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Assessment of the Potential for Mercury Biomagnification from Sediment in the St. Lawrence River (Cornwall) Area of Concern

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by

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ABSTRACT

Elevated concentrations of mercury exist in sediments of the Cornwall area of the St. Lawrence River. To assess the bioavailability of this mercury and its potential for effects on fish, wildlife and humans through biomagnification, a study was conducted involving (a) analyses of the relationships of total and methyl mercury concentrations in benthic invertebrates to those in sediment, and (b) predictions of concentrations of total and methyl mercury in representative consumers of benthic invertebrates and their predators using screening-level trophic transfer models.

In October 2001, sediment and 3 benthic invertebrate taxa (midges, snails, amphipods) were sampled from 34 locations in the St. Lawrence River at Cornwall, 22 of which were exposed in the past to mercury-contaminated industrial effluents and 12 of which were reference sites. Samples were analyzed for total and methyl mercury concentrations. A series of physico-chemical variables were also measured in sediment and overlying water. Exposed and reference sites were compared in terms of mercury concentrations in sediment and invertebrates. Relationships between mercury in each invertebrate taxon and mercury in sediment were evaluated by regression analysis. Physico-chemical sediment and water variables were included as additional predictors. Concentrations of total and methyl mercury in the tissues of fish and wildlife receptors (white sucker, yellow perch, walleye, great blue heron, mink) were predicted by multiplying measured body concentrations in the resident invertebrates by relevant biomagnification factors obtained from a review of pre-existing studies.

Total and methyl mercury concentrations in sediment at the majority of sites exposed to historical industrial discharges are substantially greater than concentrations at reference sites. Invertebrates from up to half of the exposed sites have total mercury concentrations above the maximum reference site concentration; for methyl mercury, this fraction is a third or less. Whereas the concentration of total mercury in sediment is strongly predictive of total mercury concentration in invertebrates (analysed without allowing gut clearance), methyl mercury in sediment is weakly correlated to methyl mercury in invertebrates and, for midges and

amphipods, is significant only after adjusting for effects of other sediment covariables (total nitrogen, % sand and Mn for midges; % silt and Mn for amphipods).

Assuming intermediate and maximum mercury exposure and uptake conditions, the trophic transfer modelling outcomes for perch and walleye indicate up to 9 exposed sites could be considered of concern because of predicted tissue concentrations of methyl mercury exceeding reference sites concentrations and the tissue residue guideline of 92 ng/g wet weight.

Results of this assessment suggest that mercury from sediment is taken up by invertebrates largely in inorganic form, but is likely not strongly incorporated into tissues as the more bioavailable and toxicologically relevant methyl form. Screening level predictions of mercury concentrations in fish receptors suggest that within several sites and zones of the study area, mercury could bioaccumulate in receptors to levels that are not protective of adverse effects. However, the likelihood of realizing this degree of mercury biomagnification is not clear due to uncertainties associated with predicting receptor mercury concentrations.

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ABBREVIATIONS, ACRONYMS AND SYMBOLS

adj	adjusted
AOC	Area of Concern
BMF	biomagnification factor
BSAF	biota-sediment accumulation factor
CI	confidence interval
CL	confidence limit
dw	dry weight
FCM	food chain multiplier
GLWQA	Great Lakes Water Quality Agreement
Hg	mercury; used where form (MeHg or THg) is unspecified
IJC	International Joint Commission
inv	invertebrate
LEL	lowest effect level
max	maximum
MeHg	methyl mercury
min	minimum
PEL	probable effect level
QA/QC	quality assurance/quality control
RAP	Remedial Action Plan
rec	receptor
ref	reference
reg	regression
sed	sediment
SEL	severe effect level
THg	total mercury
TN	total nitrogen
TOC	total organic carbon
TP	total phosphorus
TRG	tissue residue guideline
wt	weight
ww	wet weight
$[x]_i$	concentration of substance *x* in matrix *i*

1 INTRODUCTION

1.1 Background and Mandate

In the 1970s, 42 locations in the Great Lakes where the aquatic environment was severely degraded were identified as "problem areas" by the International Joint Commission (IJC). Of these, 17 are along Canadian lakeshores or in boundary rivers shared by the US and Canada. The IJC's Great Lakes Water Quality Board recommended in 1985 that a Remedial Action Plan (RAP) be developed and implemented for each problem area. The RAP approach and process is described in the 1987 Protocol to the *Great Lakes Water Quality Agreement* (GLWQA). The goal is to restore the "beneficial uses" of the aquatic ecosystem in each problem area, which were now called "Areas of Concern" (AOCs). Fourteen possible "impairments of beneficial use", which could be caused by alterations of physical, chemical or biological conditions in the area, are defined in Annex 2 of the GLWQA.

The Canadian government's commitment to the GLWQA was renewed in 2000 with the Great Lakes Basin 2020 (GL2020) Action Plan, under which the efforts of eight federal departments to "restore, conserve, and protect the Great Lakes basin" over the next five years were to be co-ordinated. Environment Canada's contribution included the funding of detailed chemical and biological assessments of sediments in each of the 16 (of the 17) remaining Canadian AOCs (1 AOC has since been delisted). The National Water Research Institute (NWRI) was given the responsibility of conducting and reporting on these assessments.

Under the terms of reference for the NWRI's mandate, the BEnthic Assessment of SedimenT (BEAST) methodology of Reynoldson et al. (1995) is to be applied to the AOC assessments. To date, the methodology has involved evaluation of sediment contaminant concentration, laboratory toxicity, and benthic invertebrate community structure. Recent reviews of the BEAST framework have recommended the inclusion of an additional line of evidence – information on the bioaccumulation of contaminants liable to biomagnification (Grapentine et al. 2002). To obtain this additional information, support has been received from the Great Lakes Sustainability Fund for work in AOCs in 2001, including the St. Lawrence River at Cornwall, Ontario. The

study described in this document was conducted to supplement existing data to complete an assessment of sediments in the Cornwall AOC that were historically exposed to industrial effluents.

1.2 Decision Framework for Sediment Assessment

The underlying philosophy of the NWRI's approach to sediment assessment is that observations of elevated concentrations of contaminants alone are not indications of ecological degradation. Rather, it is the biological responses to these contaminants that are the concern. A recommendation on remedial activity requires evidence to be provided of an adverse biological effect either on the biota resident in the sediment, or on biota that are affected by contaminants originating from the sediment, either by physical, chemical or biological relocation.

It is recognized that to make decisions on sediment quality and the need to remediate, four components of information (in addition to knowledge on the stability of sediments) are required (Krantzberg et al. 2000):

- ***Sediment chemistry and grain size*** – Quantifies the degree to which sediments are contaminated. Indicates exposure (or at least potential exposure) of organisms to contaminants (with consideration of exposure pathways). Provides information on physicochemical attributes of the sediment to assist in the interpretation of any observed biological effects.

- ***Benthic invertebrate community structure*** – Used to determine whether natural faunal assemblages in contaminated sediments differ from those in uncontaminated reference locations. Can indicate a biological response to sediment conditions. Organisms that reside in and ingest sediments experience the most ecologically relevant exposures to contaminants present, and represent important food web components.

- ***Sediment toxicity*** - Differences in resident invertebrate communities between contaminated and uncontaminated sites alone cannot be conclusively attributed to toxic chemicals.

Sediment toxicity data provide supporting evidence that responses observed in the community are associated with sediment contaminants rather than other potential stressors.

- ***Invertebrate body burdens*** - Measurements of contaminants in tissues of resident benthic fauna provide evidence of bioavailability and that the contaminants are responsible for observed effects on the organisms (Borgmann et al. 2001). In addition, the information can be used to assess the risk to higher trophic levels due to biomagnification. Some contaminants, although bioavailable, may not accumulate in benthic invertebrates to sufficient concentrations to induce effects. A few of these contaminants (e.g., mercury) have the property of biomagnifying up the food chain to produce adverse responses in higher trophic level organisms.

Overall assessment of a site is achieved by integrating the information obtained both within and among the above four lines of evidence. The decision framework was developed from the Sediment Triad (Long and Chapman 1985; Chapman 1996) and the BEAST (Reynoldson et al. 1995) frameworks, and is described in detail elsewhere (Grapentine et al. 2002).

1.3 The St. Lawrence River (Cornwall) Area of Concern

The St. Lawrence River at Cornwall AOC has been the subject of two major RAP reports – Stage 1: Environmental Conditions and Problem Formulation (St. Lawrence RAP Team 1992) and Stage 2: The Recommended Plan (St. Lawrence RAP Team 1997). The environmental issues of concern identified for the Cornwall/Massena (New York) section of the St. Lawrence River were:

- Mercury contamination,
- PCB contamination,
- Presence of other contaminants,
- Fecal bacterial contamination,
- Habitat destruction and degradation,
- Excessive growth of nuisance aquatic plants,
- Exotic species, and

- Fish and wildlife health problems related to contaminants.

Of the 14 beneficial uses evaluated for the Cornwall AOC, 7 were determined as "impaired". Four of these 7 are associated with sediment contaminants:

- Degradation of benthos,
- Restrictions on fish and wildlife consumption,
- Degradation of fish and wildlife populations, and
- Restrictions on dredging activities.

Since 1997, further assessments of sediments and contaminants in depositional areas of the Cornwall waterfront have been performed. In February 2001, the Cornwall Sediment Strategy Working Group reviewed recent environmental studies (Reynoldson 1998; Rukavina 2000). Key conclusions were:

- Sediment deposits in Zone 2 are generally stable; those in Zones 1, 3 and 4 remain to be investigated. (See Figure 1 for location of zones.) The disturbance and resuspension of sediments from human activity (e.g., boat traffic, shoreline construction) may be of concern.
- Direct toxicity of sediment-bound contaminants is not evident or low based on laboratory toxicity tests and assessment of resident benthic communities.
- Bioavailability of mercury from sediments and the potential for food chain effects is of concern and needs to be investigated.

The current chief environmental issue of concern is the elevated concentrations of mercury in sediments due to past discharges from local sources, and the potential risk to fish, wildlife and humans through biomagnification. The bioaccumulation component of the assessment framework is important to consider where concern exists for contaminants such as mercury and chlorinated organic compounds that can be highly concentrated in the food web without inducing effects on survival, reproduction or growth at the lower trophic levels (which are typically examined for sediment assessments). Measurement of invertebrate body burdens allows the assessment of the potential for effects on higher trophic level organisms (which are more

difficult to measure and typically not examined in sediment assessments) resulting from the transfer of contaminants through dietary sources.

1.4 Purpose of the Study

The purpose of this study is to determine if deleterious amounts of mercury from sediments in the Cornwall AOC could potentially be transferred through benthic invertebrates to fish, wildlife or humans. In other words: Is there evidence that mercury biomagnification is an environmental issue of concern? The results of this study should lead to one of two alternate conclusions: (a) mercury is unlikely to concentrate in the food web at levels that can cause adverse effects, or (b) mercury **could** concentrate in the food web at levels that can cause adverse effects. The determination of whether mercury biomagnification and adverse effects to higher trophic level organisms (fish, wildlife, human) are actually occurring in the Cornwall AOC is beyond the scope of this study, and would need to be addressed by a more comprehensive assessment such as a detailed risk assessment. The latter conclusion (b) is of **potential** biomagnification, but does not determine actual biomagnification.

2 OBJECTIVES AND APPROACH

2.1 Objectives of Study

The purpose of the study was achieved through two objectives:

A. Determining if benthic invertebrates in locations where mercury is elevated are a potential source of mercury to higher trophic levels.

B. Determining if the amount of mercury potentially available is of concern.

The first objective was addressed by comparing concentrations of mercury (Hg) in benthic invertebrates from test sites to those from reference sites, and by determining whether sediment Hg concentration is related to invertebrate (whole body) Hg concentration. For the second objective, the concentrations of Hg in selected trophically linked receptor species (i.e.,

consumers of benthic invertebrates and their predators) were predicted based on measured Hg in invertebrates and literature-derived biomagnification factors. (Traas et al. (2002) is an example of an application of this approach.). The predicted Hg concentrations in the selected receptors were compared to appropriate tissue mercury guidelines established for the protection of higher trophic level organisms. Whereas predictions of receptor tissue mercury concentrations focused on methyl mercury (MeHg) because it is the most toxicologically relevant and predominant form of mercury in tissues of fishes and higher trophic level receptors (USEPA 1997b; Environment Canada 2002), determinations of Hg distributions and bioaccumulation in sediment and invertebrates were made on the basis of both total mercury (THg) and MeHg to allow comparisons with results from other studies and guidelines that involve THg.

The biomagnification modelling was broken down into four steps:

- Identification of receptors of potential concern.
- Measurement of contaminant concentrations in invertebrates and sediment.
- Selection of biomagnification factors.
- Prediction of possible receptor species tissue concentrations.

Knowledge of the food web structure of a site is needed to determine relevant receptor species (fish, bird, mammal). These are identified in the following subsection. Determinations of concentrations of mercury in sediment ($[Hg]_{sed}$) and invertebrates ($[Hg]_{inv}$) are described in the sampling design and methods sections. The identified receptors determined what biomagnification factors (BMFs) to use for predicting receptor mercury concentrations and what guideline to use (e.g., guidelines for protection of wildlife consumers of aquatic biota; human health guidelines for protection from fish consumption) for comparison. The review and selection of BMFs are discussed in the data analyses (subsection 3.3.2.1.) and Appendix A, and the estimation of [Hg] in the tissues of receptor species is described in subsection 3.3.2.2.

If the predicted contaminant concentration in the receptor exceeded the guideline, a potential risk of adverse effects due to biomagnification was concluded. Alternatively, if the predicted contaminant concentration in the receptor was less than the guideline, no potential risk was concluded.

2.2 Identification of Receptors of Concern

Based on generic food webs for the Great Lakes (e.g., Diamond et al. 1994), information on fauna resident in the Cornwall AOC (St. Lawrence RAP Team 1997) and guidelines from Environment Canada (2002), receptors representative of four trophic levels were selected for biomagnification modelling:

- Benthic invertebrates (trophic level 1): ***amphipods, midges and snails***.
- Benthivorous fish (trophic level 2): ***white sucker***. Total mercury concentrations in white sucker collected from Lake St. Francis, Ontario show a decrease from late 1970s to early 1980s, and remain relatively stable (with some fluctuations) to 1994 (Dreier 2000).
- Small piscivorous fish (trophic level 3): ***adult yellow perch***. (1) Lake St. Francis. Overall, there is a decrease in the mean total mercury concentration in 25 cm yellow perch from 1977 (~650 ng/g ww) to 1994 (< 200 ng/g ww); however, decreases have been relatively minor since 1978 (St. Lawrence RAP Team 1997). More recent data collected in 1998 and 2000 show total mercury concentrations slightly higher than those reported in 1994, with means of 254 ng/g ww (1998) and 319 ng/g ww (2000) for fish between 18 and 20 cm long. (2) Lake St. Lawrence, Ontario. Perch (25 cm long) show a decrease in mean total mercury concentration from 1981 (~350 ng/g) to 1993 (~ 200 ng/g) (similar concentration to that seen in 1994 Lake St. Francis perch). Perch (18-20 cm long) collected in 1999 have a slightly lower mean total mercury concentration (188 ng/g) than that reported in 1993 (Lisa Richman, Ontario Ministry of the Environment, pers. comm).
- Large piscivorous fish (trophic levels 3 and 4): ***walleye***. (1) Lake St. Francis. Overall, mean total mercury concentrations in 50 cm long walleye show a decrease from 1976 (1000 ng/g ww) to 1982 (< 600 ng/g) and remain fairly stable from 1982 to 1994 (with a peak in 1992). (St. Lawrence RAP Team 1997; Dreier 2000). More recent data collected in 1999 show mean total mercury concentrations in walleye higher than those reported in 1994, with a mean of 980 ng/g ww; however, fish were between 25 and 69 cm long (Lisa Richman, Ontario Ministry of the Environment, pers. comm). (2) Lake St. Lawrence. Mean total mercury concentrations remain fairly stable from 1981 (< 300 ng/g ww) to 1993 (St. Lawrence RAP Team 1997).

- Piscivorous bird (trophic levels 3 and 4): ***great blue heron***. Great blue herons are widespread. Fishes (mostly <25 cm in length) are the preferred prey (Environment Canada 2002).
- Piscivorous mammal (trophic levels 3 and 4): ***mink***. Mink are associated with numerous aquatic habitats and are opportunistic feeders (Environment Canada 2002).
- ***Human fisher*** (trophic level 5).

As part of the Sport Fish Contaminant Monitoring Program, regular collections of walleye, yellow perch, and white sucker from Lakes St. Lawrence and St. Francis take place. Sport fish consumption restrictions for total mercury begin at 450 ng/g and total restriction is advised for levels above 1570 ng/g. Total mercury concentrations are found to be at levels that warrant consumption advisories for the walleye from both Lakes and for the perch and sucker from Lake St. Francis (MOE 2002). For the walleye, greater restrictions are imposed for Lake St. Francis, and commence for fish 35-45 cm long, with total restriction (>1570 ng/g) for fish 65-75 cm long. For Lake St. Lawrence, restrictions start for fish 55-65 cm long and there are no total restrictions imposed.

A model of the feeding relationships linking these receptors with each other and benthic invertebrates and sediment is shown in Figure A1 (Appendix A).

2.3 Study Area

Background information on environmental conditions in the Cornwall AOC is given in Dreier (2000). The present sediment assessment focused mainly on four depositional areas along the north channel identified by acoustic mapping of the river bed (Rukavina 2000, Figure 1). Previous sediment surveys performed by the MOE in these depositional areas reported total mercury concentrations in sediments below the provincial Lowest Effect Level (LEL) in Zone 4, above the provincial LEL (Persaud et al. 1993) in Zones 1, 2, and 3, and above the Severe Effect Level in Zone 2 (Richman 1994, 1996, 1999, 2000; Richman and Dreier 2001; Metcalfe-Smith et al. 1995; Appendix B: Table B1). Additionally, total mercury concentrations in benthic invertebrates were higher in some areas of Zone 2 than in other areas within the same zone as

well as upstream (Lake St. Lawrence) (Richman 1994). A far-field exposed area was also selected for sampling (one site). This site was located downstream of Zones 1 – 4. Flow distribution patterns in the St. Lawrence River channels indicate that this far-field area receives flow from the north channel, which contains Zones 1, 2, and 3 (St. Lawrence RAP Team 1992). Anderson (1990) and Richman (1994) reported total mercury concentrations in sediments collected from this far-field area elevated above upstream reference sites (Lake St. Lawrence) and above the LEL.

Reference areas located outside Zones 1 - 4 were selected upstream of the AOC in Lake St. Lawrence, upstream of Zone 1 (below the dam), and off the southern sides of Cornwall and St. Regis Islands where the water flow is from the south channel of the river. Stations in these areas provided data on background mercury concentrations in sediment and biota relevant to the AOC.

2.4 Experimental Design

2.4.1 Sampling design

Sampling stations were arrayed in a multiple gradient design supplemented with reference sites. Stations were positioned in Zones 1 through 4, and in upstream and downstream (outside effluent plume) reference locations. In total, 34 stations — 12 reference + 22 test (i.e., potentially exposed to effluent) — were sampled for benthic invertebrate tissue and sediment and overlying water chemistry during 9-19 October 2001. A list of station locations is provided in Table 1, and sites are shown in Figure 2. The location of stations were selected on the basis of (a) representing the widest range of mercury concentrations in sediment, (b) representing least contaminated/reference conditions in the area, and (c) overlapping locations of previous studies.

This mixed (gradient + control/impact) sampling design allowed two types of comparisons for assessing the distribution of mercury in sediment in biota. Using all sites, relationships between sediment [Hg] and biota [Hg] levels were examined. In addition, Hg concentrations in biota collected from locations exposed to Hg-containing effluent in the past were compared to Hg concentrations in biota collected from the reference locations.

2.4.2 Measurement endpoints

Invertebrates (snails, amphipods, and midges) and sediment were collected from locations of sediment deposits exposed to past discharges of mercury-containing effluent, as well as from unexposed reference locations. Sediment was obtained from the top 0 - 10 cm layer of river/lake bed. This layer includes the vertical home range of most benthic invertebrates. Two distinct invertebrate taxa were targeted for collection from each location. It was not possible to collect the same two taxa from all locations. Although midges were obtained from all sites, amphipods were absent from 58% of the reference sites and 41% of the test sites, and present in low numbers at the remaining sites. Snails were collected at 100% of test sites but were absent from 42% of reference sites. Analyses of total and methyl mercury were performed on samples composited from organisms within each of two or three taxa (i.e., taxa were analyzed separately). Invertebrates were not allowed time to clear sediment from their guts because predators consume whole organisms. Mercury associated with sediment, as well as that incorporated into tissues, is potentially available for transfer through the food chain.

2.4.3 Assumptions

For the prediction of Hg concentrations in the tissues of upper trophic level biota, bioaccumulation is considered to occur predominantly through dietary pathways. This is suggested by several experimental and modelling studies (Bodaly et al. 1997; Downs et al. 1998). In modelling the exposure to and uptake of Hg by receptors, several conservative assumptions (i.e., maximum potential exposure to Hg) have been made. These include:

- For fish receptor
    - Fish consume invertebrates only from the site.
    - Fish feed on the same invertebrate taxa as those collected in field sampling.
- For wildlife receptor
    - 100% of the diet is fish.
    - Fish are consumed only from the site in question.
    - Fish consume invertebrates only from the site.
    - Fish feed on the same invertebrate taxa as those collected in field sampling.
- For human receptor

- maximum observed proportion of diet is fish.
- Fish are consumed only from the site in question.
- Fish consume invertebrates only from the site.
- Fish feed on the same invertebrate taxa as those collected in field sampling.

In addition, the flux of mercury between sediment, water and biota compartments were considered in equilibrium.

3 METHODS

3.1 Sample Collection and Handling

Prior to sediment collections, temperature, conductivity, pH and dissolved oxygen were measured in the water column approximately 0.5 m above the bottom using Hydrolab apparatus. Water samples were then collected (for alkalinity and nutrients) from 0.5 m above the bottom using a van Dorn sampler. Total phosphorus samples (125 mL) were preserved with I mL of 30% sulphuric acid. Water samples were stored at 4°C for later analysis.

A mini-box core sampler was used to collect the top 10 cm of sediment at the majority of sites. At Sites 1321 (reference), and Sites 27 and 31 (Zone 2), where a mini-box core could not be employed due to the nature of the sediment, a Ponar sampler was used. At each site, a representative sample of the top 10 cm sediment was collected from each box-core or Ponar grab and set aside in a glass tray. The remaining top 10 cm of sediment was placed in a 68 L tub. When the tub was full, the sediment set aside in the glass tray was homogenized and distributed to containers for individual analyses. Sediment collected for determination of total and methyl mercury was dispensed in pre-cleaned polyethylene bottles. Variables measured at each tissue collection site are listed in Table 2. All samples were kept at 4°C, with the exception of the sediment and biota for mercury analyses, which were frozen (-20°C).

Invertebrate biota was removed from the top 10 cm of sediment (in the 68 L tubs) by wet sieving with river water using 12" stainless steel sieves (500-µm mesh). Biota collected on the sieve

were sorted into separate taxa in glass trays using stainless steel instruments, rinsed with deionized water and placed in pre-weighed and pre-cleaned (10 % HCL) 5 mL scintillation vials, weighed, and frozen on site (-20°C). A layer of parafilm was placed between vial and cap. Biota samples were later freeze-dried and reweighed. The wet:dry ratios were used in converting mercury concentrations in biota from a dry weight to wet weight basis (see section 3.4.2.2).

Stainless steel sieves and instruments were detergent washed between stations. If persistent organic matter remained on the sieve after the detergent wash (on visual inspection), a more aggressive cleaning solution was implemented (caustic ethanol). Homogenizing and sorting trays and scoops were detergent washed, rinsed in 20% HCl , and rinsed with Lake/River water.

3.2 Sample Analyses

Analyses of alkalinity, total phosphorus, nitrate+nitrite-N, ammonia-N and total Kjeldahl N in water samples were performed by the Environment Canada's National Laboratory for Environmental Testing (NLET) (Burlington, ON) by procedures outlined in Cancilla (1994) and NLET (2000). Concentrations of trace metals (including total mercury), major oxides, total phosphorus, total nitrogen and total organic carbon in sediment were measured by Caduceon Environmental Laboratory, Ottawa, ON, following procedures outlined by USEPA/CE (1981). Particle size analysis (percents clay, silt, sand, gravel) was performed by the Sedimentology Laboratory, NWRI, Burlington, ON, following the procedure of Duncan and LaHaie (1979). Mercury (total and methyl) analyses of sediment and biota were performed by Flett Research Ltd. (Winnipeg, MB). Procedures for mercury analyses, which were based on Bloom and Crecelius (1983), Horvat et al. (1993) and Liang et al. (1994), are summarized below.

3.2.1 Total mercury in sediment

Flett laboratory: Between 100 and 1000 mg of thawed sediment sample (or spiked sediment, blanks or reference material) was digested overnight (16-18 hours) in 3 mL of 7:3 nitric/sulfuric acid at 150°C. After cooling, the sample was diluted to 25 mL with low-mercury deionized water, spiked with BrCl and allowed to react. The residual BrCl was then destroyed by addition of hydroxylamine hydrochloride. An aliquot of the sample (100 µL – 2 mL) was placed into a

sparging vessel, to which was added stannous chloride. The elemental mercury produced was purged onto a gold trap with Hg-free nitrogen. The gold trap was heated with UHP argon carrier gas passing through it, and the mercury released was measured by a Brooks-Rand CVAFS model-2 detector. The detection limit was 1-5 ng /g.

Caduceon laboratory: Freeze dried sediments (0.5 g) were digested with HNO_3:HCl for two hours. $SnCl_2$ was added to reduce Hg to volatile metallic form. If there was high organic material, $KMnO_4$ was added to the digestion solution to destroy organo-mercury bonds. Hydroxyl amine hydrochloride was then added to neutralize $KMnO_4$ excess so $SnCl_2$ could react with Hg in solution. Digestion was followed by measurement using a cold vapour atomic absorption spectrometer. The detection limit was 5 ng/g.

3.2.2 Total mercury in biota

The same procedure as described for analysis of total mercury in sediment by Flett Research was used for biota, with the following differences in the sample digestion: up to 100 mg of invertebrate sample (or spikes, blanks or reference material) was digested for 6 hours in 10 mL of 1:2.5 nitric/sulfuric acid at 250°C; after cooling, the sample was diluted to 25 mL with low mercury deionized water, spiked with BrCl and allowed to react.

3.2.3 Methyl mercury in sediment

Sediment was prepared for analysis by distilling 200-300 mg of homogenized sample (or spikes or blanks) in ~45 mL of low-mercury deionized water. Approximately 40 mL of distillate was collected and acidified with KCl/H_2SO_4. (Note: Since some methyl mercury results were ≤0.1% of the total mercury results, a methylene chloride extraction was carried out on some of the highest total mercury samples. No significant difference in methyl mercury concentrations was observed between results obtained by either method. Therefore, it is assumed that insignificant methyl mercury production was occurring in the distillation process and thus all samples were processed by distillation.) An aliquot of the prepared sample (1-2 mL, depending on observed interferences from the matrix) was ethylated in solution (final volume ~ 40 mL) using sodium tetraethyl borate. The solution was buffered to pH 5.5. The resulting ethylmethyl mercury was

purged onto a Tenax trap with mercury-free nitrogen. The trap was heated, purged with UHP argon onto a GC column (for separation of the ethylmethyl mercury from Hg° and diethyl mercury), run through a pyrolizer (to reduce all mercury to Hg°), and then sent to a cold vapour atomic fluorescence analyser for detection. (GC oven: Perkin Elmer 8410 GC; column: chromasorb WAW-DMSC 60/80 mesh with 15% OV-3; detector: Brooks-Rand CVAFS model-2). The detection limit was 0.027 ng/g.

