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Can mercury in fish be reduced by water level management? Evaluating the effects of water level fluctuation on mercury accumulation in yellow perch (*Perca flavescens*)

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Abstract Mercury (Hg) contamination of fisheries is a major concern for resource managers of many temperate lakes. Anthropogenic Hg contamination is largely derived from atmospheric deposition within a lake's watershed, but its incorporation into the food web is facilitated by bacterial activity in sediments. Temporal variation in Hg content of fish (young-of-year yellow perch) in the regulated lakes of the Rainy–Namakan complex (on the border of the United States and Canada) has been linked to water level (WL) fluctuations, presumably through variation in sediment inundation. As a result, Hg contamination of fish has been linked to international regulations of WL fluctuation. Here we assess the relationship between WL fluctuations and fish Hg content using a 10-year dataset covering six lakes. Within-year WL rise did not appear in strongly supported models of fish Hg, but year-to-year variation in maximum water levels (ΔmaxWL) was positively associated with fish Hg content. This WL effect varied in magnitude among lakes: In Crane Lake, a 1 m increase in ΔmaxWL from the previous year was associated with a 108 ng increase in fish Hg content (per gram wet weight), while the same WL change in Kabetogama was associated with only a 5 ng increase in fish Hg content. In half the lakes sampled here, effect sizes could not be distinguished from zero. Given the persistent and wide-ranging extent of Hg contamination and the large number of regulated waterways, future research is needed to identify the conditions in which WL fluctuations influence fish Hg content.

Keywords Mercury · Water level fluctuations · Fish · Water regulation

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Introduction

Anthropogenic methylmercury (MeHg) contamination of recreational and commercial fisheries is an on-going concern in many temperate lakes of eastern North America due to impacts on both human and wildlife health (Wiener et al. 2003; Driscoll et al. 2007, 2013). The source of this MeHg is primarily derived from atmospheric deposition of material volatilized by anthropogenic activities (e.g., coal burning power plants; Driscoll et al. 2007). Mercury (Hg) is incorporated into aquatic food webs after inorganic Hg delivered via atmospheric deposition to a watershed or directly to a lake is converted into MeHg by bacteria (Benoit et al. 2003), and so relationships between simple atmospheric deposition of Hg and Hg accumulation in fish are often complex (Munthe et al. 2007; Harris et al. 2007). Much of this methylation is done by sulfate-reducing bacteria (SRB; Benoit et al. 2003), and thus among-system variation in the supply of Hg to SRB and the spatial distribution of favorable habitats for SRB may lead to spatial variation in the availability of MeHg to food webs (Wiener et al. 2006).

Favorable conditions for SRB include anoxic, carbon-rich sediments (Benoit et al. 2003) such as those commonly occurring in wetlands (Bodaly et al. 1984; Kelly et al. 1997), or in areas that have been recently inundated (Gilmour et al.

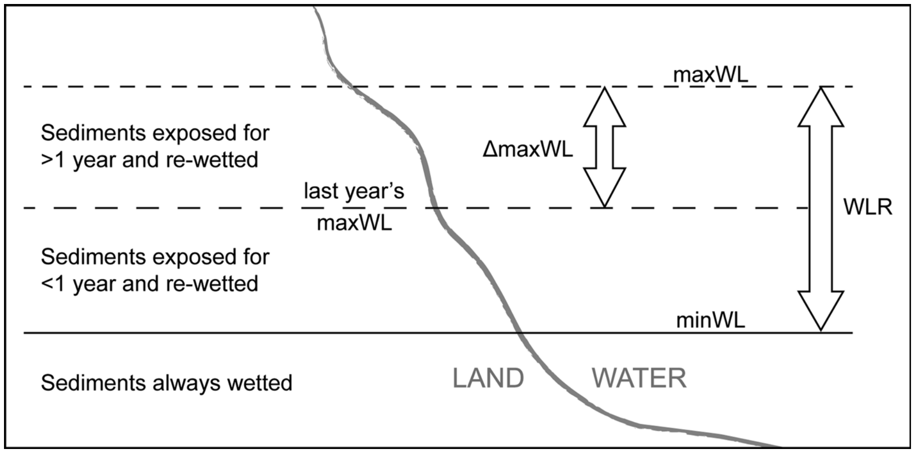

Fig. 1 Conceptual figure of hypothetical sediment classes created by WL fluctuations. Production of MeHg by SRB is hypothesized to be greatest (per unit area) in sediments that have been exposed for more than 1 year, because those sediments have accumulated relatively large quantities of sulfate and organic matter. *maxWL* maximum water level elevation, *minWL* minimum water level, *WLR* water level rise, *ΔmaxWL* change in maximum water level from last year to this year

