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Water striders (family Gerridae): mercury sentinels in small freshwater ecosystems

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Water striders accurately reflect the entry of mercury in food chains of small freshwater systems.

Abstract

To circumvent some of the previous limitations associated with contaminant-monitoring programs, we tested the suitability of the water strider (Hemiptera: Gerridae) as a mercury sentinel by comparing total mercury concentrations in water striders and brook trout (*Salvelinus fontinalis*) from a variety of stream sites in New Brunswick, Canada. There was a strong association between the two variables across sites ($r^2 = 0.81$, $P < 0.001$) in systems where both atmospheric deposition and a point source (an abandoned gold mine) were likely contributing to ambient mercury levels. In a small stream draining the gold mine tailings pile, water striders had mercury concentrations an order of magnitude higher than those from reference locations. Temporal variation at three southern New Brunswick stream sites was non-significant. These results suggest that water strider mercury levels accurately quantify food chain entry of the element. The use of sentinel species holds great potential for expanding contaminant-monitoring programs.

Keywords: Sentinel; Brook trout; Streams; Gold mine; Atmospheric deposition

1. Introduction

Given that concentrations of contaminants, such as mercury, have increased in certain areas of the world over the past century (Schuster et al., 2002), there is a growing need to identify geographic areas of concern and associated ecological pathways (Pilgrim et al., 2001). To achieve these goals, it is necessary to have consistent measurement indices, allowing comparisons to be made locally, regionally, and globally. One approach to unifying otherwise diverging monitoring efforts is through the use of sentinel species (Beeby, 2001). Sentinel species accurately reflect ambient contaminant concentrations, and can be routinely and conveniently collected and analyzed as part of monitoring programs. In this regard, invertebrate species are commonly used as environmental sentinels, as exemplified by extensive monitoring of contaminant levels in marine bivalves (O'Connor, 1998; Rebelo et al., 2003). In freshwater, shellfish have also been employed to measure contaminant concentrations along perceived

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gradients (Cope et al., 1999). However, shellfish are rarely abundant in small headwater streams, in which case benthic macroinvertebrates have been utilized. Some limitations surrounding the use of stream benthos include difficulty in taxonomic resolution and the attainment of adequate tissue for analysis, reduced distribution across sites of interest (Cain et al., 1992), and poor relationships between concentrations in insects and direct contaminant sources (Croteau et al., 2003).

Water striders (Hemiptera: Gerridae) are relatively long-lived, predatory invertebrates that feed on aquatic and terrestrial insects (Pennak, 1978). They meet much of the criteria outlined by Beeby (2001) as appropriate sentinel (accumulator) species. These include: ubiquity and abundance, large body size, long life span, ease of identification and ageing, and a relatively sedentary lifestyle. They occur over a wide range in North America and the world, and are abundant in many pond, lake, and river ecosystems. Each generation can survive up to 1 year (Merritt and Cummins, 1996), and wingless forms show low mobility during summer months (Wilcox and Maier, 1991).

Mercury build-up in aquatic food chains can, in part, result from atmospheric deposition (Bodaly et al., 1993) and from local point sources due to local geological deposits, or be introduced by industrial and residential processes and mining activities (Mol et al., 2001). Many such sources are present in Canada's Atlantic provinces (Pilgrim et al., 2000). In this study, we attempted to calibrate and validate water striders as an environmental sentinel (Beeby, 2001) by relating mercury levels in the family to those of small brook trout, an important recreational species, in New Brunswick streams. We also sought to determine the ability of water striders to accumulate high mercury levels (i.e. an order of magnitude above comparable reference sites) to evaluate their potential for future use in identifying mercury areas of concern on a local, regional, and global scale.

2. Materials and methods

Water striders and brook trout were collected simultaneously from small streams in New Brunswick, Canada from May to July 2002 and 2003 (Table 1, Fig. 1). Water striders were captured with D-frame kick nets, while brook trout were fished with a backpack electroshocker. Collection sites were generally second and third order streams approximately 1–5 m wide. HAB, MCB, and PLB are small coastal basins (<50 km^2) along the north shore of the Bay of Fundy. DCB, EAB, GHB, and TRB are small streams on Grand Manan, an island 35 km off southern New Brunswick in the Bay of Fundy. 18A, 18M, 18B, GCU, and SEU are stream sections in the headwaters of the Southeast Upsalquitch River in northern New Brunswick, and drain a former gold mine tailings pile. Water striders were also captured opportunistically at two locations (DEB and NWA) in the Little Southwest and Northwest Miramichi River, respectively, in north central New Brunswick (Table 1).

