Ecosystem Responses to Mercury Contamination

Indicators of Change

Based on the SETAC North America Workshop on Mercury Monitoring and Assessment

14-17 September 2003
Pensacola, Florida, USA

Edited by
Reed Harris • David P. Krabbenhoft
Robert Mason • Michael W. Murray
Robin Reash • Tamara Saltman

Coordinating Editor of SETAC Books
Joseph W. Gorsuch
Gorsuch Environmental Management Services, Inc.
Webster, New York, USA

CRC Press
Taylor & Francis Group
Boca Raton London New York

CRC Press is an imprint of the
Taylor & Francis Group, an informa business

Published in collaboration with the Society of Environmental Toxicology and Chemistry (SETAC)
1010 North 12th Avenue, Pensacola, Florida 32501
Telephone: (850) 469-1500 ; Fax: (850) 469-9778; Email: setac@setac.org
Web site: www.setac.org
ISBN-10: 1-880611-86-4 (SETAC Press)
ISBN-13: 978-1-58488-661-7 (SETAC Press)

No claim to original U.S. Government works
Printed in the United States of America on acid-free paper
10 9 8 7 6 5 4 3 2 1

International Standard Book Number-10: 0-8493-8892-9 (Hardcover)
International Standard Book Number-13: 978-0-8493-8892-7 (Hardcover)

Library of Congress Cataloging-in-Publication Data

Ecosystem responses to mercury contamination : indicators of change / edited by Reed Harris ... [et al.].
p. cm.
Includes bibliographical references (p.).
ISBN 0-8493-8892-9 (alk. paper)
1. Mercury--Environmental aspects. 2. Environmental indicators. 3. Environmental monitoring. I. Harris, Reed.

QH545.M4E26 2006
577.27'5663--dc22 2006049169

Visit the Taylor & Francis Web site at
http://www.taylorandfrancis.com

and the CRC Press Web site at
http://www.crcpress.com

and the SETAC Web site at
www.setac.org

4 Monitoring and Evaluating Trends in Methylmercury Accumulation in Aquatic Biota

James G. Wiener, R.A. Bodaly, Steven S. Brown, Marc Lucotte, Michael C. Newman, Donald B. Porcella, Robin J. Reash, and Edward B. Swain

ABSTRACT

The monitoring of mercury in aquatic food webs supporting the production of fish and wildlife is directly relevant to concerns about health and ecological risks of methylmercury (MeHg) exposure. We present a framework for monitoring concentrations of mercury in aquatic biota, with emphasis on assessing responses to changes in loadings of mercury from atmospheric deposition and other sources. In this chapter, we (1) identify specific attributes (criteria) of indicators that would be useful for discerning temporal trends and spatial patterns in the concentration of mercury in aquatic biota, (2) critically evaluate and rank candidate biological indicators useful for monitoring trends in mercury, (3) outline approaches for sampling and analysis of recommended biological indicators, (4) identify ancillary data needs and potential confounding factors that should be considered or documented to ensure the defensible interpretation of data on monitored biological indicators, and (5) consider the environmental settings (waterbody type and geographic location) that would be most sensitive for detecting changes in atmospheric deposition of mercury. Criteria were applied to ensure that the biological indicators selected are useful, relevant, and sufficiently diagnostic to detect a change in mercury bioaccumulation in response to altered mercury loadings. Toxicological problems with mercury in aquatic ecosystems result from biotic exposure to MeHg, a highly toxic compound that readily accumulates in exposed organisms and can biomagnify to high concentrations in organisms atop aquatic food webs. Biotic monitoring should, therefore, focus on assessing trends in bioaccumulation of MeHg; in samples from trophic levels below fish, this requires the determination of MeHg. We considered six general groups of

aquatic biological indicators: piscivorous fish, prey fish, benthic invertebrates, zooplankton, phytoplankton, and periphyton. Piscivorous fish and 1-year-old prey fish, all analyzed individually, are considered the preferred aquatic biological indicators for trend monitoring. For piscivorous fish, total-mercury determinations on axial muscle (preferably without skin), sampled annually, would indicate gradual (multiyear) trends in MeHg that are directly relevant to humans who eat sport fish. For prey fish, annual sampling and analysis of either whole fish or axial muscle for total mercury should indicate annual changes in exposure to MeHg. In North America, the historical record for MeHg in piscivorous fish (from data on total mercury in filets or axial muscle) extends about 35 years, much longer than the comparatively sparse historical record for MeHg in water and aquatic biota of lower trophic levels. The analytical method (cold vapor atomic absorption spectrophotometry) that produced most of the historic data on total mercury in piscivorous fish is valid, and the potential utility of these existing data for trend analysis merits careful consideration. Benthic invertebrates have been monitored and analyzed for mercury more extensively in estuarine systems than in fresh waters. The consumption of estuarine macroinvertebrates, such as oysters, clams, shrimp, and crabs, is also a direct pathway for human exposure to MeHg. Determination of MeHg in shellfish and macrocrustaceans could, therefore, be useful for trend monitoring in estuaries. The importance of MeHg uptake and transfer at the base of the food web is recognized. However, the utility of periphyton, phytoplankton, zooplankton, and freshwater benthic invertebrates for trend monitoring is diminished by interpretational complexities associated with large temporal variation in the biotic composition and MeHg content among samples, and by our limited understanding of the processes and variables that affect concentrations of MeHg in these groups. Many anthropogenic and natural factors, independent of the bulk loading of mercury from atmospheric deposition, can strongly influence the concentrations of MeHg in aquatic biota. Our ability to discern linkages between MeHg concentrations in aquatic biota and changing external loadings of mercury to aquatic systems will depend on knowledge of such factors and on the minimization of their confounding effects in biotic monitoring programs.

4.1 INTRODUCTION

Elevated mercury concentrations in fish tissue adversely affect the quality of fishery resources in many inland, coastal, and marine waters. In the United States, mercury was responsible for 76% of all fish-consumption advisories in 2004, and 44 states and 1 territory had advisories attributed to mercury (USEPA 2005). Growing awareness of the mercury problem has prompted increasing efforts to survey mercury in fish, and the number of statewide fish-consumption advisories issued for coastal waters, lakes, and rivers in the United States has increased substantially during the past decade (Wiener et al. 2003; USEPA 2005). In Canada, mercury accounted for more than 97% (2572) of all fish-consumption advisories listed in 1997 (USEPA 2001). Nearly all of the mercury in fish is methylmercury (MeHg), a highly toxic compound that readily crosses biological membranes, accumulates in exposed organisms, and can biomagnify to high concentrations in aquatic food webs (Grieb et al. 1990; Bloom 1992; Francesconi and Lenanton 1992; Wiener et al. 2003).

Atmospheric deposition is an important source of total mercury in many surface waters (Fitzgerald et al. 1998; Bindler et al. 2001; Lin et al. 2001; Lamborg et al. 2002), and it has been widely inferred that a significant portion of the MeHg bioaccumulated in many aquatic ecosystems is derived from mercury entering the surface water or its watershed in atmospheric deposition (Johansson et al. 1991; Watras et al. 1994; Rolfhus and Fitzgerald 1995; Jackson 1997; Monteiro and Furness 1997; Downs et al. 1998). In many remote and semi-remote areas of the Northern Hemisphere that lack in-watershed sources of anthropogenic mercury, the rate of mercury accumulation in lacustrine sediments has increased by a factor of 2 to 5 or more since the mid-1800s or early 1900s, based on analyses of dated cores of sediment and peat (Swain et al. 1992; Lucotte et al. 1995; Lockhart et al. 1998; Lorey and Driscoll 1999; Bindler et al. 2001; Lamborg et al. 2002; Shotyk et al. 2005). Some cores from semi-remote sites show evidence of recent declines in mercury deposition, possibly associated with decreasing emissions of anthropogenic mercury (Engstrom and Swain 1997; Benoit et al. 1998; Bindler et al. 2001; Shotyk et al. 2003, 2005).

We present a framework for monitoring concentrations of mercury in aquatic biota, with emphasis on assessing responses to changes in loadings of mercury from atmospheric deposition and other sources. The monitoring of mercury in aquatic food webs supporting the production of fish and wildlife is directly relevant to societal concerns about this toxic metal. Much of the scientific effort on mercury contamination of aquatic food webs has been prompted by the human health risks of MeHg exposure (Myers and Davidson 1998; Mahaffey 2000; Clarkson 2002), given that the consumption of finfish and shellfish is the primary exposure pathway in humans (NRC 2000; Mahaffey et al. 2004). Consumption of fish is also an important pathway of MeHg exposure for wildlife atop aquatic food webs (Heinz 1996; Wolfe et al. 1998; Wiener et al. 2003).

4.2 OBJECTIVES

This chapter focuses on monitoring trends in bioaccumulation in relation to anticipated changes in emissions of mercury from anthropogenic sources. Aquatic biota, however, are exposed to mercury from multiple sources, including historic anthropogenic, current anthropogenic, and natural sources. We identify aquatic biological indicators that can provide evidence of a temporal change in bioaccumulation of mercury (estimated from concentrations in tissue or whole organisms) from all sources. The objectives of this chapter are fivefold:

1) To identify specific attributes (criteria) of indicators that would be useful for discerning temporal trends and spatial patterns in the concentration of mercury in aquatic biota
2) To critically evaluate and rank candidate biological indicators useful for monitoring trends in mercury
3) To outline approaches for sampling and analysis of recommended biological indicators

4) To identify ancillary data needs and potential confounding factors that should be considered or documented to ensure the defensible interpretation of data on monitored biological indicators
5) To identify the environmental settings (water body type and geographic location) that would be most sensitive for detecting changes in atmospheric deposition of mercury in a trend-monitoring program

Many factors other than the bulk loading of mercury from atmospheric deposition can strongly influence the concentrations of MeHg in aquatic biota. Our ability to discern trends in MeHg concentrations in aquatic biota that are linked to changing loadings of mercury will depend on knowledge of such factors and on the minimization of their confounding effects in biotic monitoring programs for mercury.

4.3 AQUATIC BIOLOGICAL INDICATORS

4.3.1 Criteria to Select Indicators

The selection of biological indicators should be guided by criteria to ensure that the indicators are relevant, useful, and sufficiently diagnostic to detect a change in the concentration of mercury in whole organisms or specific tissue(s) over multi-year or decadal time scales. We identified 9 criteria for the selection of biological indicators:

1) *Relevance.* A key criterion in the selection of biological indicators is relevance to human and ecological health and to the development of policy. Fish are directly relevant, for example, given that consumption of fish is the primary pathway for exposure to MeHg. The concentration of MeHg in fish is also a key variable in the issuance of fish-consumption advisories.
2) *Historical data on the indicator.* Existing information on the statistical variation, bias, and other interpretational attributes of potential biological indicators should be examined and considered in the design of a sampling program for assessing trends in mercury bioaccumulation.
3) *Clear recognition of confounding factors.* Many human and natural factors that are unrelated to bulk loadings of mercury from the atmosphere or other sources can strongly influence concentrations of mercury in aquatic biota. A *confounding factor* is here defined as a variable that interferes with the isolation of the effects of mercury loading on temporal trends in mercury concentrations in monitored biota within a water body or group of waters. Our ability to discern biotic trends in mercury concentrations linked to changing loadings of mercury to aquatic systems will require both knowledge of potential confounding factors and the minimization of their confounding effects in monitoring programs (see Section 4.6.2).
4) *Knowledge of intrinsic co-variables.* Concentrations of mercury in fish are typically correlated with age or body size. An understanding of, and ability to account for, the effects of such intrinsic variables is essential for evaluating contaminant trends.

5) *Broad geographic distribution.* When feasible, monitoring should focus on bioindicator species or taxa having a broad geographic range, to assess trends and patterns across large geographic areas.
6) *Importance in trophic transfer of MeHg.* Preference should be given to bioindicator organisms that have an important role in the trophic transfer of MeHg within food webs.
7) *Constrained feeding ecology and trophic position.* Temporal shifts in feeding habits or trophic position can alter dietary MeHg uptake, complicating the interpretation of trends in mercury concentration. An *a priori* knowledge of feeding ecology should facilitate the selection of suitable candidate bioindicator species during the design of a monitoring program. If a bioindicator is known to undergo ontogenetic shifts in diet, it would be advisable to limit sampling and analysis to a given life stage.
8) *Temporal response to changes in mercury loadings.* Long-lived bioindicators with long response times (several years) to changing mercury loadings could be more susceptible to the influence of confounding factors, possibly reducing their utility for detecting effects of altered loadings. Thus, short-lived (≤1-year old), as well as long-lived, organisms should be considered when selecting potential bioindicators.
9) *Impacts of sampling on the target population.* Continued removal of individuals could eventually affect the concentration of mercury in members of the sampled population, creating artifacts in trend data. The potential effects of sampling on the target population, and potential approaches for reducing such effects, should be considered. Nonlethal techniques for sampling of fish tissue (Baker et al. 2004; Peterson et al. 2005) may be required when sampling protected populations or when sampling in protected environments, such as national parks.

4.3.2 Candidate Aquatic Biological Indicators

Six general groups of aquatic biological indicators were considered: 1) piscivorous fish, 2) prey fish, 3) benthic invertebrates, 4) zooplankton, 5) phytoplankton, and 6) periphyton. We begin with a brief summary of our understanding of mercury bioaccumulation and trophic transfer in aquatic ecosystems, to provide essential background information for the subsequent sections on aquatic biological indicators. This summary is based on selected recent reviews (Jackson 1998; Morel et al. 1998; Benoit et al. 2003; Wiener et al. 2003) and the original reports cited.