3.2.4 Methyl mercury in biota

Freeze dried biota (5-10 mg of homogenized sample, spike, blank or reference material) were digested overnight with ~500 µL of KOH/methanol at 75 °C. Sample aliquots (50-60 µL) were then treated and analysed as described above for the ethylation and subsequent steps in the determination of methyl mercury in sediment. The detection limit was 0.51 ng/g.

3.3 Biota-sediment accumulation factors (BSAFs)

A BSAF was calculated for each invertebrate taxa and site combination, for total and methyl mercury. The BSAF equation used was that defined by Thomann et al. (1995), and is the ratio of the metal concentration in the organism to that in the sediment:

$$BSAF = [Hg]_{inv}/[Hg]_{sed}$$

BSAFs assume that the concentration of contaminant in the organism is a linear function of the contaminant concentration in the sediment.

3.4 Data Analyses

3.4.1 Mercury distribution in sediment and biota

Sites in which concentrations of Hg in invertebrates ($[Hg]_{inv}$) were significantly elevated above background levels for the study area were identified by comparing $[Hg]_{inv}$ for effluent-exposed sites to the 99^{th} percentile value (= maximum) for the reference locations. This was done separately for MeHg and THg and for each invertebrate taxon.

Relationships between concentrations of Hg in sediment and invertebrates were determined using regression analysis, again separately for MeHg and THg and for each invertebrate taxon. The approach was to estimate the degree to which Hg in biota is predictable from Hg in sediment, with and without environmental covariables. Simple linear regression (ordinary least squares) was used for the single predictor ($[Hg]_{sed}$) model. Stepwise and "best subset" multiple linear regression procedures (Draper and Smith 1998; Minitab 2000) were used for the fitting of multiple predictor models. Initially, a subset of environmental variables expected to potentially influence uptake of Hg from sediment by biota (based on reviews such as Braga et al. 2000; Lawrence and Mason 2001) were selected from the group of measured variables (Table 2). These included sediment concentrations of total organic C, total P, total N, Fe, and Mn; sediment particle size fractions of sand, silt and clay; overlying water concentrations of total P, nitrate/nitrite-N, ammonia-N, total Kjeldahl N, dissolved O_2; and overlying water alkalinity, pH and conductivity. (Overlying water variables were not used as predictors for the midge models because, being mainly infaunal, they are more likely to be exposed to porewater than to overlying water.) To increase normality of data distributions and linearity of relations between variables, some data were transformed: log(x) for THg and MeHg in sediment and biota; log(x) for nutrients, Fe and Mn in sediment; and arcsine-square root(x) for the particle size fractions. Normality and linearity of the water column data were not generally improved by transformations, so these were analyzed untransformed.

Stepwise and best subset regressions were performed as both complementary and as corroborative procedures. All models fitted to the data included $[Hg]_{sed}$ as a predictor. The specific null hypothesis of interest was that "the effect of $[Hg]_{sed}$ on $[Hg]_{inv} = 0$, after accounting for effects of other predictors". For the stepwise regressions, the criteria for entry and removal of additional variables were a P-value ≤ 0.15 for the partial F-test. For the best subset regressions, models were fitted for all combinations of predictors. Determination of the "best" model was based on several criteria (in roughly decreasing order of importance):

- maximum $R^2_{adjusted}$
- significance of partial F-tests (= t-tests) for predictors (especially $[Hg]_{sed}$)
- significance of F-test for regression
- variance inflation factors (VIFs) for predictors < 10

- homoscadastic and normally distributed residuals
- Mallow's C_p statistic not >> number of predictors

Lack-of-fit tests for curvature in response-predictor relationships and interactions between predictors were performed and examined for nonsignificance. Observations having large standardized residuals or large influence on the regression were also considered in model evaluations. The best model was identified based on the overall meeting of these criteria. Both single and multiple predictor models were then examined for the degree to which $[Hg]_{sed}$ predicts $[Hg]_{inv}$, as indicated by the significance of the *t*-test of the coefficient for $[Hg]_{sed}$.

3.4.2 Prediction of mercury concentrations in receptors

3.4.2.1 Review and selection of biomagnification factors

A review of information on BMFs was conducted using typical methods of electronic database and chain-of-citation searches as well as consultation with leading researchers in the field of mercury ecotoxicology and risk assessment. Details on the methods and the results of the review are described in Appendix A. A summary is provided below.

The search was focused on the period 1996-2002, as a thorough review of the literature was carried out in 1997 by USEPA (1997a,b,c). The information required to estimate mercury concentrations in receptors was obtained by reviewing published literature, unpublished reports, databases, web pages and any other sources of data on BMFs relevant to the benthic invertebrate taxa and receptors; assessing the quality of the BMF data, and; tabulating BMFs and estimates of their variability, together with information on the BMF determinations (e.g., location of study, organisms involved, proportion of receptor's diet that is invertebrates, effects of cofactors (if any), assumed ingestion rates and home ranges). The following criteria were applied to screen literature to obtain either BMFs or candidate datasets for calculating BMFs, after Suedel et al. (1994) and Gobas and Morrison (2000):

- If organisms that were presented were not from a logical food chain, or no evidence was presented that the feeding relationship between predator and prey was a functional feeding relationship, the data were not used. One exception to this rule was made in selecting a study

of mink fed diets of different proportions of contaminated and uncontaminated fish (Halbrook et al. 1997), since there was a reasonable likelihood that these fish species would have been part of their diet.

- Mean concentrations of total Hg or MeHg needed to be presented for both predator and prey, and in comparable units.
- BMFs involving Hg concentrations in feathers or fur of predators were excluded.
- Unless evidence of comparability could be found, studies from non-freshwater systems or with non-comparable species were not used. More information is presented below on the assessment of comparability of different systems and species.

There were few studies that quoted BMF estimates specifically for the receptor species and feeding relationships defined in Figure A1. Of the small number of studies that calculated BMFs which were directly comparable in part to the food chain model, most were from freshwater pelagic foodwebs. Some were also studies in different ecosystems (marine, temperate montane freshwater, tropic freshwater). Thus, it was necessary to use the most relevant studies to obtain BMFs and document the relative comparability of different species and ecosystems to those presented in the study design for this assessment. Information to support substitutions of receptor with comparable species from the literature (in applying BMF estimates) is presented in Tables A3 - A12. Species were considered the most qualitatively similar when they occupied similar habitats, had similar feeding habits and dietary composition, similar range, similar feeding substrate, and similar food ingestion:body weight ratio. Sources for this information were CCME (1999a), CWS (2002), Sample and Suter (1999), Scott and Crossman (1973), and USEPA (1997c). A breakdown of the number of BMFs obtained/calculated per feeding relationship and the range of corresponding BMF values is presented in Table A1.

3.4.2.2 Calculation of receptor tissue mercury concentrations

It is widely recognized that mercury is transferred through trophic levels primarily in the methyl form (USEPA 1997b). It is also accepted that mercury in the tissues of fishes and higher trophic level organisms is almost entirely in the organic (methyl) form. Environment Canada (2002) states that "total mercury" concentrations in piscivorous fishes are probably ~99% methyl mercury, and note that Bloom (1992) suggests that previous studies reporting methyl mercury

fractions in fishes less than 95% were likely in error. Therefore, mercury concentration in receptors were predicted on a MeHg basis, using (a) MeHg measurements in invertebrates and (b) combined THg and MeHg BMF values (assuming that reported THg concentrations largely represent MeHg concentrations).

Concentrations of MeHg in the tissues of receptors were predicted by multiplying measured body concentrations in the resident invertebrates by the food chain multiplier relevant for the receptor:

$$C_{rec} = FCM \times C_{inv}$$

where:

C_{rec} = mean contaminant concentration in the consumer (receptor) species

C_{inv} = mean contaminant concentration in invertebrates

FCM = food chain multiplier

The FCM represents the cumulative biomagnification of a substance from one trophic level to a higher trophic level (USEPA 1997c). Whereas a BMF applies to only one trophic level transfer, a FCM refers to one or more, and may be a multiple of more than one BMF. Thus, FCM = $BMF_1 \times BMF_2 \times BMF_3 \times \ ... \times \ BMF_n$, where 1,2,3,…, n are transfers of one trophic level. The BMFs used to obtain FCMs and calculate C_{rec} values are in Table A1, which shows the low, medium and high BMFs from the literature review for each transfer between trophic levels as shown in Figure A1. In Table 3, the FCM for transfer from benthic invertebrates to each receptor is estimated by multiplying the BMFs for the intermediate steps from Table A1. Low, medium and high FCM values are obtained from use of all minimum, all medium or all maximum estimates for each BMF. In instances where only a single BMF value is available for a particular receptor, the low, medium and high FCM is the same. For the walleye, heron and mink, it is recognized that they could be trophic level 3 as well as trophic level 4 predators. Therefore, FCMs were estimated for both food chain pathways.

Invertebrate methyl Hg concentrations used in the predictions of Hg in receptors include observed $[Hg]_{inv}$ values for 2-3 taxa collected from a site. These were used to obtain minimum and maximum observed $[Hg]_{inv}$ for the taxa collected from the site. “Medium” $[Hg]_{inv}$ for the site

was calculated as the mean of the values. Since fish contaminant data are reported for the most part on a wet weight basis, and the guidelines used in this study are also based on wet weights, methyl Hg concentrations in invertebrates were converted to a wet weight basis. Biota comprised on average between 82.3 to 88.4% water. The ratio of wet to dry weight was determined for each individual sample submitted for analysis (rather than using an overall average ratio for each taxon). Wet weights were determined using the following conversion:

$[Hg]_{inv}$ (ng/g dry weight) / (ratio of wet: dry weight) = $[Hg]_{inv}$ (ng/g wet weight)

Total and methyl mercury concentrations in each invertebrate taxon, converted to wet weights, are shown in Appendix C, Tables C1 and C2.

For each site, minimum, intermediate and maximum concentrations of MeHg for each receptor were predicted by:

$$[Hg]_{rec} = FCM \times [Hg]_{inv}$$

using corresponding low, medium and high $[MeHg]_{inv}$ and FCMs. For the walleye, heron and mink, FCMs for both food chain pathways were combined. From the available values, the lowest and the highest FCMs were used for the minimum and maximum predictions, and the mean of the two medium values was used for the intermediate prediction. The predicted MeHg concentrations in receptors are generic in that they are not specific to particular tissues.

3.5 Quality Assurance/Quality Control

3.5.1 Field

Four randomly chosen sites (5, 171, 184, and 1332) were designated as QA/QC stations. At these stations, triplicate sediment and water samples were collected for determination of within-site and among-sample variability.

3.5.2 Laboratory

Flett Research Ltd. conducted determinations of total and methyl mercury in sediment and benthic invertebrates. QC evaluation for these procedures included analyses of sample duplicates, matrix spikes and certified reference materials, as well as evaluations of sample recoveries. For sediment, sample duplicates were analyzed at least once every 15 samples, and matrix spikes were performed on every tenth sediment sample to determine mercury recoveries. The NRC certified sediment reference material "MESS-2" was concurrently digested and analysed for total mercury. For biota, duplicate "DORM-2" reference material, "MQAP fish check samples", and spiked matrix duplicates were analyzed for total and methyl mercury with each lot of 10 - 20 samples. Each of the three invertebrate taxa was represented in the analyses of sample duplicates and matrix spikes.

Caduceon Environmental Laboratory analyzed sediment for trace metals (including total mercury), major oxides, total phosphorus, total nitrogen and total organic carbon. QA/QC procedures involved control charting of influences, standards, and blanks. Reference material was used in each analytical run. Calibration standards were run before and after each run. Run blanks and reference standards were run 1 in 20 samples. Sample duplicates were analyzed once every 16 samples.

An inter-laboratory comparison of analyses for total Hg was conducted based on results from Flett Research and Caduceon Laboratory for sediment subsampled from the same sample. Data for the 34 samples were compared by regression analysis. The slope of the regression line is a measure of the overall agreement in [THg] determinations, whereas the scatter of points about the line should indicate joint laboratory measurement error.

4 RESULTS

4.1 Quality Assurance/Quality Control

4.1.1 Field

Variability among site triplicates in a measured analyte has three sources: natural within-site heterogeneity in the distribution of the analyte in sediment or water, differences in handling

among samples, and laboratory measurement error. Among-triplicate variability indicates the overall "error" associated with quantifying conditions at a site based on a single sample. Variability is expressed as the coefficient of variation (CV = standard deviation / mean × 100). Results for particle size, TOC and mercury, nutrients, metals, and water chemistry concentrations for the field-replicated stations (5, 171, 184, and 1332) are shown in Tables D1 and D2, Appendix D.

Differences in variability are seen among sites and among the parameters from the same site. Overall, variability among sediment samples is highest for total and methyl mercury, with CVs ranging from 8 to 48% and from 2 to 65%, respectively. Variability is highest at Site 184 for total and methyl Hg. The CVs are low for the water quality parameters (range from 0.1 to 15%), metals (range from 1 to 19%), nutrients (range from 0.1 to 13%), and % clay (4 to 5%). Higher CVs are noted for the silt and sand fractions of the sediment (range from 2 to 24%, and from 2 to 44%, respectively).

The CVs for total Hg in sediments are slightly higher than those reported by Richman (1999) for replicate cores taken from the Cornwall area (12 to 23%). However, QA/QC samples for this study were taken from three separate drops of the box core, whereas the replicate core samples were collected from the same box core drop in the Richman (1999) study.

4.1.2 Laboratory

Data for Flett laboratory duplicates and repeat analyses for mercury in sediment and biota are given in Tables 4 to 6. There is good agreement between sample duplicates and repeats. Mean CVs for duplicate analyses are 13, 10, 11 and 11% for $[THg]_{sed}$, $[THg]_{inv}$, $[MeHg]_{sed}$ and $[MeHg]_{inv}$, respectively. These are lower than those reported for other studies using gas chromatography and cold-vapour atomic fluorescence spectroscopy (Paterson et. al. 1998). Repeat analyses, performed for $[MeHg]_{inv}$, have a mean CV of 12%. Recoveries for analyses of sediment and biota samples, matrix spikes and certified reference materials are shown in Table E3. Mean recoveries range from 89.6 to 98.7% for the samples, 89.4 to 103.4% for the matrix spikes, and 93.5 to 102.3% for the reference materials. Lowest mean recoveries in samples and matrix spikes are obtained for $[MeHg]_{sed}$. The overall range of spike recoveries (74.4 to 119.8%)

is comparable to that obtained by Lawrence and Mason (2001), who used similar analytical methods.

Duplicate measurements of sediment metals, major oxides and nutrients, and corresponding analyses of reference materials for Caduceon Laboratory are shown in Tables D4 and D5, respectively. The mean CV for the sample duplicate measurements (two sites) is 3.1% (range: 0 to 7.9%), and 8.1% (range 0 to 15.7%). Recoveries for reference materials are mostly between 90 and 100%, but range from 75 to 129%.

The inter-laboratory comparison for analyses of total mercury in sediment is described in Appendix D. Results show a strong agreement between measurements: the slope of Flett $[Hg]_{sed}$ vs Caduceon $[Hg]_{sed}$ is 1.02. The percent explained variability (r^2) is 79%.

4.2 Mercury Levels

4.2.1 Sediment

4.2.1.1 Total mercury

Flett laboratory

On a dry weight basis, the lowest mercury concentrations are found in the reference sediments (range 16 – 174, median 75 ng/g), followed by sediments collected from Zone 4 (range 53 – 127, median 93 ng/g; Table 4, Figure 3). The highest mercury concentrations are found in sediments collected from Zone 2 (range 379 - 5568, median 1845 ng/g), followed by Zone 3 (range 610 - 2879, mean 1745 ng/g), Zone 1 (range 378 - 1646, median 576 ng/g) and the far-field location (Site 171) (253 ng/g).

The LEL for mercury (200 ng/g) is not exceeded at any of the reference stations, nor in the sediments collected from Zone 4. The LEL is exceeded at all sites in Zones 1, 2, 3, as well as downstream (d/s) Site 171. The SEL (2000 ng/g) is exceeded at Site 108 in Zone 3, and at five sites (5-2, 9, 64, 19, & 31) in Zone 2. The highest mercury concentration is at Sites 19 and 31 in Zone 2.

Caduceon Laboratory

On a dry weight basis, total mercury concentrations are higher on average than those reported by Flett; however, trends are similar (Table 4). The lowest mercury concentrations are found in the reference sediments (range 44 – 268, median 117 ng/g), followed by sediments collected from Zone 4 (range 133 – 908, median 149 ng/g). The highest mercury concentrations are found in sediments collected from Zone 2 (range 780 - 14300, median 3400 ng/g), followed by Zone 1 (range 724 - 4880, median 2705 ng/g), Zone 3 (range 1490 - 3310, mean 2400 ng/g), and the far-field location (391 ng/g). The highest mercury concentration is at Site 5-2, 19, and 31 in Zone 2.

4.2.1.2 Methyl mercury - *Flett laboratory*

Methyl mercury concentrations (Table 4, Figure 4) range from 0.2 to 1.1 ng/g dry wt (median 0.6 ng/g) at reference sites and from 0.4 to 5.4 ng/g at test sites (median 1.5 ng/g). The highest concentrations occur at sites located in Zone 2, ranging from 0.7 to 4.8 ng/g (median 3.2 ng/g), followed by Zone 3 (median 2.4 ng/g), Zone 1 (median 1.5 ng/g), d/s (median 0.9 ng/g), and Zone 4 (median 0.7 ng/g). The fraction of methyl mercury relative to total mercury is low (mean = 0.6 % with a 95% confidence interval of 0.4 – 0.8 %; but at two outlying Sites – 1320 and 179 – the percent methyl mercury is 2.4 and 3.1%, respectively).

Regression analysis of log transformed data showing the relationship between methyl mercury and total mercury in the sediment is shown in Figure 5. There is a significant positive correlation ($P < 0.001$) between the total and methyl mercury concentrations in the sediment.

4.2.1.3 Comparison of sediment mercury at reference sites to exposed sites

Since Flett laboratory performed total and methyl mercury analysis in the sediment (and biota) samples and Caduceon laboratory performed only total mercury analysis in the sediments, comparisons of sediment mercury at reference to exposed sites are based on Flett data.

For total mercury, all sites in Zones 1, 3 and 2 and the far-field location exceed the maximum reference site concentration, whereas in Zone 4 all sites are below (Figure 3). Exceedences range

up to 11.9×, 20.9×, 40.4× and 1.8× the reference site maximum for Zones 1, 3 and 2, and the far-field site, respectively.

A similar pattern is observed for methyl mercury (Figure 4). All sites in Zones 1, 3 and all but two sites in Zone 2 exceed the maximum for the reference sites. In Zone 4, MeHg at one site is above the maximum for the reference sites, while MeHg concentration at the far-field site is below. Exposed site MeHg concentrations exceed the reference site maximum by lesser amounts than is the case for THg: up to 4.7×, 2.3×, 4.2× and 1.4× for Zones 1, 3, 2 and 4, respectively.

4.2.1.4 Comparison of mercury in sediment with historical data

Figure 6 compares the 2001 total Hg concentrations in Zone 2 sediment to data from surveys in 1994 and 2000 (Appendix B; Richman 1996, 2000). At all sites, total mercury concentration shows a decrease from 1994 to 2001, with the exception of station 27. Station 31 shows a slight increase from the 2000 data (but an overall decrease from 1994).

4.2.2 Invertebrates

4.2.2.1 Total mercury

On a whole-body, uncleared-gut basis, midges (chironomids) show the greatest range of total Hg concentrations (17 – 1642 ng/g, median 92 ng/g, n = 34), followed by the snails (35 – 735 ng/g, median 134 ng/g, n = 29) and amphipods (64 – 400 ng/g, median 237 ng/g, n = 10; Table 5). Comparing the 10 sites where complete data exist for all three taxa, the midges accumulate the most total Hg at 50% of the sites, followed by the snails (30%) and amphipods (20%). Comparing the 29 sites where data exist for midges and snails, the midges accumulate more total Hg than snails at 55 % of the sites.

4.2.2.2 Methyl mercury

The midges also show the greatest range of methyl Hg concentrations (1.7 – 34.9 ng/g, median 8.5 ng/g, n = 34), followed by the snails (7.1 - 108 ng/g, median 24.5 ng/g, n = 29) and amphipods (13.8 – 76.1 ng/g, median 32.5 ng/g, n = 10; Table 6). Looking at the 10 sites where

complete data exist for all three taxa, the amphipods accumulate the most methyl Hg at 60% of the sites, followed by the snails (40%), and midges (0%). Comparing midges and snails at the 29 sites, the snails accumulate more methyl Hg than midges at 28 of the 29 sites.

4.2.2.3 Comparison of mercury in biota at reference sites to exposed sites

Figures 7 -12 show concentrations of total and methyl mercury in midges, snails and amphipods at the 22 sites potentially exposed to mercury-bearing effluents compared to concentrations at the reference sites.

Midges – Total Hg Sites above the maximum for reference sites include all in Zones 1 and 3, and 7 of 10 sites in Zone 2 (Figure 7). The order of decreasing mean total Hg concentrations in midges for the exposed areas is: Zone 3 > Zone 2 > Zone 1 > Far-field > Zone 4. Total Hg concentrations in midges from exposed sites range up to 15.1× the reference site maximum.

Midges – Methyl Hg Sites exceeding the maximum for reference sites include both sites in Zone 3, 1 of 10 in Zone 2, and 1 of 5 in Zone 4 (Figure 8). The order of decreasing mean methyl Hg accumulation is: Zone 3 > Zone 4 > Zone 2 > Far-field > Zone 1, which differ from that observed for total Hg. Methyl Hg concentrations in midges from exposed sites range up to 2.5× the reference site maximum.

Snails – Total Hg Sites above the maximum for reference sites include 2 of 4 in Zone 1, both in Zone 3; 5 of 10 in Zone 2, and 1 of 5 in Zone 4 (Figure 9). The order of decreasing mean total Hg accumulation in snails is: Zone 3 > Zone 1 > Zone 2 > Zone 4 > Far-field. Total Hg concentrations in snails from exposed sites range up to 4.4× the reference site maximum.

Snails – Methyl Hg Sites above the maximum for reference sites include 1 of 4 in Zone 1, both in Zone 3; 3 of 10 in Zone 2, and 1 of 5 in Zone 4 (Figure 10). The order of decreasing mean methyl Hg accumulation is the same as that observed for total Hg. Methyl Hg concentrations in snails from exposed sites range up to 3.2× the reference site maximum.

Amphipods – Total Hg All eight exposed sites from which amphipods were obtained are above both of the reference sites (Figure 11). The order of decreasing mean total Hg accumulation is: Zone 2 > Zone 3 > Zone 1 > Zone 4. Total Hg concentrations in amphipods from exposed sites range up to 5.6× the reference site maximum.

Amphipods – Methyl Hg All exposed sites except the Zone 1 site and 1 of 5 in Zone 2 are higher in amphipod methyl Hg concentration than the maximum for reference sites (Figure 12). The order of decreasing mean methyl Hg accumulation is: Zone 3 > Zone 2 > Zone 4 > Zone 1, which differs from that observed for total Hg. Methyl Hg concentrations in amphipods from exposed sites range up to 4.3× the maximum for reference sites.

4.2.3 Biota-sediment accumulation factors (BSAFs)

BSAFs for total and methyl mercury are shown by zone for each taxon in Figure 13. For total mercury, BSAFs are < 1 in all zones except at the reference sites and Zone 4 sites. Highest BSAFs are observed at the reference sites for the midges and at Zone 4 sites for the snails and amphipods. The reference sites and Zone 4 are the areas of lowest total mercury sediment concentrations (Figure 3). Methyl mercury accumulate in biota to much higher concentrations than that found in sediment at some sites in all zones and at the reference sites. The greatest accumulation (relative to sediment concentration) occurs at reference sites and in Zone 4 for midges (same as that observed for total Hg), and at reference sites and Zone 3 sites for snails and amphipods.

4.3 Supplementary Physico-Chemical Conditions of Sediment and Overlying Water

4.3.1 Sediment nutrients

Total phosphorus (TP), total nitrogen (TN), and total organic carbon (TOC) in sediments (dry weight) are shown in Table E1 (Appendix E). TOC is lower at reference sites, ranging from 0.9 to 4.9% (median 3.1%) and from 2.6 to 21.2% at exposed sites (median 3.8%). Highest TOC is noted for Sites 183 and 184 in Zone1. Total nitrogen ranges from 816 to 4990 µg/g at reference sites (median 3340 µg/g) and from 1310 to 4178 µg/g at the exposed sites (median 3150 µg/g),

and TP ranges from 650 to 1497 µg/g at reference sites (median 1105 µg/g) and from 727 to 1190 µg/g at exposed sites (median 1040 µg/g). The SEL is exceeded at Sites 183 and 184 (Zone 1) for TOC, and at Site 1326 (reference) for TN.

4.3.2 Sediment particle size

Particle size data for Cornwall sediments are shown in Table E1 (Appendix E). In general, sediment in the study area consist of silt (ranging from 10 to 69%) and clay (ranging from 10 to 52%), or silt and sand (ranging from 0.2 to 80%). At reference sites, the median percentage silt (57%) and clay (33%), is higher than at exposed sites (43 & 23%, respectively), while the median percentage sand at reference sites (4%) is lower than at exposed sites (32%). Six of the 10 stations from Zone 2 (16, 17, 19, 27, 31, and 54) contain gravel (ranging from 0.3 to 4%). There is no gravel at reference sites.

4.3.3 Other metals

Concentrations (dry weight) of other metals analysed in the sediment (Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn), and the corresponding provincial LELs and SELs are shown in Table E2 (Appendix E). In general, concentrations of Cr, Cu, Fe, Ni, Pb, and Zn are greater than the LEL at most sites in the study area including the reference sites. The SEL is exceeded at Site 9 (Zone 2) for Pb and Zn and at station 64 (Zone 2) for Cu, Pb, and Zn.

Comparing metal concentrations at reference sites and test sites, percent iron is highest at the reference sites, ranging 0.9 to 3.7% (median 2.3%), and ranging from 0.8 to 2.1% (median 1.3%) at test sites. Median concentrations of copper (37 µg/g), lead (33 µg/g), and zinc (121 µg/g) at reference sites, are comparable to that at test sites (40, 31, & 124 µg/g respectively).

4.3.4 Overlying water chemistry

Conditions of overlying water 0.5 m above the sediment (Table E3, Appendix E) are similar at reference and test sites, with overlapping ranges and very similar medians for each variable. The ranges across sites are all low: temperature 2.8°C, dissolved oxygen 1.50 mg/L, alkalinity 4.00 mg/L, pH 0.5 units, conductivity 18 µS/cm, TP 0.05 mg/L, TKN 0.13 mg/L, NO_3NO_2 0.12 mg/L, and ammonia 0.02 mg/L. This suggests a homogeneous water mass across sampling sites. The

reference sites are slightly shallower than test sites with a median depth of 6.2 and 8.8 m, respectively (Table 1).