Water striders were analyzed whole or pooled (two to 20 individuals) for a single analysis and brook trout muscle tissue or whole bodies were analyzed depending on size (Table 1). Total mercury (μg/g dry weight) levels were measured using cold vapour atomic fluorescence spectroscopy (Tekran, model 2600) after freeze-drying for 48 h, 7:3 nitric/sulphuric acid digestion, and heating (90 °C for 4h). QA/QC was monitored using internationally accepted standards (Horvat et al., 1997). Each run of 48 analyses included four blanks, four HgCl standards, four DORM-2 standards, and samples. Runs were accepted if sample peaks were greater than four times the blank peaks and standards were within 10% of their relative standard deviations. Samples were corrected based on DORM-2 conversion. Ranges of 95–98% conversions were found for spiked samples. Sample replicates varied by less than 10%.

A 500 ml surface water sample was collected at a subset of sites directly into a clean fluoropolymer bottle following sample handling techniques designed for collection of mercury at trace levels (US EPA, 1996). A 0.45 μm, 15 mm diameter capsule filter (*Aquaprep® 12175*), fluoropolymer tubing and a peristaltic pump were used for sampling. Samples were preserved with 0.5% BrCl solution immediately upon collection. Field blanks with De-Ionized water and the same reagents were obtained for every set of samples. Total mercury concentrations were determined following US EPA Method 1631B (US EPA, 1999). Quality was assured through calibration and testing of the oxidation, purging and detection system. Surface water samples were analyzed for trace levels of mercury at the class-100 mercury analytical laboratory of Fisheries and Oceans Canada in Dartmouth, Nova Scotia. Samples having high (>0.5 μg/l) total mercury concentrations were analyzed at the University of New Brunswick on a cold-vapor atomic absorption spectrometer following the same procedure.

All statistical analyses were conducted using SYSTAT 9.0 software (SPSS, Inc.). Correlations of water strider total mercury levels with brook trout total mercury levels and water mercury levels were analyzed using linear regression. To reduce heterogeneity of variance across sites, water strider, brook trout and water mercury levels were log-transformed for tests involving the site draining the gold mine tailings pile. One-way analysis of variance (ANOVA) was used to compare mercury levels in water striders at three sites potentially influenced by the gold mine tailings with

Table 1
Sites and dates of brook trout (n, number of individuals analyzed) and water strider (n, number of composite samples analyzed with individuals per sample in parentheses) collections in New Brunswick streams

Site	Abbreviation	Date	n	FL (mm)	Brook trout regression equation	r^2	Gerridae, n
Hay Brook	HAB	21-May-03	5	142 ± 22	Hg = −0.0072 × FL + 2.3748	0.05	3 (1)
		*7-Jul-03	3	137 ± 34	–	–	3 (3)
McLaughlin Brook	MCB	20-May-03	2	73 ± 31	–	–	4 (2)
		*7-Jul-03	3	170 ± 27	–	–	7 (2–4)
Point Lepreau Brook	PLB	20-May-03	5	87 ± 39	Hg = 0.0013 × FL + 0.1657	0.71	2 (1)
		*7-Jul-03	5	152 ± 17	Hg = −0.0008 × FL + 0.4724	0.04	3 (2–3)
Deep Cove Brook	DCB	*9-Jul-03	5	122 ± 17	Hg = 0.0078 × FL − 0.0062	0.76	7 (2–4)
East Brook	EAB	*9-Jul-03	5	156 ± 35	Hg = 0.0001 × FL + 0.4596	<0.01	4 (2–6)
Grand Harbour Brook	GHB	*9-Jul-03	4	115 ± 49	–	–	6 (12–20)
Trout Brook	TRB	*10-Jul-03	4	112 ± 11	–	–	6 (4–5)
18-Mile Brook above Gossan Creek	18A	27-Jun-02	15	106 ± 27	Hg = 0.0005 × FL + 0.1404	0.05	NC
		*28-Jun-03	5	117 ± 15	Hg = −0.0018 × FL + 0.5877	0.05	8 (2)
18-Mile Brook at mouth of Gossan Creek	18M	27-Jun-02	18	129 ± 30	Hg = 0.0012 × FL + 0.2056	0.08	NC
		*28-Jun-03	5	117 ± 21	Hg = 0.0011 × FL + 0.6761	0.02	6 (2–3)
18-Mile Brook below Gossan Creek	18B	27-Jun-02	15	95 ± 19	Hg = 0.0025 × FL + 0.0906	0.27	NC
		*28-Jun-03	5	76 ± 21	Hg = −0.0015 × FL + 0.7089	0.03	9 (2–3)
Gossan Creek—near tailings	GCU	28-Jun-03	NP	NP	–	–	3 (2)
Southeast Upsalquitch River	SEU	27-Jun-02	14	92 ± 15	Hg = 0.0045 × FL + 0.1459	0.24	NC
Little Southwest Miramichi—Devil's Brook	DEB	12-Jun-03	NC	NC	–	–	5 (2–3)
Northwest Miramichi—above salmon barrier	NWA	5-Jun-03	NC	NC	–	–	4 (2)