In a toxicological sense, the primary problem with mercury in aquatic ecosystems can be defined as biotic exposure to, or bioaccumulation of, MeHg. Trend monitoring of mercury in aquatic biota should accordingly focus on MeHg, which readily crosses biological membranes and accumulates to concentrations in aquatic organisms that vastly exceed those in surface water. In fish, concentrations of MeHg commonly exceed those in water by a factor of 10^6 to 10^7 or more. Most of the mercury in the aquatic environment is inorganic, yet nearly all (>95%) of the mercury accumulated in fish is MeHg (Grieb et al. 1990; Bloom 1992; Francesconi and Lenanton 1992), obtained almost entirely via dietary uptake (Rodgers 1994; Hall

et al. 1997; Harris and Bodaly 1998). Relative to MeHg, inorganic mercury is less readily transferred through successive trophic levels and does not biomagnify (Watras et al. 1998).

Methylmercury biomagnifies in aquatic food webs, and patterns in biomagnification are similar even among aquatic systems that differ in type of water body, mercury source, and pollution intensity. The transfer of MeHg in the upper trophic levels of aquatic food webs is almost entirely via dietary uptake, whereas direct uptake from water can be important for some lower food-chain organisms, such as phytoplankton and zooplankton. The concentration of MeHg increases up the food web from water and lower trophic levels to fish and piscivores, and the fraction of total mercury present as MeHg also increases with increasing trophic level through fish. The fraction of total mercury present as MeHg can vary greatly within trophic levels below fish. In aquatic invertebrates, for example, the MeHg fraction can range from about 10% to more than 90% of total mercury. It is, therefore, essential to determine MeHg (rather than total mercury) in biological samples from trophic levels below fish, including phytoplankton, periphyton, benthic invertebrates, and zooplankton. In fish, determination of total mercury, which requires less analytical effort and expense than MeHg, provides reliable estimates of MeHg concentration.

The greatest increase in MeHg concentration occurs in the trophic step between water and algae. Bioaccumulation factors between water and seston, for example, often range from about 10^5 to about 10^6, whereas ratios of MeHg concentrations between successive trophic levels above algae are generally less than 10^1. Within an assemblage of fish, concentrations of MeHg increase with ascending trophic level, and variation in trophic position accounts for much of the variation in mercury concentration among species within a given water body. Concentrations of MeHg in fish also increase with increasing age or size because of the very slow rate of elimination relative to the rapid rate of uptake and because larger fish can consume larger prey with higher concentrations of MeHg. Much of the MeHg accumulated in fish is stored in skeletal muscle, tightly bound to sulfhydryl groups in protein.

Although the entry of MeHg into the base of the food web and its subsequent transfer in the lowest trophic levels are poorly understood, it is evident that the concentration of MeHg in all trophic levels is strongly correlated with its supply from methylating environments. In fish, for example, much of the modern spatial variation in mercury concentrations (within a given trophic level) can be attributed to variation in factors and processes that affect the microbial production of MeHg and its entry into oxic waters.

4.3.2.1 Fish

We consider 2 groups of fish as candidate biological indicators for monitoring trends in MeHg: 1) piscivorous (fish-eating) fish and 2) small prey fish (often termed "forage fish"). Analyses of piscivorous fish reflect the concentrations of MeHg near the top of aquatic food webs, providing a useful measure of potential dietary exposure in humans who eat fish. Small prey fish are eaten by a variety of piscivorous fish, birds, and mammals, but are generally not important in the diet of humans. Humans eat large omnivorous fish; however, the substantial confounding effects of the large

spatial and temporal variation in the diet and trophic position of omnivorous fish diminish their potential utility for trend analysis.

4.3.2.1.1 Piscivorous Fish

Piscivorous fish can accumulate high concentrations of MeHg, and information on MeHg in piscivorous fish is directly relevant to the public and the policy community. Piscivorous fish are present in most surface waters and can be obtained with moderate sampling effort with a variety of active and passive gear. Sampling would generally not affect target populations except in very small lakes and streams, where nonlethal sampling would lessen impacts on target populations.

In the United States, the threshold mercury concentration for commercial sale of fish is determined by the Food and Drug Administration, whereas consumption advice for recreational (noncommercial) fish is developed by individual states and tribes. Mercury data collected for development of fish-consumption advisories are typically from analyses of filets (axial muscle tissue, with or without skin) for total mercury, with concentrations expressed on a wet-weight basis. Analysis of filets for total mercury yields a valid estimate of MeHg concentration (Grieb et al. 1990; Bloom 1992), whether the analyzed sample consists of a large filet or a small mass of tissue obtained with a biopsy needle (Cizdziel et al. 2002; Baker et al. 2004).

Many piscivorous fish are important recreationally or commercially. The sampling and analysis of heavily exploited fish stocks are not recommended for trend monitoring, because intensive fishing pressure — over a period of years to decades — can substantially reduce mercury concentrations in members of heavily exploited populations (Section 4.6.1). Interpretation of temporal trends in mercury concentrations in fish, relative to changes in mercury loadings, will be more defensible if applied to fish populations and water bodies that are not subjected to intensive fishing pressure. Many commercially and recreationally important marine fish have been significantly depleted by over-fishing (Pauly et al. 2003; Coleman et al. 2004; Hutchings and Reynolds 2004). The application of trend data on mercury concentrations in heavily exploited marine fishes, such as yellowfin tuna *Thunnus albacares* (Kraepiel et al. 2003), has questionable validity as an approach for assessing temporal changes in the abundance of MeHg in the ecosystem.

In summary, piscivorous fish are present in most surface waters, require moderate sampling effort, and are the primary pathway for dietary exposure of humans to MeHg. Analyses of filets or axial muscle for total mercury should indicate gradual (multi-year) trends in the supply of MeHg. Given these attributes, nonmigratory piscivorous fish are priority candidates for monitoring MeHg. In very small lakes or streams, nonintrusive sampling methods should be used to reduce impacts on sampled populations.

4.3.2.1.2 Prey Fish

Prey fish are here defined as small, usually short-lived, finfish. In North American fresh waters, many are members of the cyprinid (minnow), percid (perch), and centrarchid (sunfish) families. Prey fish are widely distributed, common, and important in the transfer of MeHg to higher trophic levels, such as piscivorous fish and many fish-eating birds. MeHg concentrations in prey fish of uniform age are less

susceptible to certain, potential confounding factors, such as variation in trophic position, than are concentrations in long-lived, piscivorous fish.

Mercury concentrations in prey fish are useful indicators of relative MeHg levels in food webs supporting the production of sport fish and wildlife, information relevant to the public and the policy community. There is a sizable scientific literature on MeHg in prey fish, but they have been monitored less extensively than sport fish. Effects of removal sampling on target populations would be insignificant in all but the very smallest lakes.

This group of fish includes species that feed on zooplankton, benthic invertebrates, and occasionally, periphyton (Roseman et al. 1996; Bodaly and Fudge 1999; Gorski et al. 1999). The size of young prey fish can vary seasonally because measurable growth occurs throughout much of the year. Small prey fish, such as yearling yellow perch (*Perca flavescens*), finescale dace (*Phoxinus neogaeus*), or mimic shiner (*Notropis volucellus*), typically grow rapidly during the summer in temperate lakes, increasing their biomass by 2- to 5-fold. Mercury concentrations are usually increasing or stable during this period of growth; consequently, the total body burden of mercury in individual fish increases substantially during summer (Bodaly and Fudge 1999; Gorski et al. 1999). Concentrations of MeHg in prey fish can be expected to vary seasonally (Figure 4.1), and such variation should be considered when crafting sampling protocols for trend monitoring. Prey fish require little to moderate sampling effort, and samples taken in the early spring or fall, when growth is slow and temporal variation in mercury concentration is less, may provide the best comparisons among years and surface waters. Analysis of prey fish for total mercury or MeHg would reveal inter-annual variation in MeHg exposure (Frost et al. 1999; Gorski et al. 1999).

In summary, prey fish are present in most surface waters, require moderate sampling effort, are important in the trophic transfer of MeHg in aquatic food webs, and probably indicate annual changes in exposure to MeHg. Given these attributes,

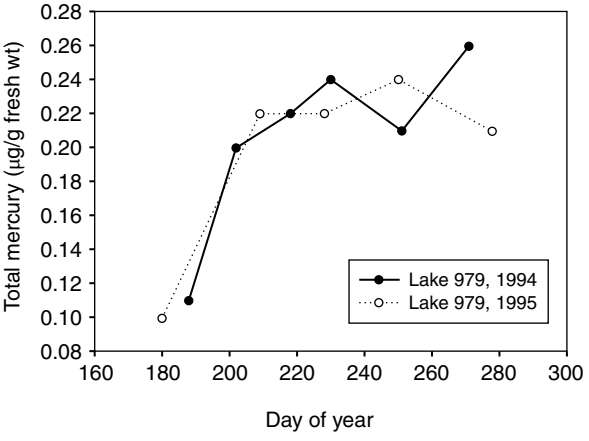

FIGURE 4.1 Whole-body concentrations of total mercury (present largely as MeHg) in caged finescale dace, showing seasonal increases in mercury concentrations during summer in Lake 979, an experimental reservoir in northwestern Ontario that was flooded during the Experimental Lakes Area Reservoir Project. (*Source:* Modified from Bodaly and Fudge 1999.)

prey fish are appropriate candidates for monitoring of MeHg. Seasonal variation should be considered carefully in the design of trend-monitoring protocols for prey fish.

4.3.2.1.3 Example of a Prey-Fish Indicator: Yellow Perch

Analyses of total mercury in whole bodies or axial muscle tissue of age-1 yellow perch have provided a useful measure of MeHg concentrations in food webs of many North American lakes. This widely distributed species inhabits lakes and reservoirs across much of the north-central, northeastern, and eastern United States and across the central and eastern provinces of Canada (Scott and Crossman 1973; Becker 1983). An ecologically similar congeneric species, the Eurasian perch (*Perca fluviatilis*), is distributed across much of Europe and northern Asia (Thorpe 1977).

During their first year, yellow perch have a small gape (jaw opening), which limits their diet largely to small zooplankton and small zoobenthos (Roseman et al. 1996; Lyons et al. 2000). Thus, the trophic position of age-1 yellow perch is not expected to vary substantially among sites. Generally abundant in lakes within much of its geographic range, the yellow perch is a preferred prey of certain piscivores, such as walleye (*Sander vitreus*) and common loons (*Gavia immer*), and is an important link in the food-web transfer of MeHg (Colby et al. 1979; Barr 1996). Concentrations of total mercury in age-1 or age-2 yellow perch are strongly and positively correlated with concentrations in coexisting piscivorous fish, including walleye, black bass (*Micropterus* spp.), and northern pike (*Esox lucius*) (Suns et al. 1987; Cope et al. 1990; JG Wiener, unpublished data for northern pike). Statistical analyses have shown strong relations between the total mercury concentration and burden of age-1 yellow perch and ecosystem characteristics (e.g., lake chemistry, wetland influence) or whole-lake manipulations (e.g., experimental acidification) that are known to influence the production of MeHg and its abundance in aquatic food webs (Grieb et al. 1990; Suns and Hitchin 1990; Wiener et al. 1990; Simonin et al. 1994; Frost et al. 1999; Wiener et al. 2003). Substantial mercury data are also available for the Eurasian perch (Metsaelae and Rask 1989; Andersson et al. 1995; Haines et al. 1995; Porvari 1998; Svobodova et al. 1999; Lindestroem 2001).

One-year-old yellow perch can be readily sampled in spring with small-mesh trap nets, seines, or small electroshockers fished in littoral habitat without significantly affecting their abundance or year-class strength. Age-1 fish obtained in spring have resided in a sampled lake for about 1 year. A target sample size of 15 to 30 whole, age-1 yellow perch (analyzed individually) from a given lake typically yields a standard error of the mean in the range of 1 to 6 ng/g wet weight, providing a precise estimate of mean whole-body concentration (JG Wiener, University of Wisconsin–La Crosse, data from Wisconsin and Minnesota lakes; corresponding mean concentrations range from about 20 to 200 ng/g wet weight). At age-1, the age of yellow perch can be accurately determined by examining scales taken near the area of insertion of the left pectoral fin.

4.3.2.2 Benthic Invertebrates

Benthic invertebrates are macroscopic animals that live at or near the sediment/water interface. Some benthic invertebrates, particularly mussels, readily accumulate metals, prompting their use as biological indicators of mercury contamination (Smith

and Green 1975). Benthic invertebrates have been widely used in biological monitoring of freshwater and marine habitats (Resh and McElvary 1993; Southerland and Stribling 1995; Weigel et al. 2003), providing a foundation for their use in trend-monitoring programs.

The dietary importance of benthic invertebrates to many species of fish, birds, and mammals (Vander Zanden and Vadeboncoeur 2002) signifies their importance in the trophic transfer of MeHg and their potential relevance as biological indicators. Some benthic invertebrates (e.g., oysters, clams, shrimp, crabs, and crayfish) are consumed by humans, providing a direct pathway for exposure to MeHg. In the United States, shellfish rank below fish as a source of dietary MeHg in the human population (NRC 2000; Schober et al. 2003).

