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Potential risk to common loons (*Gavia immer*) from methylmercury exposure in acidified lakes

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Abstract

Piscivorous birds and mammals in areas remote from point sources of Hg contamination may be exposed to dietary methylmercury concentrations that are sufficiently high to cause reproductive impairment. Common loons (*Gavia immer*) were observed to show aberrant nesting behavior and low overall reproductive success when Hg concentrations in prey (small fish and crayfish) averaged $>0.3\ \mu g\ g^{-1}$ wet weight (Barr, 1986), levels known to occur in fish from many lakes in central Ontario. We used data on Hg in Ontario fish to estimate the proportion of lakes where fish small enough for loons to eat (<250 g) had Hg concentrations that exceeded estimated thresholds for reproductive impairment. Up to 30% of lakes exceeded thresholds for reproductive impairment, depending on the species of fish and the threshold Hg concentrations chosen. There was a significant negative correlation between fish-Hg concentration and lake pH in most fish species examined. For these species, reductions in sulfate deposition rates are predicted to result in a corresponding reduction of lakes in Ontario having fish with potentially toxic concentrations of Hg.

Introduction

It is now well established for several fish species that fish from low pH, low alkalinity lakes tend to have higher mercury (Hg) concentrations than fish of the same species and size from alkaline, circumneutral lakes (Bjorklund *et al.*, 1984; McMurtry *et al.*, 1989; Scheider *et al.*, 1979; Suns & Hitchin, 1990; Wiener *et al.*, 1990b; Wren *et al.*, 1991). The net microbial production of methylmercury in both the water column and at the water-sediment interface is inversely correlated with lake pH (Winfrey & Rudd, 1990; Xun *et al.*, 1987), and a causal relationship between lake acidification and increased accumulation of Hg in yearling perch has been established (Wiener *et al.*, 1990a). The effects of environmental acidification on the bioaccumulation of metals, including Hg, by fish have been reviewed by Spry & Wiener (1991).

Because the predominant chemical form of Hg in freshwater fish is monomethylmercury (Grieb *et al.*, 1990; Huckabee *et al.*, 1979), the phenomenon of elevated Hg concentrations in fish from acidified environments has raised concerns for the health of both human consumers (Clarkson, 1990) and piscivorous wildlife (Scheuhammer, 1991). In order to be able to assess the risk for methylmercury toxicity in fish-eating wildlife, it is necessary to know the dietary levels of methylmercury required to produce a toxic response, as well as the concentrations of Hg in fish of a size range and species composition most likely to be used as food by the particular species of wildlife under consideration. One of the most sensitive biological processes affected by methylmercury exposure is reproduction. Reproductive success in birds can decrease by 35–50% in response to dietary methylmercury exposure insufficient to cause obvious signs of intoxication in adults (Heinz, 1974; Scheuhammer, 1987). As reviewed by Scheuhammer (1991), significant reproductive effects of dietary methylmercury exposure in captive birds typically occur in the range of 2–5 μg Hg g^{-1} (dry wt.) in food. For common loons (*Gavia immer*), it has been reported that reproductive impairment characterized by decreased use of potential territories and a decline in egg laying occured when mean Hg (predominantly as methylmercury) in prey items was ≥ 0.3 μg g^{-1} wet wt. (approx. ≥ 1.2 μg g^{-1} dry wt.) (Barr, 1986). These are the lowest dietary concentrations of Hg reported to cause adverse effects in an avian species. It is also known that adult loons typically eat fish weighing <250 g (Barr, 1973).

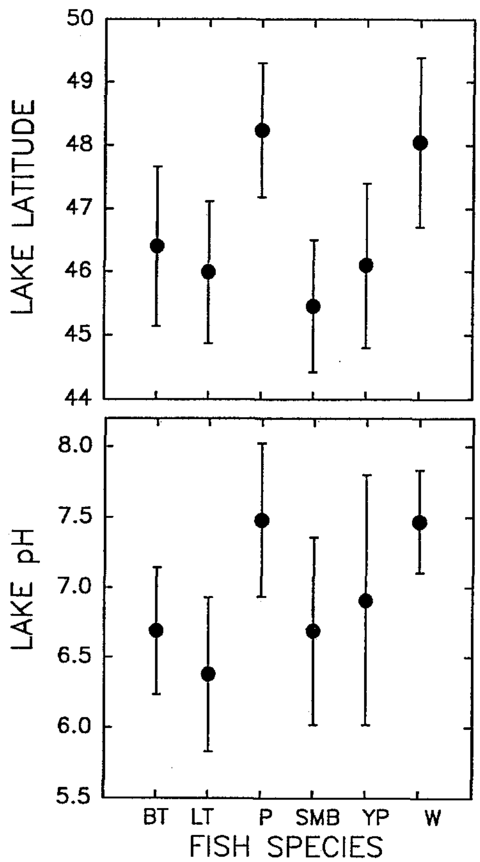

Fig. 1. Latitude (degrees) and pH distribution of lakes for which there were data on Hg concentrations in small fish, pH, and DOC in the OMOE/OMNR sportfish database (Sudbury lakes excluded). Means ± SD for lakes containing BT (brook trout; $n = 16$), LT (lake trout; $n = 51$), P (pike; $n = 37$), SMB (smallmouth bass; $n = 37$), YP (yellow perch; $n = 12$), and W (walleye; $n = 104$) are plotted.