Mean fork lengths (± 1 SD) and regression equations describing mercury variability with fish size are included.
NC, not collected; NP, not present at site; asterisks indicate samples used in trout vs. strider regression.

three reference sites. Linear regression analysis was performed on mercury concentrations in brook trout and water striders in relation to distance downstream from the gold mine tailings pile, and on water striders in relation to water mercury levels. A general linear model (GLM) with three factors was used to determine the effect of sampling period (May vs. July) and site (HAB, MCB, PLB) on water strider and brook trout mercury levels. To test for interaction, the model was initially run with all possible interactions (including three-way). Interaction terms were removed in a stepwise fashion by excluding in order of significance level (i.e. highest P value first) and the model was re-analyzed with remaining terms. Interactions were considered non-significant at $\alpha = 0.05$.

3. Results

Associations between brook trout fork length (typically used as a covariate; Somers and Jackson, 1993) and mercury levels were generally weak, with low r^2 values and conflicting slope directions at given sites (Table 1), possibly reflecting the relatively small size range of trout from collection sites (range 35–201 mm for all sites). Therefore, we used mean brook trout mercury concentrations as representative of each particular site.

There was a strong correlation between water strider total mercury levels and brook trout total mercury levels ($r^2 = 0.81$, $P < 0.001$, Fig. 2). Highest concentrations in both trout and striders were measured in Hay Brook (HAB), a second order stream on the Fundy mainland coast of southwest New Brunswick. Lowest concentrations were found at Point Lepreau Brook (PLB, a second order stream located $\sim$10 km southwest of HAB) and at 18-Mile Brook above the confluence of Gossan Creek (18A, a pristine site located in northern New Brunswick). Intermediate values were found in stream sites on Grand Manan (DCB, EAB, GHB, TRB), an island 35 km from mainland New Brunswick in the Bay of Fundy.

Immediately downstream of the gold mine tailings pile in a first order steam (GCU) at the headwaters of the Southeast Upsalquitch River in northern New Brunswick, we were unable to capture any fish species or benthic macroinvertebrates, despite comparable efforts electrofishing and kick netting, respectively, relative to other sites. The only taxon present at GCU was surface dwelling water striders, and mercury concentrations in these striders were an order of magnitude higher than those at reference sites (Fig. 3). Mercury concentrations in brook trout and water striders decreased in a downstream direction away from the tailings pile ($r^2 = 0.38$, $P < 0.001$, Fig. 3). However, when comparing water striders from the mine-affected reach (GCU, 18M, 18B) with local (18A) and regional (DEB, NWA, both in the Miramichi, a neighbouring basin) reference sites (Fig. 4), only those water striders