Many benthic invertebrates have short life spans (≤1 year) but little is known about how quickly mercury concentrations in such organisms respond to changes in *external* loadings of mercury to the aquatic ecosystem. Bed sediment can be an important sink for mercury in aquatic systems if sediment-associated mercury is isolated from active biogeochemical cycling (Henry et al. 1995; Wiener and Shields 2000). Sediment can also serve as a source of MeHg in freshwater and estuarine ecosystems, given that the oxic/anoxic interface in the sediment is an important zone of mercury methylation (Gilmour et al. 1998; Benoit et al. 2003). Benthic organisms have physical contact with bed sediment, and the relative contributions of mercury from in-place sedimentary sources and of mercury from current external sources to their MeHg burdens will influence their sensitivity as indicators to altered mercury loadings from external sources. In this regard, sediment-dwelling invertebrates that feed on particles from the overlying water column — a group including many clams and aquatic insects — may be more useful than deposit feeders as indicators of external mercury loadings. The kinetics of MeHg bioaccumulation (ingestion, assimilation, and elimination) in benthic invertebrates have received little study but the limited available information should nonetheless be applied to the selection of candidate bioindicator species and to the interpretation of trend data.

The trophic position of benthic invertebrates varies widely among species. The diet is their primary pathway of contaminant exposure, and the feeding ecology of a benthic species largely determines its exposure to dissolved and particulate sedimentary contaminants (Brown et al. 2000). The dietary assimilation efficiency for MeHg (55–70%) in marine invertebrates is much higher than that for inorganic mercury (2–22%; Wang and Fisher 1999). In marine bivalves, exposure can occur through multiple pathways, including direct uptake from the overlying water or pore water and dietary uptake via ingestion of plankton and detritus from water or sediment (Thomann et al. 1995).

Benthic invertebrate communities are taxonomically and trophically complex, and their abundance and species composition in a water body often vary seasonally and among years. Sediment-dwelling invertebrates can be readily sampled but considerable effort is often required to remove benthic organisms from grab samples of sediment, to determine their taxonomic composition, and to obtain sufficient sample mass of a target taxon for analysis. Sampling would not substantially affect target populations.

In summary, benthic invertebrates are important in the trophic transfer of MeHg. The sensitivity of benthic organisms to altered mercury loadings from external sources will presumably depend on the relative contributions of mercury from in-place sediment and external sources to their total MeHg uptake. Moreover, the large effort required to produce samples that are well-defined taxonomically and trophically reduces their desirability as biotic indicators for trend monitoring in freshwater systems. Conversely, in estuarine systems, there has been more extensive monitoring and analysis of mercury in shellfish and macrocrustaceans, which could be useful in monitoring of estuaries.

4.3.2.3 Zooplankton

Zooplankton are small, often microscopic crustaceans that live in the water column. They are widely distributed, common, and important in pelagic food webs. Zooplankton are eaten by many fish and by early life stages of some fish that become piscivorous as juveniles or adults. Zooplankton are not eaten by humans but are an appropriate and relevant candidate indicator because of their importance in the trophic transfer of MeHg to fish. Sampling would not significantly affect populations or assemblages of zooplankton, even in small lakes.

Zooplankton vary seasonally and annually in abundance (Rusak et al. 2002), and respond within hours or days to changes in the supply of MeHg to the water column (Herrin et al. 1998; Paterson et al. 1998; Tsui and Wang 2004). Zooplankton can accumulate significant quantities of MeHg from food or water (Monson and Brezonik 1999; Peech Cherewyk 2002; Tsui and Wang 2004). Spatio-temporal variation in the taxonomic composition and abundance of zooplankton is large in temperate lakes (Rusak et al. 2002). Consequently, the trophic position of bulk zooplankton samples can vary greatly because these assemblages are composed of trophically diverse taxa that can eat phytoplankton, bacteria, detritus, and other zooplankton. Species assemblages can change rapidly, and a target species or taxon may not be available at certain times of the year.

The MeHg content of zooplankton varies among taxa (Back and Watras 1995), a complicating factor that can be eliminated by the determination of MeHg in individual taxa (Back et al. 1995). Zooplankton are readily sampled but samples should be checked and processed to remove phytoplankton and detritus. A single bulk sample from a plankton net can be used to characterize the open-water community of zooplankton in a lake at a particular time. The processing of samples can require substantial effort, and bulk samples of zooplankton from some surface waters contain particles that are extremely difficult to remove, diminishing the integrity of the sample.

Existing data on MeHg in zooplankton are few, and there has been little long-term monitoring of MeHg in zooplankton. In northern lakes, the concentrations of MeHg in zooplankton vary seasonally and annually (Figure 4.2), typically increasing in summer and declining in autumn (Herrin et al. 1998; Paterson et al. 1998). In thermally stratified lakes, the concentration of MeHg in zooplankton can increase markedly after the fall overturn (Figure 4.2), when MeHg in anoxic hypolimnetic

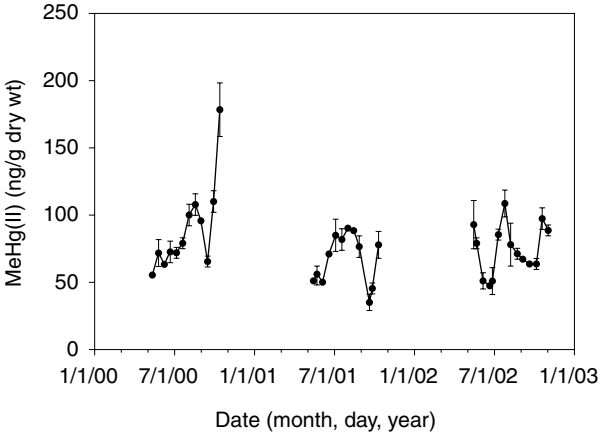

FIGURE 4.2 Concentrations of MeHg (mean ± 1 standard error) in zooplankton from Lake 240 of the Experimental Lakes Area (northwestern Ontario, Canada), showing seasonal variation during summer and pronounced rapid increases in mean concentration after the fall overturn. (*Source:* Michael J. Paterson, Fisheries and Oceans Canada, Winnipeg, Manitoba, unpublished data.)

waters becomes mixed throughout the water column (Herrin et al. 1998). Such seasonal variation merits careful attention in sampling protocols for zooplankton in a trend program for mercury, regardless of whether bulk samples or specific taxa are used as a bioindicator, and may require multiple sampling events during the year. Samples taken in mid-summer would reflect conditions for bioaccumulation of MeHg during the period of maximal growth of fish but would miss the effect of the fall overturn in stratified lakes. Given such large intra- and inter-annual variation, we expect that many years of sampling and analysis would be needed to discern long-term trends in MeHg concentrations in zooplankton.

In summary, zooplankton are present in all lakes, are readily sampled, are important in the trophic transfer of MeHg, and would respond rapidly to changes in the supply of MeHg to the water column. Seasonal and interannual changes in MeHg concentration would complicate the timing of sampling and the interpretation of trend data. Pronounced short-term variability may reduce their suitability for assessing trends on the multi-year time scales expected for reductions in mercury emissions.

4.3.2.4 Phytoplankton

Phytoplankton are microscopic plants (algae) that live in the water column and are trophically situated at or near the base of pelagic food webs. Phytoplankton are taxonomically diverse, and their abundance and species composition vary seasonally, annually, and spatially. Phytoplankton are present in all aquatic systems and are considered important in the trophic transfer of MeHg. It has been generally assumed that zooplankton receive most of their MeHg via consumption of phytoplankton; however, recent research suggests that direct uptake of MeHg by zooplankton from water may be more important than previously thought (Monson and Brezonik 1999;

Peech Cherewyk 2002; Tsui and Wang 2004). Phytoplankton have a constrained trophic position; are not consumed by fish, wildlife, or humans, and are indirectly relevant to the public or the policy community. They would respond very rapidly — within minutes or hours — to changes in MeHg concentrations in water. Phytoplankton obtain MeHg directly from water (Mason et al. 1996), and algal density can influence concentrations of MeHg in phytoplankton via biomass dilution (Pickhardt et al. 2002).

Phytoplankton are easily sampled with fine-mesh nets. However, samples require considerable processing before analysis to remove zooplankton, detritus, or other particulates. Sampling would not measurably affect target populations, even in the smallest lakes.

Little is known about MeHg in phytoplankton, particularly freshwater phytoplankton (Becker and Bigham 1995). Concentrations of MeHg in freshwater phytoplankton are related to those in water but the partitioning of MeHg between water and phytoplankton is strongly affected by concentrations of dissolved organic matter (Watras et al. 1998).

In summary, phytoplankton are present in all lakes, are trophically well defined, and are a candidate indicator for trend monitoring. They would rapidly respond to changes in the supply of MeHg. However, discrete samples of phytoplankton are not easily obtained and phytoplankton are not a recommended indicator for trend monitoring, largely because of the complexities associated with interpretation of trend data for phytoplankton. Large intra- and inter-annual variations in the MeHg content of phytoplankton would complicate the interpretation of trend data and the identification of long-term trends.

4.3.2.5 Periphyton

Periphyton are microscopic and macroscopic algae that attach to and grow on solid surfaces, such as lake bottoms, rooted aquatic vegetation, and submerged woody debris. Periphyton form part of the base of littoral food webs in lakes. Periphyton communities are taxonomically diverse and the attached communities contain other organisms, such as bacteria and zooplankton, as well as detrital material. Periphyton vary seasonally and annually in both abundance and species composition.

Periphyton are widely distributed and common; however, the trophic position of the overall community is complex and variable. Periphyton have been studied little with regard to mercury cycling but would be expected to respond rapidly to changes in the supply of MeHg. Periphyton can be directly eaten by fish, and periphyton mats may be an important site for mercury methylation in some ecosystems (Cleckner et al. 1999). Methylmercury typically comprises a very small, but quite variable, fraction of the total mercury in periphyton (Cleckner et al. 1998; Bowles et al. 2001). Periphyton are relevant neither to the public nor the policy community, because they are not consumed by people or wildlife, and their importance in the cycling of mercury is unclear. Periphyton would be relatively easy to sample but samples would contain a complex mixture of plants, small invertebrates, and detritus, complicating interpretation of the MeHg concentrations therein. Sampling would not significantly affect target populations.

In summary, periphyton are present in all lakes, easy to sample, and would respond rapidly to changes in the abundance of MeHg. Periphyton are sometimes eaten directly by fish. The diverse, complex, and variable nature of the periphyton community, however, would complicate interpretation of mercury concentrations in periphyton in a monitoring program.

4.3.3 Recommended Aquatic Biological Indicators

The criteria listed in Section 4.3.1 were applied to the selection of biological indicators to ensure their relevance and utility for assessing trends in the bioaccumulation of MeHg associated with altered loadings of mercury to aquatic systems. This evaluation, based largely on the discussion in Section 4.3.2, is summarized in Table 4.1.

We consider prey fish, piscivorous fish, and estuarine benthic invertebrates (listed in order of preference) suitable aquatic biological indicators for trend monitoring. One-year-old prey fish and piscivorous fish (analyzed individually) are the preferred biological indicators for freshwater systems. Determination of total mercury in axial muscle (preferably without skin) from piscivorous fish would indicate gradual (multi-year) trends in MeHg that are directly relevant to humans who eat sport fish. For 1-year-old prey fish, the annual sampling and analysis of either whole fish or axial muscle for total mercury would indicate annual changes in MeHg exposure in freshwater and marine ecosystems. In coastal estuaries, 1-year-old prey fish (analyzed for total mercury), as well as shellfish and macrocrustaceans (analyzed for MeHg), are recommended as biological indicators for trend monitoring. Adult piscivorous fish from heavily exploited populations or heavily fished water bodies are not recommended as biological indicators because of the potentially large confounding effects of intensive fish harvest on MeHg concentrations in such populations.

The sampling of aquatic biological indicators of different size, age, and trophic position can enhance the interpretation and understanding of temporal patterns in MeHg concentration. Concentrations of MeHg in 1-year-old prey fish will be sensitive to interannual variations in controlling factors and processes (such as mercury loadings, temperature, and hydrology), whereas older piscivorous fish will integrate and reflect temporal changes across multi-year time scales. Based on observed chronologies of mercury concentrations in aquatic biota in reservoirs during the first 30 years after flooding (Bodaly et al. 1997), we anticipate that decreases in MeHg concentrations in response to decreased atmospheric loadings would first be evident in 1-year-old prey fish, whereas decreases in concentrations in long-lived piscivores would lag a few years later.

The importance of MeHg uptake and transfer at the base of the food web is recognized and should be the subject of focused, intensive research. The utility of periphyton, phytoplankton, zooplankton, and freshwater benthic invertebrates for trend monitoring, however, is hampered by procedural and interpretational complexities associated with the large spatio-temporal variation in species assemblages, sample heterogeneity, and MeHg concentration. The utility of these groups for trend monitoring is also hampered by our very limited understanding of the processes, variables, and confounding factors that affect their bioaccumulation of MeHg.

