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Mercury in the Pelagic Food Web of Lake Champlain

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Abstract

Lake Champlain continues to experience mercury contamination resulting in public advisories to limit human consumption of top trophic level fish such as walleye. Prior research suggested that mercury levels in biota could be modified by differences in ecosystem productivity as well as mercury loadings. We investigated relationships between mercury in different trophic levels in Lake Champlain. We measured inorganic and methyl mercury in water, seston, and two size fractions of zooplankton from 13 sites representing a range of nutrient loading conditions and productivity. Biomass varied significantly across lake segments in all measured ecosystem compartments in response to significant differences in nutrient levels. Local environmental factors such as alkalinity influenced the partitioning of mercury between water and seston. Mercury incorporation into biota was influenced by the biomass and mercury content of different ecosystem strata. Pelagic fish tissue mercury was a function of fish length and the size of the mercury pool associated with large zooplankton. We used these observations to parameterize a model of mercury transfers in the Lake Champlain food web that accounts for ecosystem productivity effects. Simulations using the mercury trophic transfer model suggest that reductions of 25 to 75% in summertime dissolved epilimnetic total mercury will likely allow fish tissue mercury concentrations to drop to the target level of $0.3 \mu\text{g g}^{-1}$ in a 40-cm fish in all lake segments. Changes in nutrient loading and ecosystem productivity in eutrophic segments may delay any response to reduced dissolved mercury and may result in increases in fish tissue mercury.

Keywords

Lake Champlain; aquatic; mercury; zooplankton; fish

Introduction

Lake Champlain continues to experience mercury contamination problems resulting in elevated mercury levels in zooplankton and fish (Shanley et al. 1999; Kamman and Chen unpublished data) leading to public advisories to limit or curtail human consumption of top trophic level fish such as walleye. However, Lake Champlain and other large lakes do not fit the standard model of dissolved organic carbon (DOC)-rich, low pH lakes at high risk for elevated mercury in biota (Driscoll et al. 1994; Driscoll et al. 2007; Simonin et al. 2008). Lake Champlain exhibits lower water column total mercury (THg) (Gao et al. 2006) and lower methyl-mercury (MeHg) (Jackson et al. 2009) compared to smaller lakes with similar biotic mercury levels (see Kamman et al. 2004a, 2005). Part of the explanation for the difference between Lake Champlain and smaller lake systems may be Lake Champlain's long food chain lengths which result in efficient biomagnification of mercury level from primary producers to top-level consumers (Stemberger and Chen 1998) and the long life spans of top-level consumers in the Lake.

Food web chain length and long top-consumer life spans do not completely explain the similar top-trophic level mercury burdens under the highly contrasting biogeochemical conditions found across Lake Champlain segments. Mercury concentrations in biota at low trophic levels in Lake Champlain are comparable to those in smaller lakes, despite very low water column THg and MeHg levels. For example, the 63 to 202 μm size range zooplankton (typically herbivores or trophic level 2) in Lake Champlain (Shanley et al. 1999) exhibit similar ($410 \pm 110 \text{ ng g}^{-1}$) THg compared to the mean THg ($432 \pm 23 \text{ ng g}^{-1}$) for all zooplankton (including $> 202 \mu\text{m}$, herbivores and omnivores) from a spatially representative sampling of smaller VT and NH lakes (Kamman et al. 2004). The larger zooplankton in Lake Champlain ($> 202 \mu\text{m}$, omnivores) exhibited mercury concentrations of $760 \pm 20 \text{ ng g}^{-1}$ on the main lake (Shanley et al. 1999) and 1300 to 2300 ng g^{-1} in Malletts Bay (Chen et al. 2011). However, the mean surface water THg value for Lake Champlain was reported to be approximately 0.37 ng L^{-1} (Gao, et al. 2006) while the spatially representative mean value for small lakes in VT and NH was $1.78 \pm 0.05 \text{ ng L}^{-1}$ (Kamman et al. 2004a,b) and lakes in NY was $1.24 \pm 0.09 \text{ ng L}^{-1}$ (Simonin et al. 2008). Thus, the increased mercury bioavailability in Lake Champlain seems to arise at the base of the food web.

The similarity of biotic mercury levels and the contrast in pH, DOC, and surface water THg and MeHg levels between systems suggest a possible difference between Lake Champlain (and possibly other large lakes) and small lakes in the factors that govern mercury availability and uptake by lower-trophic level organisms. One important difference between the two classes of lake systems influencing mercury availability and uptake may be the relative importance of direct atmospheric deposition of THg and MeHg to the lake surface (Gao et al. 2006).

Planktonic bioaccumulation of Hg differs between eutrophic and oligotrophic lakes where plankton in more productive lakes appear to biodilute the Hg resulting in lower Hg concentrations than in oligotrophic lakes (Pickhardt et al. 2002; Chen and Folt 2005; Chen et al. 2005). Plankton blooms have also been shown to reduce aqueous concentrations of other metals (Chen and Folt 2000). Trophic status in Lake Champlain varies from oligotrophic (Mallets Bay) to eutrophic (Missisquoi Bay, South Lake) due to large spatial variation in nutrient inputs and the restricted circulation of certain lake segments. Therefore, we anticipated that ecosystem productivity as reflected in the biomass of different ecosystem compartments would influence mercury transfers between trophic strata.

The goals of this investigation were to develop an empirical mercury trophic transfer model functional over the broad range of environments present in Lake Champlain and to demonstrate how such a model could be used to evaluate the effects of nutrient and mercury reductions on fish tissue mercury levels of sport fish in the lake.

Study area and methods

Lake characteristics

Lake Champlain is a large (1127 km²) inland lake, 193 km long and 19 km maximum width with depths up to 122 meters (LCBP 2011). The lake has 5 major segments (Figure 1), two with restricted circulation due to constructed causeways. The drainage basin is 21,326 km² and contains varied land use including forested, agricultural, and urban areas. Contrasting land use across the basin in conjunction with limitations on circulation leads to a wide range in ecosystem productivity or trophic status across the basin (Table 1). Eutrophic conditions prevail in the agricultural and narrow southern lake and agriculturally dominated Missisquoi and St. Albans bays of the northern Lake. Mesotrophic conditions are found over much of the central lake. Oligotrophic conditions exist in restricted Mallets Bay.

Sampling design

We sampled water, seston, and zooplankton at 13 stations representing the full range of trophic conditions in the lake (Figure 1). Samples were collected between August 7 and September 11, 2009 during scheduled sampling of designated water quality monitoring stations conducted by the Vermont Agency of Natural Resources (VTANR) Lake Champlain Monitoring Program (LCMP). Five stations were sampled twice during the sampling period. The VTANR-LCMP conducted their standard water quality sampling at the same time as samples for mercury were collected. Water quality sampling included measurements of Secchi depth, lake thermal and oxygen profiles, collection of samples for Chl-a, nitrogen and phosphorus fractions, and samples for phytoplankton characterization. The detailed methods for sampling and analysis by the VTANR-LCM are provided elsewhere (VTDEC 2011).

Sampling for mercury analysis was conducted in triplicate for each component sampled. As water was always circulating under the sampling vessel due to wind-generated currents and because considerable handling time elapsed between samplings, the triplicate samples are considered to be true replicate samples for each station-day. Samples were collected off the

windward bow rail of the fiberglass research vessel such that current carried water to the sampler before any interaction with the vessel.