Methods

We used the database on Hg concentrations in fish muscle, fish size, and lake water chemistry characteristics developed jointly by the Ontario Ministries of the Environment and of Natural Resources. Details concerning the geographical distribution of lakes, fish collection methods, Hg analysis, and lake water chemistry analyses have been presented in other reports (McMurtry *et al.*, 1989; Wren *et al.*, 1991). Figure 1 depicts the latitude and pH characteristics of the subset of lakes having data for small fish (<250 g) chosen for the majority of the statistical analyses that we performed. All lakes in the dataset were non-impounded drainage lakes.

All statistical analyses were performed using SAS (SAS Institute Inc., 1988). For the initial analysis, we selected all lakes from the dataset having fish-Hg values for small fish (20–250 g) of at least one of the following fish species: walleye (*Stizostedion vitreum vitreum*), northern pike (*Esox lucius*), yellow perch (*Perca flavescens*), smallmouth bass (*Micropterus dolomieui*), largemouth bass (*Micropterus salmoides*), lake trout (*Salvelinus namaycush*), brook trout (*Salvelinus fontinalis*), whitefish (*Coregonus clupeaformis*), or white sucker (*Catostomus commersoni*). We then calculated the percentage of lakes with small fish of each species having mean Hg concentrations above a 'toxic threshold'. The threshold values chosen were 0.4 and 0.5 μg g^{-1} (wet wt.) in axial muscle, roughly equivalent to the threshold of 0.3 μg g^{-1} in whole fish reported by Barr (1986) for reproductive impairment in free-living loons. (Hg concentrations in fish muscle are marginally greater than those in whole fish). The guideline value for human consumption is 0.5 $\mu g\ g^{-1}$ (OMOE/OMNR 1988).

Because several studies have reported anomalously low Hg concentrations in various biota collected from the Sudbury, Ontario vicinity (Scheuhammer, 1991; Wren & Stokes, 1988; Wren *et al.*, 1986) we considered removing Sudbury area lakes from the dataset before performing any further statistical analyses. We used the 'Sudbury Zone' delineated by Neary *et al.* (1990) to designate lakes as either within or outside the Sudbury area. To test the hypothesis that fish from lakes near Sudbury had significantly lower Hg concentrations than those from other lakes of similar water chemistry, we used SAS to create a population of lakes with equivalent mean $\pm$ SD pH and dissolved organic carbon (DOC) to the Sudbury lakes, then compared Hg concentrations in fish from the two populations of lakes.

We performed multiple linear regression analyses to determine whether fish-Hg concentrations (ln Hg) were related to fish size (ln length [cm]), lake latitude, DOC (ln DOC), or lake pH. For these analyses, we included all lakes having both fish-Hg and water chemistry (pH, DOC) data. Finally, using an acidification model developed for eastern Canada (Jones *et al.*, 1990; Marmorek *et al.*, 1990), we predicted how changes in SO_4 deposition rates might affect the proportion of lakes in the province having fish <250 g with high Hg concentrations.

Results

Proportion of Ontario lakes having small fish with high Hg concentrations

Depending on fish species, up to almost 20% of sampled lakes in central and northern Ontario contained small fish with mean Hg concentrations $>0.5\ \mu g\ g^{-1}$ (Fig. 2). This percentage increased to about 30% if a lower Hg threshold ($0.4\ \mu g\ g^{-1}$) was used. These highest percentages were associated with lakes containing walleye. Between 8% and 18% of perch, pike, and small-