TABLE 4.1
Recommended criteria for selection of aquatic biological indicators for monitoring and assessment of methylmercury (MeHg), and their application to candidate biological indicators

		Extent to which the aquatic biological indicator satisfies the criterion:					
Criterion	**Importance of criterion**	**Periphyton**	**Phytoplankton**	**Zooplankton**	**Benthic invertebrates**	**Prey fish**	**Piscivorous fish**
Relevance to the MeHg problem	To ensure relevance to human health, ecological risk, and development of policy	Low	Low	Low	Low in fresh waters; High in coastal estuaries	Medium to High	High and direct
Availability of historic MeHg data	To document the utility of the indicator and its probable reliability for detecting change in mercury loading	Low	Low	Low	Low to Medium	Low to Medium	High
Recognition of extrinsic confounding factors	To facilitate the defensible interpretation of monitoring results on MeHg	Low	Low	Medium	Medium to High	Medium to High	High
Knowledge of intrinsic co-variables	To ensure knowledge of organismal attributes that can affect MeHg concentration and complicate interpretation of results	Low	Low	Medium	Low to Medium	Medium	High

TABLE 4.1 (continued)
Recommended criteria for selection of aquatic biological indicators for monitoring and assessment of methylmercury (MeHg), and their application to candidate biological indicators

		Extent to which the aquatic biological indicator satisfies the criterion:					
Criterion	**Importance of criterion**	**Periphyton**	**Phytoplankton**	**Zooplankton**	**Benthic invertebrates**	**Prey fish**	**Piscivorous fish**
Broad geographic distribution	To select biotic indicators that have broad spatial coverage in a regional, national, or multi-national monitoring program	Widespread, but spatially and temporally variable assemblages	Widespread, but spatially and temporally variable assemblages	Widespread, but spatially and temporally variable assemblages	Several widespread species and genera	Several widespread species and genera	Several widespread species and genera
Important in trophic transfer of MeHg	To select biotic indicators with a significant role in the trophic transfer of MeHg in aquatic food webs	Unclear	Important	Highly important	Highly important	Highly important	Highly important
Constrained food habits or trophic position	Spatio-temporal variation in trophic position can confound and complicate interpretation of trends in MeHg concentration in the indicator	Highly constrained	Variable and complex	Variable and complex; constrained in some filter feeders	Variable and complex	Constrained	Ontogenetic shifts with increasing size; less variable in adults

Temporal response to changes in abundance of MeHg	A slow rate of response to altered MeHg loading could increase the potential for interference by confounding factors, whereas a rapid response, coupled with high intraannual variability would hinder identification of multi-year trends	Hours to days	Hours to days	Hours to days	Days to seasonal in small benthos; annual to multi-annual in large benthos	Annual	Multi-annual
Effects of sampling on target population or community	To reduce effects of sampling on monitored populations and biotic communities, and to limit potential confounding biotic responses to sampling	Nonintrusive	Nonintrusive	Nonintrusive	Nonintrusive	Nonintrusive	Generally nonintrusive, except in some small lakes and streams

4.4 MONITORING AND TREND ANALYSIS

We recommend that biological indicators be sampled annually to assess responses of MeHg concentrations to changes in mercury loadings and that such sampling be limited to nonmigratory species. Although sampling at less frequent intervals (e.g., every 2 or 3 years) would decrease effort and cost per monitored water body, it would substantially reduce statistical power to detect temporal trends and would delay the detection of changes in mercury concentration (Hebert and Weseloh 2003; Bignert et al. 2004).

Freshwater piscivorous fish and prey fish should be sampled in several water bodies because data from multiple sites (i.e., clusters of sites) are needed to describe trends within a given geographic area. The primary response to decreased mercury deposition within a given area may be decreased mercury concentrations in fish, yet concentrations may remain stable or increase in some water bodies because of other factors that influence mercury cycling, MeHg production, and bioaccumulation. In Minnesota, for example, analysis of state-wide trend data on total mercury in filets of standard-sized game fish from 176 lakes showed that 49% of the lakes had statistically significant ($p < 0.05$) decreases in fish-mercury concentrations, 25% had significant increases, and 26% did not change (Bruce A. Monson, Minnesota Pollution Control Agency, St. Paul, Minnesota, personal communication). Significantly more of the Minnesota lakes exhibited decreasing mercury concentrations in fish than could be attributed to chance alone (χ^2 test, $p < 0.05$). Concentrations have also declined significantly in walleyes inhabiting boreal lakes of central Canada in the last 20 to 30 years (Johnston et al. 2003), and variation among lakes in fish-mercury trends is also evident in the Canadian data (Figure 4.3).

Concentrations of MeHg in individual fish typically increase with increasing size and age, and data on the length, weight, and age of individual fish are needed to interpret trends in mercury concentration. Piscivorous fish should, therefore, be analyzed individually. Temporal trends in mercury concentrations in piscivorous fish can be biased by variation in the size or age composition of samples taken at different times. It is, therefore, useful to estimate mercury concentrations in fish of a given age or size. In Florida, mercury concentrations in axial muscle of 3-year-old largemouth bass (*Micropterus salmoides*) have been successfully used to examine variation in fish contamination among lakes (Lange et al. 1993) and to monitor temporal trends in the Everglades (Atkeson et al. 2003). For piscivorous fish, stratified sampling by predefined length interval could be a useful framework for sampling a target population.

One widely used approach has been to sample individual fish from a target population across a range of lengths and to apply linear regression between mercury concentration and length to estimate the concentration in a fish of some standardized length; examples include 50-cm walleye and 60-cm northern pike (Parks and Hamilton 1987; Johnston et al. 2003). This procedure yields a concentration adjusted to a specific length, along with an associated confidence interval around the estimated value. Statistical power analysis of initial data can be used to estimate the sample size (number of fish) needed to detect a change of given magnitude (Exponent 2003).

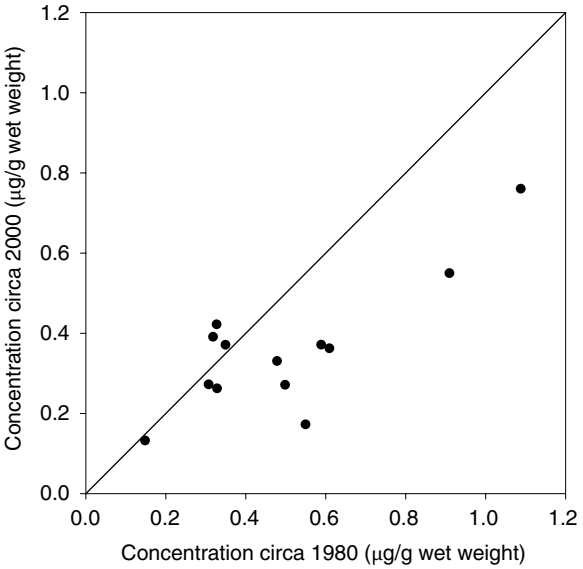

FIGURE 4.3 Recent changes in concentrations of total mercury in axial muscle of walleyes from 13 boreal lakes in northwestern Ontario and Manitoba, Canada. Standardized concentrations for 50-cm walleye sampled during 1996–2000 are plotted against standardized concentrations for fish sampled during 1977–1983 (data are for reference lakes reported in Johnston et al. 2003). Each point below the diagonal line, which has a slope of 1.0, represents a lake where the standardized concentration declined between the 2 sampling intervals.

Polynomial regression with indicator variables is another recommended statistical method for analysis of fish-mercury data. This procedure, described by Tremblay et al. (1998), allows rigorous statistical comparison of mercury-to-length relations among years and is considered superior to simple linear regression and analysis of covariance for analysis of data on mercury-length relations in fish.

For piscivorous fish, we recommend that axial muscle without skin be sampled and analyzed to avoid the additional variation that would result from differing proportions of skin and axial muscle tissue among samples. Concentrations of mercury are less in skin than in axial muscle, and the inclusion of skin in a filet can reduce the concentration of total mercury or MeHg in the sample by 5 to 12 percent (Glass et al. 1998; Serdar et al. 2001).

The historic data on total mercury in piscivorous fish are generally considered valid, and it could be useful to perform trend monitoring in carefully selected fresh waters for which historic data are acceptable and sufficient for statistical comparisons. In the late 1960s and early 1970s, the discovery of mercury-contaminated fish prompted widespread surveillance and monitoring of mercury in fish. The earliest surveys focused largely on surface waters that received mercury in polluted wastewater, but often included fish sampled from presumed "reference sites" that did not receive mercury from industrial point-source discharges. The surveillance of mercury

in fish was expanded spatially with growing awareness of the importance of atmospheric deposition as a potentially significant source of mercury in surface waters lacking on-site anthropogenic sources (Wiener et al. 2003). Historic mercury data for fish from surface waters that did not receive point-source discharges of mercury may be useful for assessing decadal trends related to atmospheric deposition. Historic fish-mercury data from analyses of composite samples (containing subsamples from multiple fish), however, has limited statistical utility for trend analysis (Exponent 2003).

We recommend that prey fish be sampled in early spring or fall, when growth is slow and temporal variation in mercury concentration is less pronounced, to facilitate comparisons among years and surface waters. Both axial muscle and whole fish have been successfully used in research applications, and analysis of either matrix would be suitable in a monitoring program for mercury. Determination of total mercury would be appropriate for muscle tissue, given that nearly all (≥95%) of the mercury in fish muscle is MeHg. Most (>70%) of the mercury in whole prey fish is MeHg (Francesconi and Lenanton 1992; Hill et al. 1996; Bodaly and Fudge 1999; Hammerschmidt et al. 1999), and we recommend that all whole prey fish be analyzed for total mercury and that a subset (from 5 to 20% of the fish, selected at random) be analyzed for MeHg.

If whole prey fish are analyzed, data should be reported as whole-body concentration and burden. Mercury *burden*, defined as the total mass of mercury accumulated in a whole fish, is calculated as the product of body weight and whole-body concentration. In prey fish of known age (e.g., age-1), mercury burden is an ecologically relevant measure of bioaccumulation, representing the mass of mercury accumulated by a fish during its residence in a monitored water body. The burden also represents the mass of mercury that a predator would ingest when eating the prey fish. Prey fish should be analyzed individually, and ancillary measurements include the total length (to nearest millimeter), fresh weight (to 0.01 g), and age of individual fish. Age can be estimated by examination of scales or other bony structures (DeVries and Frie 1996).

The trophic position of candidate benthic invertebrates for trend monitoring should be known or estimated so that samples with equivalent trophic status can be analyzed and compared. Stable-isotope methods are available for estimating trophic position (Fry 1991; Broman et al. 1992) and for assessing the influence of trophic position on mercury concentrations (Cabana and Rasmussen 1994; Kidd et al. 1995). The application of $\delta^{15}N$ and associated metrics in monitoring programs would enhance our ability to understand trends and strengthen inferences concerning linkages between trends in biotic concentrations and mercury loadings. The corrections and enhancements of Phillips (2001) and Phillips and Gregg (2001) should be considered when using data on stable isotopes to estimate the trophic status of benthic species.

The technique that has been widely applied for analyzing total mercury in aquatic biota since the late 1960s (cold vapor atomic absorption spectrophotometry) remains a valid analytical method. Accordingly, we infer that most of the historical data for total mercury in fish tissues are valid. Moreover, the historical data on total mercury concentrations in fish tissues provide defensible estimates of prior MeHg concentrations.

This is in contrast to most data collected before 1990 on total mercury and MeHg in water; most of these early data for water are not considered valid because sensitive analytical methods were not sufficiently developed and samples were typically contaminated during handling. There are some historical data on total mercury in aquatic invertebrates, but comparatively few estimates of MeHg in historic samples. Thus, the historical mercury record for piscivorous fish in North America is now about 35 years long, whereas the much sparser historical record for MeHg in water and aquatic biota in lower trophic levels is much shorter, perhaps 10 to 20 years in most regions. In comparison, the historic record of total-mercury deposition that can be examined by analyses of dated cores of sediment, peat, and glacial ice spans 2 to several centuries (Engstrom and Swain 1997; Martínez-Cortizas et al. 1999; Bindler et al. 2001; Schuster et al. 2002; Shotyk et al. 2003).

4.5 ANCILLARY DATA

The defensible interpretation of trend data in a biological monitoring program for mercury will be enhanced by the concurrent collection of relevant information on monitored airsheds, watersheds, and surface waters (see Chapters 2 and 3). The interpretation of fish-mercury data and the identification of factors causing temporal shifts in MeHg concentrations in aquatic biota would be strengthened by co-locating trend monitoring for airsheds, watersheds, and biological indicators at intensive study sites (Driscoll et al. this volume (Chapter 2), Krabbenhoft et al. this volume (Chapter 3)). Important metrics for monitored airsheds and watersheds include annual atmospheric deposition of total mercury, MeHg, and sulfate; interannual variations in rainfall and temperature; watershed area; land cover, abundance of hydrologically connected wetlands; and annual export of total mercury and MeHg from catchments to monitored surface waters. For lakes, key limnological data include lake morphometry (area, maximum depth, mean depth, percent littoral area); physicochemical characteristics of water (pH, dissolved organic carbon, sulfate, total suspended solids, chlorophyll, acid neutralizing capacity, color, and phosphorus); depth profiles of temperature and dissolved oxygen during summer stratification; hydrologic type (e.g., seepage or drainage lake); and concentrations of total mercury and MeHg in oxic water (preferably sampled when lakes are mixed). Information on the depositional chronology of total mercury in dated sediment cores would be useful for lakes with a multi-decadal, historic record of mercury concentrations in fish. Ancillary site-specific biological data include harvest of fish and shellfish; results of biotic and fishery surveys; and the age, growth rate, condition, sex, and trophic position (inferred from stable-isotope and dietary analyses) of aquatic biological indicators.