2004). Studies have generally shown that a combination of water quality parameters associated with the supply of nutrients to SRB (e.g., sulfate concentrations) and the abundance of wetlands (which provide anoxic, carbon-rich sediments) explains much of the considerable spatial variation in fish Hg levels (Wiener et al. 2006). Many areas otherwise considered pristine (i.e., aquatic habitats draining relatively undisturbed watersheds) have high levels of Hg in their food webs because abundant wetlands provide ideal conditions for methylation (Driscoll et al. 2007).

Sorensen et al. (2005) highlighted a link between water level (WL) fluctuations and the MeHg concentrations in young-of-year (YOY) yellow perch (*Perca flavescens*) in Voyageurs National Park (VNP) and surrounding lakes, an area considered relatively pristine. Previous work has established the effect of reservoir establishment (i.e., permanent inundation) on MeHg fluxes into the food web (Bodaly et al. 1984; Kelly et al. 1997), but Sorensen et al. (2005) suggested sediments that have been exposed for months or years and are then re-wetted are a significant source of Hg inputs to the food web (Snodgrass et al. 2000; Selch et al. 2007). The drying and re-wetting of these sediments is controlled by variation in WLs, and Sorensen et al. (2005) found WL fluctuation to have a linear, positive relationship with temporal change in fish Hg content (which is mostly in the form of MeHg; Sandheinrich and Wiener 2011). Indeed, other predictors that are often found to be related to spatial variation in fish Hg were not good predictors of temporal variation in fish Hg after WL fluctuations were taken into account (e.g., total organic carbon content, pH, and Secchi depth). This finding is particularly relevant because WLs in many lakes and reservoirs can be manipulated, and thus might offer a potential management tool for reducing Hg contamination in fish.

Annual WL fluctuations create three sediment classes: sediments that are permanently inundated, sediments that are dried and re-wetted on an annual basis and sediments that are dry for more than 1 year before re-wetting (Fig. 1). These sediment classes likely differ in their production of MeHg due to differences in the delivery and accumulation of sulfur and organic carbon that affect SRB (Driscoll et al. 2013). Exposed sediments receive atmospheric deposition of sulfate and accumulate organic carbon (e.g., via plant growth; Sorensen et al. 2005), both necessary for SRB activity. Sediments exposed for longer time periods (>1 year) may be particularly important, as they not only accumulate more sulfate, but also accumulate organic carbon throughout the growing season. Based on this conceptual understanding, we predicted that either the within-year water level rise (WLR) or the change in maximum water level (ΔmaxWL) from 1 year to the next were likely to be correlated to annual MeHg production and thus annual variation in YOY yellow perch Hg content.

As is the case with many other north temperate lakes, Hg accumulation in fish is a major management issue for the U.S. National Park Service in lakes at VNP (Kallemeyn et al. 2003). WLs in Rainy Lake and the Namakan Reservoir complex (Namakan, Kabetogama, Sand Point, Crane, and Little Vermilion lakes) are actively managed via dams at the outlet of the Namakan Reservoir complex and Rainy Lake (Fig. 2) for hydroelectric power generation (at the outlet of Rainy Lake) and other recognized uses (Kallemeyn et al. 2003). WL management in this system is directed by the International Joint Commission (IJC), a regulatory body comprised of representatives from the United States and Canada.

For the purpose of hydroelectric power generation, WLs in the Namakan complex and Rainy Lake are manipulated