Water grab samples for mercury speciation were collected at approximately 20 cm depth in the epilimnion using the EPA “clean hands / dirty hands” method directly into pristine 2 L PTEG sampling vessels (see Jackson et al. 2009). Samples were triple bagged and placed immediately in the dark on ice in a cooler for shipment to the laboratory. Separate 2 L grab samples were collected in PTEG bottles for gravimetric analysis of seston mass per L. These samples were also stored in the dark on ice. Filtered samples were collected from the filtrate of the VTANR Chl-a sampling apparatus (0.2 μm Teflon filter) into precombusted 60 ml amber glass bottles and stored in the dark on ice for later measurement of DOC and absorbance at 254 nm.

Zooplankton samples in two size fractions for mercury speciation and biomass analyses were collected following the methods of Chen et al. (2000) with some modifications described here. Multiple tows using a bongo net frame with 45 and 202 μm nets were conducted until enough sample was obtained for analysis in each size fraction. Vertical tows were conducted from 1 meter off the bottom to the surface and length and number of tows recorded. Due to shallow water depth in Missisquoi Bay, horizontal plankton tows were taken while tow length was recorded with a GPS. Water volumes sampled for zooplankton were calculated from the net opening diameter and tow length (Stemberger and Chen 1998). Material in the net cod ends was rinsed through acid-washed and rinsed filters using ultraclean water after each tow. The contents of the 45 μm net were filtered through a 202 μm filter (with > 202 μm material discarded) to generate a sample representing the 45 to 202 μm size range. Material collected from the 202 μm net represented the > 202 μm size range. Samples were rinsed into 250 ml pristine PTEG sample containers, triple bagged, and placed on ice in the dark for transport to the laboratory.

The field sampling personnel transported samples on ice directly to the Dartmouth College Trace Element Analytical Facility laboratory within 48 hours of collection. Previous studies (Jackson et al. 2009) identified a 48-hour hold time as acceptable to maintain mercury speciation in Lake Champlain waters. The 2 L water samples were immediately filtered using precombusted Whatman™ GF/F filters to operationally separate seston from “dissolved” mercury. Filter blanks were run with each set of samples. Ionic mercury and methyl-mercury were determined in both the filtrate and particulate (seston) fractions following the ultra low level method described in Jackson et al. (2009). Briefly, the method involved species-specific isotope dilution with purge and trap preconcentration, gas chromatographic species-separation of MeHg and Hg^{2+} , and high-resolution inductively coupled plasma mass spectrometry isotope quantitation. Method detection limits were 0.003 ng L^{-1} for MeHg, and 0.074 ng L^{-1} for Hg^{2+} . The 2 L samples collected for seston mass determinations were filtered in the same manner as the mercury speciation samples with the exception that the filters were weighed pre and post filtration (after drying).

Zooplankton samples were processed at Dartmouth. Zooplankton samples were transferred to pre-weighed trace metal clean glass vials, weighed, freeze dried, and reweighed to determine the dry weight of each samples. Freeze dried samples were sent to the Dartmouth

Trace Element Analytical Facility for Hg speciation measurements using isotope dilution methods following Jackson et al. (2009) and Chen et al. (2011). Briefly, zooplankton samples were spiked with appropriate amounts of mercury containing enriched values of Hg^{199} and MeHg^{201} . The spiked samples were extracted by alkaline extraction. Isotopic analysis was conducted by high-resolution inductively coupled plasma mass spectrometry. Standard reference materials were recovered at 102% (MeHg) and 101% (total Hg) for NIST 2976 mussel ($n = 11$), and 120% (MeHg) and 118% (total Hg) for TORT-2NRC-CNRC ($n = 1$).

Samples collected for measurement of DOC and specific absorbance at 254 nm were shipped on ice to the University of New Hampshire Water Resources Research Center. Samples were held in the dark at 4°C for up to 4 weeks prior to analysis. Samples were analyzed for DOC using a Shimadzu™ TOCV-CH and EPA Method 415.1. Specific absorbance at 254 nm was measured using a Thermo Genesys™ 10uv spectrophotometer with a 1 cm cell.

Pelagic fish American eel (*Anguilla rostrata*), Atlantic salmon (*Salmo salar*), brown bullhead (*Ictalurus nebulosus*), lake trout (*Salvelinus namaycush*), northern pike (*Esox lucius*), smallmouth bass (*Micropterus dolomieu*), walleye (*Sander vitreus*), white perch (*Morone americana*), and yellow perch (*Perca flavescens*) were sampled opportunistically from 1987 to 2002 by VTANR from all segments of Lake Champlain for the purpose of establishing fish tissue mercury levels in different species and locations in the lake. The location of catch was recorded and assigned to the nearest water quality sampling station, fish species was identified and recorded, and length was measured. Fish were transported on ice to the VTANR laboratory for THg analysis following the methods described in Kamman et al. (2004 a, b). Mean recovery of a standard perch sample was 92.9% ($n = 37$). Mean relative percent difference for a standard perch sample was 5.7% ($n = 265$). Sediment standard reference materials certified at 0.72 ppm and 1.12 ppm were recovered at 101% ($n = 14$) and 1.07% ($n = 17$), respectively (see Kamman et al. 2004b). As sampling of pelagic fish was beyond the scope of our project budget, after testing for temporal variation using analysis of covariance (ANCOVA), we used the existing VTANR fish mercury data to assess lake-segment to lake-segment differences in fish tissue mercury and to relate fish tissue mercury to the mercury content of other ecosystem compartments. Due to the long life spans of the sampled fish relative to zooplankton and phytoplankton sampled in the present study and the recent temporal stability of fish tissue mercury levels in the region (see Chalmers et al. 2010) these data were suitable for establishing temporally stable associations of fish tissue mercury with other lake-segment specific data obtained in this study. As some of the sampled fish species (American eel, Atlantic salmon, brown bullhead, white perch, and yellow perch) were either not spatially well-represented or were not relevant to our sport fish modeling target they were not included the general linear model (GLM) for predicting fish tissue mercury concentrations. A total of 157 fish samples were used to calculate means representing 9 of the 13 lake segments for GLM development of the fish tissue mercury prediction equation component of the overall trophic transfer model. Three lake segments (Cumberland Bay, Isle Lamotte off Grand Isle, and Burlington Bay) lacked sufficient

numbers of representative fish species to calculate means. The Northeast Arm (8 additional fish samples) could not be included due to missing large zooplankton data.

Theory / calculation

Previous studies (e.g. Chen and Folt 2005; Chen et al. 2005; Stemberger and Chen 1998) established that the mercury concentration in pelagic fish tissue is related to the composition of the zooplankton community, nutrient status, the primary productivity (Pickhardt et al. 2002), and food quality in the system (Karimi et al. 2007), mercury concentration in water (Simonin et al. 2008) as well as mercury wet deposition (Hammerschmidt et al. 2006). These relationships have been derived both experimentally and from surveys of multiple upland lakes with varying lake characteristics. We hypothesized that differences in mercury loading rates, food web information, and ecosystem productivity between lake segments would be predictive of zooplankton and fish mercury in Lake Champlain.