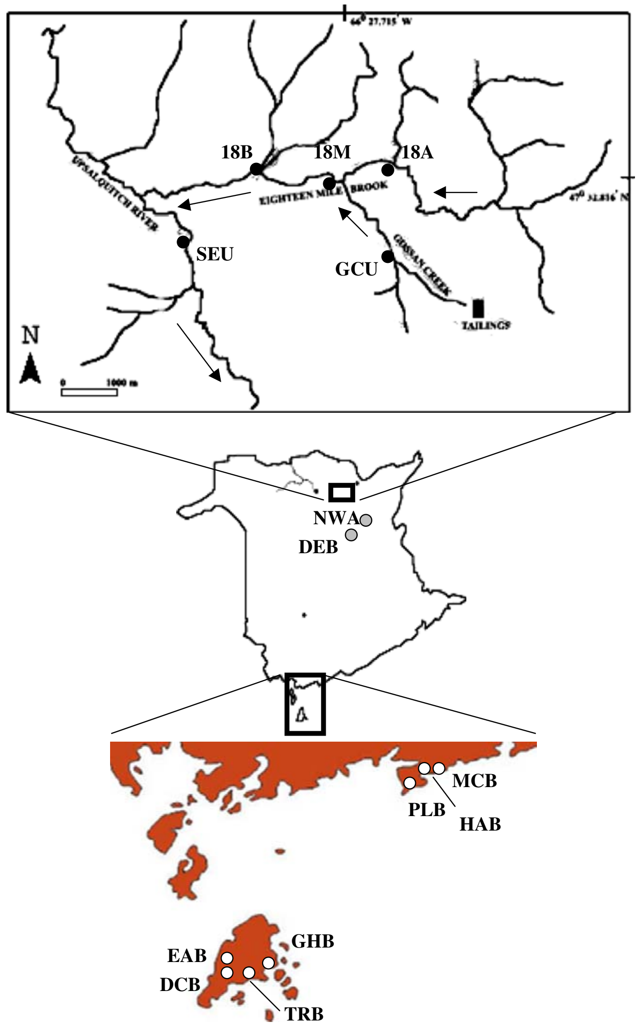

Fig. 1. Map of the two main study sites including the area surrounding a former gold mine operation (top panel) and the outer Bay of Fundy (bottom panel) in New Brunswick, Canada. Arrows indicate the direction of water flow. Sampling locations are marked with circles. See Table 1 for site codes.

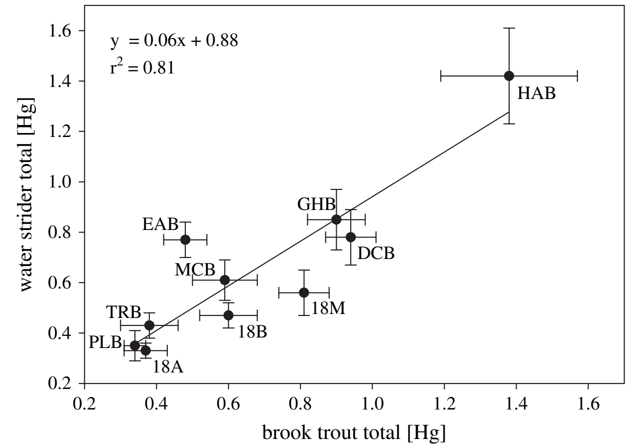

Fig. 2. Correlation between total mercury levels (μg/g dry weight) in small brook trout and water striders in New Brunswick streams, measured in late June and early July 2003. Each data point represents the mean (±1 standard error) of three to nine analyses. See Table 1 for site codes.

in the stream section immediately below the tailings pile (GCU) had significantly higher mercury levels ($P < 0.05$).

Water strider mercury levels were also associated with water mercury levels ($r^2 = 0.68$, $P = 0.006$, Fig. 5), due largely to the high water and Gerrid mercury levels in the stream section immediately below the gold mine tailings pile (GCU).

Three sites in southern New Brunswick (HAB, MCB, PLB) were used to test temporal variation in the relationship between water strider and brook trout mercury levels. The effects of taxon ($F = 0.342$, df = 1, $P = 0.562$) and sampling period ($F = 0.978$, df = 1, $P = 0.377$) were non-significant, but differences across the three streams were identified ($F = 4.729$, df = 2, $P = 0.014$, Fig. 5). HAB trout and striders had consistently highest mercury levels, with MCB trout and striders intermediate and PLB lowest (Fig. 6). No interaction terms were significant.