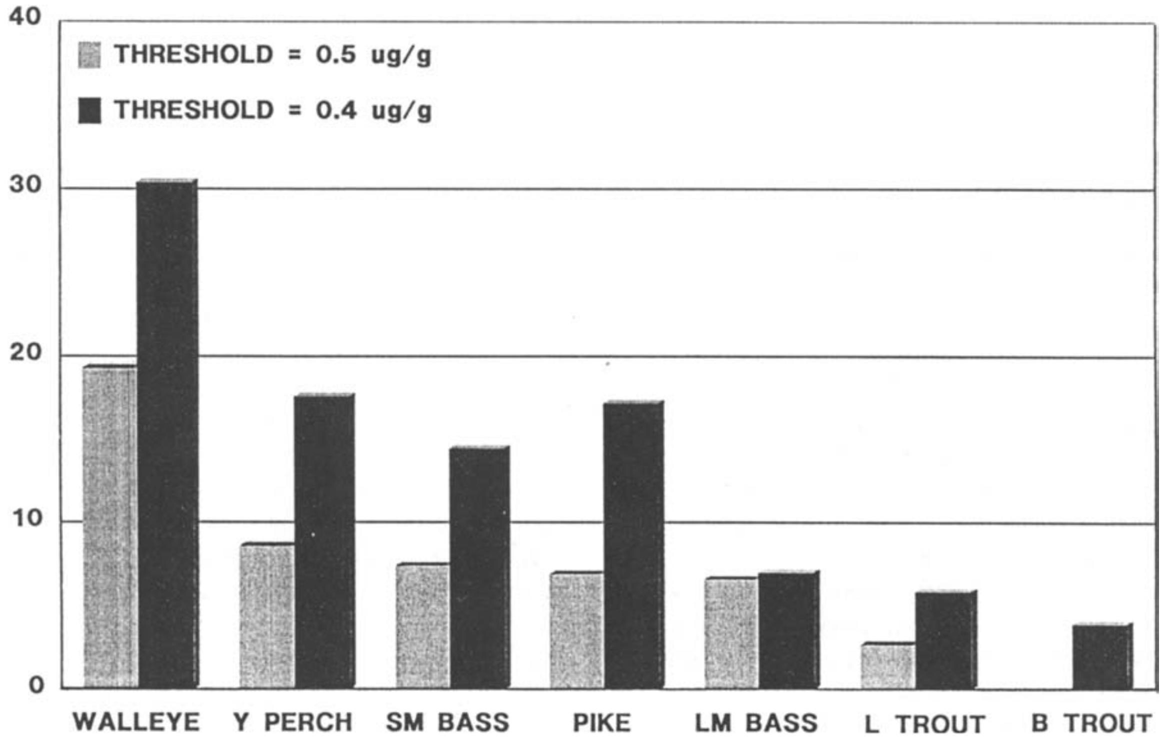

Fig. 2. Percent of Ontario lakes where mean Hg concentration in fish <250 g exceeds a specified threshold.

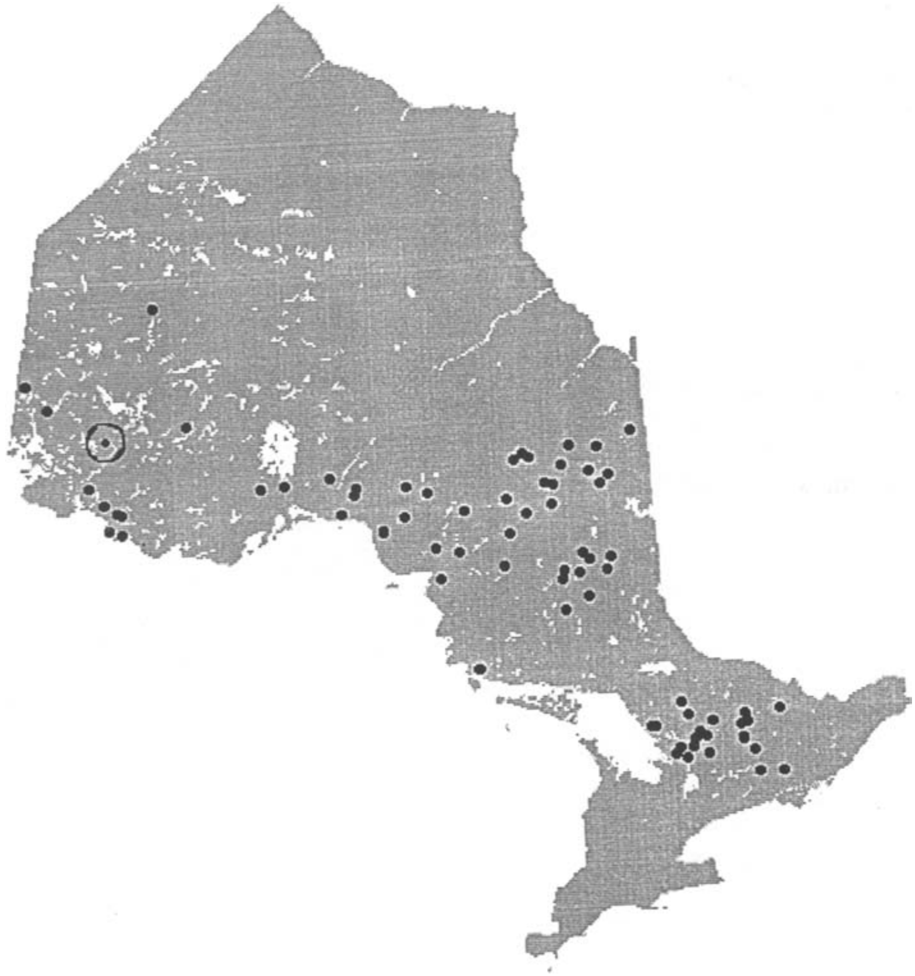

Fig. 3a. Locations of all lakes in the dataset having Hg concentration data for small walleye, pike, perch, and/or small and large-mouth bass. Circled point indicates the city of Dryden, Ontario, the location of a chlor-alkali plant responsible for Hg contamination of the English-Wabigoon river system studied by Barr (1986).