We also hypothesized that measurement and calculation of the mercury content of ecosystem compartments (e.g. the pool of Hg in the water column associated with a zooplankton fraction, or seston) would be more helpful in establishing mercury trophic transfer relationships than simple expression of the average mercury concentration (on a dry weight basis) of a sample representing an ecosystem compartment. We anticipated that pool sizes would be important because zooplankton biomass is strongly influenced by primary productivity (Masson et al. 2004), which varies significantly across Lake Champlain. To express mercury content of different ecosystem compartments as pools we measured both the concentration of mercury in a given media and the biomass or mass of the media per liter of lake water. Aquatic ecosystem biomass estimates are challenging due to the 3-dimensional spatial distributions and variability of different ecosystem components such as phytoplankton, zooplankton, and fish. In this study, the biomass-based expressions of mercury pools associated with different ecosystem components are treated as indicator metrics for the size of the pools rather than absolute measures of the size of the pools. As demonstrated below, these measures effectively capture information on the interaction of ecosystem components with mercury sources and sinks in the system.

To evaluate if the different sub-basins of Lake Champlain with contrasting mercury loading rates and/or trophic status behaved like contrasting small lakes, we tested for significant differences in the mercury concentration or content of different ecosystem compartments across lake sampling stations using ANOVA. For some ecosystem compartments such as pelagic fish, testing for differences between locations required an ANCOVA model taking into account the known dependence of fish tissue mercury content on fish length (a common proxy for age and the length of time a fish has been accumulating mercury) and species (differences in dietary preferences and metabolism).

A primary goal of the study was to construct an empirical model of mercury trophic transfers in Lake Champlain. To build this model we systematically explored possible relationships between mercury levels in different ecosystem compartments expressed as average dry mass concentrations or as compartment pools ($\mu\text{g L}^{-1}$ lake water) using ANCOVA and general linear modeling (GLM). We developed best-fit models relating specific trophic level mercury burdens to food web and lake environment parameters as well

as the mercury content of lower trophic levels and media. Mercury was evaluated as methylmercury, ionic mercury, and total mercury in each compartment to determine which mercury measure produced the best trophic transfer model.

JMP version 6.0.3 (SAS Institute) was used for all statistical analysis.

Results

Species, spatial, and temporal variation

All measured lake characteristics were significantly different across lake segments (Table 1). The eutrophic South Lake A (station 4) and Missisquoi Bay (station 51) segments were frequently significantly different from other stations in most parameters as determined by Tukey-Kramer HSD comparisons of all means at the $p = 0.1$ to $p = 0.01$ levels. These two segments had higher total P, total N, Chl-a, DOC, terrestrial source DOC (absorbance at 254 nm), seston mass, and lower Secchi depths than other segments. These segments also had significantly higher methyl and total mercury in the dissolved and particulate fractions than other segments. One difference between the two eutrophic segments was that the South Lake A station exhibited higher Gran alkalinity (70 mg L^{-1}) compared to Missisquoi Bay (40.6 mg/L). Although this difference could not be assessed for statistical significance, as only one sample per station was available during the time-window we analyzed in this study, the alkalinity differences between segments are known to be temporally stable from long-term monitoring observations (see VTDEC 2011b) and reflect differences in geology of the tributary watersheds.

A geographic cluster of stations, Mallets Bay, Cumberland Bay, and Isle Lamotte (off Grand Isle), had significantly lower total dissolved mercury concentrations than other segments. The Northeast Arm and Isle Lamotte (off Rouses Point) exhibited significantly lower particulate mercury than other segments.

Despite a lack of significant pairwise differences in most measured parameters between most mesotrophic segments (Table 1), there were significant correlations between parameters across the range of variance observed (see GLM results below).

Zooplankton biomass and mercury content varied significantly between lake segments (Table 2). Mean segment small zooplankton (45 to $202 \mu\text{m}$) biomass ranged from $3.42 \mu\text{g L}^{-1}$ to $52.77 \mu\text{g L}^{-1}$ and was significantly higher in the eutrophic lake segments (South Lake A and Missisquoi Bay). Mean segment large zooplankton ($> 202 \mu\text{m}$) biomass ranged from $0.87 \mu\text{g L}^{-1}$ to $23.00 \mu\text{g L}^{-1}$ yet was only significantly higher in Missisquoi Bay than in other lake segments due to high variance within segments. Isle Lamotte (Rouses Point) had the highest total zooplankton biomass ($53.7 \mu\text{g L}^{-1}$), due to very high small zooplankton biomass, yet also had the lowest large zooplankton biomass. The eutrophic segments South Lake and Missisquoi Bay had total zooplankton biomass values of 29.2 and $27.6 \mu\text{g L}^{-1}$, respectively. Mallets Bay had the lowest total zooplankton biomass ($9.27 \mu\text{g L}^{-1}$) with biomass evenly distributed between large and small size fractions.

The small zooplankton THg pool ranged from 0.05 pg L⁻¹ to 1.23 pg L⁻¹. The small zooplankton THg pool in Missisquoi Bay was significantly larger than in all other segments. The small zooplankton THg pool at the South Lake A station was significantly larger than all other segments except Missisquoi Bay. The large zooplankton THg pool ranged from 0.05 pg L⁻¹ to 1.33 pg L⁻¹ with Shelburne Bay exhibiting a significantly higher pool size than other stations and Isle Lamotte (off Rouses Point) exhibiting a significantly lower pool size than other stations.

Pelagic fish sampled (American eel, Atlantic salmon, brown bullhead, lake trout, northern pike, smallmouth bass, walleye, white perch, and yellow perch) ranged in length from 150 to 863 mm, averaging 447 mm. The mean fish tissue mercury concentration was 0.48 µg g⁻¹ (wet weight). Sixty-three percent of fish sampled had mercury concentrations above the EPA human health advisory criteria of 0.3 µg g⁻¹ (wet weight). An ANCOVA model using length as the covariate and species as the main effect explained 77% of the variance in fish tissue mercury concentration ($p < 0.0001$). There was no significant length by species interaction. Length alone explains 37% of the variance in fish tissue mercury concentration ($p < 0.0001$). Adding a sampling year effect to the ANCOVA model indicated that only 2.3% of total variance in fish tissue Hg was attributable to temporal variance. This small signal was driven by a single year. Only the 1995 sample year effect was significant ($p = 0.0033$). This was driven by an emphasis on sampling Shelburne Bay in 1995 and considered to be more of a spatial effect than a true temporal effect. Catches of the pelagic fish species represented were not evenly distributed by lake segment (Chisquare test, $p < 0.0001$). Length and lake segment explained 53% of the variance in fish tissue mercury ($p < 0.0001$), indicating that there were significant differences in fish tissue mercury concentrations by lake segment.

Because replicate measurements of different ecosystem compartments (e.g. water, Chl-a, zooplankton, fish) were not paired observations (meaning zooplankton replicate A could not be directly attributable to water sample replicate A), means by lake segment were calculated to facilitate general linear modeling of the relationships between different ecosystem compartments and lake characteristics. As there were significant differences between some parameters at different (repeat) sampling times at the same sampling station the empirical modeling discussed below makes use of both the spatial and temporal variation of lake characteristics to predict trophic transfers between ecosystem compartments. In some GLMs presented below there are up to 18 spatially or temporally differentiated coincident observations that may be used in modeling. In other cases only 9 appropriate coincident observations were available due to missing data for one or more of the components.

Predictive relationships between ecosystem compartments

Mean fish length and the total water column pool of THg associated with large (> 202 µm) zooplankton (definition of water column pool: g biomass zooplankton L⁻¹ x pg THg g⁻¹ zooplankton = pg zooplankton THg L⁻¹) explained 94% of the variance in mean fish tissue mercury concentration by lake segment ($p = 0.0008$, $n = 8$, Figure 2). Mallets Bay was a positive outlier to this trend (Figure 2).