4. Discussion

The strong correlation between mercury levels in water striders and small brook trout indicates that water striders are an appropriate sentinel in determining the entry of mercury in food chains of small lotic systems. The low temporal variation in the relationship also provides evidence that point-in-time estimates can provide a reliable indication of the overall pattern in the system of study. Furthermore, the ability of water striders to survive in hostile environments and accumulate high levels of mercury provides compelling evidence that the

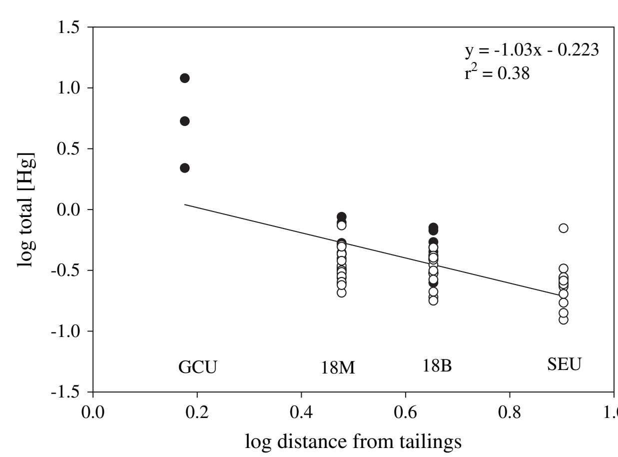

Fig. 3. Log-scale total mercury levels in water striders (solid circles) and brook trout (open circles) vs. distance downstream from a gold mine tailings pile. The solid line indicates best-fit linear regression for all data. See Table 1 for site codes.

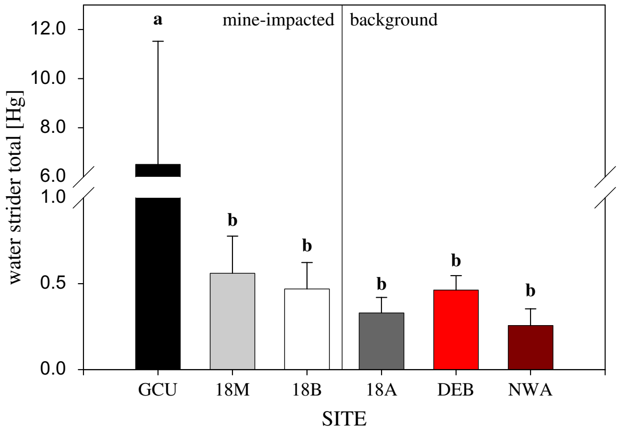

Fig. 4. Water strider total mercury levels (μg/g dry weight) at three sites below a gold mine tailings pile (GCU, 18M, 18B) and at three reference sites (18A, DEB, NWA). Different letters indicate significant differences at $\alpha = 0.05$ (one-way ANOVA). See Table 1 for site codes.

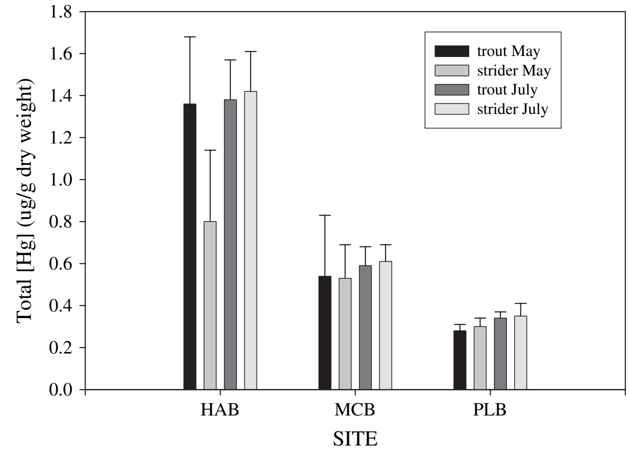

Fig. 6. Brook trout and water strider total mercury levels (μg/g dry weight) at three Fundy coastal sites (Hay Brook, McLaughlin Brook, and Point Lepreau Brook) collected on two sampling dates (May 20–21 and July 7, 2003). See Table 1 for site codes.

family is tolerant of adverse conditions and may not be able to regulate its contaminant burden, both important characteristics in choosing an environmental sentinel (Beeby, 2001).

Nummelin et al. (1998) previously concluded that water striders were suitable as bioindicators of heavy metal accumulation. These authors found differences in metal levels (Al, Fe, Mn, Zn, Cu, Cd) in striders in close proximity to a steel factory compared with reference locations. However, there was no comparison made as to how these levels matched concentrations in other matrices (e.g. water or another taxa), such as in this study.