and largemouth bass-containing lakes exceeded threshold. A smaller proportion of lakes with lake trout ($<5\%$) exceeded threshold. There were no lakes in the dataset with small brook trout, whitefish, or white sucker containing mean Hg concentrations $>0.5\ \mu g\ g^{-1}$. (Note that the data presented in Fig. 2, although suggestive of differential Hg accumulation by small fish of different species, cannot be explicitly interpreted to depict differences in mean Hg concentrations among fish species because different species did not always co-occur on the same lakes. Different subsets of lakes could differ in their morphological and/or chemical characteristics, factors that in turn could influence Hg concentrations independently of species).

Figure 3a depicts the locations of all the lakes in the dataset having Hg concentration data for small (<250 g) perch, pike, small- and largemouth bass, and/or walleye, whereas Fig. 3b shows the locations of those lakes for which mean Hg in one or more of the listed species exceeded $0.4\ \mu g\ g^{-1}$.

Sudbury lakes

When data for small fish of all species were examined together, fish from Sudbury lakes had

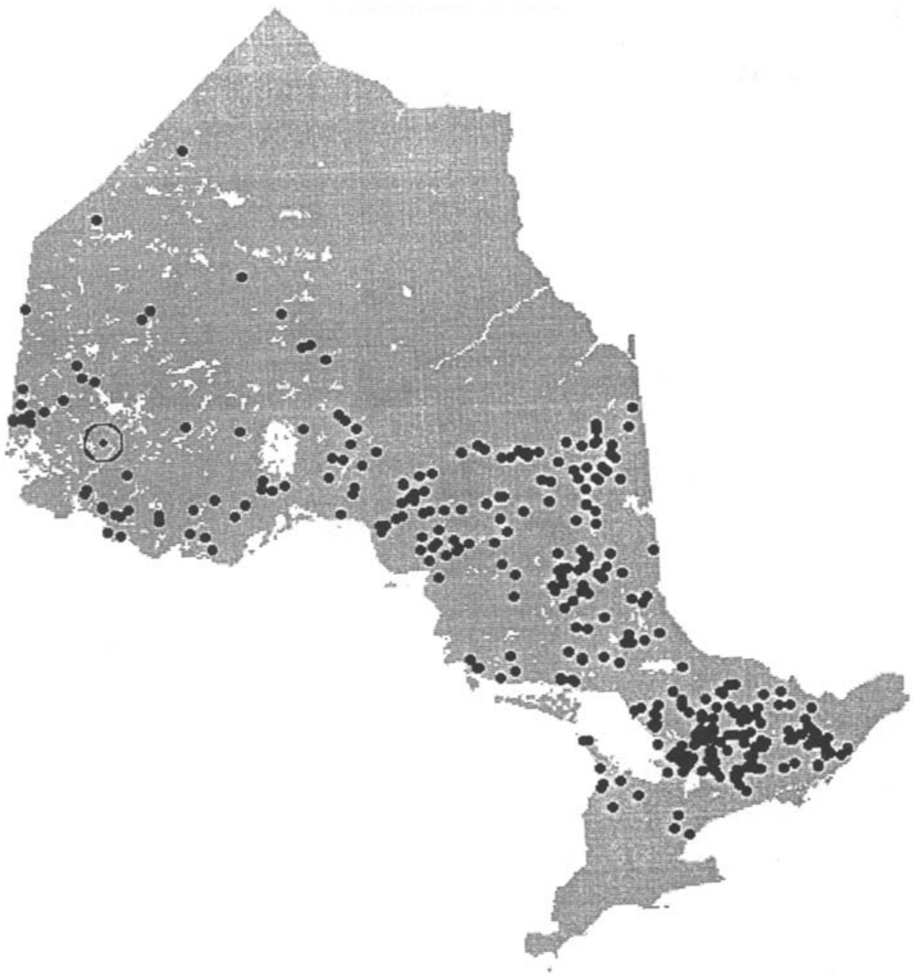

Fig. 3b. Locations of lakes where Hg concentrations in small fish of one or more of the indicated species averaged more than 0.4 $\mu g\ g^{-1}$. Circled point as in Fig. 3a.