4.4 Relationships Between Mercury Concentrations in Invertebrates and Sediment

4.4.1 Total mercury

Concentrations of total Hg in each invertebrate taxon vs total Hg in sediment are plotted in Figure 14, with fitted regression lines using sediment [THg] alone as the predictor. For all taxa, the slopes are highly significant ($P \leq 0.004$) and the adjusted r^2s are 0.243 (snails), 0.581 (midges) and 0.647 (amphipods). Prediction of biota [THg] is improved for all taxa by including sediment environmental nutrient and grain size variables as additional predictors (Table 7). These brought the R^2_{adj} values up to 0.453, 0.634 and 0.879 for the snails, midges and amphipods, respectively. For the midges and amphipods, $[THg]_{sed}$ is the strongest predictor and remains highly significant ($P \leq 0.001$), whereas for the snails $[THg]_{sed}$ becomes less significant ($P=0.041$) after TOC and TN are included in the model. Nevertheless, even after fitting other sediment (and, for snails and amphipods, overlying water) variables to the regressions, $[THg]_{inv}$ is strongly related to $[THg]_{sed}$. The slope for the midges is more than double those for the snails and amphipods, which are of similar magnitudes.

4.4.2 Methyl mercury

The relationships between MeHg in biota and MeHg in sediment (Figure 15, Table 7) are weaker than those for total Hg. With $[MeHg]_{sed}$ alone as the predictor, only the snail regression is significant ($P=0.028$). The r^2_{adj} values are 0.010, 0.117 and 0.136 for the midges, amphipods and snails, respectively. With additional predictors, the regressions are made significant (P_{reg} = 0.045, 0.002 and <0.001 for amphipods, midges and snails, respectively), with R^2_{adj} = 0.358, 0.484 and 0.571 for the midges, snails and amphipods, respectively. More importantly, $[MeHg]_{sed}$ is also predictive of $[MeHg]_{inv}$ in the multiple linear regressions, with $P = 0.037$, 0.032 and 0.013 for snails, midges and amphipods, respectively. For the midges and amphipods, $[MeHg]_{sed}$ is the best of the predictors, but for the snail regression, TOC and TN are much stronger (as for the [THg] model). Thus, invertebrate MeHg concentrations are influenced by sediment MeHg concentrations, but to a lower extent than $[THg]_{inv}$ is by $[THg]_{sed}$. The fact that

(a) the models that best predict $[MeHg]_{inv}$ include $[MeHg]_{sed}$ as a significant term and (b) the magnitudes and directions of the regression coefficients are more or less stable across various models (except MeHg for amphipods) suggest real relationships between $[MeHg]_{inv}$ and $[MeHg]_{sed}$. However, the low R^2_{adj} values for the even the multiple predictor models indicate that the effect sizes of the relationships are small. Relationships between $[MeHg]_{inv}$ and $[THg]_{sed}$, also examined, are found to be no stronger than the $[MeHg]_{inv}$ - $[MeHg]_{sed}$ ones.

4.5 Predictions of Mercury Concentrations in Receptors

4.5.1 Presentation of model outcomes

Predicted concentrations of methyl mercury in each receptor species at each sampling site, calculated by multiplying observed methyl mercury concentrations in invertebrates (wet weight values from Tables D1 and D2) by the appropriate FCMs (from Table 3), are shown in Table 8 and Figures 16 to 18. Receptor MeHg concentrations are presented separately for "minimum", "intermediate" and "maximum" levels of mercury exposure and uptake scenarios. In each of the three series of subfigures, predicted $[MeHg]_{rec}$ for the five receptors are presented in bar charts comparing reference and exposed sites. In the bar charts, which have the same logarithmic scales in all figures and subfigures, two criteria concentrations are marked: (1) the maximum (= 99^{th} percentile) of the predicted $[MeHg]_{rec}$ for the reference sites, and (2) the tissue residue guideline (TRG) for the fishes.

The TRG applies only to the fish receptors. It refers to the concentration of MeHg in the diets of wildlife that consume aquatic biota. The TRG used for MeHg is the lowest of the reference concentrations derived by Environment Canada (2002) for the protection of wildlife receptors in the AOC that consume aquatic biota: 92 ng/g ww. This pertains to the American mink (table 12 of Environment Canada 2002). The recommended TRG for the protection of *all* wildlife species – 33 ng/g ww – is not appropriate because it is based on the reference concentration for Wilson's Storm Petrel, which is not native to the Cornwall area (Environment Canada 2002).

4.5.2 Exceedences of criteria

Methyl Hg – minimum The low predictions of $[MeHg]_{rec}$ result in 4 of 22 exposed sites exceeding those for the reference sites (Figure 16). Of these, two are for sites in Zone 3, and one is for a

site in each of Zones 2 and 4. The magnitudes of the exceedences are up to 2.7× the reference maximum. The same pattern of $[MeHg]_{rec}$ values among sites occurs for all receptors. (This occurs for all three exposure and uptake scenarios.) None of the predicted $[MeHg]_{rec}$ for either exposed or reference sites exceeds the TRG.

Methyl Hg – intermediate The intermediate predictions of $[MeHg]_{rec}$ result in 9 of 22 exposed sites exceeding predictions for the reference sites (Figure 17). Of these, one site is in Zone 1, two are in Zone 3, five are in Zone 2, and one is in Zone 4. The highest magnitude of exceedences is 3.1× the reference maximum. Of the exposed site predictions, the number of sites at which the predicted $[MeHg]_{rec}$ exceeds the TRG is zero for the sucker, five for the perch, and nine for the walleye. In comparison, no receptors at any of the reference sites have predicted $[MeHg]_{rec}$ exceeding the TRG.

Methyl Hg – maximum The maximum predictions of $[MeHg]_{rec}$ result in the same nine exposed sites (as for the intermediate predictions) exceeding those for the reference sites (Figure 17). The highest magnitude of exceedences is also the same – 3.1× the reference maximum. Of the exposed site predictions, the number of sites at which the predicted $[MeHg]_{rec}$ exceeds the TRG is 0 for the sucker, 9 for the perch, and all 22 for the walleye. Among reference sites, zero, one, and eight sites have predicted $[MeHg]_{rec}$ exceeding the TRG for sucker, perch and walleye, respectively.

4.5.3 Overall patterns

Beyond the comparisons of predicted $[MeHg]_{rec}$ for exposed sites to reference sites to the TRG, patterns are evident in the differences in predicted $[MeHg]_{rec}$ among the five receptors, and among the three exposure and uptake scenarios.

Among receptors Predicted $[MeHg]_{rec}$ generally increases with the trophic level of the receptor, with mean heron or mink predictions being 2× to 50× those of the sucker. The pattern is weakest for the minimum Hg exposure and uptake scenario. Consequently, the number of sites at which $[MeHg]_{rec}$ exceeds the TRG, and the amount by which the TRG is exceeded, increases with the trophic level of the receptor. However, the number of exposed sites at which predicted

$[MeHg]_{rec}$ exceeds the maximum of reference site concentrations is the same among receptors. This is because within a series (i.e., any of the minimum/intermediate/maximum groups), all derive from the same $[MeHg]_{inv}$ values. Differences among predicted $[MeHg]_{rec}$ values reflect differences among uptake pathways in the FCMs from Table 3. The pattern of variability among sites is the same for all receptors within a scenario (i.e., the $[MeHg]_{rec}$ values are fully correlated among receptors).

Among exposure and uptake scenarios Predicted $[MeHg]_{rec}$ for a given receptor increased with severity of the exposure and effect scenario (i.e., minimum to intermediate to maximum). The ratio of mean $[MeHg]_{rec}$ (averaged across all sites) for maximum:minimum scenarios ranges from 4.6× (sucker) to 134× (mink). Differences among scenarios increase with trophic level of the receptor due to the increase in variability in the FCMs as the trophic pathway lengthens. In the minimum predictions, none of the exposed or reference site $[MeHg]_{rec}$ values exceed the TRG. For the intermediate scenario, 4-9 exposed sites have $[MeHg]_{rec}$ greater than the TRG based on perch and walleye. With the maximum scenario, 9-22 exposed sites have $[MeHg]_{rec}$ greater than the TRG based on perch and walleye.

5 DISCUSSION

5.1 Mercury Concentrations in Effluent-Exposed Sites Relative to Reference Sites

5.1.1 Sediment

Concentrations of THg in the upper 10 cm layer of sediment sampled in 2001 from all sites in Zones 1, 2 and 3 of the St. Lawrence River (Cornwall) AOC are substantially greater than [THg] in sediment from references sites upstream or outside of areas exposed to past industrial effluents (Figure 3). While the maximum $[THg]_{sed}$ observed is 5568 ng/g dry weight, most concentrations are between 400 and 3000 ng/g, compared to about 10-100 ng/g for the reference sites. These compare to background concentrations of 10-700 ng/g for the Great Lakes, and concentrations of up to 3200 and 15000 for contaminated sites in the Niagara and St. Clair Rivers, respectively (Environment Canada 1997). The CCME (1999b) freshwater sediment quality guideline (Probable Effect Level) for THg is 486 ng/g. On average, Zone 2 sites are the most contaminated, followed by Zone 3 and Zone 1. In Zone 4 sites, [THg] is similar to the higher

reference site values, and in the far-field, downstream site [THg] exceeds the reference site levels. For MeHg, the same general pattern is observed (Figure 4), except that differences between the exposed and reference sites are less than an order of magnitude (1-5 ng/g vs < ~1 ng/g for the exposed and reference sites, respectively). Sediment [MeHg] is strongly related to sediment [THg] (Figure 5), with [MeHg] making up an average of 0.6% of the [THg]. Compared to available data for the 0-10 cm layer of sediment in Zone 2 sites from previous years, [THg] in 2001 shows a decrease from 1994. The spatial pattern of these results (Figures 3B, 4B) is strong evidence for a local (as opposed to regional) source of Hg to the AOC, which is in agreement with other assessments (Anderson 1990; Richman 1994; Callaghan 1996; Filion and Morin 2000; Richman and Drier 2001).

5.1.2 Benthic Invertebrates

Both THg and MeHg are taken up by the three invertebrate taxa assessed. Biota-sediment accumulation factors (based on whole-body, uncleared-gut concentrations) are all >1 for MeHg and range up to ~ 2 for THg and 50 for MeHg (excluding outliers, Figure 13). Snails have the highest BSAFs and [Hg]s overall, followed closely by amphipods. Tremblay et al. (1996a), in a study of two reservoirs and a natural lake in Quebec, reported BSAFs for detritivorous insects to be 1.9-2.8 for THg and 5.2-22.6 for MeHg.

Gut contents are included in the mercury analyses of the biota, which could obscure true BSAFs. As the amount of sediment in the gut increases, the measured BSAF will converge to 1. A true BSAFs < 1 will be overestimated because the concentration in the sediment is greater than the tissue concentration, whereas a true BSAFs >1 will be underestimated because sediment concentrations are lower than that found in the tissue (Bechtel Jacobs 1998).

Differences in observed $[Hg]_{inv}$ between exposed and reference sites are greater for THg than MeHg. Whereas in 50% or more of the sites in Zones 1, 2 and 3 $[THg]_{inv}$ is greater than the maximum of the reference sites, $[MeHg]_{inv}$ is generally elevated in a third or less of the exposed sites. The two Zone 3 sites are consistently highest in $[MeHg]_{inv}$ for all taxa, and highest in [THg] for snails and midges. Filion and Morin (2000) measured [THg] in five separate benthic invertebrate taxa collected from littoral (0.5 – 1.1 m depth) sites in the Cornwall AOC, including

a site in each of Zones 1 and 2. Although not directly comparable to the deeper sites of the present study, their [THg] in midges (~1000 ng/g dw) and snails (~400 ng/g) in the Zone 1 and 2 sites, respectively, are similar to our maximum [THg] observed for midges (1029 ng/g) and snails (430 ng/g) in Zones 1 and 2, respectively. Amphipod [THg] in the Zone 1 site (600-700 ng/g) reported by Filion and Morin (2000) is more than double our corresponding value (239 ng/g).

5.2 Effects of Mercury in Sediment on Mercury in Invertebrates

The log-log relationships between $[THg]_{sed}$ and $[THg]_{inv}$ across sites are strong, whereas those for $[MeHg]_{sed}$ and $[MeHg]_{inv}$ are weak (Table 7, Figures 14 and 15). $[THg]_{sed}$ alone significantly predicts $[THg]_{inv}$, but $[MeHg]_{sed}$ requires environmental variables as additional predictors to significantly predict $[MeHg]_{inv}$ for the midges and amphipods. Furthermore, as can be seen from the low R^2_{adj} - values of these models, only 35.8 to 57.1% of the variation in $[MeHg]_{inv}$ is explained by the predictors together. The amount explainable by $[MeHg]_{sed}$, the partial r^2, which is proportional to the P (predictor) for $[MeHg]_{sed}$ (Table 7) would be even lower. Therefore, while $[MeHg]_{sed}$ can be a statistically significant predictor, its effect size and likely ecological significance are low.

Concentrations of Hg in the benthic invertebrates are measured without clearing their guts. Thus, a fraction of the observed $[Hg]_{inv}$ could include sediment-bound Hg in the gut. While this is relevant for assessing uptake of Hg by predators of invertebrates, which consume whole organisms, it probably accounts for the strong $[THg]_{sed}$ - $[THg]_{inv}$ relationship. Concentrations of THg in sediment are generally 2-3 orders of magnitude greater than those for MeHg and they vary more among sites. Therefore, it is not surprising that the $[THg]_{sed}$ - $[THg]_{inv}$ relationship is stronger than the $[MeHg]_{sed}$ - $[MeHg]_{inv}$ relationship.

Several other studies report significant relationships between [Hg] in sediment and [Hg] in benthic invertebrates. Bechtel Jacobs (1998) reviewed data from 15 studies of [Hg] in freshwater benthic invertebrates and sediment. In 13 of these, invertebrate guts were not cleared. Slopes of $\log[THg]_{inv}$ vs $\log[THg]_{sed}$ regressions were 0.327 ± 0.246 (mean $\pm$ standard error), and the mean

r^2 was 0.12. Slopes for our St. Lawrence River sites are between 0.242 and 0.570. Tremblay et al. (1996b) found correlation between [MeHg] in chironomids and $[MeHg]_{sed}$ of r=0.78 (P<0.005, n=18) for a series of Quebec lakes. For our St. Lawrence River sites, this relationship had an r=0.2 (P=0.25, n=34). Désy et al. (2000) sampled sediment and a single snail species from 21 sites in the St. Lawrence River downstream of Cornwall. In an analysis involving 33 snails, some depurated and some not, the correlation between log[MeHg] in snails and $log[THg]_{sed}$ was reported to be 0.83. In the present study, involving snails from several families, $log[MeHg]_{snail}$ and $log[THg]_{sed}$ were not significantly related (r=0.12, P=0.065, n=29).

Sediments of Tremblay et al. (1996b), Bechtel Jacobs (1998) and (to a lesser extent) Désy et al. (2000) were less contaminated with Hg than the sites of the present study. In an assessment of bioaccumulation by midges and amphipods from Hg-contaminated and reference sediments in the Peninsula Harbour, Marathon, Ontario AOC (Grapentine et al. 2003) using the same methods as the current study, agreement between studies for $log[Hg]_{inv}$ vs $log[Hg]_{sed}$ regressions is strong. The corresponding slope coefficients (Cornwall / Peninsula Harbour) are:

- THg in midges = 0.570 / 0.431,
- THg in amphipods = 0.284 / 0.376,
- MeHg in midges = 0.160 / 0.163,
- MeHg in amphipods = 0.334 / 0.300.

In the multiple linear regressions, there is also consistency between studies in the signs of the physico-chemical co-predictors and their relative significance. Overall, the Cornwall models explain less variation in $[Hg]_{inv}$ than those for Peninsula Harbour; however, sediments in the latter AOC are higher in [Hg] than those in the former.

In conclusion, results from this assessment indicate that $[MeHg]_{inv}$ is largely determined by factors other than $[MeHg]_{sed}$ (or [THg]). Although observing positive relationships between sediment and invertebrate mercury concentrations is evidence that mercury transfers from sediment into the food web, the lack of a strong $[MeHg]_{sed}$ – $[MeHg]_{inv}$ relationship (Figure 15), which can be viewed as a "dose – response" relationship under natural conditions, argues against a causal link between the two variables alone (USEPA 1998).

5.3 Predicted Mercury Concentrations in Receptor Species

5.3.1 Integration of prediction outcomes

Models involving a range of biomagnification conditions were used to predict potential [Hg] in receptors. Five receptor species were considered to encompass the trophic levels linking sediments to the top predators, where biomagnification is expected to be greatest. Three levels of dietary exposure and trophic transfer of Hg were assumed: minimum and maximum scenarios to bracket the range of potential outcomes and an intermediate scenario to characterize "average" conditions. Conclusions determined from overall evaluations of the model outcomes should consider:

- $[MeHg]_{rec}$ for exposed sites compared to $[MeHg]_{rec}$ for references sites;
- $[MeHg]_{rec}$ relative to the TRG;
- How many receptors are predicted to exceed the criteria at each site;
- How many of the exposure and uptake scenarios result in exceedences; and
- How many sites per zone exceed the criteria.

On the whole, a minority of the 22 exposed sites were predicted to have $[MeHg]_{rec}$ higher than the maximum reference site $[MeHg]_{rec}$: four sites in three zones for the minimum scenario, and nine sites in four zones for the intermediate and maximum scenarios (Figures 16-18). The more critical outcome of the evaluation is whether or not the predicted $[MeHg]_{rec}$ values for exposed sites exceed the appropriate TRG *in addition to* exceeding the reference site maximum $[MeHg]_{rec}$. Figure 19 shows the sites meeting this condition for all exposure and uptake scenarios for the fish receptors. For the sucker, no exposed sites were predicted to result in such "hits" for any of the scenarios. Perch $[MeHg]_{rec}$ predictions resulted in five hits for the intermediate scenario, and nine hits for the maximum scenario. The sites with hits in the intermediate scenario predictions are: Site 183 of Zone 1, Sites 101 and 108 of Zone 3, Site 31 of Zone 2, and Site 179 of Zone 4. Sites with hits in the maximum scenario predictions include these same five sites plus Sites 16, 19, 27 and 64 of Zone 2. For the walleye, minimum scenario predictions resulted in no hits; intermediate and maximum scenario predictions both flagged the same nine sites as the perch maximum predictions.

The TRG applies to concentrations of MeHg in fishes, and are for the protection of wildlife or human consumers of fishes. Some data are available for direct evaluation of the predicted tissue mercury levels for heron and mink. Wolfe et al. (1998) reviewed THg and MeHg toxicity and tissue residue data associated with adverse effects for birds and mammals. (As noted above, nearly all mercury in fishes and higher trophic level animals should be in the methyl form.) For great white heron, liver concentrations > ~6000 ng/g ww THg correlated with chronic adverse effects. A conservative residue threshold for major toxic effects in water birds was concluded to be 5000 ng THg/g ww in liver. For mink, a similar criterion of 5000 ng/g ww MeHg in muscle or brain was suggested. This value of 5000 ng/g corresponds to 3.7 on the log-scales in Figures 16 to 18. For the great blue heron receptor, the highest predicted $[MeHg]_{rec}$ in any of the scenarios is 2064 ng/g ww, and for the mink the highest $[MeHg]_{rec}$ predictions is 3036 ng/g ww (Table 8). Thus, $[MeHg]_{rec}$ in heron and mink is not predicted to exceed the tissue residue benchmarks suggested by Wolfe et al. (1998).

5.3.2 Uncertainty in the prediction of mercury concentrations in receptors

The prediction of the potential transfer of mercury from benthic invertebrates to the trophically linked receptor species involves several simplifying assumptions, each of which is associated with some degree of uncertainty in its relevance to conditions in the Cornwall AOC. While it is beyond the scope of this study to quantify these uncertainties, those considered most important are identified here.

Assumptions regarding the modelling of Hg biomagnification include those dealing with the exposure of the receptors to Hg, and those dealing with the effects of Hg on the receptors. Regarding the latter category, some of the sources of uncertainty discussed by USEPA (1997c) could apply to the present study:

- validity of the biomagnification model,
- variability of the calculated BMFs and FCMs,
- selection of the receptors of concern,
- trophic levels at which receptors feed,
- limitations of the toxicity database (with respect to the determination of TRGs), and
- effects of environmental cofactors and multiple stressors.

Among these sources, the greatest contributor to uncertainty in predicting the trophic transfer of mercury could be the large range in the selected BMF and FCM values. These range over 1-1.5 orders of magnitude between lowest and highest, and include all BMFs judged to be potentially applicable to the Cornwall AOC. Further validation of their relevance would require field studies beyond the scope of this assessment. Owing to limitations of the available data and the desire to minimize assumptions about the distributions of the data, a probabilistic approach was not applied to predict receptor mercury concentrations. Rather, low, medium and high FCMs were used to define the range of possible outcomes and intermediate values that "balance" the minimum and maximum rates of biomagnification. Another problem inherent in the literature-derived BMF data is the difficulty in assigning prey and predator species to discrete trophic levels due to omnivory. When omnivory is integrated with a continuous measurement of trophic position (e.g., using stable isotope methods), estimates of BMFs will generally be higher for each discrete trophic level (Vander Zanden and Rasmussen 1996). Correct determination of trophic levels is also limited by how well the composition of a predator's diet is quantified. Often the information necessary to clearly establish this is not available in the published studies.

Another potentially large source of uncertainty in predictions of $[MeHg]_{rec}$ relates the exposure of receptors to Hg. These assumptions (listed in Sec. 2.4.3) are recognized as being conservative and limited in their representation of natural conditions. Spatial (and perhaps temporal) heterogeneity in the distribution of THg and MeHg throughout the study area, and aspects of receptor ecology challenge the maximum exposure scenario. A particularly important source of uncertainty could be the assumption of 100% residency of all consumers in the food chain on each site. The degree to which this assumption is unrealistic is proportional to the size of the foraging areas of the receptor species relative to the area of contaminated sediment. Given that the sampling sites could be on the order of 10×10 m to 100×100 m (= 0.01 to 1.0 ha), the 100% residency assumption is likely unrealistic. According to data compiled in the Wildlife Exposure Factors Handbook (USEPA 1993), feeding territory sizes for great blue heron range from 0.6 ha to 0.98 km^2, and distances they travel from heronry to foraging grounds range from 1.8 to 8 km. Home range sizes of mink are reported as 7.8 to1626 ha, and 1.85 to 5.9 km of stream/river. These foraging/home range areas substantially exceed the site and zone boundaries

of this study. If areas outside of the Hg-contaminated zones of the Cornwall AOC are not equally Hg-contaminated, the actual $[MeHg]_{rec}$ would be lower than those predicted by the models.

5.3.3 Observed mercury levels in receptors from the Cornwall area

While comparisons with observed [Hg] in fishes, heron and mink from the Cornwall AOC are potential means of validating the predicted $[MeHg]_{rec}$, this is not straightforward. Fish and wildlife exposure histories are difficult to determine. For reasons noted above, resident receptors are unlikely to feed as assumed by the prediction model (i.e., focus on single sites). Other sources of mercury beyond Zones 1-4 of the study area could contribute to receptor mercury burdens. Nevertheless, measured [Hg] in recently sampled receptors are indications of actual, as opposed to potential, biomagnification.

Surveys of sport fish contaminant levels include collections of adult perch and walleye from Lake St. Francis (the widening of the St. Lawrence River starting about 5 km downstream of the far-field site) and Lake St. Lawrence (the upstream reference area) in 1998-2000 (Lisa Richman, Ontario Ministry of the Environment, pers. comm.). Median concentrations of THg in perch from Lake St. Francis in 1998 and 2000 were about 200 ng/g ww. Perch from Lake St. Lawrence in 1999 averaged slightly less than 200 ng/g for [THg]. Median [THg] for walleye from Lake St. Francis in 1998 were 800 ng/g. Outlying concentrations of 2000 and 3000 ng/g also recorded. These data are close to the most recent (1992-1994) data on mean THg concentrations in perch and walleye summarized by St. Lawrence RAP Team (1997), and are exceeded by the predicted [THg] of the present study only for a few sites under the maximum exposure and uptake scenarios.

Comparisons of the predicted $[MeHg]_{rec}$ with the sport fish data are difficult because adult perch and walleye are able to move throughout the AOC, and were collected from Lake St. Francis, an area with Hg-contaminated sediment of its own. More relevant are data on Hg in shorter ranging, smaller/younger fishes collected from several zones within the AOC in 2002 (Jeff Ridal, St. Lawrence River Institute of Environmental Sciences, pers. comm.). Perch up to ~14 cm in length–probably feeding more on invertebrates than fishes (Scott and Crossman 1973)–generally had [THg] ≤100 ng/g ww, except for one value of 340 ng/g for a fish from Zone 1. Brown

bullheads were also collected and analysed. While these are not one of the receptors of interest, they are ecologically similar to suckers, and likely more closely associated with sediments in diet and habits than perch (Scott and Crossman 1973). The bullheads were 15-30 cm in length, and had [THg] of ~50-150 ng/g, with a maximum of 270 ng/g, again for an individual from Zone 1. The highest predicted [MeHg] for white sucker is 60.7 ng/g.

In an assessment of contaminant burdens and biomarker responses of heron in the St. Lawrence River, Champoux et al. (2002) measured THg in kidney of fledgling great blue herons collected from Île Dickson (~2 km downstream of St. Regis Island at the entrance to Lake St. Francis) between 1991 and 1994. The mean ± standard deviation for [THg] in kidney was 554.6 ± 144.7 ng/g ww (converted from reported dw concentration and percent moisture). This value is exceeded by the predicted [MeHg] for one reference and most exposed sites with the maximum scenario; all intermediate scenario predicted [MeHg] are less than 554.6 ng/g.

Mink trapped within 3 km of the Cornwall AOC in 2000-2001 were analysed for liver [THg] by Martin and Klenavic (2003). Mean ± standard deviation of [THg] were 4860 ± 4260 ng/g ww, with a maximum value of 10930 ng/g. These levels are substantially higher that even the maximum exposure and uptake scenario predictions of [MeHg] in mink (Table 8).

Observations of [MeHg] in receptor species residing in the Cornwall AOC thus suggest that mercury does accumulate in tissues of higher trophic level members of aquatic food webs. It is also evident that, except for the maximum exposure and uptake scenario for the heron, the receptor MeHg concentrations predicted from the screening level approach of this assessment are not overshooting actual tissue levels. The key question to resolve is evaluating how much of the observed $[MeHg]_{rec}$ originates from sediments in the AOC (specifically Zones 1-4).

5.4 Potential Risk of Adverse Effects of Mercury due to Biomagnification from Sediment

Concluding that mercury originating from contaminated sediment could concentrate in the food web at levels that can cause adverse effects depends on establishing that:

(1) mercury in invertebrates from sites exposed in the past to industrial effluents is elevated relative to concentrations in invertebrates from reference sites;
(2) mercury in invertebrates is related to mercury in sediment; and
(3) predicted levels of mercury in receptors at exposed sites that exceed levels in receptors at reference sites also exceed the TRG.

Results show that at most of the exposed sites THg and, to a lesser degree, MeHg in all invertebrate taxa are significantly higher than concentrations for the reference sites (Figures 7-12). Measured mercury concentration in invertebrates is related to mercury concentration in sediment, but mainly for THg (Figure 14, Table 7). While $[MeHg]_{sed}$ is statistically predictive with other environmental predictors of $[MeHg]_{inv}$ for all taxa, the effect is not large. Alone, $[MeHg]_{sed}$ shows a relationship to $[MeHg]_{inv}$ only for snails (Figure 15). This it noteworthy because MeHg is the form important to the biomagnification process. Regarding the trophic transfer modelling, based on outcomes for perch and walleye under the intermediate and maximum mercury exposure and uptake scenarios, up to nine exposed sites could be considered "of concern" because of predicted $[MeHg]_{rec}$ exceeding reference sites conditions and the TRG (Figure 19).