The variance in the large zooplankton THg pool was best explained ($r^2 = 0.72$, $p = 0.0076$, $n = 12$) by a model that included the size of the small zooplankton THg pool ($p = 0.0039$), seston biomass ($p = 0.0049$), and the large zooplankton biomass ($p = 0.0143$). Large zooplankton MeHg concentration (pg g^{-1} dry weight) was highly correlated with large zooplankton THg concentration ($r^2 = 0.96$) with a slope of 0.73 and offset of -13.5 ($p < 0.001$) for 10 of the 12 lake segments. The Isle La Motte and Shelburne Bay segments were outliers to the trend, but fell on a line of similar slope (0.65) with a more negative offset (-30.4). Expressed on a dry-weight basis the correlation between MeHg and THg remained robust ($r^2 = 0.71$).

The variance in the small zooplankton THg pool was best explained ($r^2 = 0.97$, $p < 0.0001$, $n = 13$) by a model including small zooplankton biomass ($p = 0.0001$), the seston THg pool ($p = 0.0243$), and an interaction between the seston THg pool and the small zooplankton biomass ($p = 0.0064$).

The partitioning of THg between the dissolved pool and the seston pool was well described ($r^2 = 0.84$, $p < 0.0001$, $n = 18$) by a model including the mean seston mass ($p = 0.0001$), the dissolved THg pool ($p = 0.0691$), and the lake water Gran alkalinity ($p = 0.0102$).

To facilitate trophic-transfer modeling, and potentially reduce the cost of future surveys, predictive relationships for ecosystem compartment mass and biomass were explored. Large zooplankton biomass was reasonably well predicted ($r^2 = 0.55$, $p = 0.0058$, $n = 16$) by the sum of seston mass and small zooplankton biomass ($p = 0.0027$) and the water absorbance at 254 nm ($p = 0.0085$). Small zooplankton biomass was well predicted ($r^2 = 0.84$, $p < 0.0001$, $n = 16$) by cyanobacteria biovolume (from VTANR-LCMP) ($p = 0.0343$) and water absorbance at 254 nm ($p = 0.0271$). A simpler model using only seston mass ($p < 0.0001$) was an equally good predictor of small zooplankton biomass ($r^2 = 0.84$, $p < 0.0001$, $n = 16$). Cyanobacteria biovolume ($p = 0.0354$) and water absorbance at 254 nm ($p < 0.0001$) were also good predictors of seston mass ($r^2 = 0.93$, $p < 0.0001$, $n = 17$). Seston mass was well correlated with total phosphorus ($r^2 = 0.84$, $p < 0.0001$, $n = 16$). A model for seston mass as a function of Secchi depth ($r^2 = 0.85$, $p < 0.0001$, $n = 18$) was also obtained.

Discussion

With fish tissue mercury concentrations in nearly two-thirds of the sampled pelagic fish exceeding the EPA advisory criterion of $0.3 \mu\text{g g}^{-1}$ wet weight it is imperative to develop a working model of mercury trophic transfer in the Lake Champlain ecosystem. The empirical trophic transfer model we present here (Figure 3) allows inference about the effects of air and water management options on the mercury levels in fish. Below we discuss some of the limitations of the data set and the resulting model. We describe the model structure and employ the model to evaluate the effects of some potential management scenarios.

As anticipated due to the strong contrasts in lake environment (e.g. depths) between lake segments, catches of the pelagic fish species represented were not evenly distributed by lake segment. For example, northern pike catches were distributed evenly across all segments with the exception of the Isle La Motte segment and the Northeast Arm. In contrast, Atlantic

salmon was caught only in the Isle La Motte and Northeast Arm segments, but in no other segments. Because the sampling efforts conducted by VTANR for these fish were opportunistic, differences in sampled species representing each lake segment may or may not represent the likely pelagic fish populations of each segment. Lacking better information, we assume for the purposes of exploratory analysis and for developing a first-generation trophic transfer model that the opportunistic catches are generally representative of the different fish populations of the lake segments. Therefore, we omit consideration of fish species as a covariate for the remainder of the analysis. This assumption has the impact of embedding in the model the opportunistic fish sampling species distribution as an intrinsic lake segment environment characteristic.

Controlled for fish length, there were significant differences in fish tissue mercury concentration by segment. Part of the observed difference between lake segments is likely due to a difference between intrinsic mercury accumulation rates of the different fish communities representing each lake segment and part to due to other environmental characteristics (e.g. mercury loading, food-web, productivity) of the different lake segments. Given the nature of the fish sample it is equally possible that the significant “species effect” apparent in the ANCOVA is due to the lake segment environmental characteristics associated with a particular fish species habitat in the sample. The alternative causal factors (fish species or lake environment) cannot be disentangled using ANCOVA. However, general linear modeling (described below, see also Table 1) suggests that lake environment characteristics are important contributors to differences in fish tissue mercury concentration across lake segments, and this interpretation is supported by literature from smaller lake studies (Kamman et al. 2004; Chen et al. 2005; Driscoll et al. 2007; Simonin et al. 2008).

Productivity at the food-web base varied substantially across lake segments as did the proportion of seston mass pools attributable to autotrophs and heterotrophs. Seston mass was a function of Chl-a, terrestrial-source DOC, and cyanobacteria biovolume except in Missisquoi bay where cyanobacteria dominated during the sampling period. The partitioning of the water column mercury pool into seston increased with seston mass and decreasing water alkalinity. However, the concentration of mercury in seston decreased with increasing seston mass (bloom dilution). The biomass of small zooplankton (herbivores and detritivores) correlated with cyanobacteria biovolume and an index of terrestrial dissolved carbon (absorbance at 254 nm) suggesting that heterotroph biomass is important to the base of the food web in many lake segments. The biomass of large zooplankton (omnivores) was positively correlated with seston and small zooplankton mass and negatively correlated with dissolved terrestrial carbon suggesting differences in food quality between lake segments. Mercury in small zooplankton was a function of the mercury content of water and seston and showed dilution in relation to the amount of small zooplankton biomass. Mercury in large zooplankton was a complex function of the mercury content in seston and small zooplankton pools as well as an interaction between large and small zooplankton biomass. Mercury concentrations in top trophic level fish were highly correlated with the standing stock of mercury in large zooplankton (Figure 2), emphasizing the importance of zooplankton as a key guild in controlling bioaccumulation to fish. Fish from Mallets bay were outliers (exhibiting higher fish-tissue mercury relative to the standing stock of zooplankton mercury) from the lake-wide trend. This departure may be explained in part by the oligotrophic status

(e.g. low productivity, lack of bloom dilution) of Mallets Bay, and in part to potential differences in preyfish between Mallets Bay and other lake segments.

The best-fit functions for predictions of ecosystem compartment mercury levels (expressed as the pool of mercury in a compartment) were combined with the mass and biomass prediction functions to form an empirical mercury trophic transfer model applicable across all environments of Lake Champlain (Figure 3, Table 3). The model (Figure 3, Table 3) quantifies the response in fish-tissue mercury concentrations to changes in the primary productivity and dissolved total mercury in the water column. Predictions of future primary productivity and dissolved total mercury can be obtained from independent models (e.g. Gao et al. 2006).

We found that it was necessary to build the trophic transfer model relating the mercury content of ecosystem compartments as “pools” rather than concentrations in order to obtain statistically significant predictive functions. This confirmed our hypothesis that biomass information (effect of biodilution, see Pickhardt et al. 2002; Chen and Folt 2005) would be helpful to effectively describe trophic transfer of mercury. Biomass dilution effects were evident at all trophic levels where they could be evaluated (all levels except fish). The best-fit predictive relationships also conformed to our expectations of the relationships between trophic levels (e.g. Chen and Folt 2005) and pathways of energy and mercury transfer (e.g. Stemberger and Chen 1998; Chen et al. 2005).