mean Hg concentrations that were about 1/2 those of fish from other Ontario lakes in our dataset with statistically equivalent pH and DOC characteristics (Table 1). This difference also held for some individual species (lake trout, walleye; $p < 0.01$ [data not shown]). For some other species, sample sizes were not large enough to make confident statistical comparisons. For example, small yellow perch were sampled in only 5 Sudbury lakes, and in only 2 non-Sudbury lakes with similar pH and DOC. Nevertheless, all species tended to have higher Hg concentrations in the non-Sudbury lakes whether or not the

Table 1. Comparison of Hg concentrations in fish <250 g (all species combined) in Sudbury vs. non-Sudbury lakes having similar pH and DOC.

	n	pH	DOC	Fish-Hg[a]
Sudbury	22	6.7[b] (5.8–7.2)	3.1 (1.9–4.9)	0.21 (0.02–0.50)
Non-Sudbury	76	6.5 (5.8–7.3)	3.4 (1.9–4.3)	0.43[c] (0.07–1.61)

[a] $\mu g\ g^{-1}$ wet wt. in axial muscle.
[b] Values are means (ranges) of *n* lakes.
[c] Significantly different, $p < 0.01$.

Table 2. Slopes and overall R^2 from multiple linear regression analysis of fish-Hg vs. selected variables. (*) $p<0.05$; (**) $p<0.01$; (***) $p<0.001$.

Species (# of lakes)	pH	ln DOC	ln length	Latitude	Overall R^2
Walleye (155)	−0.208**	0.429***	1.632	−0.032	0.27
Pike (132)	−0.187**	0.393***	2.103***	−0.072*	0.37
Perch (15)	−0.385*	−1.077**	2.001**	0.148	0.66
SM bass (46)	−0.430***	0.337	1.413***	−0.152*	0.43
LM bass (8)	−0.066	1.698	1.717*	0.196	0.93
L trout (71)	−0.093	1.176***	1.233***	−0.041	0.60
B trout (18)	−0.236	0.750*	0.530	−0.041	0.35
Whitefish (15)	0.811*	0.194	−0.199	−0.399**	0.64
White sucker (15)	−0.647**	0.882*	2.456*	−0.031	0.63

difference was statistically significant. For this reason, Sudbury lakes (a total of 22 lakes) were deleted from the dataset before further statistical tests were performed.

Relationship between fish-Hg concentrations and selected variables

The results of the multiple regression analyses are presented in Table 2. There was a significant pos-

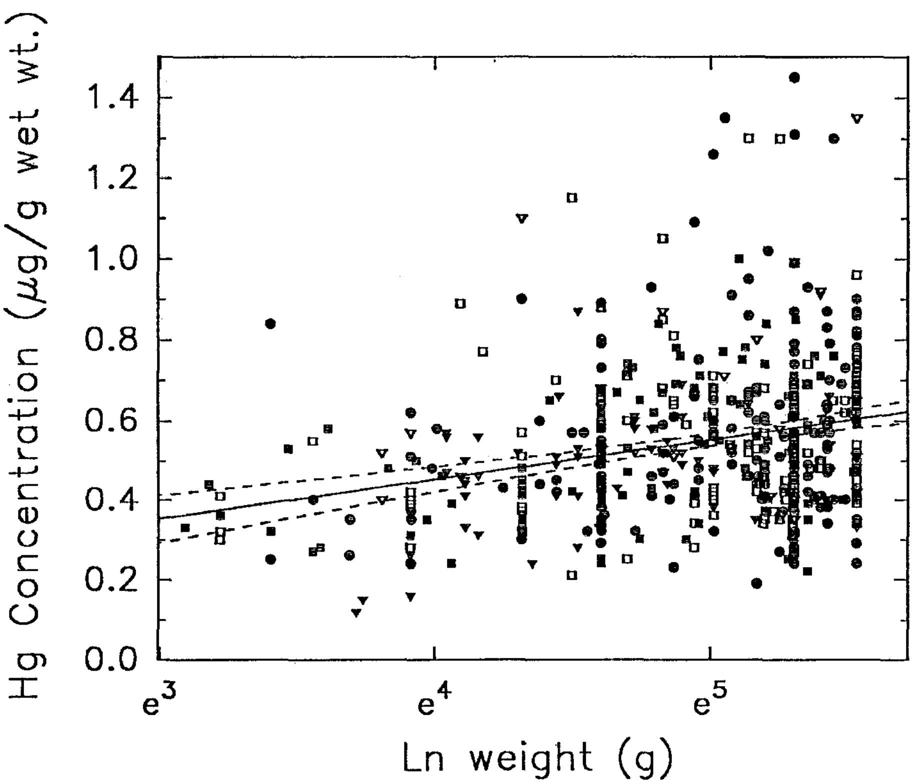

Fig. 4. Relationship between fish size (natural log weight) and Hg concentrations for 20–250 g walleye (●), pike (▼), perch (■), small-mouth bass (□), and lake trout (▽) from at-risk lakes. Data for individual fish ($n = 560$), and overall best-fit linear regression line for all data combined ($r = 0.27$, $p<0.01$) with 95% confidence intervals (dashed lines) are plotted.