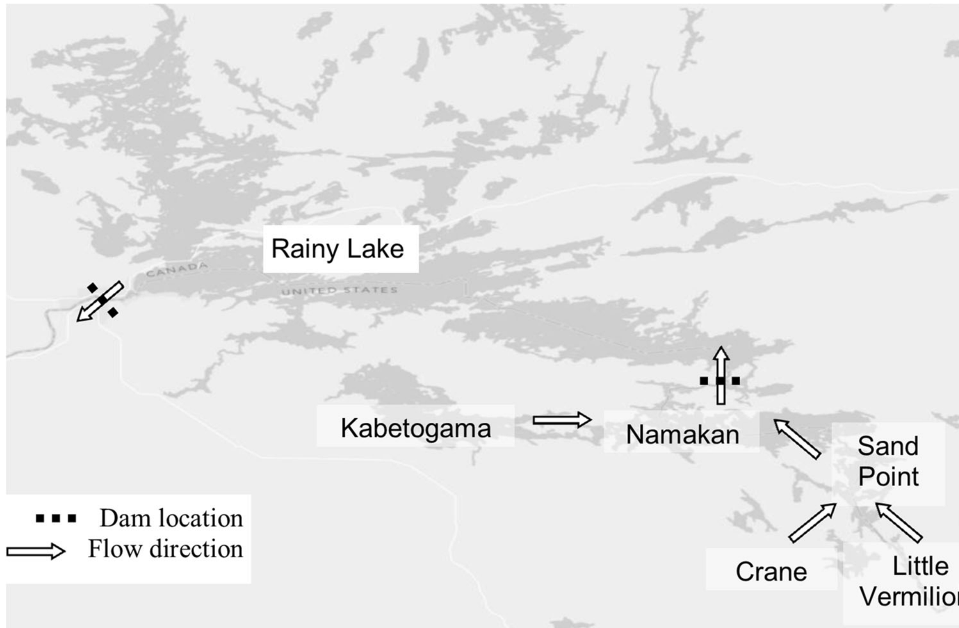

Fig. 2 Hydrologic connections and impoundments in the Rainy Lake–Namakan Reservoir complex on the border between the United States and Canada

so that WL fluctuations in Rainy Lake are reduced. This requires overwinter drawdowns of the Namakan Reservoir and a refilling of the reservoir system to capacity over the spring and summer. The IJC establishes 'rule curves' that prescribe a season-specific range of water elevation values. Dam operators then attempt to keep the water elevation within that range of values, although floods and droughts occasionally limit their success. In 2000, the IJC modified the 'rule curves' governing the range of WLs in these lakes, with the primary change being increased WLs in early spring in the Namakan Reservoir. This increase in minimum WLs also has the effect of reducing the annual WL rise, as ΔmaxWL were not significantly altered. Similar management practices occur in regulated water bodies throughout the developed world for a variety of purposes (e.g., power generation, agriculture, navigation, and waterfowl production).

The primary purpose of this study was to estimate the effects of annual WL variation on fish Hg content in regulated lakes. If the effects of WL fluctuations are evident, then the IJC's rule curves have the potential to influence Hg contamination of aquatic food webs in this reservoir system. The study by Sorensen et al. (2005) is the only analysis to document a strong relationship between annual WL fluctuations and fish Hg content. However, the Sorensen et al. (2005) study was limited to just 3 years in most lakes. Our study builds on the work of Sorensen et al. (2005) by including additional years (for a total of 10 years) on six of those lakes to evaluate whether annual WL fluctuations are related to fish Hg content.

Methods

Sample collection and analysis

Six lakes from the Rainy–Namakan complex, a subset of the lakes sampled by Sorensen et al. (2005), were sampled between 2004 and 2010 (Table 1). Fish were collected between mid-September and mid-October each year. Methods for sampling, aging and determining Hg in YOY yellow perch were the same as those reported in Sorensen et al. (2005) except that fish (between 5 and 20 per lake per year) were collected from only one site per lake. Sorensen et al. (2005) concluded that different sites within the same lake varied in the magnitude of fish Hg content, but the direction and magnitude of year-to-year variation was very similar among sites within a single lake. We incorporated the 2001–2003 data from Sorensen for the six sites we sampled for this study (site locations in S1).

Briefly, fish were collected with 15.2 or 30.5 m bag seines with 6.4 mm mesh (bar). Size thresholds for identifying YOY yellow perch were developed previously (see Appendix F1 from Sorensen et al. 2005) and those lake-specific thresholds were applied in the current dataset as well (see S1 for size thresholds).