Total mercury contents (inorganic + methyl mercury) of water, seston, and lower trophic levels were more highly correlated and provided better fits in multi-parameter models than methyl-mercury contents. Simonin et al. (2008) found that fish tissue mercury was marginally better correlated with total mercury than methyl mercury in water for smallmouth bass, largemouth bass, and yellow perch in New York. In contrast, Simonin et al. (2008) found that walleye fish tissue mercury was best correlated with water column methyl mercury. Orihel et al. (2008) demonstrated that Hg^{2+} was the limiting factor for bioavailability of mercury in lake mesocosms using isotopically spiked Hg^{2+} .

Dissolved total mercury concentrations and methyl mercury concentrations are much lower in Lake Champlain than in most smaller northeastern lakes. Methyl mercury and THg were highly correlated in large zooplankton sampled from Lake Champlain, while there tends to be no correlation between mercury forms in large zooplankton sampled from smaller lakes (Ward and Chen, unpublished data). These observations suggest there may be a tighter coupling between inorganic and bioavailable mercury in low-DOC, low-mercury waters such as Lake Champlain compared to upland lakes. These large-lake observations are also consistent with the results of mesocosm studies using Hg isotopic tracers by Orihel et al. (2006) and Orihel et al. (2008) that demonstrated inorganic mercury loading rates governed bioavailability and food web accumulation of mercury.

Using the trophic transfer model we estimated the concentration of epilimnetic total dissolved mercury required to achieve the $0.3 \mu\text{g g}^{-1}$ level in a 40-cm fish in each of the modeled segments in Lake Champlain (Table 4). In the opportunistic pelagic fish sampling the mean size fish caught was 44.7 cm. A 40-cm length fish is on the smaller end of a sport-

fishing catch on Lake Champlain. The minimum legal length for lake trout and landlocked salmon is 38.1 cm. The minimum legal length for walleye is 45.7 cm and for northern pike it is 50.8 cm. Therefore, average fish tissue mercury concentrations in fish taken of these species would be higher than indicated by our 40-cm model fish. With no change in ecosystem productivity, reductions in dissolved THg of 25–75% appear necessary to reduce 40-cm fish tissue concentrations to $0.3 \mu\text{g g}^{-1}$ (Scenario 2, Table 4) in the segments currently exceeding the advisory level. The implied required reductions in dissolved mercury concentrations can be used with the model of Gao et al. (2006) to infer possible mixtures of loading reductions (direct atmospheric, wastewater, tributary) that could be adopted to achieve the fish tissue concentration goal.

Lake Champlain has been receiving excessive phosphorus from agricultural and urban runoff and wastewater discharges for many decades, although wastewater loads have declined sharply since 1990. The accumulation of phosphorus has resulted in increased primary productivity as reflected in a reduction in Secchi depth in some segments of the lake such as Missisquoi Bay. Secchi depths have increased in the South Lake and other segments in recent years due to filtration by zebra mussels and/or phosphorus load reductions. Programs are in place that are designed to reduce phosphorus loadings and are expected to result in lower primary productivity and increased Secchi depths. Model scenario 3 in Table 4 shows the possible effect on fish tissue mercury concentrations in response to hypothetical scenarios of reduced phosphorus and improved Secchi depths across the lake.

If dissolved mercury levels are held constant, the hypothetical reduction of primary productivity generally leads to increased fish-tissue mercury concentrations (Scenario 3, Table 4). Fish tissue mercury is predicted to increase as the effect of biomass dilution is initially weakened; yet productivity remains high enough for relatively large pools of mercury to be maintained in zooplankton and transferred to fish (Table 4). If productivity falls low enough in formerly nutrient rich segments, the pool of mercury in zooplankton becomes small enough to result in decreasing fish tissue mercury (Table 4). Presumably this may occur as predators are forced to utilize a mix of prey including lower trophic strata to meet energetic requirements. As productivity decreases to very low levels (e.g. conditions in Mallets Bay) fish tissue mercury for a given length fish is likely to increase relative to mesotrophic conditions as growth rates slow due to decreasing food availability.

The integrated effect of concurrent reductions in both dissolved mercury and productivity will likely be to delay achievement of the target fish tissue mercury level and to increase the required dissolved mercury reduction (Scenario 4, Table 4) as the system adjusts to decreased productivity levels in the more nutrient rich segments. Any segments achieving oligotrophy (e.g. Mallets Bay) as a result of declining productivity will likely require greater mercury load reductions relative to mesotrophic segments to achieve desired fish tissue concentration reductions.

Summary and conclusions

Pelagic fish of similar size exhibited differences in tissue mercury concentrations in different lake segments of Lake Champlain. Differences in fish tissue mercury appear to be

due to a combination of environmental and food-web factors as well as lake-water dissolved mercury levels. Best-fit empirical trophic transfer functions took forms generally consistent with the hypothesized movement of mercury through the Lake Champlain food web. One notable exception was the finding that total dissolved mercury was a better predictor of biotic mercury levels than dissolved methyl mercury. Scenario testing with the empirical trophic transfer model suggested that fish-tissue mercury burdens would likely initially rise in mesotrophic and eutrophic segments in response to reductions in primary productivity expected as a response to anticipated reductions in nutrient loadings. As productivity levels fall toward mild mesotrophic status or lower, fish tissue mercury concentrations are anticipated to decline from peak levels. The trophic transfer functions suggest that fish tissue mercury should be responsive to reductions in mercury loadings and dissolved mercury. In the absence of productivity changes, reducing dissolved total mercury by 25 to 75% would likely achieve a management goal of reducing the tissue mercury concentration of 40-cm sport fish below $0.3 \mu\text{g g}^{-1}$ wet weight in the lake segments currently exceeding this criterion. Greater reductions would be required to bring the average fish tissue mercury of legal length walleye and northern pike below $0.3 \mu\text{g g}^{-1}$ wet weight.

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Fig. 1.

Location of lake-segments sampled on Lake Champlain. Lake Champlain is situated primarily within the United States on the New York and Vermont border, with the northern end of the lake extending into Québec Province, Canada. Missisquoi Bay (north east end) and the South Lake (southern end) segments are the most eutrophic waters. Mallets Bay (east central) contains the most oligotrophic waters in the lake system.

Fig. 2.

General linear model for (a) fish tissue mercury in $\mu\text{g g}^{-1}$ wet weight as a function of (b) fish length in $\text{mm} \times 0.0022093 \pm 0.000244$ ($p = 0.0003$) and (c) the pool of total mercury associated with large ($> 202 \mu\text{m}$) zooplankton in $\text{pg L}^{-1} \times 0.3857262 \pm 0.091475$ ($p = 0.0084$), intercept = -0.830067 ± 0.153292 ($p = 0.0029$), overall $p = 0.0008$, $n = 8$, adjusted $r^2 = 0.94$). Malletts Bay (open circle in panel a) is an outlier. The fish tissue mercury concentration in Malletts Bay is estimated from the ratio of the observed to predicted value from the general model (see Table 3).

Fig. 3.