itive correlation between fish length and fish-Hg concentration in six of nine species. There were significant negative correlations between lake pH and fish-Hg for walleye, pike, perch, smallmouth bass, and white sucker. Neither of the two trout species showed a significant relationship between mean Hg concentration and lake pH, rather Hg was positively associated with lake DOC in these 2 species. Hg in walleye, pike, and sucker showed both negative relationships with lake pH and positive relationships with lake DOC. Latitude was negatively correlated with mean fish-Hg in three species (pike, smallmouth bass, whitefish). Depending on the species, fish length, pH, DOC, and latitude together accounted for about 30–65% of the variability in Hg concentrations.

When all Hg data for small walleye, pike, perch, smallmouth bass, and lake trout from 'at-risk' lakes (lakes where mean Hg concentrations in small fish exceeded 0.4 $\mu g\ g^{-1}$) were considered, a significant correlation between fish size (weight or length) and fish-Hg was retained, but it was not the case that only heavier fish within the chosen size range had high Hg concentrations (Fig. 4). The overall mean weight of small fish (5 species combined) in at-risk lakes was 157 ± 64 g ($n = 560$), and the mean Hg concentration was $0.55 \pm 0.19\ g\ g^{-1}$ (wet wt). Ninety-five percent of at-risk lakes were of a size preferred by breeding loons (> 40 ha; Blancher *et al.*, 1992; Wayland & McNicol, 1990).

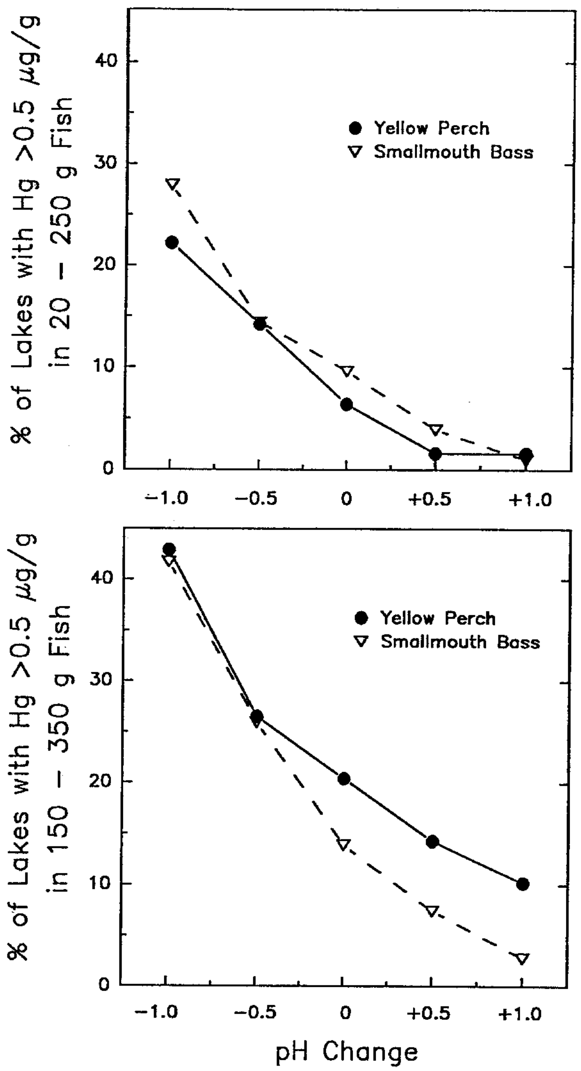

Fig. 5. Predicted changes in the percentage of Ontario lakes with perch or smallmouth bass of different size ranges having Hg concentrations above the threshold for reproductive impairment in loons in response to hypothetical changes in the mean pH of lakes.

Predicted effects of changes in lake pH and SO_4 deposition on fish-Hg concentrations

We used 2 fish species (perch and smallmouth bass) to predict changes in the proportion of lakes having small fish (20–250 g) with Hg concentrations higher than a given threshold level in response to hypothetical changes in the average pH of lakes in Ontario. From the overall regression analysis (Table 2), the calculated slopes of the ln Hg/pH relationships were −0.430 for smallmouth bass, and −0.385 for perch. As shown in Fig. 5, the predicted effect of decreasing the mean pH of perch or smallmouth bass lakes in Ontario by 0.5 pH units would be an approximate doubling of the percentage of lakes with fish-Hg concentrations above the threshold for loon reproductive toxicity. An increase in mean pH of 0.5 units would result in an amelioration of similar magnitude. For fish of a larger size range (150–350 g) such as might be consumed by larger predators (e.g. osprey, *Pandion haliaetus*), a similar but even more dramatic change in response to changes in mean pH of lakes is expected (Fig. 5).