Regarding the overall assessment of sediment conditions based on the integrated framework outlined in Section 1.2, the bioaccumulation/biomagnification line of evidence can differ from the other three lines of evidence. If fish and wildlife receptors are the concern, the appropriate spatial and temporal boundaries for assessing potential biomagnification are not the same as those for assessing sediment contaminant concentrations, sediment toxicity and benthic invertebrate communities. Activities of fishes, birds and mammals are not limited to individual sites to the same degree as contaminants and invertebrates. Whereas incorporating invertebrate contaminant bioaccumulation information into the framework works well on a site-by-site basis, fish and wildlife data require some form of spatial averaging or weighting to reflect realistic contaminant exposure conditions. On a per site basis, fish and wildlife biomagnification predictions remain "theoretical" or overly conservative.

One way of addressing the problem is to assess exposure to contaminants across areas of sediment comparable to the foraging areas of the receptors, as suggested by Freshman and Menzie (1996). Their "average concentration with area curve" exposure model involves determining the average concentration of a contaminant for increasing areas of soil, starting with the most contaminated site up to and beyond the foraging area of the receptor of interest. The average contaminant concentration for a section of soil corresponding to the foraging area is then compared to appropriate benchmark adverse effect levels. Exceedence of the benchmark by the average contaminant concentration is considered a potential impact to the receptor individual.

Application of this method requires a grid-type or other statistically suitable array of sampling sites designed to representatively quantify contaminant conditions across the study area. While this type of sampling was not done for the present study, a rough characterization of conditions across each zone can be obtained by averaging mercury concentrations for the sites within the zones. Because $[MeHg]_{rec}$ is directly related to $[MeHg]_{inv}$ (here, the contaminant source), averaging $[MeHg]_{rec}$ values is equivalent to averaging $[MeHg]_{inv}$. And since predicted $[MeHg]_{rec}$ were screened to be of concern only for the perch and walleye intermediate and maximum scenarios, only these prediction need to be considered.

If $[MeHg]_{rec}$ values are averaged for each zone (excluding geographically outlying Sites 46 and 54 from Zone 2), mean $[MeHg]_{rec}$ exceed the TRG for most zones for the intermediate perch scenario and the intermediate and maximum walleye scenarios (Table 9). The areas range in size roughly from 0.2 ha for Zone 3 to 8.5 ha for Zone 2. Fishes with foraging areas less than these sizes could potentially accumulate Hg to levels above the TRG, whereas those with larger foraging areas would be expected to accumulate less Hg.

The application of tissue Hg residue data that are associated with adverse effects in other studies to evaluate potential risks to the receptors in the present study carries some uncertainty. The data come from different tissues, species, environmental conditions and study types (e.g., field vs lab). In addition, Hg detoxification and a possible ameliorative effect of dietary selenium may contribute further uncertainty in the extrapolation of results from one set of conditions to another (USEPA 1997c). The TRGs also typically include uncertainty factors. For example, the MeHg

reference concentration (92 ng/g wet wt) incorporates an uncertainty factor of 5 (Environment Canada 2002). Considering these uncertainties, and the generally conservative ("worst case") assumption of the trophic transfer model, quantifying the probability that mercury from sediments in the Cornwall AOC could cause adverse effects to receptors is difficult. A further consideration is that sediment mercury concentrations appear to be decreasing with time. For sites from previous studies that were also sampled in 2001 (Zone 2), sediment THg concentrations are generally lower than in previous years (1994, 2000). These deposits are also considered to be stable (Rukavina 2000).

6 CONCLUSIONS

The purpose of the study was to determine if deleterious amounts of mercury could potentially be transferred from sediments through benthic invertebrates to fish, wildlife or humans in the Cornwall AOC. This is addressed by:

A. Determining if THg and MeHg are bioaccumulated by benthic invertebrates to higher concentrations in sites that were exposed to Hg-containing industrial effluents than in unexposed reference sites;
B. Testing if concentrations of THg and MeHg in invertebrates are related to concentrations in sediment; and
C. Predicting if concentrations of MeHg in consumers of benthic invertebrates and their predators (i.e., trophically linked receptor species) reach levels associated with adverse effects.

The main findings of the study are:

A. Total and methyl mercury concentrations in sediment at all or the majority of sites exposed to historical industrial discharges in Zones 1-3 are higher than those at upstream reference sites, the far-field downstream site, and at sites in Zone 4. Methyl mercury is < 1% of total mercury at all but 4 sites.

B. Total mercury concentrations in biota (midges, snails, amphipods) (analyzed with gut contents) are higher in Zones 1-3 relative to upstream reference sites and the downstream site, whereas for Zone 4, concentrations are intermediate between all other sites. Methyl mercury concentrations in biota are higher in Zone 3 and at a minority of sites in Zones 1, 2 and 4 relative to upstream reference sites and the far-field site. **This suggests that historic effluent discharges are linked to elevated invertebrate [Hg] at some sites.**

C. The concentration of total mercury in sediment is strongly predictive of concentration in benthic invertebrates (analysed without allowing gut clearance), suggesting that mercury contaminated sediments are the source, but it does not preclude other sources of mercury to the benthic invertebrates (e.g. waterborne mercury). The concentration of methyl mercury in sediment is not strongly correlated to methyl mercury in benthic biota and, for midges and amphipods, only after adjusting for effects of other sediment covariables. **This suggests that mercury from sediment is taken up by invertebrates largely in inorganic form, but is likely not strongly incorporated into tissues as the more bioavailable and toxicologically relevant methyl form. The sources and pathways of methyl mercury to invertebrates therefore remain uncertain.**

D. In the zones exposed to past industrial effluents, a minority of the 22 exposed sites were predicted to have $[MeHg]_{rec}$ higher than the maximum reference site $[MeHg]_{rec}$: four sites in three zones for the minimum exposure and uptake modelling scenario, and nine sites in four zones for the intermediate and maximum scenarios. Of these, a smaller number of predictions exceeded the tissue residue guideline for the protection fish-consuming wildlife and humans: for yellow perch, $[MeHg]_{rec}$ at five sites with the intermediate scenario and nine sites with the maximum scenario; for the walleye, $[MeHg]_{rec}$ at nine sites with both the intermediate and maximum scenarios. **This suggests that under intermediate and maximum mercury - exposure and uptake assumptions, mercury could bioaccumulate in receptors to levels that are not protective of adverse effects at a few exposed sites.** However, the likelihood of realizing this degree of mercury biomagnification is not clear, due to uncertainties associated with predicting receptor [MeHg] values and conservative assumptions of the assessment. Reducing uncertainty in the predictions of mercury biomagnification in the Cornwall AOC would be best achieved by identifying a more narrow range of appropriate BMFs, and by quantifying the actual exposures of receptors to dietary mercury.

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Table 1. Station co-ordinates (UTM NAD 83) and depth in the St. Lawrence River (Cornwall) Area of Concern.

Site	Depth (m)	Easting	Northing
Upstream Reference			
1319	2.6	506371.1	4984680.3
1320	6.4	505141.8	4983905.1
1321	5.4	514300.8	4985690.5
1322	6.0	515122.9	4983020.4
1323	6.8	515657.4	4982764.2
1325	5.8	509857.1	4985759.0
1326	8.4	507351.4	4983503.2
1331	8.0	508732.5	4983873.6
1332	10.6	515998.5	4985347.7
Downstream Reference			
1327	5.2	526730.9	4984006.7
1328	8.6	525933.5	4983770.6
A1	5.8	527311.7	4984070.2
Zone 1			
167	7.1	521173.8	4984281.9
168	7.9	521206.9	4984278.2
183	1.9	521111.5	4984161.8
184	4.0	521151.7	4984213.0
Zone 3			
101	7.1	523157.5	4984774.4
108	6.3	523196.2	4984755.9
Zone 2			
5	6.1	523952.5	4985067.8
9	6.5	523996.8	4985100.2
16	9.2	524163.8	4985100.4
17	8.9	524201.9	4985223.3
19	9.0	524252.2	4985223.2
27	10.7	524419.3	4985285.2
31	8.7	524582.0	4985396.2
46	10.1	525164.5	4985713.7
54	8.1	525459.7	4985909.0
64	3.1	524075.0	4985179.5
Zone 4			
175	13.4	525574.2	4985096.4
179	15.0	525959.6	4985031.3
173	10.5	525392.8	4985081.3
176	14.5	525662.2	4985004.5
182	11.0	526254.2	4985068.8
Farfield Downstream			
171	10.0	526920.2	4985901.2

Table 2. List of variables measured at each sampling site.

Geographical	Water	Sediment	Biota
Northing	Temperature	Total Mercury	Total Mercury
Easting	Alkalinity	Methyl Mercury	Methyl Mercury
Site Depth	Conductivity (on site)	Metals/Major Oxides	
	Total Phosphorus	Total Phosphorus	
	Nitrate+Nitrite-N	Total Nitrogen	
	Ammonia-N	Total Organic Carbon	
	Total Kjeldahl Nitrogen	% Clay, Silt, Sand, & Gravel	
	pH (on site)		
	Dissolved Oxygen (on site)		

Table 3. Literature derived biomagnification factors (BMFs) for the receptors of concern. For each receptor, the number of trophic levels removed from benthic invertebrates (Level 1) is indicated. For each transfer between trophic levels, the lowest, medium and highest estimated BMFs (from Table B1) are used in calculating the food chain multipliers (FCMs). See text for further details. Where receptors have only one BMF value, the same value is used for the low, medium, and high FCM calculations.

Receptor	Predator Type	Trophic levels of transfer	BMFs (low | med | high) of transfer	Food chain multipiers (low | med | high)
White Sucker / Forage fish	benthivorous / planktivorous fish	1 - 2	3.43	3.43
Adult Yellow Perch	small piscivorous fish	1 - 2 - 3	3.43 x 5	17.15
Walleye	large piscivorous fish	1 - 2 - 3	3.43 x (1.12 | 3.20 | 32.40)	3.84 | 10.98 | 111.1
		1 - 2 - 3 - 4	3.43 x 5 x 2.40	41.16
Great Blue Heron	piscivorous bird	1 - 2 - 3	3.43 x 6.80	23.32
		1 - 2 - 3 - 4	3.43 x 5 x (0.85 | 2.37 | 6.80)	14.58 | 40.65 | 116.6
Mink	piscivorous mammal	1 - 2 - 3	3.43 x (1.70 | 5.20 | 22.64)	5.83 | 17.84 | 77.66
		1 - 2 - 3 - 4	3.43 x 5 x (1.70 | 4.70 | 10.00)	29.16 | 80.61 | 171.5

Table 4. Total and methyl mercury in sediment (ng/g wet and dry weight) collected from the St. Lawrence River (Cornwall) Area of Concern. Within-site replicates for the four randomly selected quality assurance/quality control sites are denoted by a "-" + replicate number. (F) = Flett results, (C) = Caduceon results.

Area/Zone	Site	**Total Hg** (F) (ng/g) wet wt	**Total Hg** (F) (ng/g) dry wt	**Total Hg** (C) (ng/g) dry wt	**Methyl Hg** (F) (ng/g) wet wt	**Methyl Hg** (F) (ng/g) dry wt
Reference – upstream	1319	22	63	212	0.193	0.550
	1320	5	16	268	0.122	0.385
	1321	28	44	157	0.284	0.455
	1322	26 (33)[a]	46 (57)[a]	51	0.240 (0.221)[a]	0.418 (0.385)[a]
	1323	48	117	109	0.277	0.680
	1325	27	83	124	0.193	0.583
	1326	18	58	107	0.090	0.287
	1331	42	123	119	0.310 (0.237)[a]	0.913 (0.697)[a]
	1332-1	27 (23)[a]	127 (111)[a]	205	0.153	0.725
	1332-2	31	120	114	0.212	0.838
	1332-3	48	174	159	0.120	0.432
Reference - downstream	1327	25	55	76	0.286	0.632
	1328	40	66	44	0.140	0.231
	A1	42	137	100	0.351	1.140
Zone 1	167	128	384	869	0.426	1.280
	168	135	378	724	0.537	1.500
	183	419	1646	4880	1.360	5.350
	184-1	157	689	3210	0.323	1.420
	184-2	113 (125)[a]	442 (490)[a]	2200	0.225	0.879
	184-3	273	1195	4680	0.711	3.110
Zone 3	101	268	610	1490	1.160	2.640
	108	1402	2879	3310	1.070	2.190
Zone 2	5-1	503	1845	1870	1.050	3.870
	5-2	466	2030	14300	0.789 (0.992)[a]	3.440 (4.330)[a]
	5-3	425	1752	3960	0.915	3.770
	9	849	2481	3540	1.030	3.020
	16	438	928	1140	0.497	1.050
	17	733	1654	1310	0.616	1.390
	19	2310	5568	4100	1.420	3.410
	27	926	1745	3260	0.881 (0.713)[a]	1.660 (1.340)[a]
	31	2105	4303	5420	1.650	3.370
	46	144	545	780	1.270	4.810
	54	198 (158)[a]	421 (337)[a]	828	0.341 (0.336)[a]	0.726 (0.716)[a]
	64	702	2935	4700	0.790	3.300
Zone 4	173	33 (43)[a]	108 (140)[a]	149	0.113 (0.142)[a]	0.367 (0.462)[a]
	175	44	127	133	0.256	0.738
	176	21	93	173	0.169 (0.173)[a]	0.734 (0.751)[a]
	179	16	53	908	0.502	1.640
	182	27	65	138	0.252	0.611
Downstream	171-1	60	159	366	0.341	0.899
	171-2	92	318	413	0.341	1.180
	171-3	99	282	395	0.216	0.616

[a] laboratory duplicate

Table 5. Total mercury (ng/g dry weight) in biota collected from the St. Lawrence River (Cornwall) Area of Concern.

Area/Zone	Site	BIOTA – Total Hg		
		Chironomid	Snail	Amphipod
Ref. Upstream	1319	29	-[a]	-
	1320	33	107	-
	1321	88	56	72
	1322	43	92	64
	1323	50	-	-
	1325	62 (46)[b]	-	-
	1326	17	-	-
	1331	23	-	-
	1332	109	166	-
Ref. Downstream	1327	92	44	-
	1328	88	35	-
	A1	85	108	-
Zone 1	167	121	158	-
	168	153(142)[b]	114	-
	183	1029	466(539)[b]	-
	184	347	265(302)[b]	239
Zone 3	101	392	336	-
	108	1623	735	244
Zone 2	5	153	47	400
	9	524	150	-
	16	92	196	144
	17	31	107	235
	19	848	186	-
	27	388	259	-
	31	1642	402(457)[b]	393
	46	119(105)[b]	70	-
	54	96	102	-
	64	457	185	261
Zone 4	175	80	97	-
	179	60 (66)[b]	374	172(203)[b]
	173	106	137	-
	176	70	134	-
	182	69	51	-
Downstream	171	92	94	-

[a] no data
[b] laboratory duplicate

Table 6. Methyl mercury (ng/g dry weight) in biota collected from the St. Lawrence River (Cornwall) Area of Concern.

Area/Zone	Site	BIOTA – Methyl Hg		
		Chironomid	Snail	Amphipod
Ref. Upstream	1319	3.9	-[a]	-
	1320	8.5	18.3	-
	1321	10.4	10.6	13.8
	1322	9.3(8.0)[b](10.8)[c]	15.3	17.9
	1323	12.4	-	-
	1325	3.1	-	-
	1326	2.7	-	-
	1331	1.7	-	-
	1332	3.7	12.5	-
Ref. Downstream	1327	13.9	31.0	-
	1328	12.7	23.1	-
	A1	7.3 (7.9)[c]	34.3	-
Zone 1	167	5.2	24.5	-
	168	4.9	24.0(28.3)[c]	-
	183	10.7	93.0	-
	184	4.5	32.5(32.4)[b]	17.7
Zone 3	101	25.9	96.8	-
	108	34.9	108.0	76.1
Zone 2	5	7.0	12.9	16.9
	9	6.5	7.1	-
	16	9.2	18.1	43.8
	17	5.7	24.7	29.0
	19	13.6	37.6	-
	27	11.7	47.1	-
	31	27.9	76.4	39.6
	46	3.5	18.9(15.8)[b]	-
	54	4.2	27.2	-
	64	7.3	24.1 (24.1)[c]	53.1
Zone 4	175	13.6	12.9	-
	179	11.9	67.2	33.0(39.1)[c]
	173	10.0	19.3	-
	176	15.4	22.0	-
	182	8.1	12.2	-
Downstream	171	10.9(7.7)[b](7.7)[c]	26.8	-

[a] no data
[b] repeat analysis
[c] laboratory duplicate

Table 7. Prediction of whole body concentrations of total and methyl mercury in three invertebrate taxa based on sediment mercury concentration alone ("A" models), and sediment mercury concentration + other sediment physico-chemical variables ("B" models). The groups of multiple predictors listed are from the models that best predicted $[Hg]_{inv}$ using sediment and water (snails and amphipods) variables. $[Hg]_{sed}$ was retained in all models. All variables in the models shown were transformed: arcsine-square root (x) for the "%" variables; log(x) for the others.

Response ($[Hg]_{inv}$)	Model	Predictor ($[X]_{sed}$)	Coefficient	P (predictor)	R^2_{adj}	P (regression)
Total Hg	A	total Hg	0.570	< 0.001	0.581	< 0.001
Midges	B	total Hg	0.576	< 0.001	0.634	< 0.001
		% sand	1.135	0.018		
		% clay	2.326	0.061		
Total Hg	A	total Hg	0.242	0.004	0.243	0.004
Snails	B	total Hg	0.184	0.041	0.453	0.002
		TOC	0.996	0.002		
		TN	-1.392	0.019		
		TP	1.244	0.307		
		Fe	0.675	0.314		
Total Hg	A	total Hg	0.284	0.003	0.647	0.003
Amphipods	B	total Hg	0.285	< 0.001	0.879	< 0.001
		% clay	1.825	0.085		
Methyl Hg	A	methyl Hg	0.160	0.254	0.010	0.254
Midges	B	methyl Hg	0.368	0.032	0.358	0.002
		TN	-0.606	0.094		
		% sand	0.488	0.158		
		Mn	0.658	0.069		
Methyl Hg	A	methyl Hg	0.334	0.028	0.136	0.028
Snails	B	methyl Hg	0.345	0.037	0.484	< 0.001
		TOC	1.016	0.001		
		TN	-1.385	< 0.001		
		Mn	0.703	0.037		
Methyl Hg	A	methyl Hg	0.334	0.177	0.117	0.177
Amphipods	B	methyl Hg	0.904	0.013	0.571	0.045
		% silt	-1.701	0.025		
		Mn	1.359	0.029		

Table 8. Observed methyl mercury concentrations in invertebrates and predicted concentrations in receptor species. Units for all concentration are ng/g wet weight. Values for fishes exceeding the Environment Canada (2002) tissue residue guideline (92 ng/g ww) are highlighted.

		Invertebrates			White Sucker			Yellow Perch			Walleye			Great Blue Heron			Mink		
Area	**Site**	**min**	**med**	**max**	**min**	**med**	**max**	**min**	**med**	**max**	**min**	**med**	**max**	**min**	**med**	**max**	**min**	**med**	**max**
Reference	1319	0.5	0.5	0.5	1.7	1.7	1.7	8.6	8.6	8.6	1.9	13.0	55.6	7.3	16.0	58.3	2.9	24.6	85.8
	1320	1.0	2.6	4.1	3.4	8.7	14.1	17.2	43.7	70.3	3.8	66.5	455.5	14.6	81.6	478.1	5.8	125.5	703.2
	1321	1.0	2.0	3.4	3.4	6.9	11.7	17.2	34.3	58.3	3.8	52.1	377.7	14.6	64.0	396.4	5.8	98.5	583.1
	1322	1.3	1.8	2.1	4.5	6.2	7.2	22.3	30.9	36.0	5.0	46.9	233.3	19.0	57.6	244.9	7.6	88.6	360.2
	1323	1.3	1.3	1.3	4.5	4.5	4.5	22.3	22.3	22.3	5.0	33.9	144.4	19.0	41.6	151.6	7.6	64.0	223.0
	1325	0.4	0.4	0.4	1.4	1.4	1.4	6.9	6.9	6.9	1.5	10.4	44.4	5.8	12.8	46.6	2.3	19.7	68.6
	1326	0.3	0.3	0.3	1.0	1.0	1.0	5.1	5.1	5.1	1.2	7.8	33.3	4.4	9.6	35.0	1.7	14.8	51.5
	1331	0.2	0.2	0.2	0.7	0.7	0.7	3.4	3.4	3.4	0.8	5.2	22.2	2.9	6.4	23.3	1.2	9.8	34.3
	1332	0.4	0.9	1.3	1.4	2.9	4.5	6.9	14.6	22.3	1.5	22.2	144.4	5.8	27.2	151.6	2.3	41.8	223.0
	1327	1.3	2.9	4.5	4.5	9.9	15.4	22.3	49.7	77.2	5.0	75.6	500.0	19.0	92.8	524.7	7.6	142.8	771.8
	1328	1.5	2.8	3.8	5.1	9.1	13.0	25.7	45.4	65.2	5.8	69.1	422.2	21.9	84.8	443.1	8.7	130.5	651.7
	A1	1.0	3.4	5.7	3.4	11.5	19.6	17.2	57.5	97.8	3.8	87.3	633.3	14.6	107.2	664.6	5.8	164.9	977.6
Zone 1	167	0.6	2.7	4.7	2.1	9.1	16.1	10.3	45.4	80.6	2.3	69.1	522.2	8.7	84.8	548.0	3.5	130.5	806.1
	168	0.6	2.6	4.6	2.1	8.9	15.8	10.3	44.6	78.9	2.3	67.8	511.1	8.7	83.2	536.4	3.5	128.0	788.9
	183	1.1	7.3	13.4	3.8	24.9	46.0	18.9	124.3	229.8	4.2	189.0	1488.7	16.0	231.9	1562.4	6.4	356.9	2298.1
	184	0.5	2.5	4.8	1.7	8.6	16.5	8.6	42.9	82.3	1.9	65.2	533.3	7.3	80.0	559.7	2.9	123.1	823.2
Zone 3	101	3.1	8.9	14.7	10.6	30.5	50.4	53.2	152.6	252.1	11.9	232.0	1633.2	45.2	284.7	1714.0	18.1	438.1	2521.1
	108	2.9	10.3	17.7	9.9	35.3	60.7	49.7	176.6	303.6	11.1	268.5	1966.5	42.3	329.5	2063.8	16.9	507.1	3035.6
Zone 2	5	0.8	1.8	2.7	2.7	6.2	9.3	13.7	30.9	46.3	3.1	46.9	300.0	11.7	57.6	314.8	4.7	88.6	463.1
	9	0.9	1.2	1.4	3.1	3.9	4.8	15.4	19.7	24.0	3.5	30.0	155.5	13.1	36.8	163.2	5.2	56.6	240.1
	16	1.3	4.3	8.6	4.5	14.9	29.5	22.3	74.3	147.5	5.0	113.0	955.5	19.0	138.6	1002.8	7.6	213.3	1474.9
	17	0.6	3.1	4.9	2.1	10.6	16.8	10.3	53.2	84.0	2.3	80.8	544.4	8.7	99.2	571.3	3.5	152.6	840.4
	19	1.4	3.9	6.4	4.8	13.4	22.0	24.0	66.9	109.8	5.4	101.7	711.0	20.4	124.8	746.2	8.2	192.0	1097.6
	27	1.4	4.1	6.7	4.8	13.9	23.0	24.0	69.5	114.9	5.4	105.6	744.4	20.4	129.6	781.2	8.2	199.4	1149.1
	31	4.1	8.6	13.8	14.1	29.4	47.3	70.3	146.9	236.7	15.7	223.3	1533.2	59.8	274.0	1609.1	23.9	421.7	2366.7
	46	0.5	2.1	3.7	1.7	7.2	12.7	8.6	36.0	63.5	1.9	54.7	411.1	7.3	67.2	431.4	2.9	103.4	634.6
	54	0.6	2.7	4.7	2.1	9.1	16.1	10.3	45.4	80.6	2.3	69.1	522.2	8.7	84.8	548.0	3.5	130.5	806.1
	64	0.7	3.6	6.2	2.4	12.2	21.3	12.0	61.2	106.3	2.7	93.0	688.8	10.2	114.1	722.9	4.1	175.6	1063.3
Zone 4	175	1.7	2.0	2.3	5.8	6.9	7.9	29.2	34.3	39.4	6.5	52.1	255.5	24.8	64.0	268.2	9.9	98.5	394.5
	179	1.4	6.4	13.6	4.8	22.0	46.6	24.0	109.8	233.2	5.4	166.8	1511.0	20.4	204.7	1585.8	8.2	315.1	2332.4
	173	1.1	2.3	3.5	3.8	7.9	12.0	18.9	39.4	60.0	4.2	60.0	388.9	16.0	73.6	408.1	6.4	113.2	600.3
	176	1.4	2.6	3.7	4.8	8.7	12.7	24.0	43.7	63.5	5.4	66.5	411.1	20.4	81.6	431.4	8.2	125.5	634.6
	182	0.8	1.4	2.0	2.7	4.8	6.9	13.7	24.0	34.3	3.1	36.5	222.2	11.7	44.8	233.2	4.7	68.9	343.0
Far-field	171	1.1	3.1	5.1	3.8	10.6	17.5	18.9	53.2	87.5	4.2	80.8	566.6	16.0	99.2	594.7	6.4	152.6	874.7

Table 9. Average predicted methyl mercury concentrations (ng/g wet weight) in yellow perch and walleye for zones of sediment exposed to past industrial discharges. Predictions from intermediate and maximum mercury exposed and uptake scenarios are shown. Highlighted values exceed the tissue residue guideline of 92 ng/g ww. Area values are for the approximate amount of soft river bottom enclosed by a rectangle around the sites of the zone.