Schematic diagram of the empirical mercury trophic-transfer model developed for the pelagic ecosystem of Lake Champlain detailed in table 3. As described in the text and table 3, Secchi depth serves as proxy for primary productivity. The specific absorbance at 254 nm is related to the balance between algal vs. terrestrial sources of DOC and is thus modified by primary productivity. The dissolved total mercury (DissTHg) in the system may vary with changes in atmospheric deposition and tributary loading. Piscivore tissue mercury (PiscivoreHg ppm) is a function of fish length, primary productivity and dissolved mercury levels as modulated by the biomass dynamics of the zooplankton community.

Table 1

Location of sampling stations, means and standard errors of environmental characteristics of Lake Champlain by lake-segment sampling station.

Station Number	4	7	9	16	19	21	25	33	34	36	40	46	51
Latitude (North)	43 ° 57.10'	44 ° 07.56'	44 ° 14.53'	44 ° 25.55'	44 ° 28.26'	44 ° 28.49'	44 ° 34.92'	44 ° 42.07'	44 ° 42.49'	44 ° 45.37'	44 ° 47.12'	44 ° 56.90'	45 ° 02.50'
Longitude (West)	73 ° 24.47'	73 ° 24.77'	73 ° 19.75'	73 ° 13.92'	73 ° 17.95'	73 ° 13.90'	73 ° 16.87'	73 ° 25.09'	73 ° 13.61'	73 ° 21.30'	73 ° 09.73'	73 ° 20.40'	73 ° 07.78'
Station Name	South Lake A	Port Henry	Otter Creek	Shelburne Bay	Main Lake	Burlington Bay	Malletts Bay	Cumberland Bay	Northeast Arm	Grand Isle	St. Albans Bay	Rouses Pt	Missisquoi Bay
Alkalinity ¹ (mg L ⁻¹)	70	58.7	55.8	52.2	50.7	51.5	35.8	50.4	46.5	50.2	48.6	48.6	40.6
DOC (mg C L ⁻¹)	4.33 ± 0.12	3.31 ± 0.05	3.27 ± 0.04	3.02 ± 0.01	3.15 ± 0.09	3.26 ± 0.05	3.41 ± 0.12	3.17 ± 0.05	3.48 ± 0.09	2.93 ± 0.05	3.53 ± 0.04	2.96 ± 0.03	4.60 ± 0.15
Means Group ² , n ³	A n = 3	BCD n = 9	BCD n = 6	CD n = 3	BCD n = 6	BCD n = 3	BC n = 6	BCD n = 3	B n = 6	D n = 3	B n = 3	D n = 3	A n = 3
Specific Absorbance (abs@254nm)	0.130 ± 0.000	0.089 ± 0.002	0.092 ± 0.002	0.080 ± 0.000	0.083 ± 0.002	0.080 ± 0.000	0.088 ± 0.002	0.097 ± 0.003	0.070 ± 0.000	0.083 ± 0.003	0.080 ± 0.000	0.080 ± 0.000	0.140 ± 0.000
Means Group, n	A n = 3	BCD n = 9	BC n = 6	E n = 3	DE n = 6	E n = 3	BCDE n = 6	B n = 3	F n = 6	CDE n = 3	E n = 3	E n = 3	A n = 3
Total-Nitrogen (mg L ⁻¹)	0.408 ± 0.027	0.328 ± 0.005	0.32 ± 0.023	0.318 ± 0.004	0.306 ± 0.011	0.316 ± 0.007	0.288 ± 0.013	0.315 ± 0.021	0.258 ± 0.005	0.288 ± 0.006	0.315 ± 0.018	0.293 ± 0.009	0.483 ± 0.018
Means Group, n	B n = 5	C n = 4	C n = 4	C n = 5	C n = 5	C n = 5	C n = 6	C n = 4	C n = 4	C n = 4	C n = 4	C n = 5	A n = 5
Total-Phosphorus (µg L ⁻¹)	53.5 ± 4.7	18.6 ± 0.6	17.3 ± 1.8	13.6 ± 1.0	11.8 ± 0.5	13.4 ± 1.1	10.2 ± 0.4	14.5 ± 2.1	15.5 ± 0.6	13.9 ± 1.0	27.7 ± 2.1	20.7 ± 0.4	61.4 ± 6.6
Means Group, n	A n = 5	BC n = 4	BC n = 4	C n = 5	C n = 5	C n = 5	C n = 6	C n = 4	BC n = 4	C n = 4	B n = 4	BC n = 5	A n = 5
Chlorophyll-a (mg L ⁻¹)	10.98 ± 2.46	5.23 ± 1.32	5.36 ± 1.63	2.79 ± 0.67	2.56 ± 0.48	3.42 ± 0.60	3.47 ± 0.45	3.73 ± 0.85	3.42 ± 0.79	2.66 ± 0.40	6.17 ± 2.73	2.76 ± 0.38	7.83 ± 2.07
Means Group, n	A n = 5	AB n = 4	AB n = 4	B n = 5	B n = 5	B n = 5	B n = 6	B n = 4	B n = 4	B n = 4	AB n = 4	B n = 5	AB n = 5
Secchi-Depth (m)	1.4 ± 0.1	3.6 ± 0.5	3.8 ± 0.4	5.5 ± 0.4	6 ± 0.5	5.4 ± 0.3	5 ± 0.4	4.2 ± 0.4	5.2 ± 0.2	5.4 ± 0.1	3.8 ± 0.3	5.4 ± 0.2	1.2 ± 0.2
Means Group, n	E n = 5	D n = 4	CD n = 4	AB n = 5	A n = 5	AB n = 5	ABCD n = 6	BCD n = 4	ABCD n = 4	ABC n = 4	CD n = 4	AB n = 5	E n = 5
TSS (mg L ⁻¹)	4.55 ± 0.04	1.12 ± 0.13	1.62 ± 0.16	0.84 ± 0.03	0.85 ± 0.05	0.93 ± 0.04	0.95 ± 0.07	0.63 ± 0.04	0.79 ± 0.08	0.48 ± 0.15	1.17 ± 0.07	0.25 ± 0.08	5.33 ± 0.12
Means Group, n	B n = 3	D n = 9	C n = 6	DEF n = 3	DE n = 6	DE n = 3	DE n = 6	DEF n = 3	DEF n = 6	EF n = 3	CD n = 3	F n = 3	A n = 3
Dissolved ⁴ -MeHg (pg L ⁻¹)	32.83 ± 0.77	14.85 ± 0.77	15.36 ± 1.40	12.55 ± 0.20	12.17 ± 0.60	9.84 ± 0.14	14.63 ± 0.53	18.48 ± 1.02	7.12 ± 1.53	12.85 ± 1.65	14.33 ± 1.18	14.07 ± 0.17	17.08 ± 0.50
Means Group, n	A n = 3	BCD n = 9	BC n = 6	BCDE n = 3	CD n = 6	DE n = 3	BCD n = 6	B n = 3	E n = 6	BCD n = 3	BCD n = 3	BCD n = 3	BC n = 3
Dissolved ⁴ -Hg ²⁺ (pg L ⁻¹)	285.3 ± 4.1	401.8 ± 28.9	394.3 ± 59.2	304.1 ± 3.6	268.9 ± 5.3	276.0 ± 9.0	212.5 ± 20.1	187.4 ± 7.7	333.0 ± 53.1	164.8 ± 4.7	409.4 ± 26.6	298.1 ± 7.0	429.5 ± 4.5