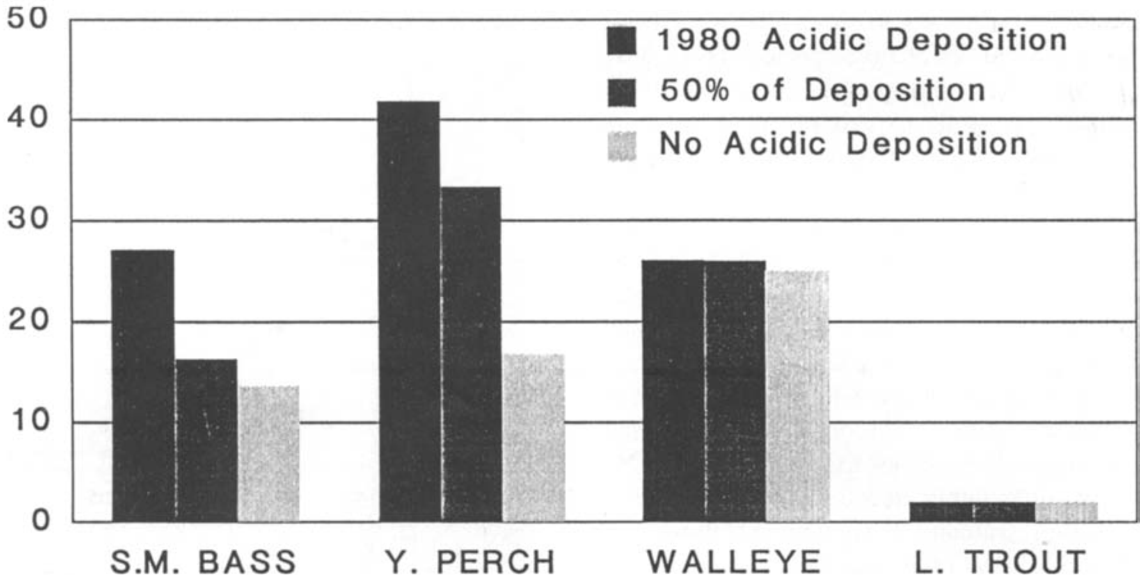

Fig. 6. Predicted changes in the percentage of Ontario lakes with small (20–250 g) smallmouth bass, perch, walleye, or lake trout having Hg concentrations $>0.5\ \mu g\ g^{-1}$ (wet wt.) in response to hypothetical changes in SO_4 deposition.

Changes in the pH of lakes can be predicted from changes in SO_4 deposition rates if the water chemistry characteristics of the lakes are known. When we used an acidification model developed for eastern Canada (Marmorek *et al.*, 1990) to predict changes in lake pH, and thence fish-Hg concentrations, in response to different scenarios of $S0_4$ deposition, we observed that mean Hg concentrations in small fish of certain species (e.g. – perch, smallmouth bass) were predicted to decrease substantially with decreasing SO_4 deposition rates, but that Hg concentrations in other species (e.g. – walleye, lake trout) would not respond significantly to decreased SO_4 loading in Ontario (Fig. 6).

Discussion

Several previously published reports have examined empirical relationships between Hg concentrations in fish and various physical and chemical variables in Ontario lakes (McMurtry *et al.*, 1989; Scheider *et al.*, 1979; Suns & Hitchin, 1990; Wren & MacCrimmon, 1983; Wren *et al.*, 1991). Two of these reports (McMurtry *et al.*, 1989; Wren *et al.*, 1991) used the same OMOE/OMNR dataset that we used in the present report. In those two previous studies, Hg data for 4 fish species (lake trout, walleye, smallmouth bass, and pike) of standardized lengths (31–54 cm, depending on the species) were examined. These standardized sizes are not within a size range used as food by most piscivorous wildlife, including loons. The present study, in which we specifically used Hg data from fish of a size range preferred as prey by adult loons (<250 g), demonstrated that a significant proportion of Ontario lakes have small fish whose Hg concentrations are sufficiently high to pose a risk to the reproductive success of loons, based on the reported reproductive toxicology of methylmercury in free-living loons (Barr, 1986). The highest proportion of lakes (20–30%) in which small fish exceeded the toxic threshold were lakes containing walleye. From 7–18% of perch-, smallmouth bass- and pike-containing lakes had small fish of those species that exceeded the threshold. Small fish of other species, such as trout, whitefish, and sucker, were seldom above the chosen thresholds.