Despite a decreasing trend of mercury concentrations in water striders and brook trout with distance from the point source of contamination (the gold mine tailings pile), mercury levels in the upper reaches of the Southeast Upsalquitch River drainage (18M, 18B) were similar to reference sites both locally (18A) and regionally (DEB, NWA). Hence the tailings contamination seems to have a localized zone of influence where mercury levels are well above background and no biota

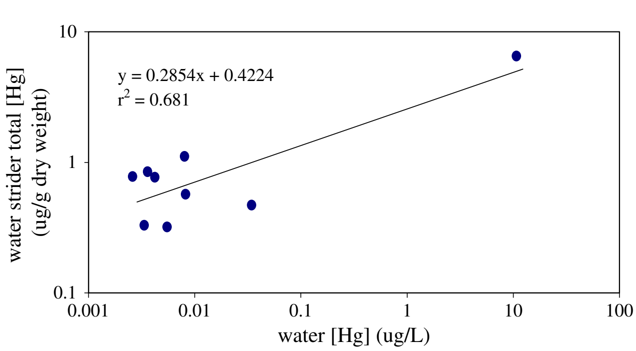

Fig. 5. Correlation between total mercury levels (μg/g dry weight) in water striders and stream water (μg/l) in New Brunswick, Canada, plotted on log-log scale.

(save for water striders) can be supported. High mercury levels have been shown to cause growth and reproductive impairments in fish (Friedmann et al., 1996; Hammerschmidt et al., 2002; Latif et al., 2001). The confounding influence of acidity in the stream immediately below the tailings pile is a potential factor governing the lack of a benthic assemblage and the presence of fish species. Acid environments have been shown to adversely affect brook trout populations (Lachance et al., 2000) and stream benthos (Courtenay and Clements, 1998). However, throughout much of its length, the pH of the stream is circumneutral (T.A. Al, unpublished data) and therefore not likely limiting colonization by invertebrates. Concentrations of other dissolved metals (Cu, Zn, Cd, Pb) are highly elevated in the stream (T.A. Al, unpublished data). These metals have been implicated previously in adversely impacting stream benthos (David, 2003; Quinn et al., 2003).

Local site differences in mercury concentrations of biota inhabiting adjacent streams draining to the Bay of Fundy, as found in our study, may be related to physical variables such as water colour/dissolved organic carbon concentrations (Watras et al., 1998), pH (Scheuhammer and Graham, 1999), and catchment characteristics such as forest cover and wetland type (Balogh et al., 2002; St. Louis et al., 1996, 2001).

The near one-to-one ratio observed between water strider and trout mercury levels raises questions about the mechanisms governing contaminant uptake in the two unrelated taxa. Feeding strategies may be similar, with a large component of the diet made up of terrestrial invertebrates (Pennak, 1978; Scott and Crossman, 1973). Stable isotope analyses of carbon ($^{13}C/^{12}C$) and nitrogen ($^{15}N/^{14}N$) ratios may therefore provide information on the comparative feeding ecology of the two groups (Peterson and Fry, 1987). The route of mercury exposure for fish is largely through the diet, with a small component entering via the gills (Hall et al., 1997). The route of uptake for water striders is unknown, but in highly contaminated environments, such as the stream draining the gold mine tailings pile (current study), airborne exposure may be a source of mercury as it evades off the surface of the water. The absolute amount of methyl-mercury is also likely greater in brook trout compared to water striders because fish generally have higher methyl-mercury to total-mercury ratios than do invertebrates (Cope et al., 1999; Hammerschmidt et al., 1999). Nummelin et al. (1998) also found differences in the accumulation of metals in water striders based on species and development stage. Striders in our study were not tested separately for the influence of these variables, and future research may involve analyses of this type.

5. Conclusions

We have demonstrated in this study the generality of the Gerridae family in its ability to reflect environmental conditions. While water striders possess a series of characteristics considered optimal for their classification as a sentinel species (Beeby, 2001), the key to their utility lies in their easy identification and capture. This characteristic will allow a wide range of investigators (government, university, community) to collect and submit samples for analysis, with great potential for the detection of areas with elevated mercury levels where risks to humans and other consumers of aquatic resources may be high (Chan et al., 2003; Risher et al., 2002).