Station Number	4	7	9	16	19	21	25	33	34	36	40	46	51
Means Group, n	ABC n = 3	A n = 9	AB n = 6	ABC n = 3	BC n = 6	ABC n = 3	C n = 6	C n = 3	ABC n = 6	C n = 3	AB n = 3	ABC n = 3	AB n = 3
Dissolved⁴-THg (pg L⁻¹)													
Means Group, n	318.1 ± 4.6 ABC n = 3	416.6 ± 29.2 A n = 9	409.7 ± 60.5 AB n = 6	316.6 ± 3.8 ABC n = 3	281.0 ± 5.1 BC n = 6	285.9 ± 8.9 ABC n = 3	227.1 ± 20.0 C n = 6	205.9 ± 8.4 C n = 3	340.1 ± 53.8 ABC n = 6	177.6 ± 6.4 C n = 3	423.8 ± 25.6 AB n = 3	312.2 ± 6.9 ABC n = 3	446.6 ± 4.0 AB n = 3
Seston-MeHg (pg L⁻¹)													
Means Group, n	13.92 ± 0.08 A n = 3	6.78 ± 0.18 B n = 9	7.66 ± 0.51 B n = 6	6.61 ± 0.31 BC n = 3	5.82 ± 0.24 BC n = 6	5.78 ± 0.49 BC n = 3	5.75 ± 0.53 BC n = 6	5.68 ± 0.13 BC n = 3	4.28 ± 0.64 C n = 6	5.62 ± 0.46 BC n = 3	6.30 ± 0.49 BC n = 3	5.83 ± 0.86 BC n = 3	12.54 ± 1.33 A n = 3
Seston-Hg²⁺ (pg L⁻¹)													
Means Group, n	123.3 7 ± 2.02 B n = 3	69.97 ± 7.65 CDE n = 9	76.49 ± 10.20 CD n = 6	43.78 ± 2.62 DEF n = 3	47.95 ± 0.74 EF n = 6	51.16 ± 2.90 CDEF n = 3	60.93 ± 7.88 CDEF n = 6	60.45 ± 1.67 CDEF n = 3	43.93 ± 1.51 F n = 6	54.55 ± 4.42 CDEF n = 3	85.97 ± 2.11 BC n = 3	33.62 ± 1.19 F n = 3	337.87 ± 3.41 A n = 3
Seston-THg (pg L⁻¹)													
Means Group, n	137.2 9 ± 1.94 B n = 3	76.76 ± 7.74 CDE n = 9	84.15 ± 10.59 CD n = 6	50.38 ± 2.46 DEF n = 3	53.77 ± 0.86 EF n = 6	56.94 ± 2.68 CDEF n = 3	66.68 ± 8.38 CDEF n = 6	66.13 ± 1.65 CDEF n = 3	48.21 ± 1.31 F n = 6	60.16 ± 4.77 CDEF n = 3	92.27 ± 2.49 C n = 3	39.45 ± 1.49 F n = 3	350.42 ± 4.19 A n = 3

¹ Alkalinity was measured on only one date without replication.

² ANOVA indicated there were significant differences across lake segments for all parameters at p < 0.0001 (excluding Alkalinity which could not be tested due to sample size). Means Groups represent the results of means comparisons at p = 0.1 using the Tukey-Kramer HSD test. Segments with the same letter are not significantly different.

³ Means of Secchi depth, total-nitrogen, total-phosphorus, and Chl-a, are means of individual samples taken at each station by VTANR on 3 to 6 dates during the period 8/1/2009 through 9/17/2009. Means of seston mass and all mercury measures are means of three replicate samples per date collected on one or more dates at each station. The total number of samples representing conditions at each station for each measurement is indicated in the table.

⁴ The “dissolved” fraction in this study is operationally defined as passing through a Whatman™ GF/F filter. Therefore, Hg²⁺ represents the inorganic divalent ionic mercury that equilibrated with a species-specific isotopic spike in the “dissolved” fraction rather than an ion activity.

Table 2

Means and standard errors of zooplankton (ZP) biomass and mercury pools in Lake Champlain by lake-segment sampling station.

Station Number	4	7	9	16	19	21	25	34	46	51
Station Name	South Lake A	Port Henry	Otter Creek	Shelburne Bay	Main Lake	Burlington Bay	Malletts Bay	Northeast Arm	Rouses Point	Missisquoi Bay
Small ZP biomass ($\mu\text{g L}^{-1}$)	19.40 $\pm$ 0.35	4.42 $\pm$ 1.46	3.42 $\pm$ 0.06	8.46 $\pm$ 0.71	2.55 $\pm$ 0.43	5.49 $\pm$ 0.70	4.04 $\pm$ 0.44	3.44 $\pm$ 0.89	52.77 $\pm$ 8.04	4.60 $\pm$ 0.15
Means Group [/]	B	C	C	C	C	C	C	C	C	A
Large ZP biomass ($\mu\text{g L}^{-1}$)	9.74 $\pm$ 0.87	10.02 $\pm$ 1.38	8.55 $\pm$ 1.62	12.55 $\pm$ 0.71	7.69 $\pm$ 0.65	8.50 (n=1)	5.23 $\pm$ 0.47	15.26 $\pm$ 3.61	0.87 $\pm$ 0.46	23.00 $\pm$ 7.33
Means Group	BCD	BCD	BCD	ABC	BCD	ABCD	CD	AB	D	A
Small ZP MeHg (pg L^{-1})	0.117 $\pm$ 0.003	0.014 $\pm$ 0.005	0.013 $\pm$ 0.000	0.033 $\pm$ 0.002	0.007 $\pm$ 0.001	0.017 $\pm$ 0.002	0.016 $\pm$ 0.004	0.003 $\pm$ 0.000	0.008 $\pm$ 0.002	0.236 $\pm$ 0.147
Means Group	B	C	C	BC	C	BC	C	BC	C	A
Large ZP MeHg (pg L^{-1})	0.439 $\pm$ 0.057	0.319 $\pm$ 0.083	0.325 $\pm$ 0.018	0.480 $\pm$ 0.046	0.299 $\pm$ 0.058	0.271 (n=1)	0.302 $\pm$ 0.078	0.118 $\pm$ 0.036	0.018 $\pm$ 0.015	0.024 $\pm$ 0.009
Means Group	AB	ABC	ABC	A	ABC	ABC	ABC	BC	C	C
Small ZP Hg ²⁺ (pg L^{-1})	0.447 $\pm$ 0.019	0.123 $\pm$ 0.049	0.106 $\pm$ 0.004	0.181 $\pm$ 0.012	0.105 $\pm$ 0.035	0.158 $\pm$ 0.029	0.163 $\pm$ 0.028	0.076 $\pm$ 0.002	0.046 $\pm$ 0.004	0.991 $\pm$ 0.177
Means Group	B	C	C	C	C	C	C	C	C	A
Large ZP Hg ²⁺ (pg L^{-1})	0.331 $\pm$ 0.061	0.240 $\pm$ 0.074	0.285 $\pm$ 0.018	0.846 $\pm$ 0.053	0.203 $\pm$ 0.019	0.267 (n=1)	0.210 $\pm$ 0.040	0.304 $\pm$ 0.055	0.035 $\pm$ 0.008	0.660 $\pm$ 0.092
Means Group	B	BC	BC	A	BC	BC	BC	B	C	A
Small ZP THg (pg L^{-1})	0.564 $\pm$ 0.021	0.138 $\pm$ 0.054	0.119 $\pm$ 0.004	0.214 $\pm$ 0.014	0.112 $\pm$ 0.036	0.175 $\pm$ 0.030	0.179 $\pm$ 0.031	0.078 $\pm$ 0.002	0.054 $\pm$ 0.005	1.227 $\pm$ 0.324
Means Group	B	C	C	C	C	C	C	C	C	A
Large ZP THg (pg L^{-1})	0.770 $\pm$ 0.004	0.559 $\pm$ 0.157	0.610 $\pm$ 0.036	1.326 $\pm$ 0.046	0.502 $\pm$ 0.072	0.538 $\pm$ (n=1)	0.512 $\pm$ 0.116	0.423 $\pm$ 0.059	0.053 $\pm$ 0.020	0.684 $\pm$ 0.096
Means Group	AB	B	B	A	BC	BC	BC	BC	C	B

[/] ANOVA indicated there were significant differences across lake segments for all parameters at $p < 0.0001$. Means Group letters identify groups of means that are not significantly different at $p = 0.1$ by Tukey-Kramer HSD means comparison of all pairs.