A number of recent experiments have shown that the reproduction and fitness of fish can be adversely affected by exposure to environmentally realistic concentrations of MeHg (Fjeld et al. 1998; Latif et al. 2001; Hammerschmidt et al. 2002; Drevnick and Sandheinrich 2003). In cases where trend monitoring reveals high MeHg concentrations in biological indicator organisms, subsequent field and laboratory studies should be considered to assess toxicological effects.

4.6 INTERPRETATION OF TREND-MONITORING DATA

Several biogeochemical processes, environmental factors, and human disturbances can influence the production of MeHg, its abundance in surface waters, and its concentrations in aquatic biota (Jackson 1998; Lucotte et al. 1999c; Benoit et al. 2003; Wiener et al. 2003). Thus, MeHg concentrations often vary spatially in aquatic organisms of the same species and size, even among nearby water bodies that receive similar atmospheric loadings of mercury (Wiener et al. 2003). The bioaccumulative responses of aquatic organisms to altered external loadings of mercury can, therefore, be expected to vary substantially among surface waters. Even within a given water body, internal ecosystem dynamics, anthropogenic activities unrelated to mercury loadings, and long-term external factors could confound or obscure responses in MeHg bioaccumulation to altered mercury loadings (e.g., Sorensen et al. 2005). Similarity of pertinent biogeochemical, environmental, and human factors affecting MeHg concentrations in aquatic organisms — among sampling intervals in a trend-monitoring program — would enhance our ability to detect responses to altered mercury loadings. Such similarity should not, however, be expected or assumed in a trend-monitoring program.

4.6.1 Sources of Variation and Potential Confounding Factors

Here we examine sources of variation and potential confounding factors that could obscure temporal changes in MeHg concentrations in aquatic biota in response to altered loadings of mercury. General sources of variation and potential confounding factors include interannual variations and dynamics of the monitored ecosystems, direct influences of anthropogenic and natural activities, and long-term external factors. Knowledge of such factors is prerequisite to discerning trends in MeHg concentrations in aquatic biota associated with altered loadings of mercury to aquatic systems. Awareness of potential confounding factors is also essential for minimizing or statistically accounting for their effects in a trend-monitoring program (see Chapter 3).

1) *Methylation and demethylation.* The methylation of inorganic mercury and demethylation of MeHg are key processes influencing the abundance of MeHg in food webs supporting the production of shellfish and fish (Benoit et al. 2003; Wiener et al. 2003). Rates of methylation and demethylation are affected by several variables, including temperature, sulfur cycling, light penetration, and the amount and quality of available organic matter (Miskimmin et al. 1992; Allan et al. 2001; Benoit et al. 2003; Kainz et al. 2003; Rencz et al. 2003). Such controlling variables can vary in response to natural events or conditions, such as water-level fluctuations and flooding, atypically cold or warm weather, increased turbidity caused by resuspension of bottom sediment, or varying abundances of allochthonous and autochthonous organic matter.
2) *Food-web and trophic dynamics.* Organisms at the base of aquatic food webs play a key role in the transfer of MeHg to upper trophic levels (Jackson and Harvey 1993; Plourde et al. 1997; Tremblay and Lucotte 1997; Bodaly and Fudge 1999). Within a given water body, interannual

variations in climatic and environmental conditions can affect the composition and abundance of pelagic and benthic invertebrates. Such variation in invertebrate assemblages can alter MeHg bioaccumulation in small and large fish at the end of the growing season (Parkman and Meili 1993; Montgomery et al. 2000; Gorski et al. 2003; Kainz et al. 2003).

Most fish are opportunistic organisms whose diets and trophic position can vary with food availability and size. Variation in environmental conditions among years (including water levels during spawning, availability of spawning sites, winter harshness, and summer oxygen depletion) can alter trophic structure of the fish community and the metabolism of fish, thereby affecting concentrations of MeHg in fish tissue (Lockhart et al. 1972; Meili 1991; Harris and Bodaly 1998; Scheuhammer and Graham 1999; Stafford and Haines 2001; Gorski et al. 2003). Interannual variation in growth rates of fish could influence the size-standardized mercury concentrations that are used to assess temporal trends in MeHg accumulation (Tremblay et al. 1998; Johnston et al. 2003; Sonesten 2003).

3) *Landscape disturbance.* Forest soils contain large inventories of mercury (Roulet et al. 1998; Lucotte et al. 1999a; Rencz et al. 2003) that can be mobilized into aquatic systems by disturbance of the forest landscape. Logging and fire can expose soil and increase the transport of mercury from soil to aquatic systems, a situation observed in northern boreal forest (Garcia and Carignan 1999; Porvari et al. 2003) and in Amazonian rainforest (Roulet et al. 1999, 2000). Agricultural practices, urban development, and road construction can also contribute to soil erosion and increased transport of mercury in soil from terrestrial to aquatic environments. Human disturbances of a watershed could also indirectly affect the production and bioaccumulation of MeHg in an aquatic system, by altering hydrologic pathways, water levels, nutrient inputs, primary production, light penetration, and trophic status.
4) *Fishing intensity.* Temporal variation in fishing intensity and harvest in a monitored water body could confound the analysis of trends in mercury concentrations in adult piscivorous fish. In Finnish and Scandinavian lakes, mercury concentrations in fish of given length declined notably a few years after intensive fishing had removed substantial numbers of the large adult fish (Göthberg 1983; Verta 1990). In experimental boreal lakes in Canada, the inventories of mercury in the water column and surface sediment were reduced little by intensive fishing (Surette et al. 2006). Intensive fishing may have affected the structure of the zooplankton communities (Masson and Tremblay 2002); however, the diets of the studied fishes did not change significantly after intensive fishing (Doire et al. 2002). Intensive fishing, however, significantly increased the growth rate of large predatory fish (Simoneau et al. 2005) and markedly decreased MeHg concentrations in fish (Surette et al. 2003).
5) *Exotic species.* The voluntary or accidental introduction of exotic species may influence the trophic structure of aquatic systems (Hrabik et al. 1998), thereby affecting MeHg concentrations in aquatic organisms in upper

trophic levels. Johnston et al. (2003), however, found that the introduction of rainbow smelt (*Osmerus mordax*) did not significantly affect concentrations of mercury in native predatory fish in lakes of central Canada.

6) *Reservoir impoundment.* The bioaccumulation of MeHg in aquatic organisms is greatly increased by the flooding of vegetated wetland or upland terrestrial habitats. This has been well documented in newly flooded reservoirs in temperate, boreal, and tropical regions (Bodaly et al. 1984; Johnston et al. 1991; Porvari 1995; Plourde et al. 1997; Seda and Kubecka 1997; Tremblay and Lucotte 1997; Lucotte et al. 1999b; Verdon and Tremblay 1999; Montgomery et al. 2000). Flooding of new reservoirs greatly increases the rate of mercury methylation, causing rapid increases (days to a few weeks) in concentrations of MeHg in water, zooplankton, and prey fish (Paterson et al. 1998; Bodaly and Fudge 1999). Concentrations of MeHg in the axial muscle of adult piscivorous fish can increase as much as 10-fold relative to pre-flood or reference values and remain elevated for decades after initial impoundment (Porvari 1998; Schetagne and Verdon 1999). New reservoirs also export MeHg, greatly increasing the concentrations of mercury in aquatic biota and fish inhabiting downstream aquatic environments (Johnston et al. 1991; Schetagne and Verdon 1999). In older reservoirs, concentrations of mercury in prey fish can fluctuate substantively in response to water-level fluctuations (Sorensen et al. 2005).
7) *Climate change.* Climate change can influence an array of environmental variables that directly and indirectly affect the biogeochemical cycling of mercury. The microbial methylation of mercury is temperature sensitive, with increasing production and concentrations of MeHg in temperate aquatic ecosystems at higher temperature (Bodaly et al. 1993). Climate change can modify the hydrological cycle, thereby altering mercury transport, transformations, and trophic structure in aquatic systems. Changes in the intensity of ultraviolet (UV) radiation at the water surface could influence both the production and photodemethylation (destruction) of MeHg in surface waters (Sellers et al. 1996, 2001; Lean and Siciliano 2003).
8) *Acidity and sulfate content of wet deposition.* Temporal variation in the acidity and sulfate content of wet deposition could modify the accumulation of MeHg in aquatic biota, given that numerous interactions between mercury transformations, MeHg uptake, sulfate, and aqueous pH have been reported (Spry and Wiener 1991; Odin et al. 1994; Frost et al. 1999; Scheuhammer and Graham 1999; Kelly et al. 2003). The biogeochemistry of mercury in waters with low acid neutralizing capacity and in methylating wetland environments could be affected by long-term chemical changes in wet deposition.

4.6.2 Steps to Constrain Confounding Factors and Enhance Interpretation

Monitoring programs should be designed to limit the effects of confounding factors that impair our ability to discern both trends in MeHg concentrations in aquatic

biota and their relation to altered atmospheric loadings of mercury. Adherence to the guidelines below should eliminate some of the pitfalls that can complicate interpretation of trend data on MeHg in aquatic biota. The application of these guidelines during the development of a monitoring program requires compilation of appropriate information on candidate study sites (surface waters and their watersheds) and their resident aquatic biota.

1) *Exclude very contaminated surface waters during site selection.* Concentrations of MeHg in biota inhabiting surface waters historically contaminated by wastes from industrial point-source discharges or historic mining activities can remain substantially elevated for decades (Latif et al. 2001; Wiener et al. 2003). In new reservoirs, the concentrations of MeHg in piscivorous fish remain elevated for several years after initial flooding, declining gradually over 2 to 3 decades (Bodaly et al. 1997). A trend-monitoring program intended to assess responses to changes in atmospheric loading should not include surface waters that are recovering from prior on-site elevation in abundance of either total mercury or MeHg.
2) *Exclude surface waters on disturbed watersheds.* Surface waters in watersheds that have recently undergone disturbance (e.g., fire or notable human development) or in watersheds where disturbance is anticipated during monitoring (e.g., logging or changing land use) should be avoided during site selection. This requires access to information on prior land use, as well as land-management plans. Given this, semi-remote surface waters in protected areas on state, provincial, or federal lands, such as national parks, would be appropriate sites for trend monitoring.
3) *Exclude surface waters subjected to intensive or highly variable fishing pressure.* Surface waters that are subjected to intensive fishing pressure or that are expected to undergo highly variable commercial or recreational harvest of fish or shellfish during monitoring should be excluded during site selection.
4) *Employ nonlethal methods when sampling small populations of piscivores.* In very small lakes or streams that support small populations, the application of procedures for nonlethal sampling (e.g., Baker et al. 2004; Peterson et al. 2005) may be advisable to avoid potential effects of removal sampling on MeHg concentrations in target populations of piscivorous fish. This determination requires advance knowledge of bioindicator organisms present at a candidate monitoring site, the approximate number (sample size) of an indicator organism to be taken during each sampling event, and the approximate size of the target population being sampled.
5) *Include multiple water bodies (clusters of sites) within each monitored geographic area.* Within a geographic area, the chosen biological indicators should be sampled in several water bodies of a given type (i.e., clusters of lakes or streams), because data from several sites will probably be needed to identify the *overall* trend within the area (Section 4.4). The direction of temporal trends can differ among individual water bodies because of spatio-temporal variation in other factors that influence mercury

cycling, net methylation, bioaccumulation, and concentrations in organisms. Statistical analyses should focus first on detecting temporal trends within individual sites and second on evaluating the overall direction of change across the cluster of sites within a monitored geographic area. Descriptive and exploratory statistical analyses should characterize monitored ecosystems that exhibit different trends (e.g., significant decrease, no change, or significant increase in MeHg concentrations) within a given area, as an initial step toward identifying potential causes of the observed differences in trends.

6) *Monitor bioaccumulation in some lakes with minimal watershed influence.* Delineating between watershed-derived mercury and atmospherically derived mercury is problematic in a biological trend-monitoring program. Wetlands can be particularly important sites of MeHg production and significant sources of total mercury, MeHg, and organic matter for adjoining surface waters (Hurley et al. 1995; St. Louis et al. 1996; Curtis 1998; Sellers et al. 2001). Concentrations of MeHg in aquatic biota should respond most rapidly to altered mercury deposition in atmospherically dominated systems, such as perched seepage lakes, which lack streams and receive little or no inflow of surface or ground water from the surrounding terrestrial environment. The production of MeHg and its bioaccumulation in such seepage lakes are therefore influenced little by watershed characteristics and processes (Watras et al. 1994; Hrabik and Watras 2002; Watras et al. 2002; Wiener et al. 2003).
7) *Ensure that contemporary and historic monitoring data are valid and comparable.* Mercury data from different biological monitoring programs often are not directly comparable because of variation in methods for sampling, sample preparation, and analyses. Contemporary monitoring efforts that intend to use historic data on biological indicators should take steps to ensure that 1) the historic data are valid and statistically useful and 2) the methods applied in contemporary and historic monitoring produce comparable data. Quality assurance procedures should be implemented to document the accuracy and ensure the reliability of analytical measurements. Increased uniformity in methods for sampling, sample preparation, chemical analyses, database management, and statistical summarization of data would facilitate comparison and synthesis of mercury data across geopolitical boundaries.
8) *Continue monitoring for a sufficient time frame.* Temporal records of mercury concentrations in aquatic biota should be sufficiently long to detect decade-scale trends that could be obscured by short-term fluctuations caused by interannual variability. This requires a commitment to sustained allocation of resources at the onset of a trend-monitoring program, for a recommended time frame of 20 to 30 years. Existing monitoring or research programs may include suitable candidate sites for monitoring, for which reliable historic data on mercury and pertinent ancillary variables are available.