Zone	Area (ha)	Average [Methyl Hg] (ng/g ww)			
		Yellow Perch		Walleye	
		med	max	med	max
Z 1	1.1	64.3	117.9	97.8	763.8
Z 3	0.2	164.6	277.8	250.3	1799.8
Z 2 (excluding sites 46 and 54)	8.5	65.3	108.7	99.3	704.1
Z 4	8.3	50.2	86.1	76.4	557.7

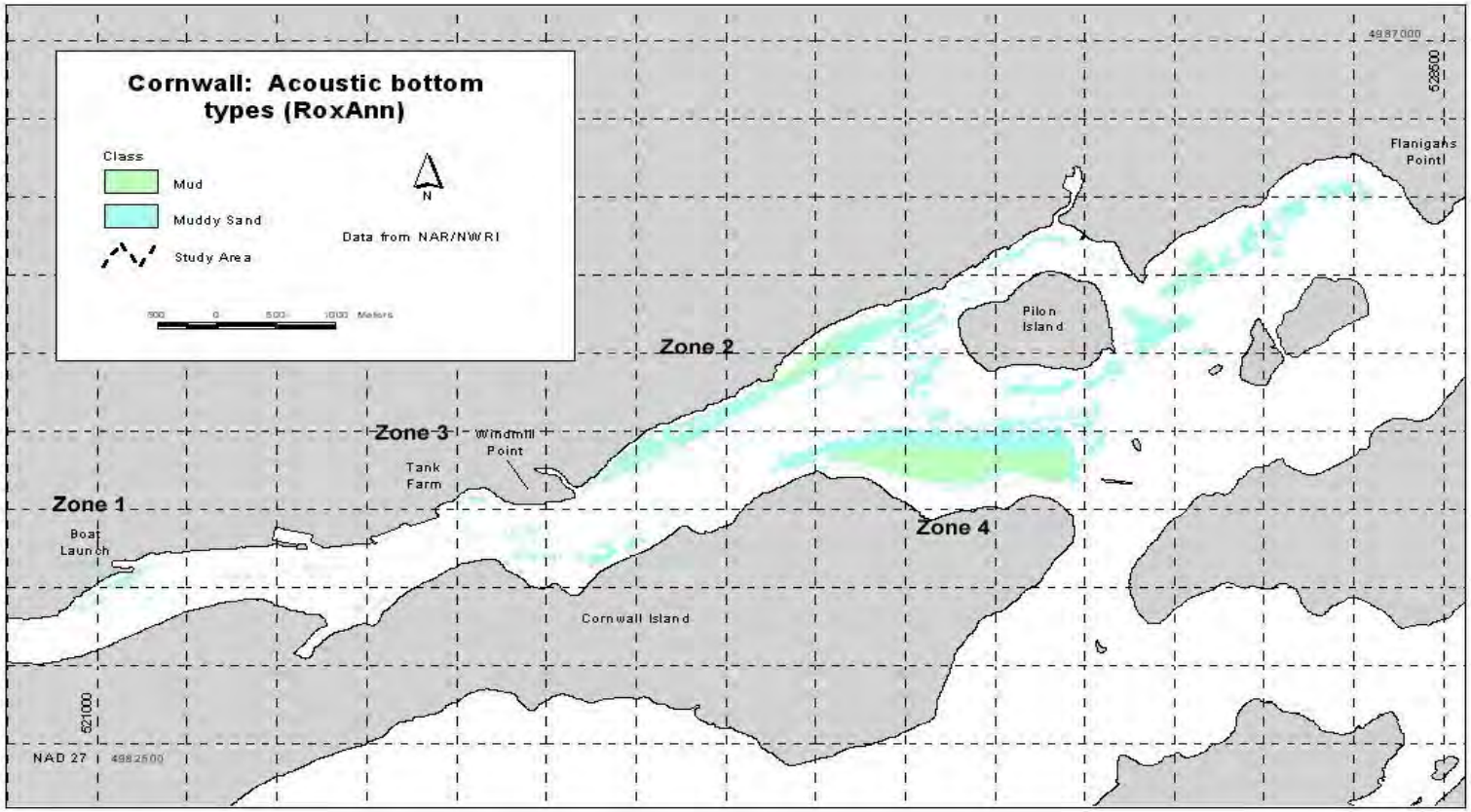


Figure 1. Fine-grained sediment deposits in study areas of the St. Lawrence River (Cornwall) Area of Concern. Colored areas show mud and muddy sand based on "RoxAnn" acoustic mapping from Rukavina (2000).

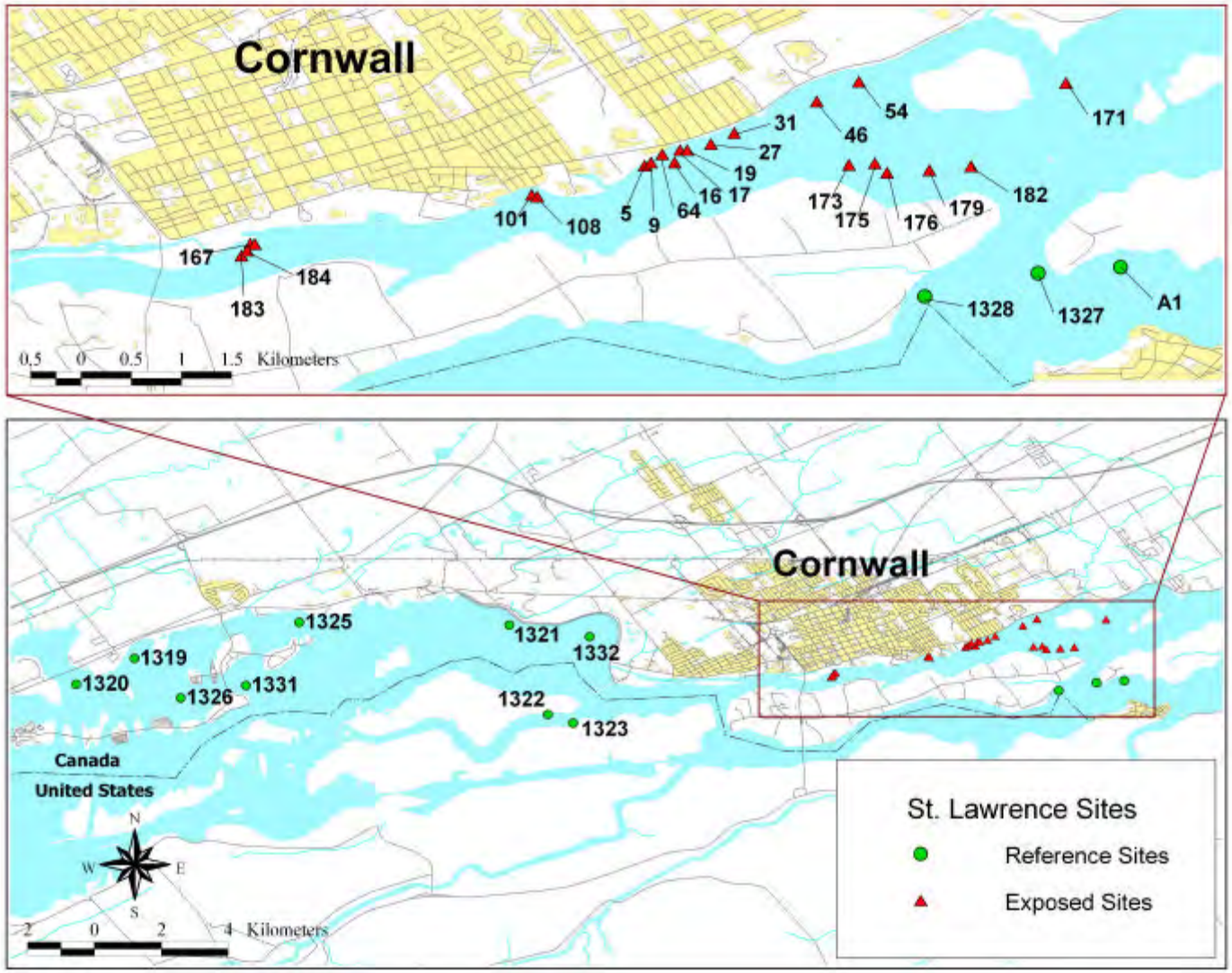


Figure 2. Invertebrate and sediment sampling locations in St. Lawrence River (Cornwall) Area of Concern. ● = reference site ▲ = exposed site.

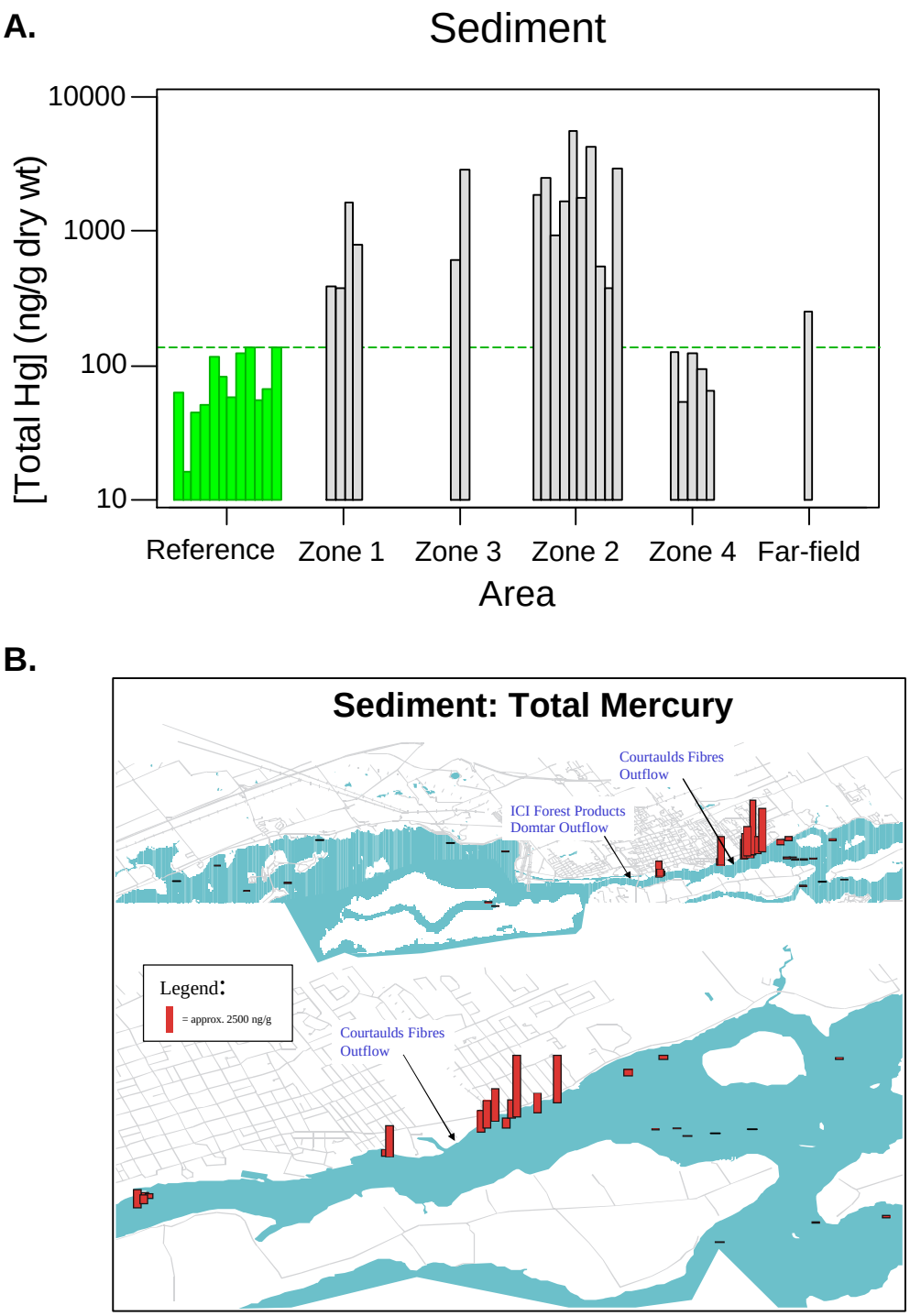


Figure 3. Total mercury in sediment from the St. Lawrence River (Cornwall) Area of Concern. A. Exposed (grey bars) compared to reference (green bars) sites. Dashed line indicates maximum reference site concentration. B. Geographic pattern of sediment mercury concentrations. The lower half of the map is an expanded view of the exposed areas shown in the upper half. Note: scaling of bars is arithmetic in B, logarithmic in A.

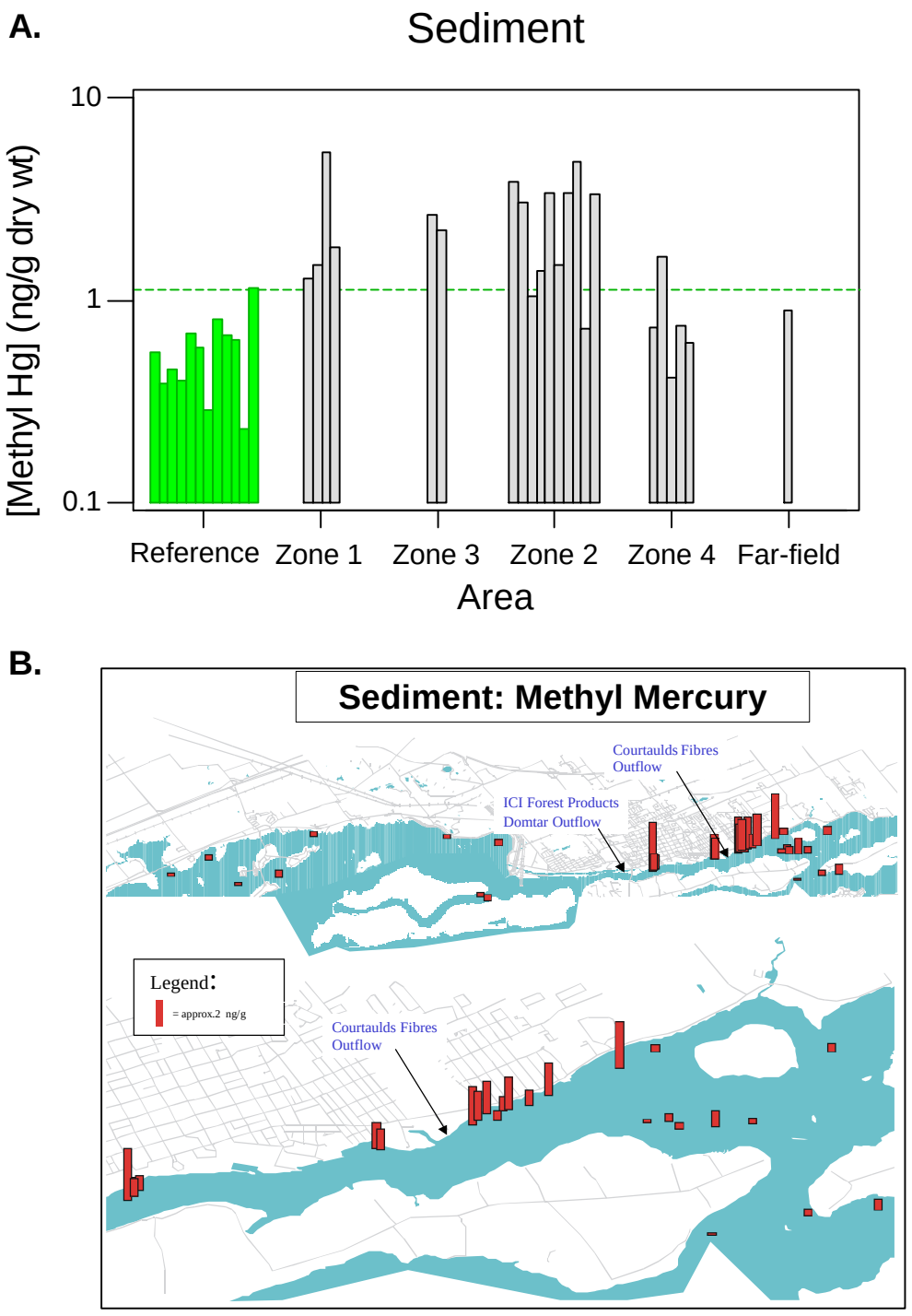


Figure 4. Methyl mercury in sediment from the St. Lawrence River (Cornwall) Area of Concern. A. Exposed (grey bars) compared to reference (green bars) sites. Dashed line indicates maximum reference site concentration. B. Geographic pattern of sediment mercury concentrations. The lower half of the map is an expanded view of the exposed areas shown in the upper half. Note: scaling of bars is arithmetic in B, logarithmic in A.

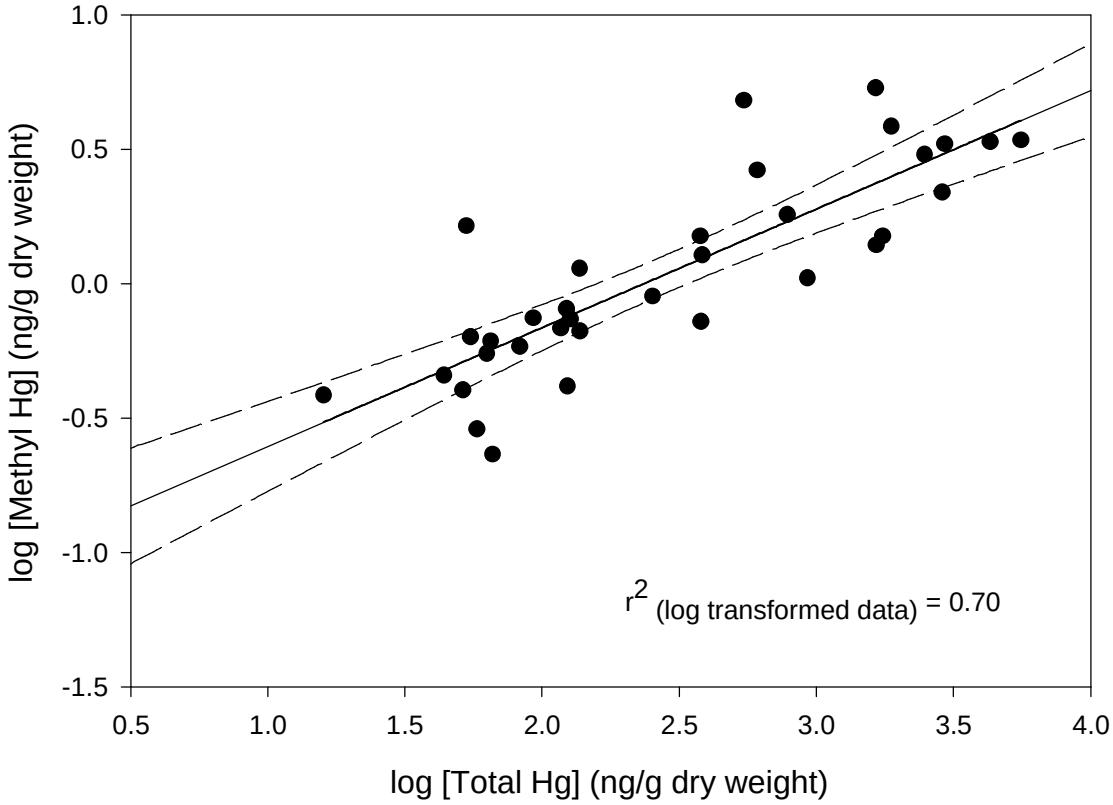


Figure 5. Relationship between methyl and total mercury in sediment. The 95% confidence interval for the regression equation is shown by the dashed lines.

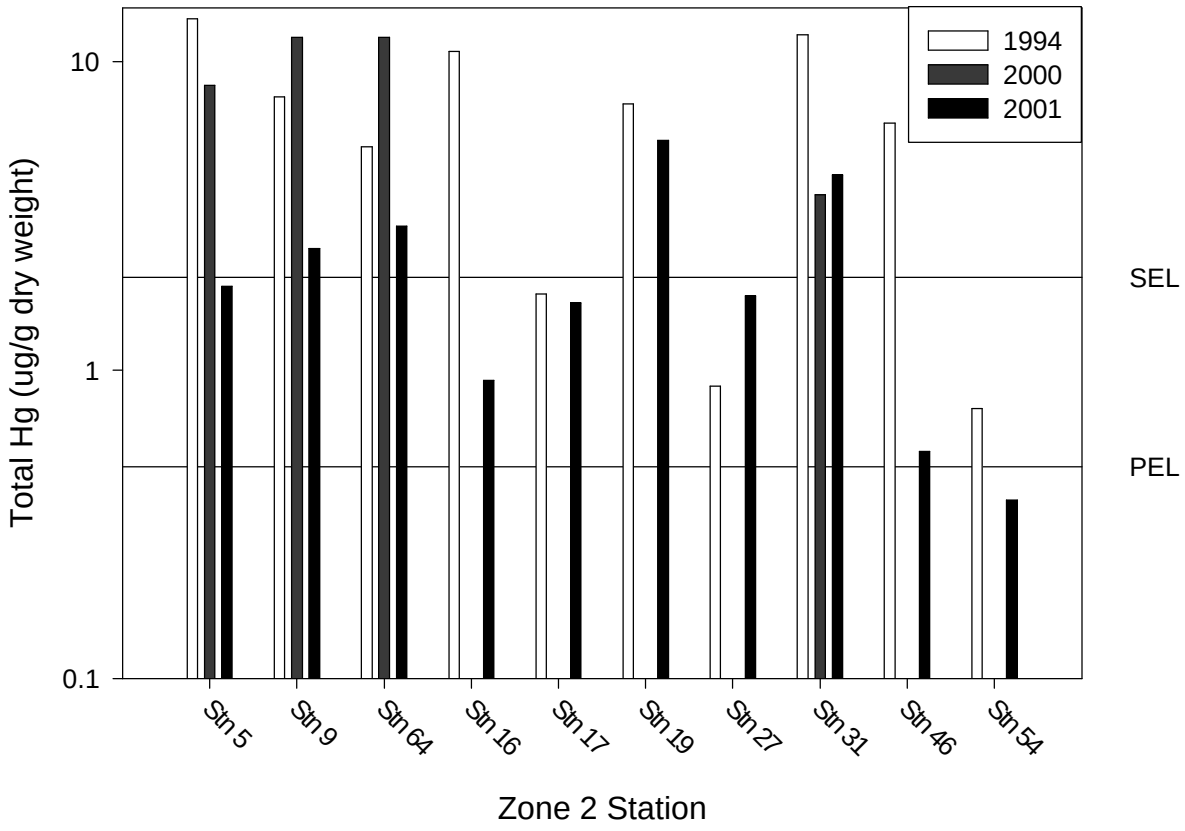


Figure 6. Comparison of 2001 mercury concentrations in sediment (0 to 10 cm depth) with historical data. Sediment quality guidelines for Ontario and Canada are shown. SEL = severe effect level (Persaud et al. 1993); PEL = probable effect level (CCME 1999b).

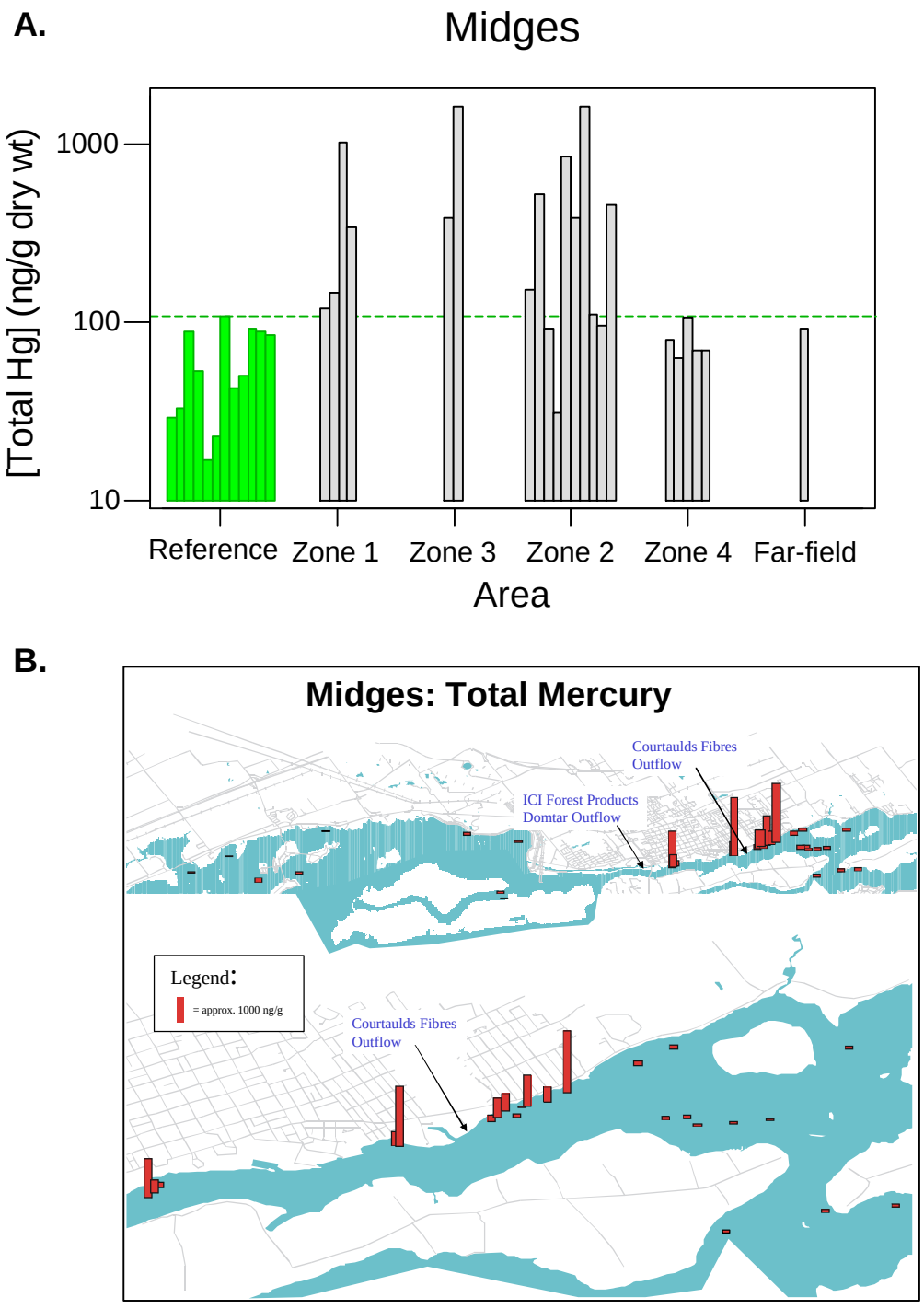


Figure 7. Total mercury in midges from the St. Lawrence River (Cornwall) Area of Concern. A. Exposed (grey bars) compared to reference (green bars) sites. Dashed line indicates maximum reference site concentration. B. Geographic pattern of mercury bioaccumulation in midges. The lower half of the map is an expanded view of the exposed areas shown in the upper half. Note: scaling of bars is arithmetic in B, logarithmic in A.

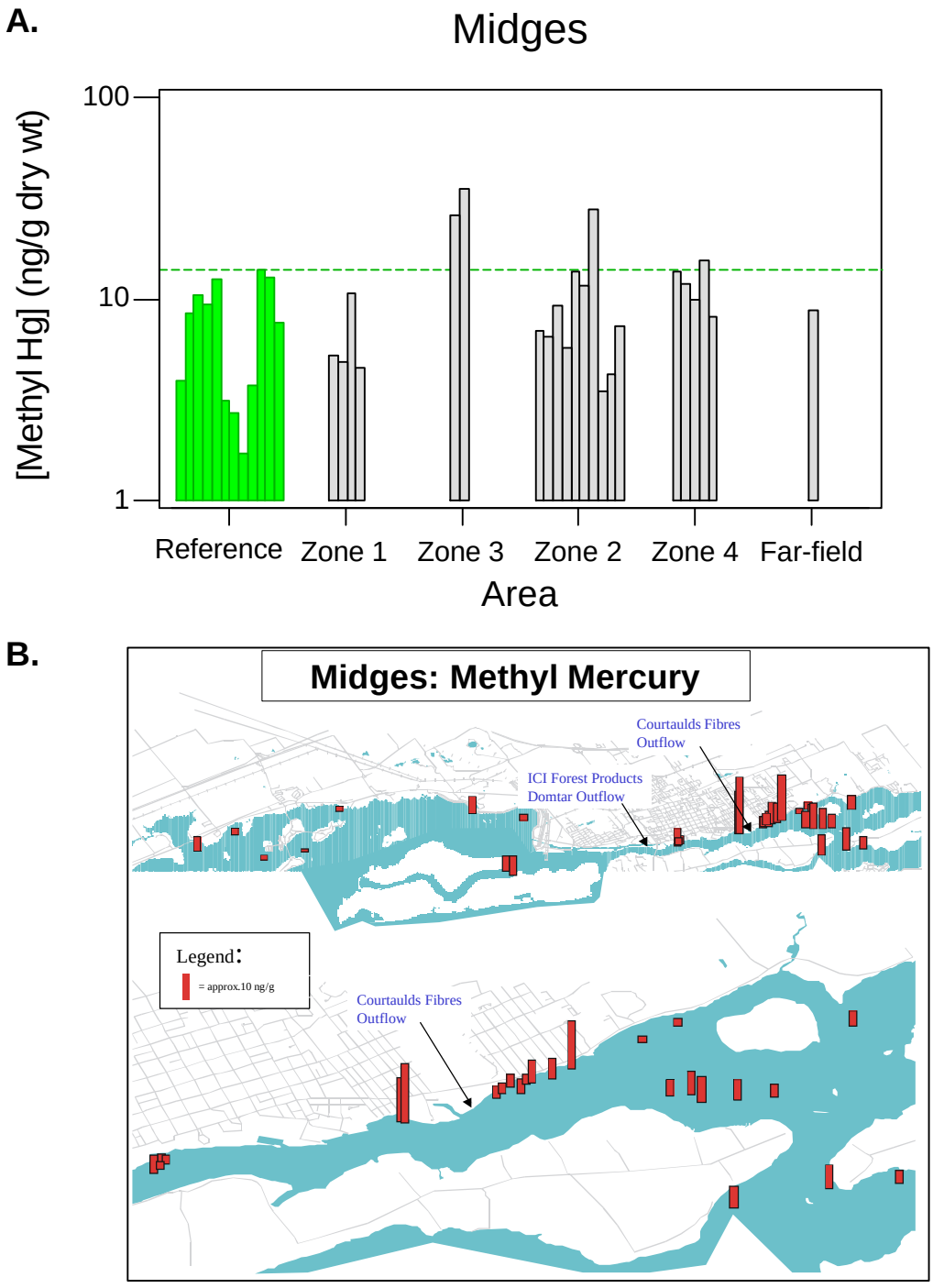


Figure 8. Methyl mercury in midges from the St. Lawrence River (Cornwall) Area of Concern. A. Exposed (grey bars) compared to reference (green bars) sites. Dashed line indicates maximum reference site concentration. B. Geographic pattern of mercury bioaccumulation in midges. The lower half of the map is an expanded view of the exposed areas shown in the upper half. Note: scaling of bars is arithmetic in B, logarithmic in A.