Table 3

Equations relating Secchi depth and seston mass and the biomass and mercury content of different ecosystem compartments in the mercury trophic-transfer model. Explanation of table structure: The name of the dependent parameter for the general linear model (GLM) is indicated in bold type in the first column followed by the equals sign. The number of observations, significance (p value), and total variance of the dependent parameter explained (adjusted r^2) by the GLM are given in the second column opposite the dependent parameter name. Each separate term on the right hand side of the general linear model (GLM) used to predict a dependent parameter is listed on a separate line indented below the dependent parameter name in the first column. The confidence level (p value) for each term of the GLM is given in the second column.

Model Parameter Equation	n, significance, variance explained
Secchi Depth (m) = 4.2922141 – 1.8128797 * ln(Seston Mass(mg L ⁻¹))	n = 18, p < 0.0001, r ² = 0.847
Seston Mass (mg L⁻¹) = exp[(Secchi Depth – 4.2922) / –1.8128797]	n = 18, p < 0.0001, r ² = 0.847
Small Zooplankton Mass (µg L⁻¹) = 0.0123775 + 4.2081298 * Seston Mass	n = 16, p < 0.0001, r ² = 0.842 p = 0.9901 p < 0.0001
Large Zooplankton Mass (µg L⁻¹) = 51.940705 + 0.0098475 * [Seston Mass + SmallZPMass] – 619.3387 * Specific Absorbance@254nm	n = 16, p = 0.0058, r ² = 0.547 p = 0.0031 p = 0.0027 p = 0.0085
SestonTHg (pg L⁻¹) / DissTHg (pg L⁻¹) = 0.5454708 + 0.1117373 * Seston Mass – 0.000335 * DissolvedTHg – 0.006613 * Alkalinity (mg L ⁻¹)	n = 18, p < 0.0001, r ² = 0.844 p = 0.0003 p < 0.0001 p = 0.0691 p = 0.0102
Small Zooplankton THg Pool (pg L⁻¹) = – 0.056662 + 0.0212303 * Small ZP Mass + 0.0021164 * SestonTHg – 0.000147 * [(SestonTHg – 85.7005) * (SmallZPMass – 6.75284)]	p < 0.0001, r ² = 0.965 p = 0.1473 p = 0.0001 p = 0.0243 p = 0.0064
Large Zooplankton THg Pool (pg L⁻¹) = 0.0877114 + 0.34296636 * SmallZPTHg – 0.383686 * Seston Mass + 0.0431152 * LargeZPMass	n = 13, p = 0.0076, r ² = 0.718 p = 0.5044 p = 0.0039 p = 0.0049 p = 0.0143
Pelagic Fish Tissue THg (µg g⁻¹ wet weight) = – 0.830067 + 0.0022093 * Fish Length (mm) + 0.3857262 * LargeZPTHg	n = 8, p = 0.0008, r ² = 0.943 p = 0.0029 p = 0.0003 p = 0.0084
Malletts Bay Pelagic Fish THg (µg g⁻¹ wet weight) = 1.667 * General Pelagic Fish Tissue THg	n = 1

Table 4

Input parameters and results for four model scenarios: 1) baseline analysis modeling the expected tissue Hg concentration in a 40-cm sport fish under current productivity (Secchi depth) and dissolved total mercury (Dis-THg) conditions; 2) calculation of the required change in dissolved total mercury necessary to reduce fish tissue mercury concentrations in 40-cm sport fish to $0.3 \mu\text{g g}^{-1}$ wet weight when productivity remains constant at observed levels; 3) response of fish tissue mercury concentrations to anticipated changes in primary productivity as reflected by changes in Secchi depth when dissolved mercury remains constant at observed levels; and 4) calculation of the required change in dissolved total mercury necessary to reduce fish tissue mercury concentrations in 40-cm sport fish to $0.3 \mu\text{g g}^{-1}$ wet weight when productivity is reduced as anticipated in scenario 3. Model output is shown in *italic* text. Fish tissue concentrations greater than the EPA criterion value of $0.3 \mu\text{g g}^{-1}$ wet weight are shown in **bold** type. The difference in dissolved mercury controls required between scenario 2 and scenario 4 is shown in the last column (MCR = more mercury control required, LCR = less mercury control required, NCR = no mercury control required).

Scenario		Model Scenario 1			Model Scenario 2			Model Scenario 3			Model Scenario 4		
Description		Observed Conditions for Productivity and Dis-THg			Reduce Dis-THg to reach $0.3 \mu\text{g g}^{-1}$ in fish tissue			Effect of anticipated productivity change on fish tissue Hg			Reduce Dis-THg to reach $0.3 \mu\text{g g}^{-1}$ in Fish Tissue with productivity change		
Lake Segment		Dissolved THg ($\mu\text{g L}^{-1}$)	Secchi Depth (m)	<i>Modeled 40-cm Fish Hg ($\mu\text{g g}^{-1}$)</i>	Required Change in Fish Hg (%)	<i>Implied Change in Dis-THg (%)</i>	Scenario Input Secchi Depth (m)	<i>Modeled 40-cm Fish Hg ($\mu\text{g g}^{-1}$)</i>	<i>Model Projected Fish Hg Change (%)</i>	<i>Implied Change in Dis-THg (%)</i>	Difference from Scenario 1		
Burlington Bay		286	4.40	0.34	-11%	-25%	6.0	0.32	-5%	-25%	no change		
Grand Isle		178	5.60	0.23	0%	0%	6.0	0.29	27%	0%	no change		
Rouses Pt		312	6.00	0.27	0%	0%	6.0	0.34	26%	-40%	MCR		
Main Lake		281	5.20	0.28	0%	0%	6.0	0.32	14%	-25%	MCR		
Malletts Bay		227	4.50	0.44	-32%	-41%	5.0	0.57	28%	-75%	MCR		
Missisquoi Bay		447	1.30	0.31	0%	0%	2.5	0.57	82%	-66%	MCR		
Northeast Arm		340	4.95	0.45	-33%	-74%	6.0	0.37	-19%	-64%	LCR		
Otter Creek		410	3.15	0.44	-31%	-58%	5.0	0.28	-36%	0%	NCR		
Port Henry		417	3.73	0.29	0%	0%	5.0	0.25	-16%	0%	no change		
Shelburne Bay		317	5.50	0.27	0%	0%	6.0	0.32	19%	-27%	MCR		
South Lake A		318	1.65	0.37	-20%	-69%	2.5	0.32	-14%	-14%	LCR		
St. Albans Bay		424	4.00	0.44	-31%	-63%	5.0	0.33	-24%	-35%	LCR		