Additional detailed guidance concerning the development of monitoring programs for bioaccumulative contaminants is available from the U.S. Environmental Protection Agency (2000) and from other chapters in this book (Chapters 2, 3, and 5). Provision of statistical guidance relevant to sampling design and data analyses is beyond the scope of this chapter, but such information is available from other sources (Gilbert 1987; Jorgensen and Pedersen 1994; Tremblay et al. 1998; Conquest 2000; U.S. Environmental Protection Agency 2000).

ACKNOWLEDGMENTS

The lead author (JGW) was supported by a Wisconsin Distinguished Professorship jointly funded by the University of Wisconsin System, the University of Wisconsin–La Crosse (UW–L) Foundation, and the UW–L College of Science and Health during the preparation of this chapter. We thank Michael Paterson (Fisheries and Oceans Canada, Winnipeg, Manitoba) for allowing the use of his unpublished data on methylmercury in zooplankton; Bruce Monson (Minnesota Pollution Control Agency, St. Paul) for sharing results from his trend analysis of mercury in game fish from Minnesota lakes; and Thomas Johnston (Fisheries and Oceans Canada, Burlington, Ontario) for providing trend data on mercury in walleyes from lakes in Ontario and Manitoba. Constructive reviews of a draft chapter were provided by Robin Stewart, Mark Brigham, Michael Murray, Reed Harris, and Tamara Saltman.

REFERENCES

Allan CJ, Heyes A, Roulet NT, St Louis VL, Rudd JWM. 2001. Spatial and temporal dynamics of mercury in Precambrian Shield upland runoff. Biogeochemistry 52:13–40.

Andersson P, Borg H, Kaerrhage P. 1995. Mercury in fish muscle in acidified and limed lakes. Water Air Soil Pollut 80:889–892.

Atkeson T, Axelrad D, Pollman C, Keeler G. 2003. Integrating atmospheric mercury deposition and aquatic cycling in the Florida Everglades: integrated summary. Tallahassee (FL): Florida Department of Environmental Protection, p. 75–90.

Back RC, Visman V, Watras CJ. 1995. Microhomogenization of individual zooplankton species improves mercury and methylmercury determinations. Can J Fish Aquat Sci 52:2470–2475.

Back RC, Watras CJ. 1995. Mercury in zooplankton of northern Wisconsin lakes: taxonomic and site-specific trends. Water Air Soil Pollut 80:931–938.

Baker RF, Blanchfield PJ, Paterson MJ, Flett RJ, Wesson L. 2004. Evaluation of non-lethal methods for the analysis of mercury in fish tissue. Trans Am Fish Soc 133:568–576.

Barr JF. 1996. Aspects of common loon (*Gavia immer*) feeding biology on its breeding ground. Hydrobiologia 321:119–144.

Becker DS, Bigham GN. 1995. Distribution of mercury in the aquatic food web of Onondaga Lake, New York. Water Air Soil Pollut 80:563–571.

Becker GC. 1983. Fishes of Wisconsin. Madison (WI): University of Wisconsin Press. 1052 p.

Benoit JM, Fitzgerald WF, Damman AWH. 1998. The biogeochemistry of an ombrotrophic bog: evaluation of use as an archive of atmospheric mercury deposition. Environ Res (Sect A) 78:118–133.

Benoit J, Gilmour C, Heyes A, Mason RP, Miller C. 2003. Geochemical and biological controls over methylmercury production and degradation in aquatic ecosystems. In: Chai Y, Braids OC, editors, Biogeochemistry of environmentally important trace elements. Washington, DC: American Chemical Society, p. 262–297.

Bignert A, Riget F, Braune B, Outridge P, Wilson S. 2004. Recent temporal trend monitoring of mercury in Arctic biota — how powerful are the existing data sets? J Environ Monitoring 6:351–355.

Bindler R, Renberg I, Appleby PG, Anderson NJ, Rose NL. 2001. Mercury accumulation rates and spatial patterns in lake sediments from West Greenland: a coast to ice margin transect. Environ Sci Technol 35:1736–1741.

Bloom NS. 1992. On the chemical form of mercury in edible fish and marine invertebrate tissue. Can J Fish Aquat Sci 49:1010–1017.

Bodaly RA, Fudge RJP. 1999. Uptake of mercury by fish in an experimental boreal reservoir. Arch Environ Contam Toxicol 37:103–109.

Bodaly RA, Hecky RE, Fudge RJP. 1984. Increases in fish mercury levels in lakes flooded by the Churchill River diversion, northern Manitoba. Can J Fish Aquat Sci 41:682–691.

Bodaly RA, Rudd JWM, Fudge RJP, Kelly CA. 1993. Mercury concentrations in fish related to size of remote Canadian shield lakes. Can J Fish Aquat Sci 50:980–987.

Bodaly RA, St. Louis VL, Paterson MJ, Fudge RJP, Hall BD, Rosenberg DM, Rudd JWM. 1997. Bioaccumulation of mercury in the aquatic food chain in newly flooded areas. In: Sigel A, Sigel H, editors, Metal ions in biological systems, Vol. 34: Mercury and its effects on environment and biology. New York (NY): Marcel Dekker Inc., p. 259–287.

Bowles KC, Apte SC, Maher WA, Kawei M, Smith R. 2001. Bioaccumulation and biomagnification of mercury in Lake Murray, Papua New Guinea. Can J Fish Aquat Sci 58:888–897.

Broman D, Naf C, Rolff C, Zebuhr Y, Fry B, Hobbie J. 1992. Using ratios of stable nitrogen isotopes to estimate bioaccumulation and flux of polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) in two food chains from the northern Baltic. Environ Toxicol Chem 11:331–345.

Brown SS, Gaston GR, Rakocinski CF, Heard RW. 2000. Effects of sediment contaminants and environmental gradients on macrobenthic community trophic structure in Gulf of Mexico estuaries. Estuaries 23:411–424.

Cabana G, Rasmussen JB. 1994. Modelling food chain structure and contaminant bioaccumulation using stable nitrogen isotopes. Nature 372:255–257.

Cizdziel JV, Hinners TA, Pollard JE, Heithmar EM, Cross CL. 2002. Mercury concentrations in fish from Lake Mead, USA, related to fish size, condition, trophic level, location, and consumption risk. Arch Environ Contam Toxicol 43:309–317.

Clarkson TW. 2002. The three modern faces of mercury. Environ Health Perspect 110(Suppl 1):11–23.

Cleckner LB, Garrison PG, Hurley JP, Olson ML, Krabbenhoft DP. 1998. Trophic transfer of methyl mercury in the northern Florida Everglades. Biogeochemistry 40:347–361.

Cleckner LB, Gilmour CC, Krabbenhoft DP, Hurley JP. 1999. Mercury methylation in periphyton of the Florida Everglades. Limnol Oceanogr 44:1815–1825.

Colby PJ, McNicol RE, Ryder RA. 1979. Synopsis of biological data on the walleye *Stizostedion vitrem vitreum* (Mitchill 1818). Rome, Italy: Food and Agriculture Organization of the United Nations. FAO Fisheries Synopsis No 119. 139 p.

Coleman FC, Figueira WF, Ueland JS, Crowder LB. 2004. The impact of United States recreational fisheries on marine fish populations. Science 305:1958–1960.

Conquest LL. 2000. Environmental monitoring: investigating associations and trends. In: Sparks T, editor, Statistics in ecotoxicology. New York (NY): John Wiley & Sons, p. 179–210.

Cope WG, Wiener JG, Rada RG. 1990. Mercury accumulation in yellow perch in Wisconsin seepage lakes: relation to lake characteristics. Environ Toxicol Chem 9:931–940.

Curtis PJ. 1998. Climatic and hydrologic control of DOM concentration and quality in lakes. In: Hessen DO, Tranvik LJ, editors, Aquatic humic substances: ecology and biogeochemistry. Berlin, Germany: Springer-Verlag, p. 93–105.

DeVries DR, Frie RV. 1996. Determination of age and growth. In: Murphy BR, Willis DW, editors, Fisheries techniques, 2nd ed. Bethesda (MD): American Fisheries Society, p. 483–512.

Doire J, Lucotte M, Fortin R, Verdon R. 2002. Influence of intensive fishing on fish diet in natural lakes from northern Québec: use of stable nitrogen and carbon isotopes (abstract). American Society of Limnology and Oceanography, Summer Meeting, Victoria, BC, Canada, June 10–14, 2002. http://www.aslo.org/meetings/victoria2002/archive/409.html

Downs SG, Macleod CL, Lester JN. 1998. Mercury in precipitation and its relation to bioaccumulation in fish: a literature review. Water Air Soil Pollut 108:149–187.

Drevnick PE, Sandheinrich MB. 2003. Effects of dietary methylmercury on reproductive endocrinology of fathead minnows. Environ Sci Technol 37:4390–4396.

Engstrom DR, Swain EB. 1997. Recent declines in atmospheric mercury deposition in the Upper Midwest. Environ Sci Technol 31:960–967.

Environmental Council of States and Clean Air Network. 2001. Mercury in the environment: states respond to the challenge. Washington, DC: Environmental Council of States. 70 p. http://www.sso.org/ecos/projects/Mercury/ECOS2%20State%20Hg%20Activities.pdf.

Exponent. 2003. Fish contaminant monitoring program: review and recommendations prepared for Michigan Department of Environmental Quality, Water Division, Lansing, MI, Doc. No. 8601969.001 0501 0103 BH29. http://www.deq.state.mi.us/documents/deq-wd-fcmp-fcmpfinal.pdf

Fitzgerald WF, Engstrom DR, Mason RP, Nater EA. 1998. The case for atmospheric mercury contamination in remote areas. Environ Sci Technol 32:1–7.

Fjeld E, Haugen TO, Vøllestad LA. 1998. Permanent impairment in the feeding behavior of grayling (*Thymallus thymallus*) exposed to methylmercury during embryogenesis. Sci Total Environ 213:247–254.

Francesconi KA, Lenanton RCJ. 1992. Mercury contamination in a semi-enclosed marine embayment: organic and inorganic mercury content of biota, and factors influencing mercury levels in fish. Mar Environ Res 33:189–212.

Frost TM, Montz PK, Kratz TK, Badillo T, Brezonik PL, Gonzalez MJ, Rada RG, Watras CJ, Webster KE, Wiener JG, Williamson CE, Morris DP. 1999. Multiple stresses from a single agent: diverse responses to the experimental acidification of Little Rock Lake, Wisconsin. Limnol Oceanogr 44(3, part 2):784–794.

Fry B. 1991. Stable isotope diagrams of freshwater food webs. Ecology 72:2293–2297.

Garcia E, Carignan R. 1999. Impact of wildfire and clear-cutting in the boreal forest on methylmercury in zooplankton. Can J Fish Aquat Sci 56:339–345.

Gilbert RO. 1987. Statistical methods for environmental pollution monitoring. New York (NY): Van Nostrand Reinhold Company. 320 p.

Gilmour CC, Riedel GS, Ederington MC, Bell JT, Benoit JM, Gill GA, Stordal MC. 1998. Methylmercury concentrations and production rates across a trophic gradient in the northern Everglades. Biogeochemistry 40:327–345.

Glass GE, Sorensen JA, Rapp GR Jr. 1998. Mercury deposition and lake quality trends. Report to the Minnesota Pollution Control Agency, St. Paul, MN.

Gorski PR, Cleckner LB, Hurley JP, Sierzen ME, Armstrong DE. 2003. Factors affecting enhanced mercury bioaccumulation in inland lakes of Isle Royale National Park, USA. Sci Total Environ 304:327–348.

Gorski PR, Lathrop RC, Hill SD, Herrin RT. 1999. Temporal mercury dynamics and diet composition in the mimic shiner. Trans Am Fish Soc 128:701–712.

Göthberg A. 1983. Intensive fishing — a way to reduce the mercury level in fish. Ambio 12:259–261.

Grieb TM, Driscoll CT, Gloss SP, Schofield CL, Bowie GL, Porcella DB. 1990. Factors affecting mercury accumulation in fish in the upper Michigan peninsula. Environ Toxicol Chem 9:919–930.

Haines TA, Komov VT, Matey VE, Jagoe CH. 1995. Perch mercury content is related to acidity and color of 26 Russian lakes. Water Air Soil Pollut 85:823–828.

Hall BD, Bodaly RA, Fudge RJP, Rudd JWM, Rosenberg DM. 1997. Food as the dominant pathway of methylmercury uptake by fish. Water Air Soil Pollut 100:13–24.

Hammerschmidt CR, Sandheinrich MB, Wiener JG, Rada RG. 2002. Effects of dietary methylmercury on reproduction of fathead minnows. Environ Sci Technol 36:877–883.