WL metrics calculated here were similar to those used in Sorensen et al. (2005). Within-year minimum WL and maximum WL were used to calculate WLR. In addition, change in maximum WL from the previous year was calculated (ΔmaxWL). All WL data were obtained from the Lake of the Woods Water Control Board. WLs in the Rainy–Namakan lake complex follow a seasonal pattern,

Table 1 Results of model selection procedure relating WL parameters and Hg content in yellow perch

Model	−2 log likelihood	AIC[a]	ΔAIC[a]
Lake + ΔmaxWL + Lake[a]ΔmaxWL	5,908.5	5,948.5	0
Lake + ΔmaxWL + WLR + Lake[a]ΔmaxWL + Lake[a]WLR	5,904.5	5,956.5	8
Lake + WLR + Lake[a]WLR	5,920.8	5,960.8	12.3
Lake + WLR	5,938.3	5,968.3	19.8
Lake + ΔmaxWL + WLR	5,936.7	5,968.7	20.2
Lake + ΔmaxWL	5,938.7	5,968.7	20.2
Lake (*null*)	5,945.8	5,973.8	25.7

[a] Smaller AIC and ΔAIC values denote greater data support for the model in question relative to the other models

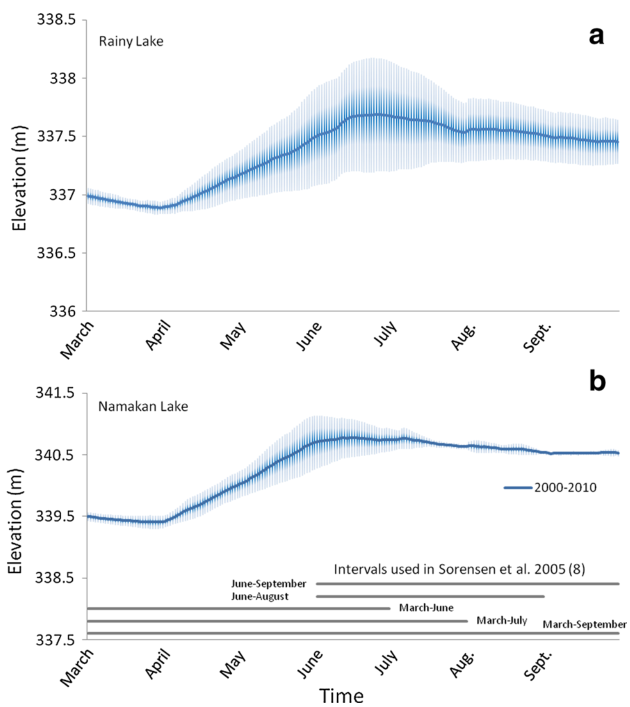

Fig. 3 Daily mean WLs (±SD) from 2000 to 2010 in **a** Rainy Lake and **b** Namakan Lake. At the bottom of **b** are visual depictions of the different intervals used in Sorensen et al. (2005) to evaluate the effect of WL fluctuations

with early-spring minimums and early to mid-summer maximums from 2000 to 2010 (Fig. 3). Sorensen et al. (2005) considered WL fluctuations over several within-year time intervals (Fig. 3). Here, we used WLR over the entire year (January–December), which turns out to be equivalent to WLR in the March–July interval used most often by Sorensen et al. (2005).

Fish Hg analysis

Methods for measuring whole-fish Hg content were identical to those used in Sorensen et al. (2005). Briefly, fish wet weight (WW) and total length are measured, then fish are dried for 24 h at 70 °C and weighed again to estimate moisture content. Length measurements were made using a ruler with 1 mm demarcations (estimates were to the nearest 0.5 mm). Weights were measured using a balance accurate to 0.1 mg. Dried fish were then shredded and ground with mortar and pestle and stored (frozen) until analysis could be completed. Total Hg was measured using USEPA method 245.6 (EPA 1991), as MeHg is generally >90 % of total Hg in fish (Sandheinrich and Wiener 2011). The detection limit for this method was ~3 ng g^{-1} WW^{-1}. Spike recovery was approximately 99 ± 4 % (SD; based on 25 spikes from 2004 to 2010 sampling efforts). Two reference standards were used: NRC Dorm2 (14 runs; recovery ranged between 98 and 105 %) and Mussel NIST 2976 (14 runs; recovery ranged between 96 and 114 %). Other details are described in Sorensen et al. (2005) for samples taken from 2001 to 2003 and reports from the analytical laboratory are available upon request.

Secchi depth, pH and chlorophyll

Secchi depth was collected at each site using methods described in Sorensen et al. (2005). Because field data sheets for 2005 have (apparently) been lost, those Secchi data were treated as missing. No pH was recorded at the time of fish sampling for years 2004–2010, so surface-water pH from mid-lake limnological sampling from 2001 to 2010 was used (US National Park Service, unpublished data). Chlorophyll concentration was also measured at a subset of these mid-lake sites as well. Chlorophyll concentrations were averaged across all sampling dates to give an index of the summer-long average. Mid-lake limnological sampling occurred monthly or bi-weekly (depending on the lake) during the growing season.