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References

Balogh, S.J., Huang, Y., Offerman, H.J., Meyer, M.L., Johnson, D.K., 2002. Episodes of elevated methylmercury concentrations in Prairie streams. Environmental Science and Technology 36, 1665–1670.

Beeby, A., 2001. What do sentinels stand for? Environmental Pollution 112, 285–298.

Bodaly, R.A., Rudd, J.W.M., Fudge, R.J.P., Kelly, C.A., 1993. Mercury concentrations in fish related to size of remote Canadian Shield lakes. Canadian Journal of Fisheries and Aquatic Sciences 50, 980–987.

Cain, D.J., Luoma, S.N., Carter, J.L., Fend, S.V., 1992. Aquatic insects as bioindicators of trace element contamination in cobble-bottom rivers and streams. Canadian Journal of Fisheries and Aquatic Sciences 49, 2141–2154.

Chan, H.M., Scheuhammer, A.M., Ferran, A., Loupelle, C., Holloway, J., Weech, S., 2003. Impacts of mercury on freshwater fish-eating wildlife and humans. Human and Ecological Risk Assessment 9, 867–883.

Cope, W.G., Bartsch, M.R., Rada, R.G., Balogh, S.J., Rupprecht, J.E., Young, R.D., Johnson, D.K., 1999. Bioassessment of mercury, cadmium, polychlorinated biphenyls, and pesticides in the Upper Mississippi River with zebra mussels (*Dreissena polymorpha*). Environmental Science and Technology 33, 4385–4390.

Courtenay, L.A., Clements, W.H., 1998. Effects of acidic pH on benthic macroinvertebrate communities in stream microcosms. Hydrobiologia 379, 135–145.

Croteau, M.-N., Hare, L., Tessier, A., 2003. Difficulties in relating Cd concentrations in the predatory insect *Chaoborus* to those of its prey in nature. Canadian Journal of Fisheries and Aquatic Sciences 60, 800–808.

David, C.P.C., 2003. Establishing the impact of acid mine drainage through metal bioaccumulation and taxa richness of benthic insects in a tropical Asian stream (The Philippines). Environmental Toxicology and Chemistry 22, 2952–2959.

Friedmann, A.S., Watzin, M.C., Brinck-Johnsen, T., Leiter, J.C., 1996. Low levels of dietary methylmercury inhibit growth and gonadal development in juvenile walleye (*Stizostedion vitreum*). Aquatic Toxicology 35, 265–278.

Hall, B.D., Bodaly, R.A., Fudge, R.J.P., Rudd, J.W.M., Rosenberg, D.M., 1997. Food as the dominant pathway of methylmercury uptake by fish. Water, Air, and Soil Pollution 100, 13–24.

Hammerschmidt, C.R., Wiener, J.G., Frazier, B.E., Rada, R.G., 1999. Methylmercury content of eggs in yellow perch related to maternal exposure in four Wisconsin lakes. Environmental Science and Technology 33, 999–1003.

Hammerschmidt, C.R., Sandheinrich, M.B., Wiener, J.G., Rada, R.G., 2002. Effects of dietary methylmercury on reproduction of fathead minnows. Environmental Science and Technology 36, 877–883.

Horvat, M., Liang, L., Azemard, S., Mandie, V., Villeneuve, J.-P., Coquery, M., 1997. Certification of total mercury and methylmercury concentrations in mussel homogenate (*Mytilus edulis*) reference material, IAEA-142. Fresenius Journal of Analytical Chemistry 358, 411–418.

Lachance, S., Berube, P., Lemieux, M., 2000. In situ survival and growth of three brook trout (*Salvelinus fontinalis*) strains subjected to acid conditions of anthropogenic origin at the egg and fingerling stages. Canadian Journal of Fisheries and Aquatic Sciences 57, 1562–1573.

Latif, M.A., Bodaly, R.A., Johnston, T.A., Fudge, R.J.P., 2001. Effects of environmental and maternally derived methylmercury on the embryonic and larval stages of walleye (*Stizostedion vitreum*). Environmental Pollution 111, 139–148.

Merritt, R.W., Cummins, K.W., 1996. An Introduction to the Aquatic Insects of North America. Kendall/Hunt, Dubuque, IA.