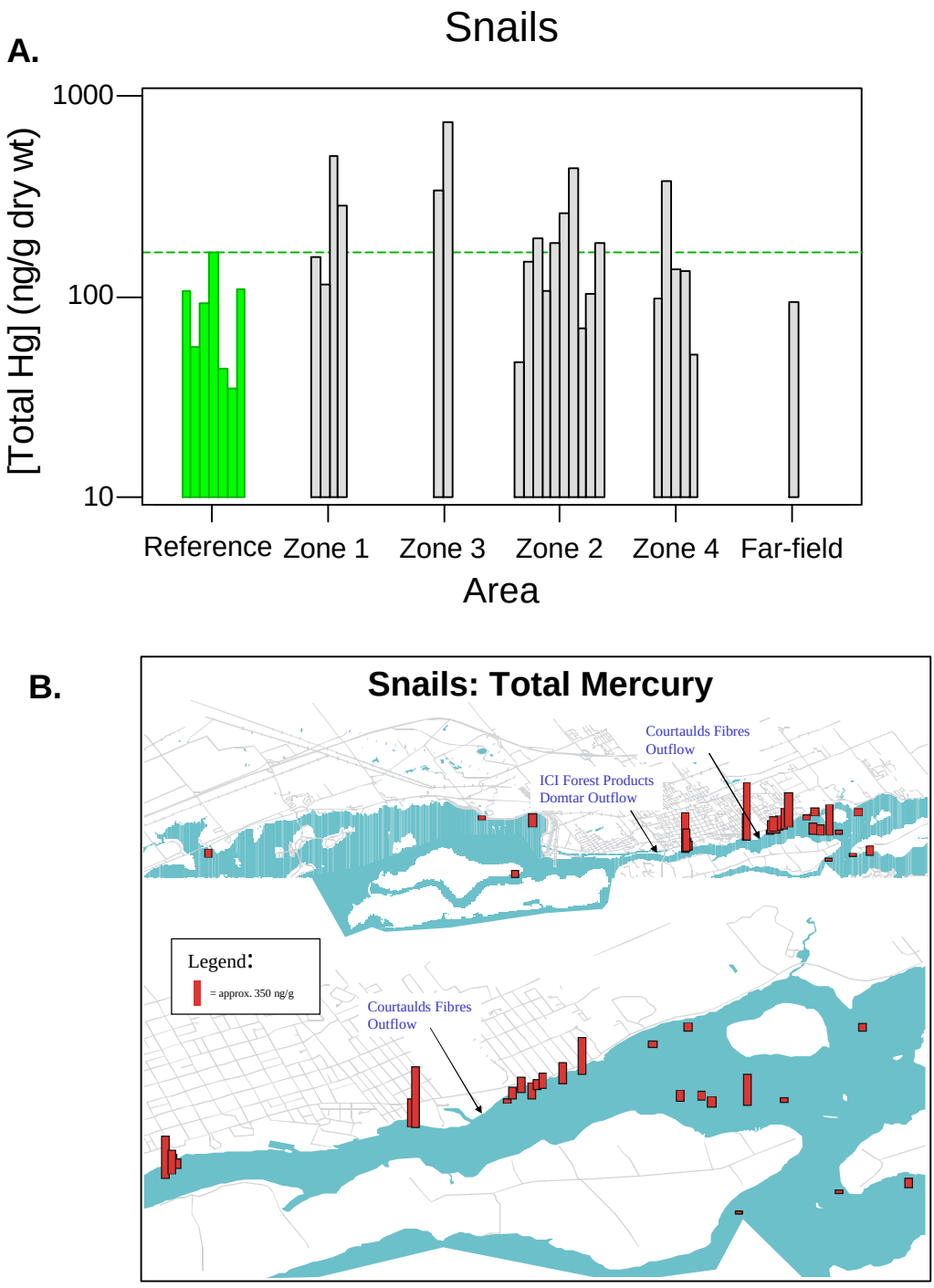


Figure 9. Total mercury in snails from the St. Lawrence River (Cornwall) Area of Concern. A. Exposed (grey bars) compared to reference (green bars) sites. Dashed line indicates maximum reference site concentration. B. Geographic pattern of mercury bioaccumulation in midges. The lower half of the map is an expanded view of the exposed areas shown in the upper half. Note: scaling of bars is arithmetic in B, logarithmic in A.

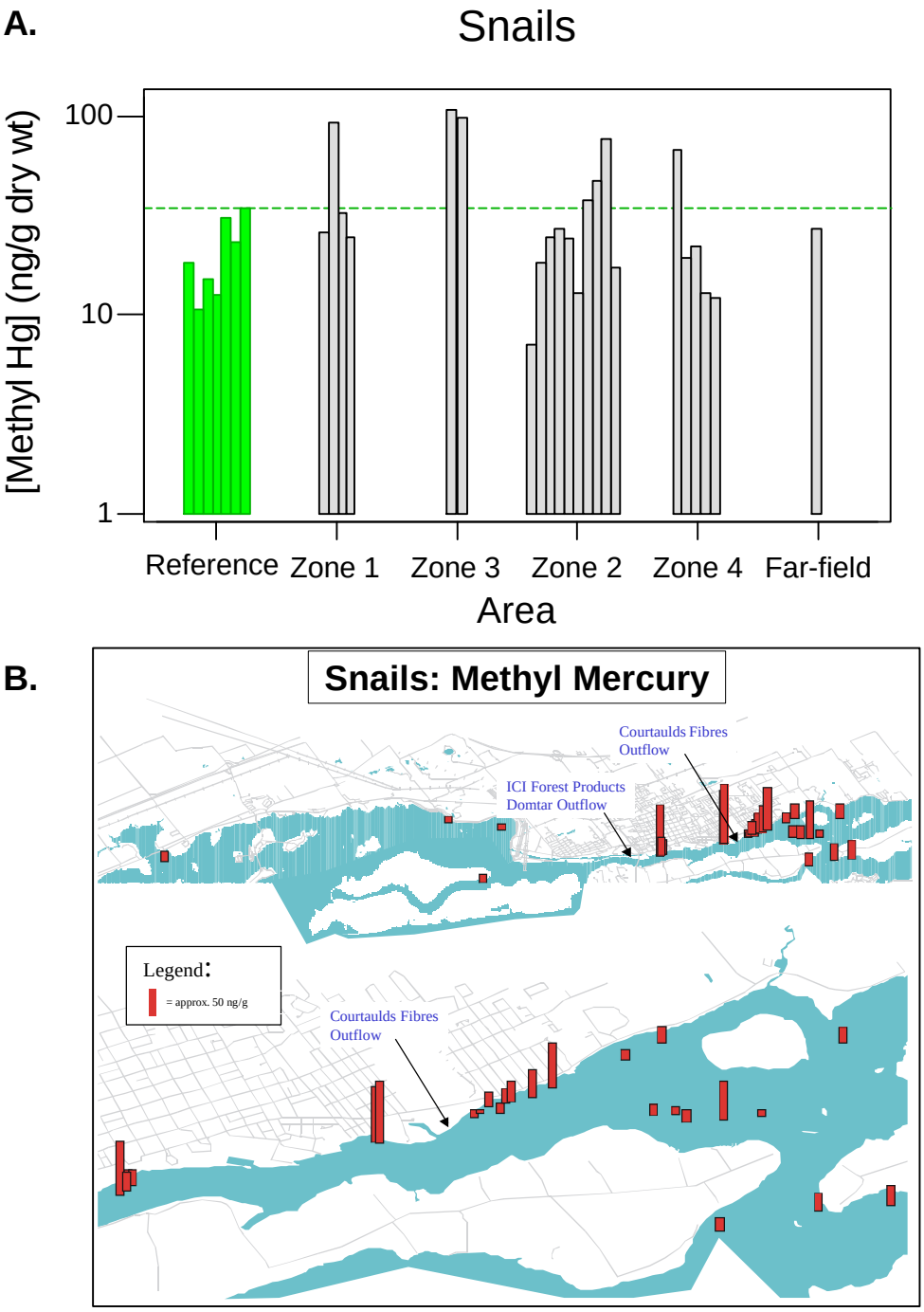


Figure 10. Methyl mercury in snails from the St. Lawrence River (Cornwall) Area of Concern. A. Exposed (grey bars) compared to reference (green bars) sites. Dashed line indicates maximum reference site concentration. B. Geographic pattern of mercury bioaccumulation in midges. The lower half of the map is an expanded view of the exposed areas shown in the upper half. Note: scaling of bars is arithmetic in B, logarithmic in A.

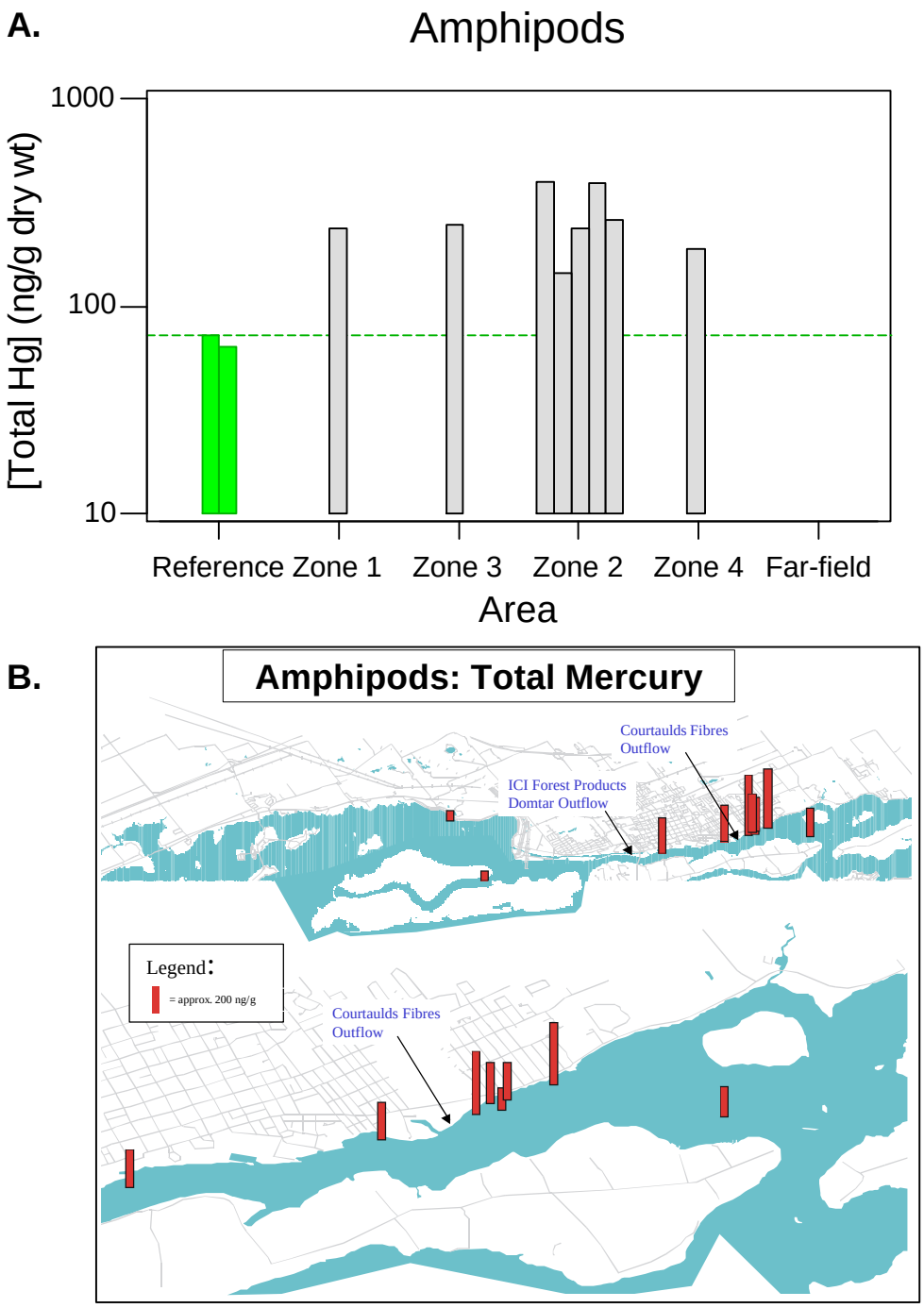


Figure 11. Total mercury in amphipods from the St. Lawrence River (Cornwall) Area of Concern. A. Exposed (grey bars) compared to reference (green bars) sites. Dashed line indicates maximum reference site concentration. B. Geographic pattern of mercury bioaccumulation in midges. The lower half of the map is an expanded view of the exposed areas shown in the upper half. Note: scaling of bars is arithmetic in B, logarithmic in A.

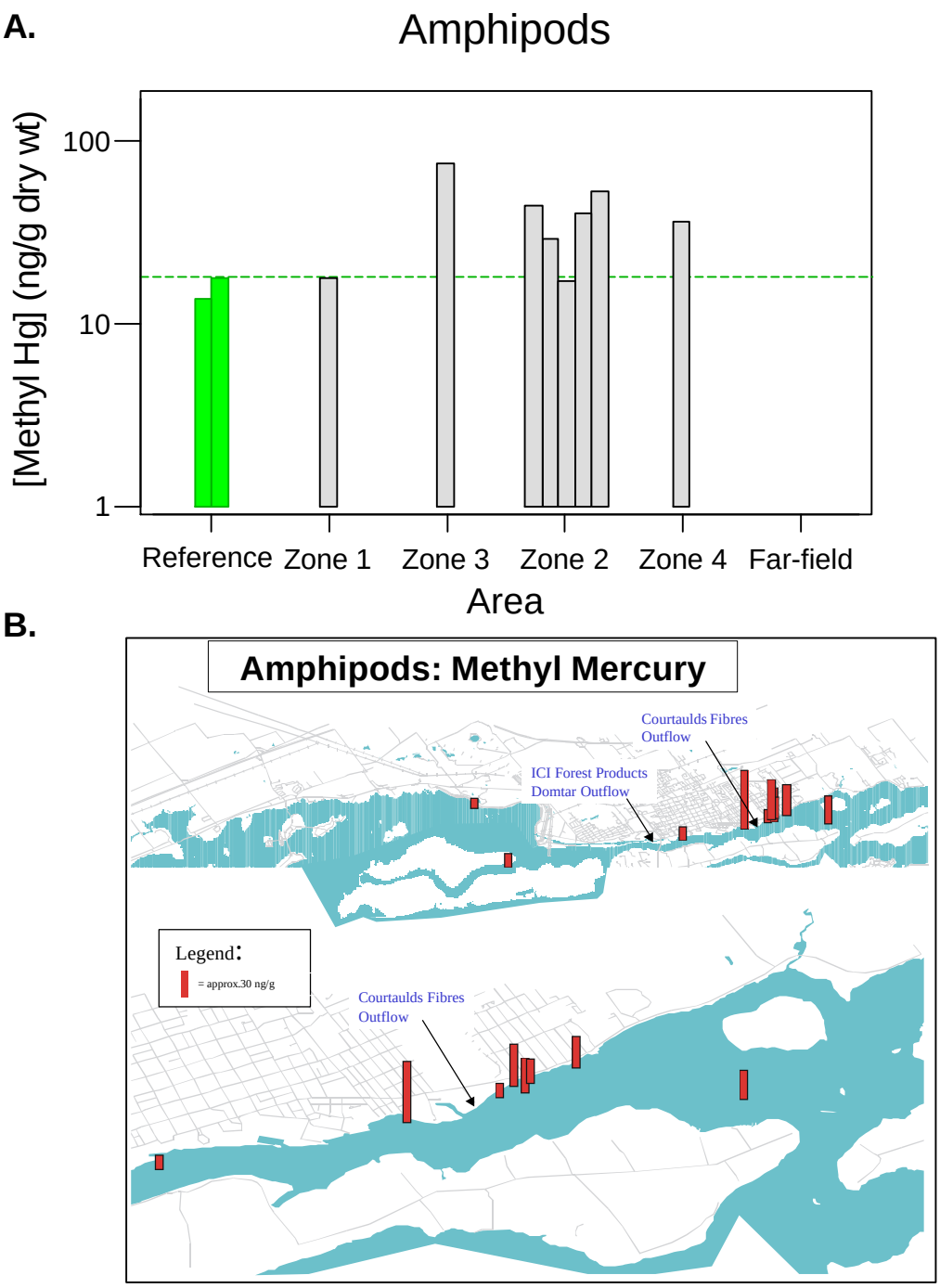


Figure 12. Methyl mercury in amphipods from the St. Lawrence River (Cornwall) Area of Concern. A. Exposed (grey bars) compared to reference (green bars) sites. Dashed line indicates maximum reference site concentration. B. Geographic pattern of mercury bioaccumulation in midges. The lower half of the map is an expanded view of the exposed areas shown in the upper half. Note: scaling of bars is arithmetic in B, logarithmic in A.

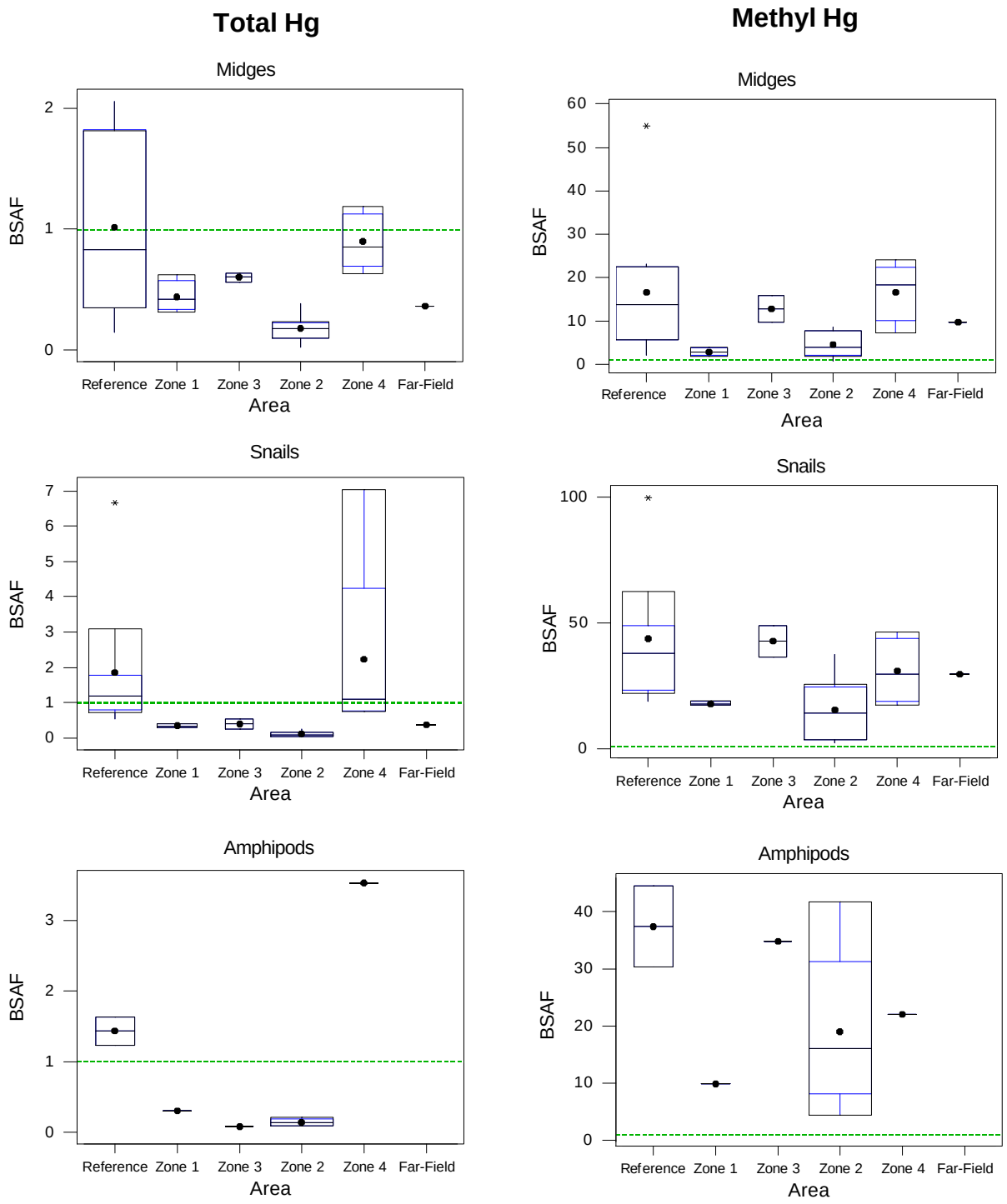


Figure 13. Biota-sediment accumulation factors (BSAFs) for invertebrate taxa from the St. Lawrence River (Cornwall) Area of Concern. Boxplots of BSAFs (=$[Hg]_{inv}$ / $[Hg]_{sed}$) for each taxon within areas show 95% Confidence Interval (CI) for median (outer, black box), inter-quartile ranges (inner, blue box [if distinct from CI box]), median (horizontal line within boxes) and mean (solid circle).

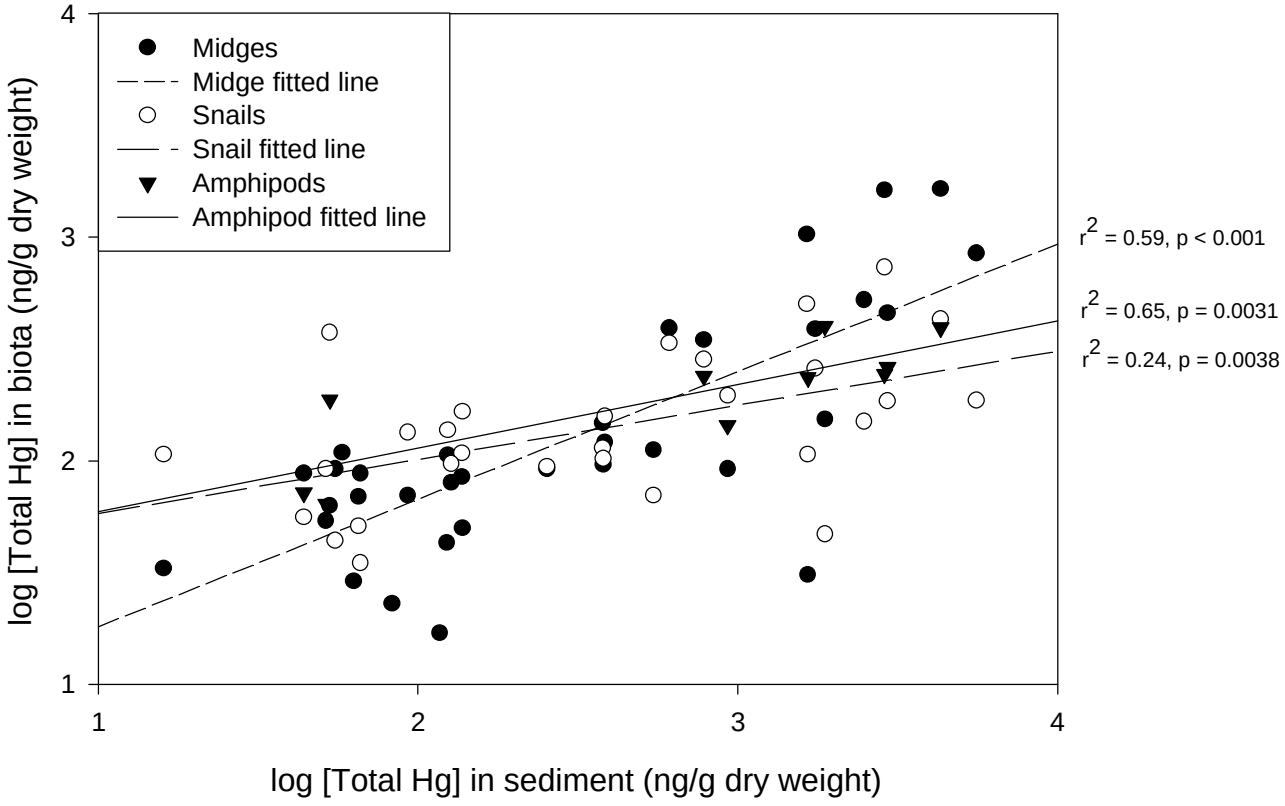


Figure 14. Relationships between total mercury in midges, snails and amphipods and total mercury in sediment. Separate regression lines are shown for each taxon.

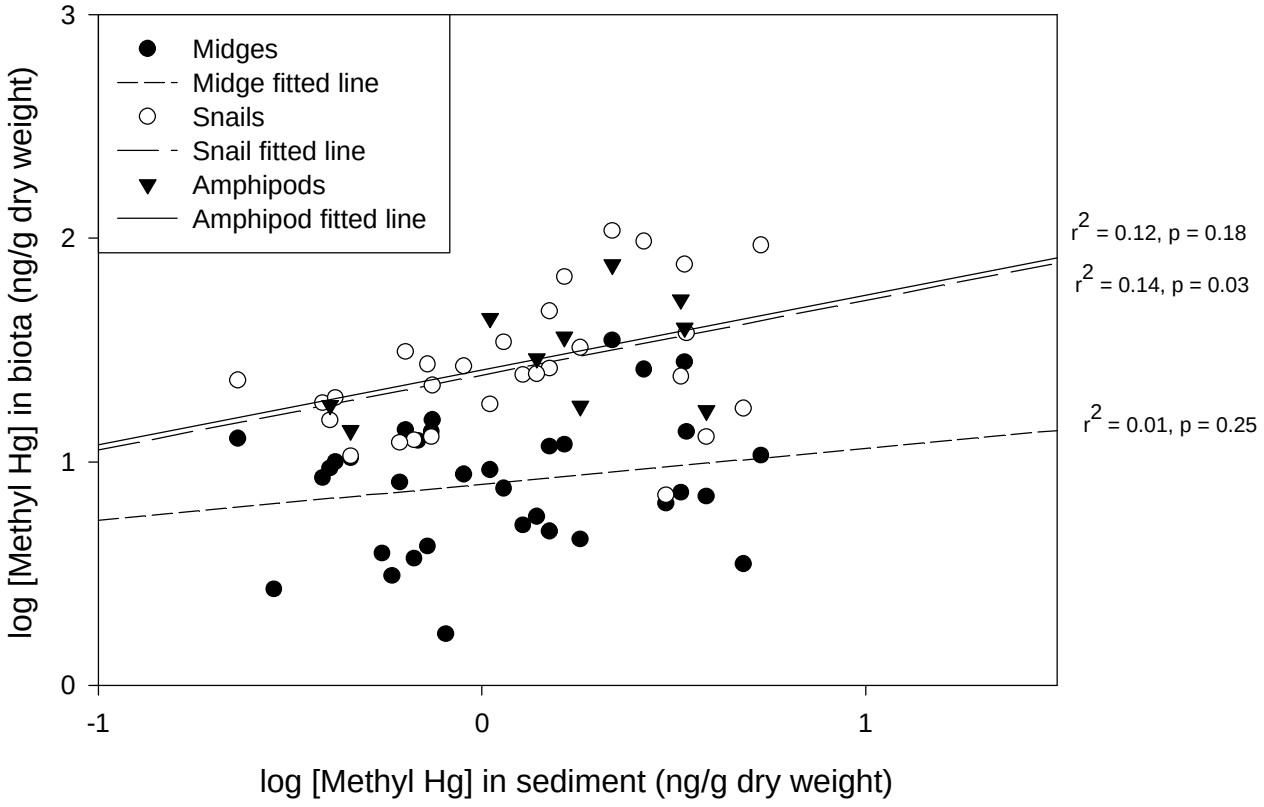


Figure 15. Relationships between methyl mercury in midges, snails and amphipods and methyl mercury in sediment. Separate regression lines are shown for each taxon.