Hammerschmidt CR, Wiener JG, Frazier BE, Rada RG. 1999. Methylmercury content of eggs in yellow perch related to maternal exposure in four Wisconsin lakes. Environ Sci Technol 33:999–1003.

Harris RC, Bodaly RA. 1998. Temperature, growth and dietary effects on fish mercury dynamics in two Ontario lakes. Biogeochemistry 40:175–187.

Hebert CE, Weseloh DVC. 2003. Assessing temporal trends in contaminants from long-term avian monitoring programs: the influence of sampling frequency. Ecotoxicology 12:141–151.

Heinz GH. 1996. Mercury poisoning in wildlife. In: Fairbrother A, Locke LN, Hoff GL, editors, Noninfectious diseases of wildlife, 2nd ed. Ames (IA): Iowa State University Press, p. 118–127.

Henry EA, Dodge-Murphy LJ, Bigham GN, Klein SM, Gilmour CC. 1995. Total mercury and methylmercury mass balance in an alkaline, hypereutrophic urban lake (Onondaga Lake, NY). Water Air Soil Pollut 80:509–518.

Herrin RT, Lathrop RC, Gorski PR, Andren AW. 1998. Hypolimnetic methylmercury and its uptake by plankton during fall destratification: a key entry point of mercury into lake food chains? Limnol Oceanogr 43:1476–1486.

Hill WR, Stewart AJ, Napolitano GE. 1996. Mercury speciation and bioaccumulation in lotic primary producers and primary consumers. Can J Fish Aquat Sci 53:812–819.

Hrabik TR, Magnuson JJ, McLain AS. 1998. Predicting the effects of rainbow smelt on native fishes in small lakes: evidence from long-term research on two lakes. Can J Fish Aquat Sci 55:1364–1371.

Hrabik TR, Watras CJ. 2002. Recent declines in mercury concentration in a freshwater fishery: isolating the effects of de-acidification and decreased atmospheric mercury deposition in Little Rock Lake. Sci Total Environ 297:229–237.

Hurley JP, Benoit JM, Babiarz CL, Shafer MM, Andren AW, Sullivan JR, Hammond R, Webb DA. 1995. Influences of watershed characteristics on mercury levels in Wisconsin waters. Environ Sci Technol 29:1867–1875.

Hutchings JA, Reynolds JD. 2004. Marine fish population collapses: consequences for recovery and extinction risk. BioScience 54:297–309.

Jackson DA, Harvey HH. 1993. Fish and benthic invertebrates: community concordance and community-environment relationships. Can J Fish Aquat Sci 50:2641–2651.

Jackson TA. 1997. Long-range atmospheric transport of mercury to ecosystems, and the importance of anthropogenic emissions — a critical review and evaluation of the published evidence. Environ Rev 5:99–120.

Jackson TA. 1998. Mercury in aquatic ecosystems. In: Langston WJ, Bebianno MJ, editors, Metal metabolism in aquatic environments. London, UK: Chapman & Hall, p. 77–158.

Johansson K, Aastrup M, Andersson A, Bringmark L, Iverfeldt Å. 1991. Mercury in Swedish forest soils and waters — assessment of critical load. Water Air Soil Pollut 56:267–281.

Johnston TA, Bodaly RA, Matias JA. 1991. Predicting fish mercury levels from physical characteristics of boreal reservoirs. Can J Fish Aquat Sci 48:1468–1475.

Johnston TA, Leggett WC, Bodaly RA, Swanson HK. 2003. Temporal changes in mercury bioaccumulation by predatory fishes of boreal lakes following the invasion of an exotic forage fish. Environ Toxicol Chem 22:2057–2062.

Jorgensen LA, Pedersen B. 1994. Trace metals in fish used for time trend analysis and as environmental indicators. Marine Pollut Bull 28:235–243.

Kainz M, Lucotte M, Parrish CC. 2003. Relationships between organic matter composition and methylmercury content of offshore and carbon-rich littoral sediments in an oligotrophic lake. Can J Fish Aquat Sci 6:888–896.

Kelly CA, Rudd JWM, Holoka MH. 2003. Effect of pH on mercury uptake by an aquatic bacterium: implications for Hg cycling. Environ Sci Technol 37:2941–2946.

Kidd KA, Hesslein RH, Fudge RJP, Hallard KA. 1995. The influence of trophic level as measured by $\delta^{15}N$ on mercury concentrations in freshwater organisms. Water Air Soil Pollut 80:1011–1015.

Kraepiel AML, Keller K, Chin HB, Malcolm EG, Morel FMM. 2003. Sources and variations of mercury in tuna. Environ Sci Technol 37:5551–5558.

Lamborg CH, Fitzgerald WF, Damman AWH, Benoit JM, Balcom PH, Engstrom DR. 2002. Modern and historic atmospheric mercury fluxes in both hemispheres: global and regional mercury cycling implications. Global Biogeochem Cycles 16(4):1104.

Lange TR, Royals HE, Connor LL. 1993. Influence of water chemistry on mercury concentration in largemouth bass from Florida lakes. Trans Am Fish Soc 122:74–84.

Latif MA, Bodaly RA, Johnston TA, Fudge RJP. 2001. Effects of environmental and maternally derived methylmercury on the embryonic and larval stages of walleye (*Stizostedion vitreum*). Environ Pollut 111:139–148.

Lean DRS, Siciliano SD. 2003. Production of methylmercury by solar radiation. J Phys IV, Proc 12th Internat Conf on Heavy Metal in the Environment 107:743–747.

Lin CJ, Cheng MD, Schroeder WH. 2001. Transport patterns and potential sources of total gaseous mercury measured in Canadian high Arctic in 1995. Atmos Environ 35:1141–1154.

Lindestroem L. 2001. Mercury in sediment and fish communities of Lake Vaenern, Sweden: recovery from contamination. Ambio 30:538–544.

Lockhart WL, Uthe JF, Kenney AR, Mehrle PM. 1972. Methylmercury in northern pike (*Esox lucius*): distribution, elimination, and some biochemical characteristics of contaminated fish. J Fish Res Board Can 29:1519–1523.

Lockhart WL, Wilkinson P, Billeck BN, Danell RA, Hunt RV, Brunskill GJ, DeLaronde J, St. Louis V. 1998. Fluxes of mercury to lake sediments in central and northern Canada inferred from dated sediment cores. Biogeochemistry 40:163–173.

Lorey P, Driscoll CT. 1999. Historical trends of mercury deposition in Adirondack lakes. Environ Sci Technol 33:718–722.

Lucotte M, Montgomery S, Bégin M. 1999b. Mercury dynamics at the flooded soil-water interface in reservoirs of Northern Québec: *in situ* observations. In: Lucotte M, Schetagne R, Thérien N, Langlois C, Tremblay A, editors, Mercury in the biogeochemical cycle. Berlin, Germany: Springer, p. 193–214.

Lucotte M, Montgomery S, Caron B, Kainz M. 1999a. Mercury in natural lakes and unperturbed terrestrial ecosystems of northern Québec. In: Lucotte M, Schetagne R, Thérien N, Langlois C, Tremblay A, editors. Mercury in the biogeochemical cycle. Berlin, Germany: Springer, p. 55–88.

Lucotte M, Mucci A, Hillaire-Marcel C, Pichet P, Grondin A. 1995. Anthropogenic mercury enrichment in remote lakes of northern Québec (Canada). Water Air Soil Pollut 80:467–476.

Lucotte M, Schetagne R, Thérien N, Langlois C, Tremblay A, editors. 1999c. Mercury in the biogeochemical cycle. Berlin, Germany: Springer. 334 p.

Lyons J, Cochran PA, Fago D. 2000. Wisconsin fishes 2000 — status and distribution. Madison (WI): University of Wisconsin Sea Grant Institute. 87 p.

Mahaffey KR. 2000. Recent advances in recognition of low-level methylmercury poisoning. Current Opinion Neurol 13:699–707.

Mahaffey KR, Clickner RP, Bodurow CC. 2004. Blood organic mercury and dietary mercury intake: National Health and Nutrition Examination Survey, 1999 and 2000. Environ Health Perspect 112:562–570.

Martínez-Cortizas A, Pontevedra-Pombal X, Garcia-Rodeja E, Nóvoa-Muñoz JC, Shotyk W. 1999. Mercury in a Spanish peat bog: archive of climate change and atmospheric metal deposition. Science 284:939–942.

Mason RP, Reinfelder JR, Morel FMM. 1996. Uptake, toxicity, and trophic transfer of mercury in a coastal diatom. Environ Sci Technol 30:1835–1845.

Masson S, Tremblay A. 2002. Effects of intensive fishing on the structure of zooplankton communities and mercury levels. Sci Total Environ 304:377–390.

Meili M. 1991. Mercury in boreal forest lake ecosystems. Comprehensive summaries of Uppsala Dissertations from the Faculty of Science, Report No. 336, Uppsala University, Sweden.

Metsaelae T, Rask M. 1989. Mercury concentrations of perch, *Perca fluviatilis* L., in small Finnish headwater lakes with different pH and water colour. Aqua Fennica 19:41–46.

Miskimmin BM, Rudd JWM, Kelly CA. 1992. Influence of dissolved organic carbon, pH, and microbial respiration rates on mercury methylation and demethylation in lake water. Can J Fish Aquat Sci 49:17–22.

Monson BA, Brezonik PL. 1999. Influence of food, aquatic humus, and alkalinity on methylmercury uptake by *Daphnia magna*. Environ Toxicol Chem 18:560–566.

Monteiro LR, Furness RW. 1997. Accelerated increase in mercury contamination in North Atlantic mesopelagic food chains as indicated by time series of seabird feathers. Environ Toxicol Chem 16:2489–2493.

Montgomery S, Lucotte M, Cournoyer L. 2000. The use of stable isotopes to evaluate the importance of fine suspended particulate matter in the transfer of methylmercury to biota in boreal flooded environments. Sci Total Environ 261:33–41.

Morel FMM, Kraepiel AML, Amyot M. 1998. The chemical cycle and bioaccumulation of mercury. Annu Rev Ecol Systematics 29:543–566.

Myers GJ, Davidson PW. 1998. Prenatal methylmercury exposure and children: neurologic, developmental, and behavioral research. Environ Health Perspect 106(Suppl 3):841–847.

NRC 2000. Toxicological effects of methylmercury. National Research Council Committee on the Toxicological Effects of Methylmercury, Washington, DC: National Academies Press. 344 p.

Odin M, Feurtet-Mazel A, Ribeyre F, Boudou A. 1994. Actions and interactions of temperature, pH and photoperiod on mercury bioaccumulation by nymphs of the burrowing mayfly *Hexagenia rigida*, from the sediment contamination source. Environ Toxicol Chem 13:1291–1302.

Parkman H, Meili M. 1993. Mercury in macroinvertebrates from Swedish forest lakes: influence of lake type, habitat, life cycle and food quality. Can J Fish Aquat Sci 50:521–534.

Parks JW, Hamilton AL. 1987. Accelerating recovery of the mercury-contaminated Wabigoon/English River system. Hydrobiologia 149:159–188.

Paterson MJ, Rudd JWM, St Louis VL. 1998. Increases in total and methylmercury in zooplankton following flooding of a peatland reservoir. Environ Sci Technol 32:3868–3874.

Pauly D, Alder J, Bennett E, Christensen V, Tyedmers P, Watson R. 2003. The future for fisheries. Science 302:1359–1361.

Peech Cherewyk K. 2002. Methylmercury bioaccumulation in zooplankton: an assessment of exposure routes and accumulation in newly flooded reservoirs. MS thesis, University of Manitoba, Winnipeg, Canada, 90 p.

Peterson SA, Van Sickle J, Hughes RM, Schacher JA, Echols SF. 2005. A biopsy procedure for determining filet and predicting whole-fish mercury concentration. Arch Environ Contam Toxicol 48:99–107.

Phillips DL. 2001. Mixing models in analyses of diet using multiple stable isotopes: a critique. Oecologia 127:166–170.

Phillips DL, Gregg JW. 2001. Uncertainty in source partitioning using stable isotopes. Oecologia 127:171–179.

Pickhardt PC, Folt CL, Chen CY, Klaue B, Blum JD. 2002. Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. Proc Nat Acad Sci 99:4419–4423.

Plourde Y, Lucotte M, Pichet P. 1997. Contribution of suspended particulate matter and zooplankton to methylmercury contamination of the food chain in mid-northern Québec (Canada) reservoirs. Can J Fish Aquat Sci 54:821–831.

Porvari P. 1995. Mercury levels of fish in Tucurui hydroelectric reservoir and in River Moju in Amazonia, in the state of Para, Brazil. Sci Total Environ 175:109–117.

Porvari P. 1998. Development of fish mercury concentrations in Finnish reservoirs from 1979 to 1994. Sci Total Environ 213:279–290.

Porvari P, Verta M, Munthe J, Haapanen M. 2003. Forestry practices increase mercury and methyl mercury output from boreal forest catchments. Environ Sci Technol 37:2389–2393.

Rencz AN, O'Driscoll NJ, Hall GEM, Peron T, Telmer K, Burgess NM. 2003. Spatial variation and correlations of mercury levels in the terrestrial and aquatic components of a wetland dominated ecosystem: Kejimkujik Park, Nova Scotia, Canada. Water Air Soil Pollut 143:271–288.