Mol, J.H., Ramlal, J.S., Lietar, C., Verloo, M., 2001. Mercury contamination in freshwater, estuarine, and marine fishes in relation to small-scale gold mining in Suriname, South America. Environmental Research 86A, 183–197.

Nummelin, M., Lodenius, M., Tulisalo, E., 1998. Water Striders (Heteroptera, Gerridae) as bioindicators of heavy metal pollution. Entomologica Fennica 8, 185–191.

O'Connor, T.P., 1998. Mussel watch results from 1986 to 1996. Marine Pollution Bulletin 37, 14–19.

Pennak, R.W., 1978. Fresh-Water Invertebrates of the United States. Wiley, New York.

Peterson, B.J., Fry, B., 1987. Stable isotopes in ecosystem studies. Annual Review of Ecology and Systematics 18, 293–320.

Pilgrim, W., Poissant, L., Trip, L., 2000. The Northeast states and Eastern Canadian provinces mercury study: a framework for action: summary of the Canadian chapter. The Science of the Total Environment 261, 177–184.

Pilgrim, W., Eaton, P., Trip, L., 2001. The need for integrated linkages and long-term monitoring of mercury in Canada. Environmental Monitoring and Assessment 67, 57–68.

Quinn, M.R., Feng, X., Folt, C.L., Chamberlain, C.P., 2003. Analyzing trophic transfer of metals in stream food webs using nitrogen isotopes. The Science of the Total Environment 317, 73–89.

Rebelo, M.D., do Amaral, M.C.R., Pfeiffer, W.C., 2003. High Zn and Cd accumulation in the oyster *Crassostrea rhizophorae*, and its relevance as a sentinel species. Marine Pollution Bulletin 46, 1354–1358.

Risher, J.F., Murray, E.H., Prince, G.R., 2002. Organic mercury compounds: human exposure and its relevance to public health. Toxicology and Industrial Health 18, 109–160.

Scheuhammer, A.M., Graham, J.E., 1999. The bioaccumulation of mercury in aquatic organisms from similar lakes with differing pH. Ecotoxicology 8, 49–56.

Schuster, P.F., Krabbenhoft, D.P., Naftz, D.L., Cecil, L.D., Olson, M.L., Dewild, J.F., Susong, D.D., Green, J.R., Abbott, M.L., 2002. Atmospheric mercury deposition during the last 270 years: A glacial ice core record of natural and anthropogenic sources. Environmental Science and Technology 36, 2303–2310.

Scott, W.B., Crossman, E.J., 1973. Freshwater Fishes of Canada. Fisheries Research Board of Canada, Ottawa, Ontario.

Somers, K.M., Jackson, D.A., 1993. Adjusting mercury concentrations for fish-size covariation: a multivariate alternative to bivariate regression. Canadian Journal of Fisheries and Aquatic Sciences 50, 2388–2396.

St. Louis, V.L., Rudd, J.W.M., Kelly, C.A., Beaty, K.G., Flett, R.J., Roulet, N.T., 1996. Production and loss of methylmercury and loss of total mercury from boreal forest catchments containing different types of wetlands. Environmental Science and Technology 30, 2719–2729.

St. Louis, V.L., Rudd, J.W.M., Kelly, C.A., Hall, B.D., Rolfhus, K.R., Scott, K.J., Lindberg, S.E., Dong, W., 2001. Importance of the forest canopy to fluxes of methyl mercury and total mercury to boreal ecosystems. Environmental Science and Technology 35, 3089–3098.

US EPA, 1999. Method 1631: Mercury in water by oxidation, purge and trap, and CVAFS. US Environmental Protection Agency, Office of Water, Office of Science and Technology, Engineering and Analysis Division, Washington, DC.

US EPA, 1996. Method 1669: Method for sampling ambient water for determination of metals at EPA ambient criteria levels. US Environmental Protection Agency, Office of Water, Office of Science and Technology, Engineering and Analysis Division (4303), Washington, DC.

Watras, C.J., Back, R.C., Halvorsen, S., Hudson, R.J.M., Morrison, K.A., Wente, S.P., 1998. Bioaccumulation of mercury in pelagic freshwater food webs. The Science of the Total Environment 219, 183–208.

Wilcox, R.S., Maier, H.A., 1991. Facultative estivation in the water strider *Gerris remigis*. Canadian Journal of Zoology 69, 1412–1413.