Atmospheric deposition of Hg and sulfate

Hg and sulfate deposition were measured by the National Atmospheric Deposition Program. For Hg, annual data from the Fernberg monitoring location (MN18) were obtained from their website (NADP 2012a). For sulfate, annual data from the Sullivan Bay monitoring location (MN32) were collected from their website (NADP 2012b).

Temperature data

Water temperature data are only available from mid-lake locations in these lakes and not available each year in each lake. Vertical water temperatures were taken either monthly or bi-weekly (depending on the lake). Each vertical water temperature profile was averaged, and then all of the measurements taken from May to October were averaged for an estimate of summer-long average water temperature (US National Park Service, unpublished data; water temperature profiles include in S1).

Statistical analysis

YOY yellow perch mean Hg concentration was modeled as a function of lake, year and WL using measurements of individual fish with linear mixed models. In the base model, lake effects were treated as fixed while year and lake × year effects were treated as random (variation at the observation scale occurred by lake). This was compared with models that elaborated the base model by inclusion of WLR, ΔmaxWL and their interactions with lake. We considered the base model to be an appropriate null model for comparisons to models that included WL associations. Data support for the selected models was evaluated using Aikaike's information criterion (AIC; Burnham and Anderson 1998). Smaller AIC values indicate relatively greater data support for given models; the difference between a particular model and the best model is used to rank the models (ΔAIC). Small ΔAIC values ($\leq$2) denote strong support (relative to other models) by the data while ΔAIC values greater than ten indicate essentially no such support (Burnham and Anderson 1998). Models were fitted using maximum likelihood, and SAS's linear mixed modeling procedure (Proc Mixed; SAS 2011).

Associations between ancillary data (water quality, water temperature, Chl *a*, and atmospheric deposition) and YOY perch Hg content were not evaluated in this model selection framework due to the poor data-to-variable ratio and numerous gaps in the ancillary data. Instead a graphical approach was used that allows qualitative evaluation of the effects of ancillary data. Simply put, residuals from the base (null) model and the best model (as selected by AIC) were plotted against ancillary variables.

We estimated associations between fish length and total Hg content using Pearson's correlation statistic (both within individual lakes and among all lakes). The correlation statistic and other descriptive statistics were calculated using R (the "cor()" function). Other descriptive statistics (means, etc.) were also calculated using R. For the purpose of display, annual means were calculated for each lake-year combination: these were strongly correlated to the median (Pearson's $r = 0.997$).

Results

The Hg content of YOY perch collected during the study period varied considerably both among lakes and years (Fig. 4; S1). This variation was most strongly related to a model that included a lake-specific ΔmaxWL association with fish Hg content (Table 1; S1). Other models were only weakly supported (ΔAIC $\geq$ 8; Table 1). In the best model, the slope of the association between ΔmaxWL and fish Hg content varies by lake (Fig. 5; S1). Although that slope was always positive, the magnitude of the slope varied among lakes and 95 % confidence intervals overlapped zero in half the lakes sampled (Kabetogama, Little Vermilion, and Rainy; Fig. 5). In Sand Point, for example, the linear relationship between ΔmaxWL and fish Hg content had a slope of $\sim$100 (Fig. 5f). This means that in Sand Point an increase in ΔmaxWL of 0.5 m from the previous year was associated with an increase of $\sim$50 ng Hg g^{-1} WW^{-1} (a 50 % increase over the mean Hg content for this lake). The same increase in ΔmaxWL in Kabetogama (where the slope was $\sim$5, Fig. 5b) would have lead to an increase of just $\sim$2.5 ng Hg g^{-1} WW^{-1} (a 6 % increase over the mean Hg content for this lake). Overall, estimates of the magnitude (slope) of the association between ΔmaxWL and fish Hg content were greater than zero in Crane, Namakan and Sand Point lakes (Fig. 5).

YOY yellow perch growth did not appear to be strongly correlated with fish Hg content in our dataset (Pearson's $r < 0.4$ between fish length and fish Hg content) and graphical analysis suggested pH, Secchi depth, chlorophyll concentration, and summer-long average water temperature were not strongly related to model residuals. Visual inspections comparing atmospheric deposition of Hg and sulfate to residuals of the best model or the base model do not suggest these parameters are likely to explain a large portion of the annual variation in fish Hg content (see S2–S13).