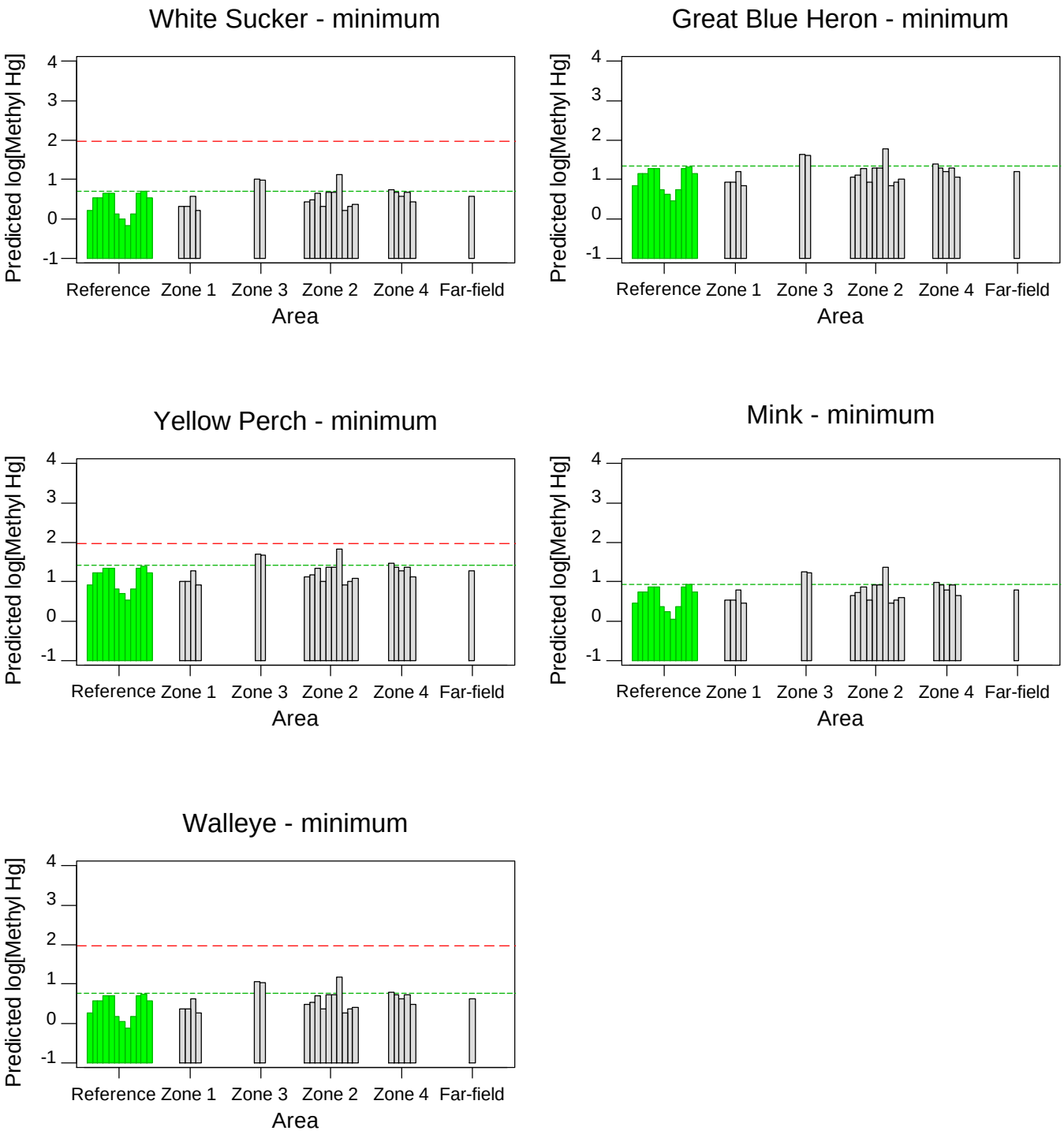


Figure 16. "Minimum" predictions of methyl mercury concentrations (ng/g wet weight) in five receptor species for St. Lawrence River (Cornwall) Area of Concern sites. These are from calculations using minimum $[MeHg]_{inv}$ and minimum biomagnification factors. Charts compare predicted [MeHg] among receptors and between reference (green bars) and exposed (gray bars) sites. Highest predicted [MeHg] for reference sites is indicated by a green dotted line. The tissue residue guideline (92 ng/g ww, CCME 2000), where applicable, is shown by a red dashed line.

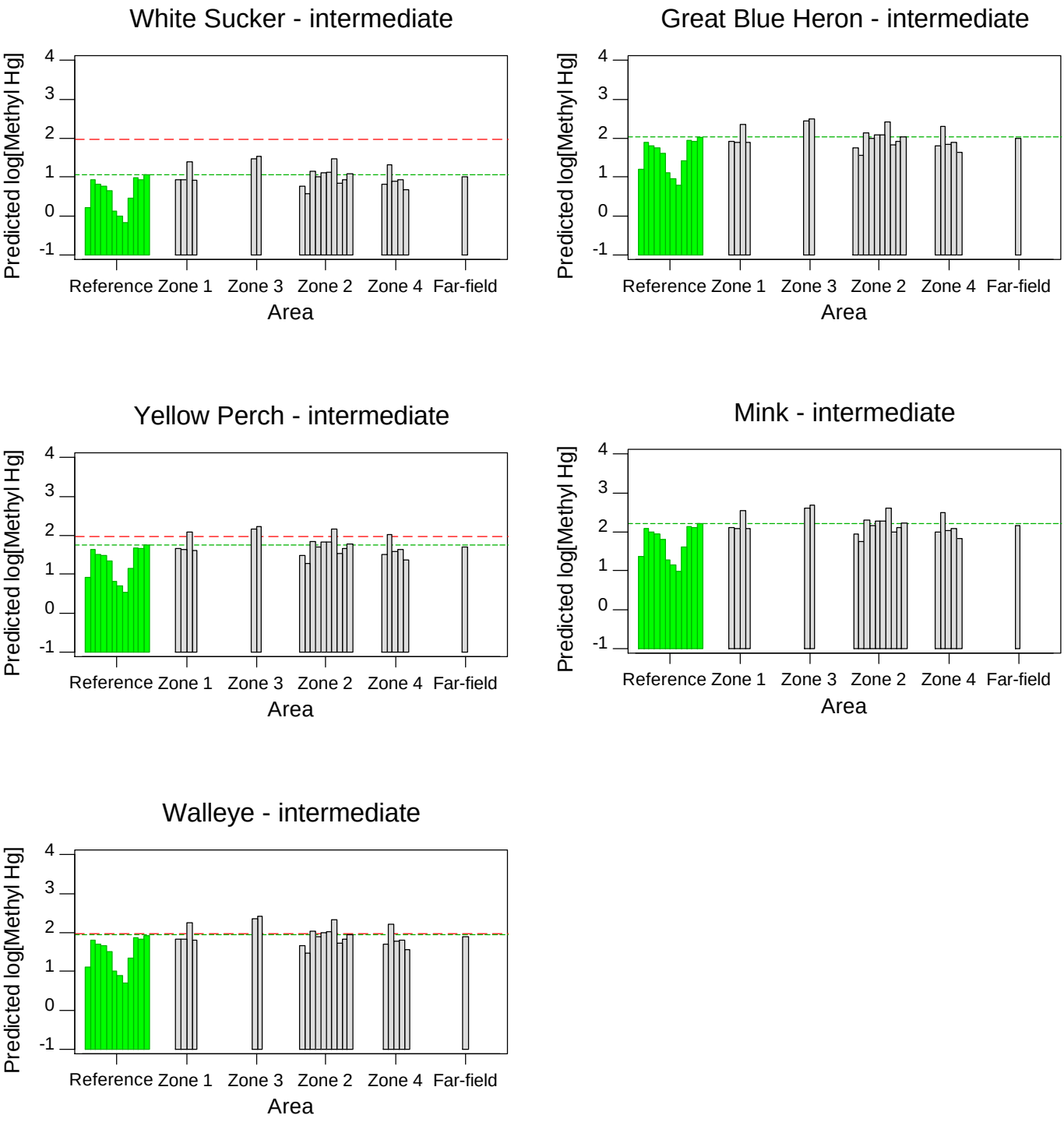


Figure 17. "Intermediate" predictions of methyl mercury concentrations (ng/g wet weight) in five receptor species for St. Lawrence River (Cornwall) Area of Concern sites. These are from calculations using mean $[MeHg]_{inv}$ and medium biomagnification factors. Charts compare predicted [MeHg] among receptors, and between reference (green bars) and exposed (gray bars) sites. Highest predicted [MeHg] for reference sites is indicated by a green dotted line. The tissue residue guideline (92 ng/g ww, CCME 2000), where applicable, is shown by a red dashed line.

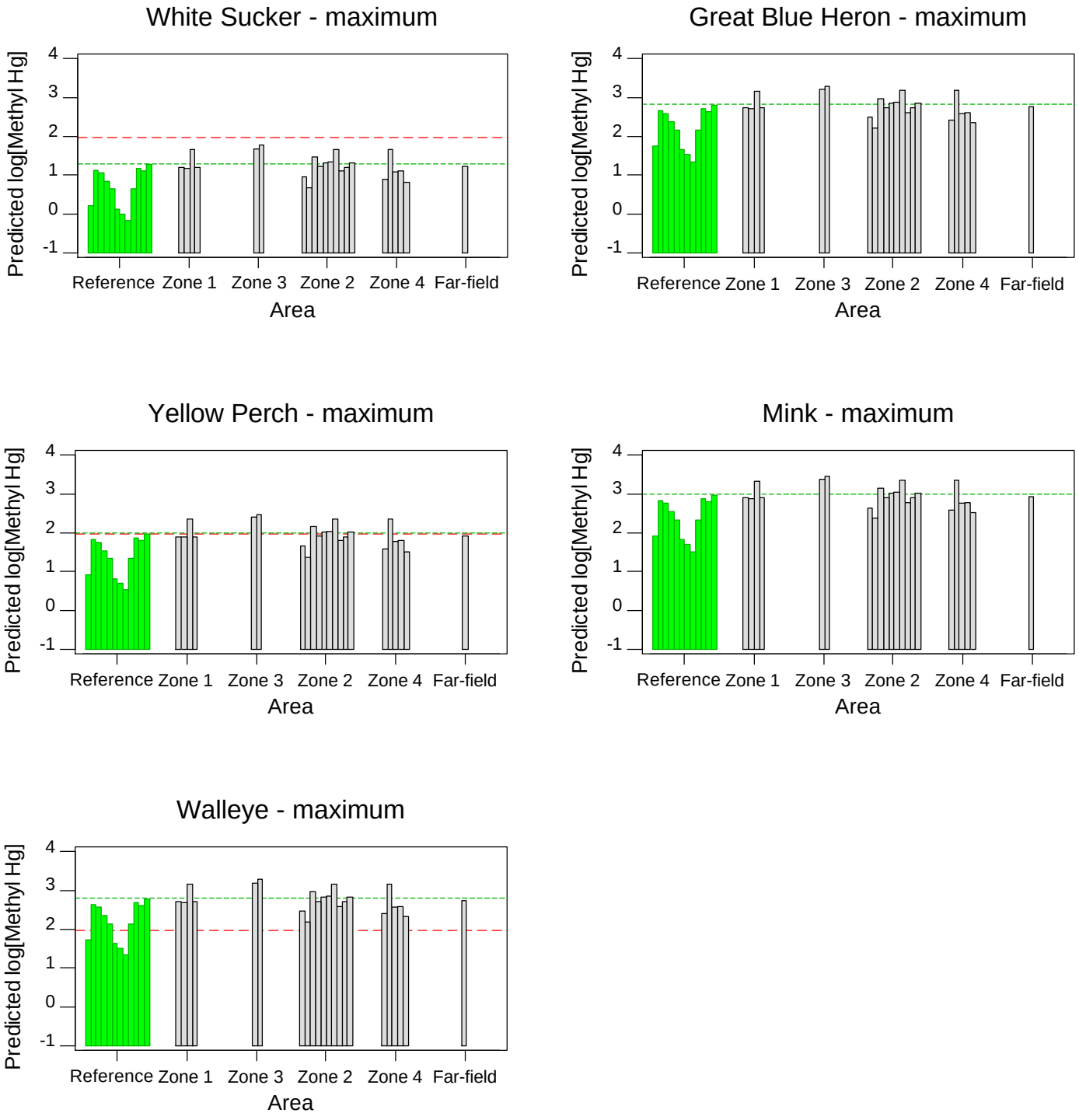
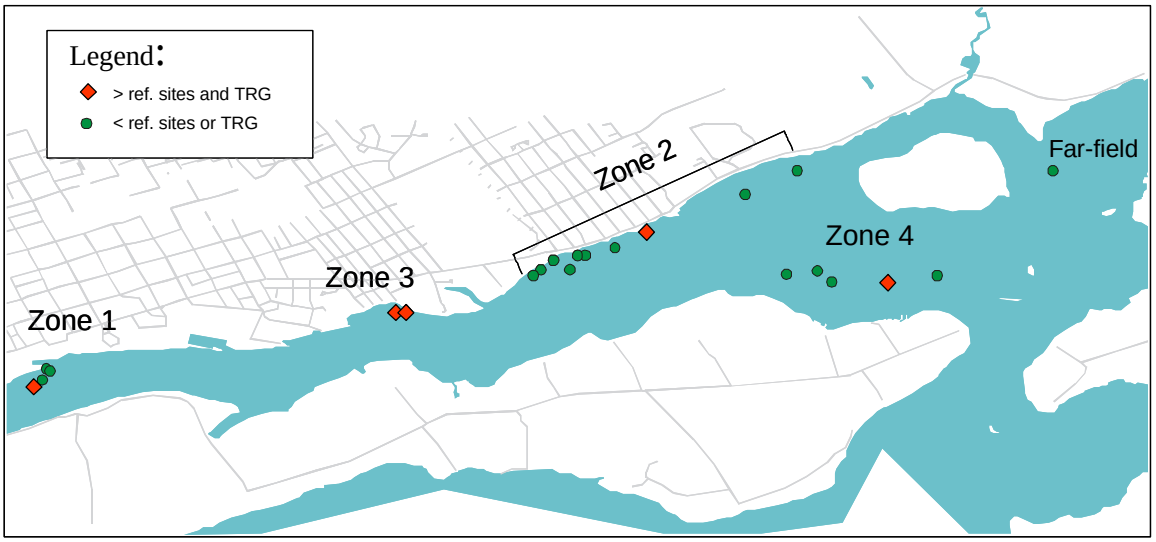


Figure 18. "Maximum" predictions of methyl mercury concentrations (ng/g wet weight) in five receptor species for St. Lawrence River (Cornwall) Area of Concern sites. These are from calculations using maximum $[MeHg]_{inv}$ and maximum biomagnification factors. Charts compare predicted [MeHg] among receptors, and between reference (green bars) and exposed (gray bars) sites. Highest predicted [MeHg] for reference sites is indicated by a green dotted line. The tissue residue guideline (92 ng/g ww, CCME 2000), where applicable, is shown by a red dashed line.

Yellow Perch: Intermediate Predicted [MeHg]



Yellow Perch: Maximum Predicted [MeHg]
Walleye: Intermediate and Maximum Predicted [MeHg]

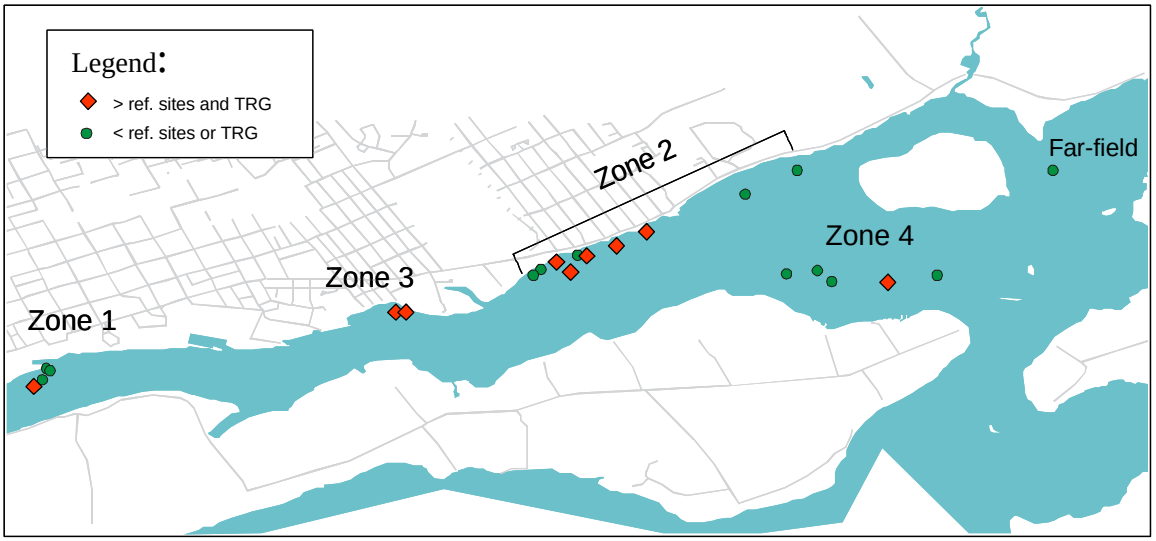


Figure 19. Geographic patterns of significantly high predicted receptor [MeHg] for exposed sites in the Cornwall AOC. Site symbol area and colour indicate relation to reference site predictions and the applicable tissue residue guideline: green solid circle (●) = $[MeHg]_{rec}$ less than ***either*** the maximum for the reference sites or the TRG; red solid diamond (◆) = $[MeHg]_{rec}$ greater than ***both*** the maximum for the reference sites and the TRG.

APPENDIX A. *Literature review of biomagnification factors (BMFs) for total and methyl mercury*

1.0 Introduction

This literature review was carried out to provide supporting information for the assessment of risk of biomagnification of mercury from contaminated sediments in Cornwall, Ontario. Biomagnification factors (BMFs), predator-prey factors (PPFs), and trophic transfer coefficients (TTCs) were obtained or derived from the literature for the calculation of total mercury and methylmercury concentrations in different trophic levels of a simple benthic freshwater food chain model (Figure A1).

1.1 Terminology

Biomagnification is the process at by which the chemical concentration in an organism exceeds that in the organism's diet, due to dietary absorption (Gobas and Morrison 2000). The biomagnification factor (BMF) is an empirically-derived measure of the rate of contaminant transfer between the organism's diet and the organism, and is expressed as the ratio of chemical concentration in the organism to the concentration in its' diet (Gobas and Morrison 2000). The synonymous terms predator-prey factor (PPF) and trophic transfer coefficient (TTC) are also found in the literature (USEPA 1997a; Suedel et al. 1994). A food chain multiplier (FCM) is used to quantify the increase in contaminant body burden through uptake from the food chain, but is defined as the factor by which a substance at higher trophic levels exceeds the bioconcentration factor (BCF) at trophic level 1 (NCASI 1999; USEPA 1997a). Therefore, it does not necessarily apply to a specific trophic transfer, and may be a multiple of more than one BMF. BMFs, TTCs, and PPFs are unitless, and the concentrations used to derive them are usually expressed in units of mass of chemical per kg of the organism, and mass of chemical per kg of food, respectively (Gobas and Morrison 2000). These concentrations can be expressed on a wet weight or dry weight basis (Gobas and Morrison 2000). BMFs, TTCs, and PPFs can be applied to specific trophic levels, as well as individual species in a food chain (USEPA 1997b). The term BMF will be used in this document in reference to biomagnification factors, predator-prey factors, and trophic transfer coefficients acquired from the literature.

2.0 Methods

2.1 Literature Search

The literature search was done using typical methods of electronic database and chain-of-citation searches as well as consultation with leading researchers in the field of mercury ecotoxicology and risk assessment. The following electronic databases were used to search primary literature, secondary literature, grey literature, and internet resources:

- ISI Current Contents Connect
- CSA Aquatic Sciences and Fisheries Abstracts (ASFA)
- CSA TOXLINE
- MEDLINE
- National Research Council of Canada (NRC) Research Press database
- US Environmental Protection Agency (USEPA)- various databases of government publications
- US Army Corp. of Engineers (USACE)- various databases of government publications
- Integrated Risk Information System (IRIS)
- Environmental Fate Database (EFDB)
- Oak Ridge National Laboratory (ORNL) publications

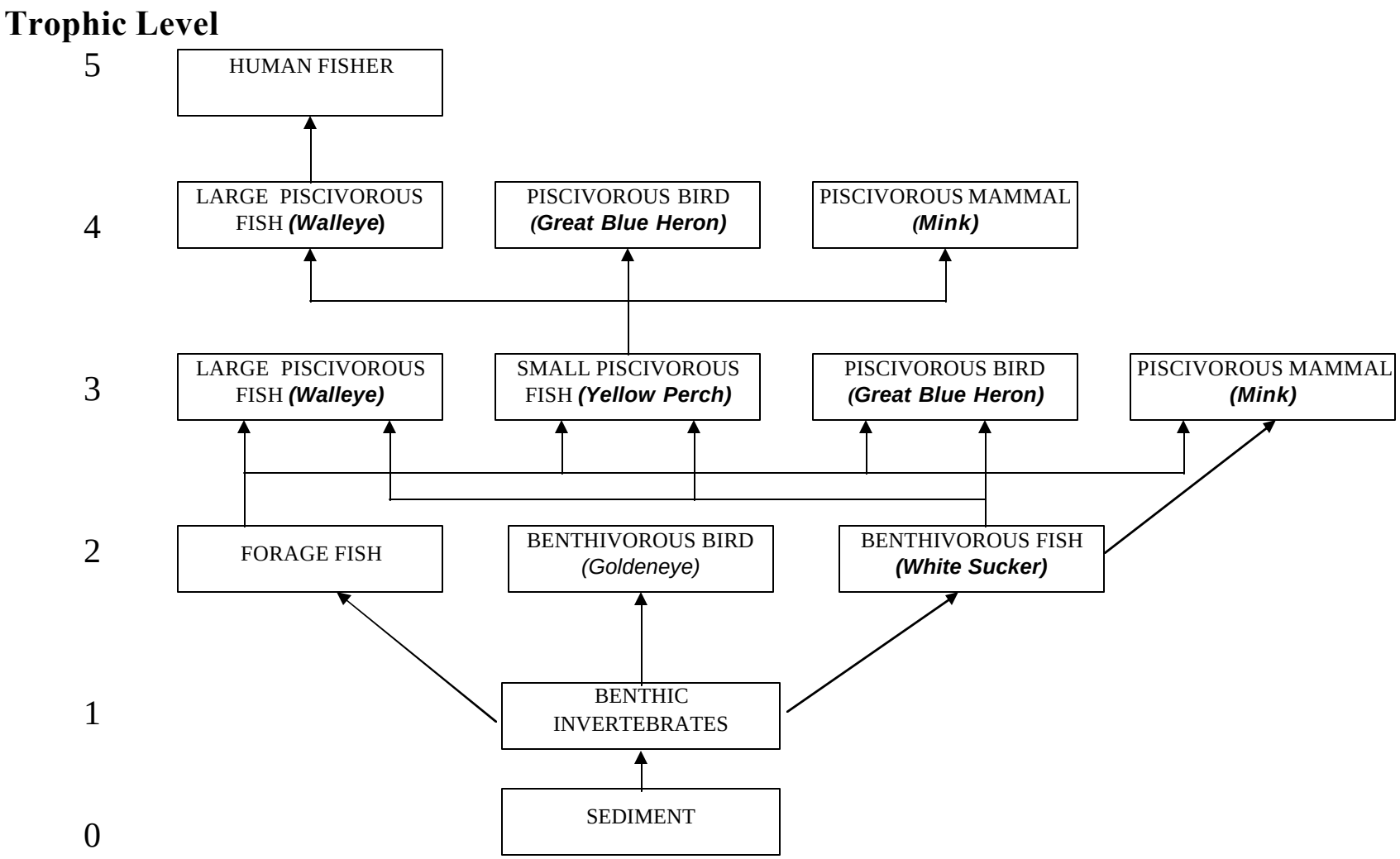


Figure A1: Generalized foodweb model for the assignment of trophic level to biomagnification factor estimates

In addition, the following journals were individually searched for recent and upcoming articles:

- *Archives of Environmental Contamination and Toxicology*
- *Archives of Environmental Health*
- *Bulletin of Environmental Contamination and Toxicology*
- *Canadian Journal of Fisheries and Aquatic Sciences*
- *Chemosphere*
- *Environmental Pollution*
- *Environmental Research*
- *Hydrobiologia*
- *Journal of Great Lakes Research*
- *Science of the Total Environment*
- *Water, Air, and Soil Pollution*
- *Water Research*

Several researchers active in mercury bioaccumulation studies were also contacted as part of the literature search.

The search was focused on the period 1996-2002, as a thorough review of the literature was carried out in a 1997 USEPA document entitled "Mercury Study Report to Congress" document (USEPA 1997a,b,c).

2.2 Assigning Trophic Levels to Receptor Species

Discrete trophic levels were applied using the food chain model (Figure A1). This was done to allow comparison of BMFs from different systems/foodwebs, as well as to conceptualize the transfer and magnification of mercury in the Cornwall scenario. However, the use of discrete trophic levels may lead to lower estimates of BMFs. An excellent discussion about the effects of omnivory on trophic position is found in Vander Zanden and Rasmussen (1996). In short, omnivory is common in aquatic communities (for example, up to 50% in pelagic food webs), and the use of discrete variables to represent trophic position will not adequately account for omnivory. When omnivory is integrated with the use of a continuous measurement of trophic position (ie- using stable isotope methods), estimates of BMFs will generally be higher for each discrete trophic level (Vander Zanden and Rasmussen 1996). Unfortunately, this literature survey did not yield any stable isotope studies on benthic freshwater food webs, and therefore system-specific BMFs based on continuous trophic position could not be obtained for lower trophic levels. Two such estimates for trophic levels 3 and 4 respectively, were obtained from pelagic foodweb studies.