Consistent with other studies that have reported anomalously low Hg concentrations in various biota in the Sudbury, Ontario vicinity (Scheuhammer 1991; Wren & Stokes, 1988; Wren *et al.*, 1986), fish from a group of 22 lakes in the Sudbury area had significantly lower Hg concentrations than fish from a group of 76 other Ontario lakes with pH and DOC characteristics statistically indistinguishable from the Sudbury lakes. The reasons for this phenomenon are not well understood, but could involve selenium (Se), an element known to have been emitted in high concentrations from Sudbury smelters (Nriagu & Wong, 1983), and known to inhibit the bioaccumulation of Hg by fish and aquatic invertebrates (Rudd *et al.*, 1980). For this reason, we chose to delete data from Sudbury lakes before performing further statistical analyses.

Although differences in water chemistry (pH, DOC) were significantly correlated with inter-lake differences in Hg concentrations in fish, the relative importance of these water chemistry parameters was species-specific (Table 2). Relationships similar to those listed in Table 2 between fish-Hg and pH or DOC have been reported for larger sized Ontario lake trout, smallmouth bass, pike, and walleye (McMurtry *et al.*, 1989; Wren *et al.*, 1991). The biogeochemical processes responsible for these relationships are still poorly understood; however it has been demonstrated that the production, and thus the availability, of methylmercury in the water column of lakes is enhanced at low pH (Miskimmin *et al.*, 1992; Xun *et al.*, 1987). It is also known that increasing the organic content of natural waters enhances the net bacterial production of methylmercury (Hecky *et al.*, 1991; Miskimmin *et al.*, 1992), and that humic substances likely mobilize Hg from watersheds (Mierle, 1990). Thus, variability in the Hg concentrations of fish can, in part, be explained by differences in water chemistry parameters in addition to biological factors such as the feeding ecology of the species, size and age differences among individual fish, growth rate differences, and differences in Hg excretion rates (Jackson, 1991).

A causal link between low alkalinity/low pH conditions and increased Hg accumulation in perch has been established (Wiener *et al.*, 1990a). Based on the calculated relationships between pH and Hg concentration in small fish of several species from the Ontario lakes dataset that we analysed (Table 2), we predicted that, for some fish species, an increase in the mean pH of lakes in the province would result in a substantially lower percentage of lakes having fish of those species with Hg concentrations above a toxic threshold (Fig. 5). Since the acidity of many Ontario lakes can be linked to patterns of anthropogenic acid SO_4 deposition (Neary & Dillon, 1988), we used a predictive model developed by Marmorek *et al.* (1990) to estimate how changing $S0_4$ deposition rates might ultimately affect Hg concentrations in small perch, bass, walleye, and lake trout in Ontario lakes. Our results indicate that different fish species will probably react differently to changing SO_4 deposition patterns (Fig. 6). The greatest response is predicted to occur in those species for which a significant negative correlation between pH and fish-Hg exists, and which are prevalent in lakes sensitive to acid deposition (e.g. – perch, smallmouth bass). Håkanson *et al.* (1990) performed similar simulations to predict the effect of changing sulfur emissions on Hg concentrations in Swedish pike and concluded that 35–40% of the occurrence of high Hg values ($> 1\ \mu g\ g^{-1}$) in standard size (1 kg) fish was likely the result of acidic deposition.

For walleye, we observed a negative relationship between pH and fish-Hg (Table 2), yet the acidification model predicted that decreasing SO_4 emissions would not result in fewer lakes containing walleye with high Hg concentrations (Fig. 6). This is because the small walleye for which we had Hg concentration data were sampled mainly from high alkalinity, high pH lakes (Fig. 1). The acidification model predicts that lakes with water chemistry characteristics of this nature will not respond, or will respond minimally, to increasing or decreasing SO_4 deposition. The Hg content of walleye in low alkalinity, acidified lakes, however, should respond to decreasing SO_4 deposition in a manner similar to that of perch or bass. For other species, such as lake trout, that failed to show a significant relationship between fish-Hg and lake pH, the acidification model again predicts a lack of effect of changing SO_4 deposition patterns on the percentage of lakes containing small fish with Hg concentrations exceeding a toxic threshold.

In conclusion, the results of the present investigation indicate that: 1) a significant proportion of Ontario lakes in which walleye, pike, perch, and smallmouth bass are found contain small fish (<250 g) of these species with Hg concentrations greater than levels linked to reproductive dysfunction in freeliving loons; 2) Hg concentrations in small fish of these species are negatively correlated with lake pH; 3) 'at-risk' lakes are of a size class preferred by breeding loons (>40 ha); and 4) decreases in anthropogenic SO_4 emissions are predicted to result in a decrease in the proportion of lakes in the province having fish, including those species preferred as prey by loons [e.g. perch; Barr, (1973)], with high Hg concentrations. Behavioral and reproductive studies of breeding loons in Ontario are needed to determine if an elevated exposure to methylmercury, and/or a significant impairment of reproduction is occurring on 'high Hg' lakes.

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