Discussion

These results make a subtle but important contribution to our understanding of the relationship between WL fluctuations and fish Hg content. Although the strongest model did include WL fluctuations as a predictor variable, that model also found that WL associations with Hg were limited to a subset of the sampled lakes. Earlier analysis was not able to compare the magnitude of WL associations among lakes (Sorensen et al. 2005), but the current analysis shows that WL associations occur in only a subset of lakes.

Variation in YOY perch Hg content was associated with the year-to-year change in maximum WL, presumably because of variation in the re-wetting of sediments exposed by low WLs. Variation among lakes in WL effects could be

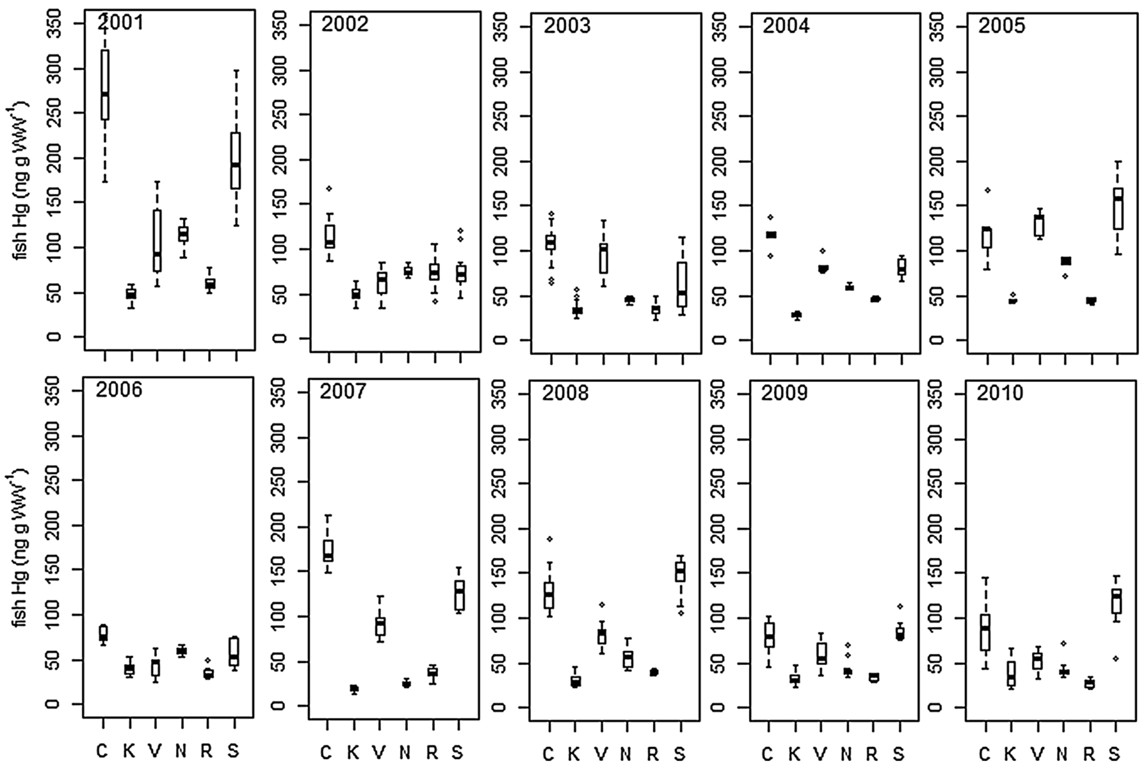

Fig. 4 *Box and whisker plots* showing median Hg content of YOY perch in lakes of the Rainy–Namakan complex from 2001 to 2010. *Boxes* encompass the first and third quartile. The *lines* (*whiskers*) show the largest or smallest observation that falls within 1.5 times the box size. Observations that fall outside the lines are shown individually. *C* Crane, *K* Kabetogama, *V* Little Vermilion, *N* Namakan, *R* Rainy, *S* Sand Point

caused by differences in shoreline characteristics of these lakes. The relationship between WL and the areal extent of exposed sediments is probably not linear in many locations. Locations or lakes with shallow nearshore bathymetries would seem more likely to be strongly influenced by WL fluctuations simply because more sediments are exposed per unit of WL decline. Detailed bathymetry of areas with <1.5 m of depth (areas most likely to be affected) are lacking for the lakes sampled here, but variation in littoral area (defined as area with <4.6 m) of these lakes does not correspond directly to variation in WL effects on fish Hg content. For example, Sand Point and Kabetogama have a similar proportion of littoral areas (Table 2) and very different associations between fish Hg content and WL fluctuations (Fig. 5). Whether "littoral" areas and the area of exposed sediments are directly correlated is unknown. Future efforts to evaluate the effects of WL in these lakes should focus on determining the areal extent of sediments exposed or inundated by WL fluctuations and characterization of those sediments.

