements to MERGANSER should strive to incorporate predictors such as topographic indices, lake depth, and trophic status. Though MERGANSER was developed for New England, the predictors could be derived and applied to any region. We caution, however, that controls may shift in other regions, e.g., in the southeastern USA, where there are few natural lakes.

MERGANSER AS A POLICY AND RISK COMMUNICATION TOOL

MERGANSER has utility for local as well as regional/national scale managers and policymakers. At the local scale, managers can use MERGANSER to evaluate probabilities of mercury threshold exceedance for any New England lake, allowing for lake-specific fish consumption advisories (i.e., risk avoidance). MERGANSER can also identify lakes in New England that may not comply with the Northeast Regional Mercury Total Maximum Daily Load (TMDL), or point to additional lakes that should be addressed by the TMDL. For policymakers, the scenario test results suggest that reduced Hg deposition and/or forest cover (which are interrelated because forests scavenge atmospheric Hg) may reduce fish and loon Hg. While we applied the scenarios regionwide, one could use MERGANSER to evaluate the effects of Hg deposition reductions or land use shifts targeted to specific areas.

ASSOCIATED CONTENT

Supporting Information

A table with all predictors tested and statistics on their values and frequency distribution plots of the significant predictors, plots of model residual distributions, results of the validation run, and scenario testing maps of modeled fish Hg. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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