Resh VH, McElvary EP. 1993. Contemporary quantitative approaches to biomonitoring using benthic macroinvertebrates. In: Rosenberg DM, Resh VH, editors. Freshwater biomonitoring and benthic macroinvertebrates. New York (NY): Chapman & Hall, p. 159–194.

Rodgers DW. 1994. You are what you eat and a little bit more: bioenergetics-based models of methylmercury accumulation in fish revisited. In: Watras CJ, Huckabee JW, editors, Mercury pollution: integration and synthesis. Boca Raton (FL): Lewis Publishers, p. 427–439.

Rolfhus KR, Fitzgerald WF. 1995. Linkages between atmospheric mercury deposition and the methylmercury content of marine fish. Water Air Soil Pollut 80:291–297.

Roseman EF, Mills EL, Forney JL, Rudstam LG. 1996. Evaluation of competition between age-0 yellow perch (*Perca flavescens*) and gizzard shad (*Dorosoma cepedianum*) in Oneida Lake, New York. Can J Fish Aquat Sci 53:865–874.

Roulet M, Lucotte M, Canuel R, Farella N, Courcelles M, Guimarães J-R, Mergler D, Amorim M. 2000. Increase in mercury contamination recorded in lacustrine sediments following deforestation in the central Amazon. Chem Geol 165:243–266.

Roulet M, Lucotte M, Farella N, Serique G, Coelho H, Sousa Passos CJ, de Jesus da Silva E, Scavone de Andrade P, Mergler D, Amorim M. 1999. Effects of recent human colonization on the presence of mercury in Amazonian ecosystems. Water Air Soil Pollut 112:297–313.

Roulet M, Lucotte M, Saint-Aubin A, Tran S, Rheault I, Farella N, de Jesus da Silva E, Dezencourt J, Sousa Passos CJ, Santos Soares G, Guimarães JR, Mergler D, Amorim M. 1998. The geochemistry of mercury in central Amazonian soils developed on the Alter-do-Chão formation of the lower Tapajós River valley, Pará state, Brazil. Sci Total Environ 223:1–24.

Rusak JA, Yan ND, Somers KM, Cottingham KL, Micheli F, Carpenter SR, Frost TM, Paterson MJ, McQueen DJ. 2002. Temporal, spatial, and taxonomic patterns of crustacean zooplankton variability in unmanipulated north-temperate lakes. Limnol Oceanogr 47:613–625.

Schetagne R, Verdon R. 1999. Post-impoundment evolution of fish mercury levels at the La Grande complex, Québec, Canada (from 1978 to 1996). In: Lucotte M, Schetagne R, Thérien N, Langlois C, Tremblay A, editors, Mercury in the biogeochemical cycle. Berlin, Germany: Springer, p. 235–258.

Scheuhammer AM, Graham JE. 1999. The bioaccumulation of mercury in aquatic organisms from two similar lakes with differing pH. Ecotoxicology 8:49–56.

Schober SE, Sinks TH, Jones RL, Bolger, PM, McDowell M, Osterloh J, Garrett ES, Canady RA, Dillon CF, Sun Y, Joseph CB, Mahaffey KR. 2003. Blood mercury levels in US children and women of childbearing age, 1999–2000. J Am Med Assoc 289:1667–1674.

Schuster PF, Krabbenhoft DP, Naftz DL, Cecil LD, Olson ML, DeWild JF, Susong DD, Green JR, Abbott ML. 2002. Atmospheric mercury deposition during the last 270 years: a glacial ice core record of natural and anthropogenic sources. Environ Sci Technol 36:2303–2310.

Scott WB, Crossman EJ. 1973. Freshwater fishes of Canada. Ottawa, Ontario: Fish Res Board Can Bull 184. 966 p.

Seda J, Kubecka J. 1997. Long-term biomanipulation of Rimov Reservoir (Czech Republic). Hydrobiologia 345:95–108.

Sellers P, Kelly CA, Rudd JWM, MacHutchon AR. 1996. Photodegradation of methylmercury in lakes. Nature 380:694–697.

Sellers P, Kelly CA, Rudd JWM. 2001. Fluxes of methylmercury to the water column of a drainage lake: the relative importance of internal and external sources. Limnol Oceanogr 46:623–631.

Serdar D, Johnston J, Mueller K, Patrick G. 2001. Mercury concentrations in edible muscle of Lake Whatcom fish. Olympia (WA): Washington State Department of Ecology. Pub No 01-03-012. 28 p. + appendices. http://www.ecy.wa.gov/pubs/0103012.pdf

Shotyk W, Goodsite ME, Roos-Barraclough F, Frei R, Heinemeier J, Asmund G, Lohse C, Hansen TS. 2003. Anthropogenic contributions to atmospheric Hg, Pb and As accumulation recorded by peat cores from southern Greenland and Denmark dated using the ^{14}C "bomb pulse curve." Geochim Cosmochim Acta 67:3991–4011.

Shotyk W, Goodsite ME, Roos-Barraclough F, Givelet N, Le Roux G, Weiss D, Cheburkin AK, Knudsen K, Heinemeier J, van Der Knaap WO, Norton SA, Lohse C. 2005. Accumulation rates and predominant atmospheric sources of natural and anthropogenic Hg and Pb on the Faroe Islands. Geochim Cosmochim Acta 69:1–17.

Simoneau M, Lucotte M, Garceau S, Laliberté D. 2005. Fish growth rates modulate mercury concentrations in walleye (*Sander vitreus*) from eastern Canadian lakes. Environ Res 98:73–82.

Simonin HA, Gloss SP, Driscoll CT, Schofield CL, Kretser WA, Karcher RW, Symula J. 1994. Mercury in yellow perch from Adirondack drainage lakes (New York, U.S.). In: Watras CJ, Huckabee JW, editors, Mercury pollution: integration and synthesis. Boca Raton (FL): Lewis Publishers, p. 457–469.

Smith AL, Green RH. 1975. Uptake of mercury by freshwater clams (family Unionidae). J Fish Res Board Can 32:1297–1303.

Sonesten L. 2003. Fish mercury levels in lakes — adjusting for Hg and fish-size covariation. Environ Pollut 125:255–265.

Sorensen JA, Kallemeyn LW, Sydor M. 2005. Relationship between mercury accumulation in young-of-the-year yellow perch and water-level fluctuations. Environ Sci Technol 39:9237–9243.

Southerland MT, Stribling JB. 1995. Status of biological criteria development and implementation. In: Davis WS, Simon TP, editors, Biological assessment and criteria: tools for water resource planning and decision making. Boca Raton (FL): Lewis Publishers, p. 81–96.

Southworth GR, Peterson MJ, Ryon MG. 2000. Long-term increased bioaccumulation of mercury in largemouth bass follows reduction of waterborne selenium. Chemosphere 41:1101–1105.

Spry DJ, Wiener JG. 1991. Metal bioavailability and toxicity to fish in low-alkalinity lakes: a critical review. Environ Pollut 71:243–304.

St. Louis VL, Rudd JWM, Kelly CA, Beaty KG, Flett RJ, Roulet NT. 1996. Production and loss of methylmercury and loss of total mercury from boreal forest catchments containing different types of wetlands. Environ Sci Technol 30:2719–2729.

Stafford CP, Haines TA. 2001. Mercury contamination and growth rate in two piscivore populations. Environ Toxicol Chem 20:2099–2101.

Suns K, Hitchin G. 1990. Interrelationships between mercury levels in yearling yellow perch, fish condition and water quality. Water Air Soil Pollut 50:255–265.

Suns K, Hitchin G, Loescher B, Pastorek E, Pearce R. 1987. Metal accumulations in fishes from Muskoka-Haliburton lakes in Ontario (1978–1984). Rexdale, Ontario, Canada: Ontario Ministry of the Environment. Technical Report. 38 p.

Surette C, Lucotte M, Doire J, Tremblay A. 2003. Effects of intensive fishing in three natural lakes of northern Québec, Canada, J Phys IV, Proc 12th Internat Conf on Heavy Metals in the Environment 107:1443.

Surette C, Lucotte M, Tremblay A. 2006. Influence of intensive fishing on the partitioning of mercury and methylmercury in three lakes of Northern Québec. Sci Total Environ 368(1):248–261.

Svobodova Z, Dusek L, Hejtmanek M, Vykusova B, Smid R. 1999. Bioaccumulation of mercury in various fish species from Orlik and Kamyk Water Reservoirs in the Czech Republic. Ecotoxicol Environ Safety 43:231–240.

Swain EB, Engstrom DR, Brigham ME, Henning TA, Brezonik PL. 1992. Increasing rates of atmospheric mercury deposition in midcontinental North America. Science 257:784–787.

Thomann RV, Mahoney JD, Mueller R. 1995. Steady-state model of biota sediment accumulation factor for metals in two marine bivalves. Environ Toxicol Chem 14:1989–1998.

Thorpe J. 1977. Synopsis of biological data on the perch *Perca fluviatilis* (Linnaeus, 1758) and *Perca flavescens* (Mitchill, 1814). Rome, Italy: Food and Agriculture Organization of the United Nations. FAO Fisheries Synopsis No 113. 138 p.

Tremblay G, Legendre P, Doyon J-F, Verdon R, Schetagne R. 1998. The use of polynomial regression analysis with indicator variables for interpretation of mercury in fish data. Biogeochemistry 40:189–201.

Tremblay A, Lucotte M. 1997. Accumulation of total mercury and methylmercury in insect larvae of hydroelectric reservoirs. Can J Fish Aquat Sci 54:832–841.

Tsui MTK, Wang WX. 2004. Uptake and elimination routes of inorganic mercury and methylmercury in *Daphnia magna*. Environ Sci Technol 38:808–816.

[USEPA] US Environmental Protection Agency. 2000. Guidance for assessing chemical contamination data for use in fish advisories. Vol. 1: Fish sampling and analysis, 3rd ed. Washington, DC: USEPA Office of Water. EPA 823-B-00-007. http://www.epa.gov/waterscience/fishadvice/volume1/index.html

[USEPA] US Environmental Protection Agency. 2001. Update: national listing of fish and wildlife advisories. Washington, DC: USEPA, Office of Water. Fact Sheet EPA-823-F-01-010.

[USEPA] US Environmental Protection Agency. 2005. 2004 National listing of fish advisories. Washington, DC: USEPA, Office of Water. Fact Sheet EPA-823-F-05-004.

Vander Zanden MJ, Vadeboncoeur Y. 2002. Fishes as integrators of benthic and pelagic food webs in lakes. Ecology 83:2152–2161.

Verdon R, Tremblay A. 1999. Mercury accumulation in fish from the La Grande complex: influence of feeding habits and concentrations of mercury in ingested prey. In: Lucotte M, Schetagne R, Thérien N, Langlois C, Tremblay A, editors, Mercury in the biogeochemical cycle. Berlin, Germany: Springer, p. 215–233.

Verta M. 1990. Changes in fish mercury concentrations in an intensively fished lake. Can J Fish Aquat Sci 47:1888–1897.

Wang XF, Fisher NS. 1999. Assimilation efficiencies of chemical contaminants in aquatic invertebrates: a synthesis. Environ Toxicol Chem 18:2034–2045.

Watras CJ, Back RC, Halvorsen S, Hudson RJM, Morrison KA, Wente SP. 1998. Bioaccumulation of mercury in pelagic freshwater food webs. Sci Total Environ 219:183–208.

Watras CJ, Bloom NS, Hudson RJM, Gherini S, Munson R, Claas SA, Morrison KA, Hurley J, Wiener JG, Fitzgerald WF, Mason R, Vandal G, Powell D, Rada R, Rislove L, Winfrey M, Elder J, Krabbenhoft D, Andren AW, Babiarz C, Porcella DB, Huckabee JW. 1994. Sources and fates of mercury and methylmercury in Wisconsin lakes. In: Watras CJ, Huckabee JW, editors, Mercury pollution: integration and synthesis. Boca Raton (FL): Lewis Publishers, p. 153–177.

Watras CJ, Morrison KA, Kratz TK. 2002. Seasonal enrichment and depletion of Hg and SO_4 in Little Rock Lake: relationship to seasonal changes in atmospheric deposition. Can J Fish Aquat Sci 59:1660–1667.

Weigel BM, Wang LZ, Rasmussen PW, Butcher JT, Stewart PM, Simon TP, Wiley MJ. 2003. Relative influence of variables at multiple spatial scales on stream macroinvertebrates in the Northern Lakes and Forest ecoregion, USA. Freshwat Biol 48:1440–1461.

Wiener JG, Fitzgerald WF, Watras CJ, Rada RG. 1990. Partitioning and bioavailability of mercury in an experimentally acidified Wisconsin lake. Environ Toxicol Chem 9:909–918.

Wiener JG, Krabbenhoft DP, Heinz GH, Scheuhammer AM. 2003. Ecotoxicology of mercury. In: Hoffman DJ, Rattner BA, Burton GA Jr, Cairns J Jr, editors, Handbook of ecotoxicology, 2nd ed. Boca Raton (FL): CRC Press, p. 409–463.

Wiener JG, Shields PJ. 2000. Mercury in the Sudbury River (Massachusetts, USA): pollution history and a synthesis of recent research. Can J Fish Aquat Sci 57:1053–1061.

Wolfe MF, Schwarzbach S, Sulaiman RA. 1998. Effects of mercury on wildlife: a comprehensive review. Environ Toxicol Chem 17:146–160.