Many other lake characteristics influence fish Hg content. The two lakes with large WL effects (Sand Point and Crane) are also the lakes with the highest mean fish Hg content (and highest among-year variability), so perhaps other conditions for high fish Hg content are necessary to detect significant WL effects. Watershed characteristics (Wiener et al. 2006), sediment characteristics (Verta et al. 1986; Bodaly et al. 2004; Gilmour et al. 2004), productivity and fish growth (Essington and Houser 2003) are all known to influence fish Hg content and vary among the lakes sampled here. Watershed and sediment characteristics probably do not change rapidly enough to drive the interannual variation under investigation here (and these data are rarely available on a yearly timescale). Fish growth and productivity certainly do vary among the lakes. For example, Kabetogama is two to three times as productive as the other large lakes in VNP (Kallemeyn et al. 2003). However, we did not observe obvious connections between Hg content and fish length in this study, and model residuals were not well correlated to a surrogate for productivity (Chl *a*). Still, these factors might influence the degree to which MeHg production in sediments contribute to MeHg in the food web, and therefore the magnitude of the WL effect on fish Hg content. Identifying the factors that control the magnitude of WL effects on fish Hg content should be the focus of future research in this area.

Each lake in this study was characterized by a single site (per results in Sorensen et al. 2005). Given our results (i.e., among-lake variation in WL effects), it may be worthwhile

Fig. 5 Annual average Hg content of YOY perch (Hg ng g^{-1} WW^{-1}) as a function of change in ΔmaxWL in lakes of the Namakan–Rainy complex. Data includes samples from 2001 to 2010. *Error bars* denote standard deviation. *Lines* are derived from the best model relating Hg content in YOY perch to ΔmaxWL (see Table 2) with 95% confidence intervals. The slope of the relationship between ΔmaxWL and annual average Hg content of YOY perch (Hg ng g^{-1} WW^{-1}) is included on each figure with 95 % confidence interval. Intervals that do not overlap zero are highlighted in *bold*. Note altered axis scales for Rainy (**e**)

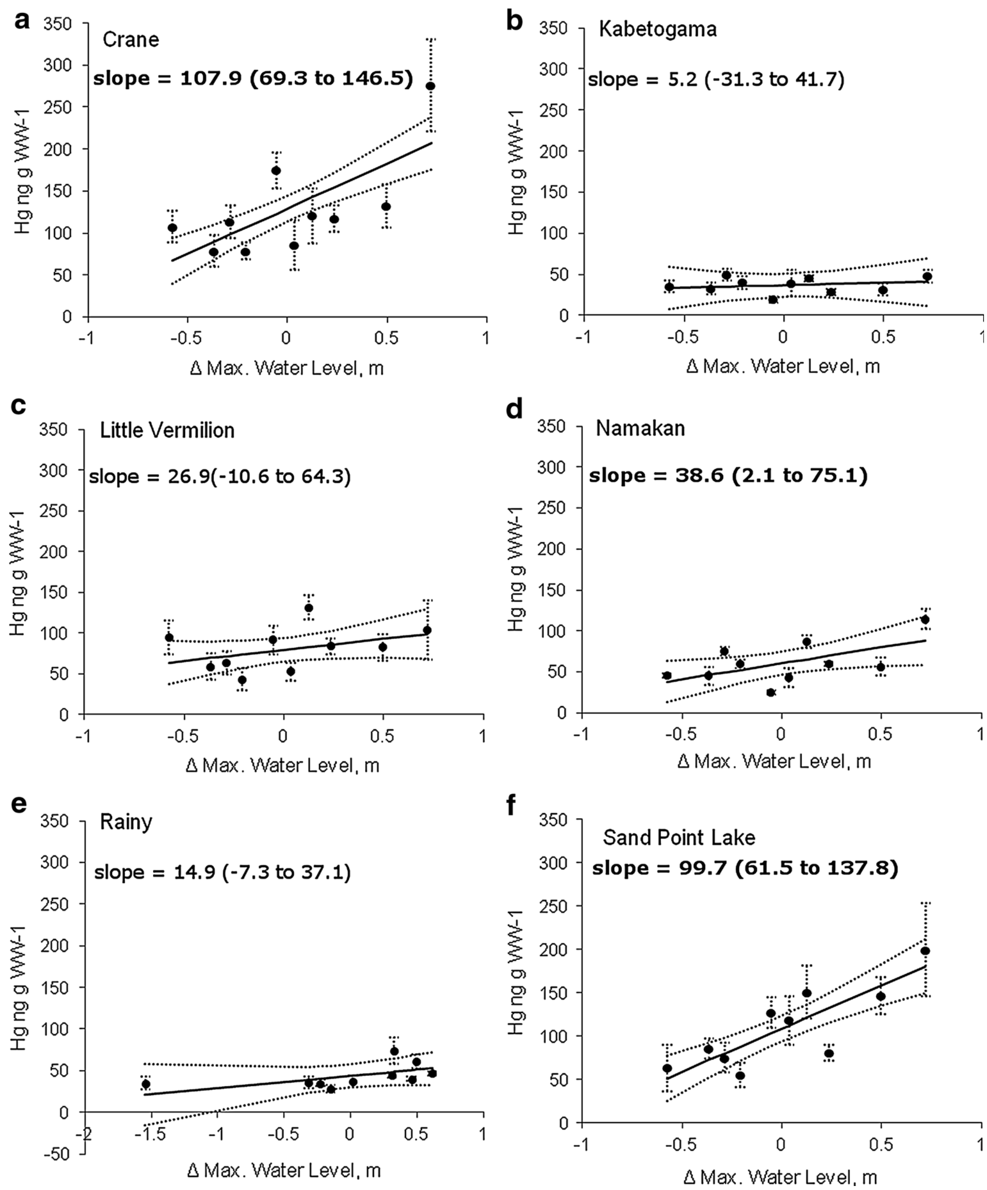

Table 2 Physical properties of lakes sampled for this study

Lake	Surface area[a] (ac)	Littoral area[a,b] (ac)	Maximum depth[a] (m)	Littoral:total area[a]	Average chlorophyll concentration (μg L^{-1})
Crane	2,920	618	24	0.21	3.21
Kabetogama	24,034	7,440	24	0.31	7.36
Little Vermilion	1,288	231	16	0.18	3.10
Namakan	24,066	5,026	46	0.21	2.10
Rainy	230,301	18,949	49	0.08	2.20
Sand Point	8,526	2,847	56	0.33	3.18

Average chlorophyll concentration was calculated from mid-lake limnological samples collected by the National Parks Service

[a] These data are from the Minnesota Department of Natural Resources

[b] Littoral area was calculated as areas of the lake with depth of <4.6 m (15 ft)

to examine more carefully whether site-specific (as opposed to lake-wide) characteristics influence WL associations with fish Hg content. If spatial controls over variation in the associations between fish Hg content and WL were understood, then estimates of fish Hg content under different WL management strategies could be made both in the Rainy–Namakan complex and in other regulated systems.

Conclusion

WL management for the purpose of reducing Hg accumulation in YOY perch and by proxy the rest of the aquatic food web is an attractive concept (Mailman et al. 2006). Certainly, the flooding of reservoirs leads to increased Hg in fish and other aquatic biota, apparently due to large quantities of terrestrial Hg being rapidly incorporated into the aquatic food web (Bodaly et al. 2004; Driscoll et al. 2007). Reducing the overall area of flooded terrestrial soils has thus been considered a viable strategy to reduce fish Hg contamination (Mailman et al. 2006). Mechanistically this seems similar to the drying and re-wetting of shallow sediments (Snodgrass et al. 2000; Evers et al. 2007). However, large differences in the effects of flooding on fish Hg content exist even during the initial flooding of reservoirs (when effects would be strongest) due to differences in organic matter content and morphology (Bodaly et al. 2004; Evers et al. 2007). Drying shallow sediments for a few months or a few years does appear associated with fish Hg content in some systems (Verta et al. 1986; Snodgrass et al. 2000; three lakes in this study), but appears unimportant in others (three lakes studied here). The current data are insufficient to identify the conditions necessary for annual WL fluctuations to influence fish Hg content, and thus it is not yet possible to quantitatively estimate how much different WL management regimes might influence overall Hg contamination.

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Conflict of interest The authors declare that they have no conflict of interest.

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