2.3 Selecting Biomagnification Factor Estimates or Candidate Datasets from the Literature

The following criteria were applied to screen literature to obtain either BMFs or candidate datasets for calculating BMFs, after Suedel et al. (1994) and Gobas and Morrison (2000):

- If organisms that were presented were not from a logical food chain, or no evidence was presented that the feeding relationship between predator and prey was a functional feeding relationship, the paper was not used. One exception to this rule was made in selecting a study of mink fed diets of different proportions of contaminated and uncontaminated fish (Halbrook et al. 1997), since there was a reasonable likelihood that these fish species would have been part of their diet.
- Mean concentrations of total Hg or MeHg needed to be presented for both predator and prey, and in comparable units.
- Unless evidence of comparability could be found, studies from non-freshwater systems or with non-comparable species were not used. More information is presented below on the assessment of comparability of different systems and species.

2.4 Calculation of Biomagnification Factors from Candidate Datasets

Biomagnification factors were calculated from mean concentrations of total mercury and/or methylmercury from the literature using the equation (Gobas and Morrison 2000):

$$BMF = C_B/C_D$$

where:
C_B= mean contaminant concentration in the consumer (receptor) species
C_D= mean contaminant concentration in the diet of the organism

In all cases where BMFs were calculated from mean concentrations, the calculation was for the mean concentrations from two trophic levels with a functional feeding relationship, which was defined and demonstrated in the study. Where results were presented for a number of different locations (ie - several different lakes), BMFs were calculated for each location and then averaged, as opposed to averaging the mean concentrations from all locations to calculate a BMF. In three cases (Hughes et al. 1997; Neumann and Ward 1999; Suedel et al. 1994), a mean BMF was calculated by averaging several reported BMFs. Summaries of these calculations are presented in Tables A3 - A12.

2.5 Comparability of Species and Systems

There were very few studies that quoted BMF estimates for the receptor species and feeding relationships defined in Figure A1. Of the small number of studies which calculated BMFs that were directly comparable in part to the food chain model, most were from freshwater pelagic foodwebs. Some were also studies in quite different ecosystems (marine, temperate montane freshwater, tropic freshwater). Thus, it was important to document the relative comparability of different species and ecosystems to those presented in the study design for this assessment. Information to support substitutions of receptor species for comparable species from the literature (in applying BMF estimates) is presented in Tables A3 - A12. Species were considered the most qualitatively similar when they occupied similar habitats, had similar feeding habits and dietary composition, similar range, similar feeding substrate, and similar food ingestion:body weight ratio. Sources for this information were CCME (1999a,b), CWS (2002), Sample and Suter (1999), Scott and Crossman (1973), and USEPA (1997c).

Applying BMFs calculated from one system to another is controversial, since rates of trophic transfer of mercury are thought to vary due to abiotic and biotic factors (USEPA 2001). The USEPA, in developing national bioaccumulation factors to assess the risk to human health of mercury exposure, indicated that these factors are poorly understood and are likely to be system and site-specific (USEPA 1997b; USEPA 2001). Abiotic factors which may influence the chemistry of mercury include pH, temperature, and dissolved organic carbon in the waterbody, and these are usually determined by watershed characteristics which in turn affect inputs, bioavailability, speciation, and methylation of mercury in the sediments and water column (Downs et al. 1998; Greenfield et al. 2001; Meyer 1998; Mason et al. 2000; USEPA 2001; Watras et al. 1998). Biotic factors include food chain length, horizontal food web structure, feeding mechanisms of organisms at lower trophic levels, and the age/size/weight or metabolic rates of individuals in the sample used to calculate a given BMF (Environment Canada 1997; Power et al. 2002; USEPA 2000). However, no single factor has been correlated with extent of bioaccumulation in all cases examined (USEPA 2001).

It was also suggested (as discussed above) that much of the uncertainty around applying BMFs from different systems may be due to an oversimplification of predator-prey relationships by using discrete trophic levels (Vander Zanden and Rasmussen 1996). One stable isotope study was found from Papua, New Guinea whose results indicated similar magnitude of biomagnification to temperate and arctic foodwebs (Bowles et al. 2001). Another stable isotope study from an arctic foodweb indicated that age did not affect bioaccumulation of mercury in the muscle of ringed seals or clams (Atwell et al. 1998). A third from a subarctic lake found a higher rate of biomagnification (BMF=5.4 versus 3.0) than for a comparable freshwater temperate system (Power et al. 2002).

Unless the relative comparability to temperate freshwater systems was demonstrated, studies from marine, arctic marine, and tropic freshwater were not used to select or derive BMFs.

3.0 Results

A total of 80 references were examined in detail to yield BMFs, datasets to calculate BMFs, or to provide supporting information in applying BMFs. Results are broken down as follows:

- Primary literature - 61 references

- Secondary literature - 5 references
- Grey literature - 14 references

Of those 80, only 11 yielded appropriate BMFs or datasets, following guidelines set out in section 2 above. However, a number of the references (Cantox Environmental Inc. 2001; Suedel et al. 1994; USEPA 1997a) were reviews that synthesized BMFs from several sources. Along with BMF estimates, the following supporting information was gathered:

- Range, standard deviation, or standard error of BMF estimates
- Trophic level of predator/receptor
- Type of study (field, laboratory, modeling, review)
- Prey species
- Predator species
- Mercury parameter (total Hg or MeHg)
- Scope of study (ie - number of lakes sampled)
- Location of study
- Biological medium sampled
- Relative age/size of organisms sampled
- Reference from which BMF or dataset came from
- Comments

These results are reported in Table A2.

A breakdown of the number of BMFs obtained/calculated per feeding relationship, and the range of corresponding BMF values is presented in Table A1.

Table A1- Breakdown of results of literature review for each hypothetical feeding relationship

Feeding Relationship	Trophic levels of transfer	# of Estimates	Total and Methyl Hg BMFs			Comments
			Low	Medium *	High	
Benthic invertebrates to forage or benthivorous fish	1 - 2	1	3.43	3.43	3.43	High BMF calculated from benthos [THg] values which are below DL excluded.
Benthivorous or forage fish to small piscivorous fish	2 - 3	1	5	5	5	
Benthivorous or forage fish to large piscivorous fish	2 - 3	8	1.12	3.20	32.4	
Benthivorous or forage fish to piscivorous bird	2 - 3	1	6.80	6.80	6.80	High THg value from heron with ambiguous feeding relationship dropped.
Benthivorous or forage fish to piscivorous mammal	2 - 3	10	1.70	5.20	22.64	High THg value from fur/hair excluded. Hg form given as total and methyl for most values.
Small piscivorous fish to large piscivorous fish	3 - 4	1	2.40	2.40	2.40	
Small piscivorous fish to piscivorous bird	3 - 4	6	0.85	2.37	6.80	High THg values from plumage excluded.
Small piscivorous fish to piscivorous mammal	3 - 4	9	1.70	4.7	10.00	Hg form given as total and methyl for most values.

* "Medium" = datum if n = 1, median if n > 2

Table A2- Summary of Literature-Derived Biomagnification Factors by Trophic Level

Value	Range	Trophic Level	Type of Study	Prey Species	Predator Species	Hg Parameter	Scope	Location	Sample Medium	Age/Size of Sample	Reference	Comments
2.14	0.3-7	2	Review	"Primary consumers" (aquatic)	"Secondary consumers" (aquatic)	Total Hg					Suedel et al., 1994	Values reported as TTCs
3.43	0.5-10.5	2	Review	"Primary consumers" (aquatic)	"Secondary consumers" (aquatic)	MeHg					Suedel et al., 1994	Values reported as TTCs
17.13	Not calculated	2	Field	Benthos	Carp and bullhead	Total Hg	One estuary	Old Woman Creek, Lake Erie	Skinless fillets (carp), whole body (bullhead)	>30 cm in length	Francis et al., 1998	BMFs calculated from mean concentrations and feeding relationships reported in paper.
1.12	0.2-1.9	3	Review	"Secondary consumers" (aquatic)	"Top predators" (aquatic)	Total Hg					Suedel et al., 1994	Values reported as TTCs
1.51	Not calculated	3	Field	Lake chubsucker	Redfin pickerel	Total Hg	Nine wetlands	Savannah River Site, South Carolina	Whole body	Chubsucker mean length/weight=79 mm/4g Pickerel mean length/weight= 106 mm/3g	Snodgrass et al., 2000	Mean BMF calculated from individual wetland BMFs, which were calculated from geometric mean concentrations in each species for each wetland. Feeding relationship implied by results cited from other studies from that area.
1.55	1.2-1.8	3	Field	Groove-snouted catfish (omnivore) and seven-spotted archerfish (insectivore)	Barramundi, giant freshwater anchovy, Sepik garpike	MeHg	One lake	Papua, New Guinea	Whole body		Bowles et al., 2001	Stable isotope (δ15N) study. Results suggest that the biomagnification power of the food web is similar to that of temperate-lake and arctic-marine systems. Range of BMFs based on BMFs calculated from +/- 1 SD from mean MeHg concentrations.
1.70	Not reported	3	Review	Only reported as "concentration of MeHg in diet"	Otter	Total Hg and MeHg	Pooled results of twelve studies.	Ontario (3 studies), Georgia (3), Louisiana (1), Manitoba (2), Wisconsin (2), Norway (1)	Muscle	Not reported	Cantox Environmental Inc., 2001	Sampling details from Wren et al., 1986. BMF calculated by Cantox Environmental Inc.
2.40	1-4	3	Field	Bluegill, black crappie, yellow perch	Chain pickerel, largemouth bass	Total Hg	Two lakes	Connecticut	Axial muscle (whole fillets)	Fish aged 2-5 years	Neumann and Ward, 1999	
2.70	Not reported	3	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Georgia	Muscle	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
3.00	Not reported	3	Review	Only reported as "concentration of MeHg in diet"	Otter	Total Hg and MeHg	One lake, N=20 for fish sample, N=4 for otter sample	Tadenac Lake, Muskoka, Ontario	Muscle	Not reported	Cantox Environmental Inc., 2001	Sampling details from Wren et al., 1983. BMF calculated by Cantox Environmental Inc.
3.40	Not reported	3	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Not reported	Liver	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
4.00	Not reported	3	Modelling	Pelagic forage fish (smelt, ciscoes, coregonids, alewife, ninespine stickleback)	Lake trout	Total Hg	96 lakes, >10 individuals/species, period 1975-84 (source= MOE sportfish contaminants monitoring)	Canadian Shield lakes, Ontario	Whole skinless fillets (smaller fish), axial muscle (larger fish)	Pooled results	Vander Zanden and Rasmussen, 1996	BMF corrected by authors for omnivory from original value of 2.0 defined by Cabana et al., 1994. Correction based on results of δ15N stable isotope study of trophic position and effects of omnivory on trophic position. Sampling details from Cabana et al
4.70	Not reported	3	Review	Only reported as "concentration of MeHg in diet"	Otter	Total Hg and MeHg	Pooled results of twelve studies.	Ontario (3 studies), Georgia (3), Louisiana (1), Manitoba (2), Wisconsin (2), Norway (1)	Liver	Not reported	Cantox Environmental Inc., 2001	Sampling details from Wren et al., 1986. BMF calculated by Cantox Environmental Inc.
5.00	Geometric SD=1.47	3	Review	"Forage fish"	"Piscivorous fish"	MeHg	14 studies	Michigan (2 studies), Ontario (5), Manitoba (1), Wisconsin (1), New York (1), Norway (1), Sweden (2), Brazil (1)	Various	Various	USEPA, 1997	BMF is geometric mean of values from literature review. Selected values from the literature used in the calculation of the average BMF are presented in attached "USEPA, 1997" worksheet.
5.40	Not reported	3	Field	Forage fish (burbot, cisco, northern lake chub, round whitefish, threespine stickleback) and benthivores (longnose sucker, slimy sculpin)	Lake trout	Total Hg	One lake	Stewart Lake, northern Labrador	Dorsal muscle	All age classes	Power et al., 2002	BMF reported in study. Stable isotope study of a subarctic freshwater lacustrine system.
5.70	Not reported	3	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Georgia	Liver	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
6.80	Not reported	3	Review	Bluntnose minnow, rainbow smelt	Common loon	Total Hg	One lake, N=20 for fish sample, N=1 for loon sample	Tadenac Lake, Muskoka, Ontario	Whole skinless fillet (fish), breast muscle (birds)	Pooled sample of fish from beach seining (fish). Loon= 5 kg	Cantox Environmental Inc., 2001	Sampling details from Wren et al., 1983. BMF calculated by Cantox Environmental Inc.
10.00	Not reported	3	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Not reported	Not reported	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
10.00	Not reported	3	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Not reported	Liver	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
14.29	Not calculated	3	Field	Carp, bullhead, catfish (<30 cm length)	Bowfin, catfish (>30 cm length)	Total Hg	One estuary	Old Woman Creek, Lake Erie	Skinless fillets (carp, bowfin, catfish), whole body (bullhead)	Piscivores= >30 cm in length, benthivores= <30 cm length	Francis et al., 1998	BMFs calculated from mean concentrations and feeding relationships reported in paper.
32.40	0.1-141	3	Review	"Secondary consumers" (aquatic)	"Top predators" (aquatic)	MeHg					Suedel et al., 1994	Values reported as TTCs
85.56	Not calculated	3	Field	Carp, bullhead, catfish (<30 cm length); gizzard shad, black crappie	Great blue heron	Total Hg	One estuary	Old Woman Creek, Lake Erie	Skinless fillets (carp, catfish, crappie), whole body (bullhead, gizzard shad)	Benthivores= <30 cm length, heron (N=1) size not reported	Francis et al., 1998	BMFs calculated from mean concentrations and feeding relationships reported in paper.
87.81	82-96	3	Field	Freshwater and intertidal fishes	Otter	Total Hg	One coastal creek and estuary (N= 32 otters)	Prince William Sound, Alaska	Fur	Juveniles to old adults (four age categories)	Ben-David et al., 2001	BMF calculated from mean concentrations and standard errors presented in paper. The feeding relationship with freshwater fishes was supported by stable isotope measurements.
Liver- 13.92 Kidney- 22.64 Hair- 108.23	Liver- 12-17 Kidney- 20-25 Hair- 87-149	3	Controlled field	Benthivores	American mink	Total Hg	50 female farmed mink	Oak Ridge National Laboratory, Tennessee	Liver, kidney, and fur	Female adults	Halbrook et al, 1997	BMFs calculated from mean concentrations in different tissues and different specific dietary mixes of contaminated and uncontaminated fish.

Table A2- Summary of Literature-Derived Biomagnification Factors by Trophic Level (continued)

Value	Range	Trophic Level	Type of Study	Prey Species	Predator Species	Hg Parameter	Scope	Location	Sample Medium	Age/Size of Sample	Reference	Comments
1.70	Not reported	4	Review	Only reported as "concentration of MeHg in diet"	Otter	Total Hg and MeHg	Pooled results of twelve studies.	Ontario (3 studies), Georgia (3), Louisiana (1), Manitoba (2), Wisconsin (2), Norway (1)	Muscle	Not reported	Cantox Environmental Inc., 2001	Sampling details from Wren et al., 1986. BMF calculated by Cantox Environmental Inc.
1.93	1-4	4	Field	Yellow perch	Osprey	Total Hg (osprey), MeHg (yellow perch)	Five osprey nesting areas	St. Mary's R., Georgian Bay, Kawartha Lakes, New Jersey	Eggs	Freshly laid and addled eggs	Hughes, 1997	
2.40	1-4	3	Field	Bluegill, black crappie, yellow perch	Chain pickerel, largemouth bass	Total Hg	Two lakes	Connecticut	Axial muscle (whole fillets)	Fish aged 2-5 years	Neumann and Ward, 1999	
2.70	Not reported	4	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Georgia	Muscle	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
3.00	Not reported	4	Review	Only reported as "concentration of MeHg in diet"	Otter	Total Hg and MeHg	One lake, N=20 for fish sample, N=4 for otter sample	Tadenac Lake, Muskoka, Ontario	Muscle	Not reported	Cantox Environmental Inc., 2001	Sampling details from Wren et al., 1983. BMF calculated by Cantox Environmental Inc.
3.40	Not reported	4	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Not reported	Liver	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
4.70	Not reported	4	Review	Only reported as "concentration of MeHg in diet"	Otter	Total Hg and MeHg	Pooled results of twelve studies.	Ontario (3 studies), Georgia (3), Louisiana (1), Manitoba (2), Wisconsin (2), Norway (1)	Liver	Not reported	Cantox Environmental Inc., 2001	Sampling details from Wren et al., 1986. BMF calculated by Cantox Environmental Inc.
5.70	Not reported	4	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Georgia	Liver	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
6.80	Not reported	4	Review	Smallmouth bass, northern pike, lake trout	Common loon	Total Hg	One lake, N=20 for fish sample, N=1 for loon sample	Tadenac Lake, Muskoka, Ontario	Dorso-lateral muscle (fish), breast muscle (birds)	Pooled sample of fish from gill netting (fish). Loon= 5 kg	Cantox Environmental Inc., 2001	Sampling details from Wren et al., 1983. BMF calculated by Cantox Environmental Inc.
10.00	Not reported	4	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Not reported	Not reported	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
10.00	Not reported	4	Review	Fish (species not reported)	Otter	Total Hg and MeHg	Not reported	Not reported	Liver	Not reported	Cantox Environmental Inc., 2001	BMF calculated by Cantox Environmental Inc.
10.00		4	Review	Predatory fishes	American mink	MeHg	Not reported	Not reported	Not reported	Not reported	USEPA, 2000	
14.50	12-16	4	Field	Yellow perch	Osprey	Total Hg (osprey), MeHg (yellow perch)	Five osprey nesting areas	St. Mary's R., Georgian Bay, Kawartha Lakes, New Jersey	Feathers- wing/mantle/tail	Pooled sample from chicks and adults	Hughes, 1997	
Liver-2.81 Kidney- 3.81 Brain- 0.85 Muscle- 1.40 Feathers- 55.7	Not calculated	4	Field	Northern pike, coregonids, walleye, suckers	Osprey	Total Hg	130 nests in three major watersheds in areas impacted and not impacted by hydroelectric development	James Bay/Hudson Bay areas, Quebec	Liver, kidney, brain, breast muscle, and feathers of osprey	Chicks and adults	Des Granges et al., 1998	BMFs calculated from mean concentrations in different tissues and weighted mean concentrations in main fish species consumed in the diet. Evidence for feeding relationship established in the paper.

Table A3- Data summary and calculations from Hughes (1997).

Location	Feather/YP (4-5)	Feathers/YP (20)	Eggs/YP (4-5)	Eggs/YP (20)
St. Mary's River	12.33	15.74	1.07	1.36
Georgian Bay	12.00	21.71	2.05	3.71
Kawartha Lakes	13.58	11.64	1.83	1.57
Mean BMF	**14.50**		**1.93**	

Notes- YP=yellow perch. (4-5)=yellow perch aged 4-5 years, (20)= 20 cm yellow perch. Data presented are unitless BMFs. Mean BMFs are for mercury in feathers and eggs, averaged for both groups of prey each. Mercury concentrations used to derive BMFs were ug/g dry weight total Hg.

Table A4- Data summary and calculations from Neumann and Ward (1999).

		BMF @ age				
Lake	Species	Age 2	3	4	5	Lake Average BMF
Pickerel	Black crappie->TP	3.7	3.1	2.7	2.2	2.88
	Bluegill->TP	2.4	2.6	2.9	3.4	
Lillinonah	Yellow perch->TP	1.4	1.4	1.3	1.2	1.93
	Bluegill->TP	1.9	2.3	2.7	3.2	
					Mean	**2.40**

Notes- TP=top predators- largemouth bass, smallmouth bass, and chain pickerel. Mercury concentration values used to derive BMFs were expressed in ug/g dry weight total Hg.

Table A5- Data summary and calculations from Suedel et al. (1994).

Parameter	Trophic Level 2	Trophic Level 3
BMF Total Hg	0.3	0.2
	0.3	0.4
	1.6	1
	1.7	1.4
	6.8	1.8
		1.9
Mean	**2.14**	**1.12**
BMF MeHg	0.5	0.1
	0.7	0.2
	2	0.3
	10.5	0.7
		4.5
		80
		141
Mean	**3.425**	**32.4**

Note- data from literature used to derive BMFs (reported as trophic transfer coefficients (TTCs)) were exp
comparable units measured in organisms which were part of functional food chains/feeding

Table A6- Data summary and calculations from Bowles et al. (2001).

Species	Trophic Level	Mean [MeHg]	+1SD	-1SD
Arius berneyi	2	0.18	0.33	0.03
Toxotes chatareus	2	0.29	0.44	0.14
Mean [MeHg] TL2		0.24	0.38	0.09
Strongylura kreffti	3	0.38	0.63	0.14
Thryssa scratchleyi	3	0.34	0.66	0.02
Lates calcarifer	3	0.46	0.76	0.16
Mean [MeHg] TL3		0.39	0.68	0.10
BMFs	**2 --> 3**	**1.67**	**1.78**	**1.20**
Mean BMF		**1.55**		

Note- *A. bernyi*=groove-snouted catfish *T. chatareus*=seven-spotted archerfish *S. kreffti*=Sepik garpike, *T.scratchleyi*=giant freshwater anchov
L. calcarifer=barramundi. All concentrations used to derive BMFs were expressed as ug/g wet weight MeHg.

Table A7- Summary of BMFs used in USEPA's (1997) PPF calculation

BMF	Predator	Prey	Location
2.75	lake trout	bloater	L. Michigan
3.5	northern pike, largemouth bass	yellow perch, white sucker	35 lake aggregate, upper michigan
3.6	northern pike, largemouth bass	rainbow smelt, whitefish	L. Tyrifjorden, Norway
4	northern pike, walleye	specific weighted diets	L. Simcoe
5	lake trout (60 cm)	rainbow smelt (15 cm)	9 lake aggregate, Ontario
5.06	northern pike, walleye	white sucker, cisco	average of 6 Canadian Shield lakes
5.22	walleye (age 5)	yellow perch (age 2)	10 lake aggregate, Wisconsin
5.63	smallmouth bass, walleye	gizzard shad, bluegill	Onandaga Lake, New York
6.8	northern pike	yellow perch	43 lake aggregate, Sweden
7.1	largemouth bass	silversides	Clear L., California
7.4	northern pike	yellow perch	25 lake aggregate, Sweden
9.8	northern pike	spottail shiner, yellow perch	4 lake average, Manitoba

Table A8- Data summary and calculations from Ben-David et al. (2001).

Trophic Transfer	Mean [total Hg]	+1 SE	-1 SE	Comments
Jackpot Bay freshwater fishes	0.12	0.14	0.1	Dolly Varden, coastrange sculpin, sticklebacks
Jackpot Bay intertidal fishes	0.085	0.092	0.07	Rockfish, kelp greenling, crescent gunnels, intertidal sculpins,
Mean Jackpot Bay fishes	0.1025	0.116	0.085	
Jackpot Bay otters	9	9.5	8.2	River otter
BMF	**87.80**	**81.90**	**96.47**	

Note- all mercury concentrations used to calculate BMFs were expressed as mg/kg dry weight total Hg. Standard errors used were those reported in the study. Both intertidal and freshwater fish Hg concentrations were used due to stable isotope dietary analysis which indicated a significant portion of intertidal fish in diet.

Table A9- Data summary and calculations for Des Granges et al. (1998).

Type of Habitat	mean [Fish]	mean [Liver]	mean [Kidney]	mean [Brain]	mean [Muscle]	mean [Feathers]
Developed	1.420	3.610	5.280	1.010	1.790	58.090
Natural	0.234	0.720	0.910	0.230	0.360	16.470

BMF per Habitat	Liver	Kidney	Brain	Muscle	Feathers
BMF Developed	2.542	3.718	0.711	1.261	40.908
BMF Natural	3.080	3.893	0.984	1.540	70.460
Mean BMF	**2.811**	**3.806**	**0.848**	**1.400**	**55.684**

Note- concentrations are expressed in mg/kg dry weight total Hg. "Developed" areas are nesting sites on hydroelectric reservoirs.

Table A10- Data summary and calculations from Halbrook et al. (1997).

Diet	mean [Diet]	mean [Liver]	mean [Kidney]	mean [Hair]
B	0.05	0.61	1.25	7.43
D	0.15	1.93	3.47	13.44
E	0.22	3.67	4.35	19.03

Diet	BMF Liver	BMF Kidney	BMF Hair
B	12.20	25.00	148.60
D	12.87	23.13	89.60
E	16.68	19.77	86.50
Mean BMF	13.92	22.64	108.23
Range	12-17	20-25	87-149

Table A11- Data summary and calculations for Snodgrass et al. (2000).

Wetland	Gmean[total Hg] benthivore	Gmean[total Hg] top predator
40	0.18	0.26
41	0.32	0.49
42	0.19	0.32
77	0.63	1.05
97	0.27	0.24
136	0.33	0.68
139	0.28	0.35
142	0.2	0.31
Mean		

Note- benthivore= lake chubsucker, top predator= redfin pickerel, Gmean=geometric mean. All concentrations are expressed in ug/g dry weight total Hg.

Table A12- Data summary and calculations from Francis et al. (1998).

Receptor	Mean [Total Hg]	Mean [MeHg]	Cutoff
Benthos	0.003		
Carp Sm	0.019	0.015	<30 cm
Carp Lg.	0.100	0.101	>30 cm
Catfish Sm.	0.066	0.064	<30 cm
Catfish Lg.	0.199	0.199	>30 cm
Bullhead	0.003	0.003	
Bowfin	0.636	0.613	
Great Blue Heron	1.620		
Crappie	0.003	0.001	
Gizzard Shad	0.004	0.002	

Trophic Transfer	Trophic Level	BMF	Details
Benthos-Benthivores	2	17.128	mean[large carp+bullhead]/[benthos]
Benthivores-Large Piscivores	3	14.294	mean[bowfin+large catfish]/mean[small carp+bullhead+small catfish]
Benthivores-Piscivorous Birds	4	85.563	[heron]/mean[small carp+bullhead+small catfish+crappie+gizzard shad]

Note- Benthos= oligochaetes, larval Chironomids, Ceratopogonidae, Chaoboridae. Carp and catfish were grouped into small and large size classes to reflect their variable trophic level with size. Functional feeding relationships were defined in the study. BMFs were only derived for total Hg. Mercury concentrations were expressed as ug/g wet weight of total Hg and MeHg.

Table A13- Summary information to compare alternate species to receptor species

Trophic Level	Latin Name	Common Name	Receptor Species Comparison	Habitat	Range include Cornwall?	Food Type	Food Substrate	Feeding Technique	Food Ingestion: Body Weight Ratio	Food Size Class	Source	Other
2	*Bucephala clangula*	Common goldeneye	Common goldeneye	Lakes/ponds/rivers	Yes	Omnivore	Freshwater benthic	Bottom forager	0.3		CCME, 1999; CWS, 2002	
2	*Bucephala albeola*	Bufflehead	Common goldeneye	Lakes/ponds/rivers	No, but in Great Lakes	Omnivore	Freshwater benthic	Gleaner	0.36		CCME, 1999; CWS, 2002	
2	*Aythya valisineria*	Canvasback	Common goldeneye	Marshes	Yes	Omnivore	Freshwater benthic	Bottom forager			CWS, 2002	Regionally very rare.
2	*Melanitta fusca*	White-winged scoter	Common goldeneye	Lakes/ponds/rivers	No, but in Great Lakes	Molluscovore/ crustaceovore	Freshwater benthic	Gleaner			CWS, 2002	Regionally rare.
2	*Aythya affinis*	Lesser scaup	Common goldeneye	Lakes/ponds/rivers	Yes	Omnivore	Freshwater benthic	Bottom forager	0.31		CCME, 1999; CWS, 2002	
2	*Catastomous commersoni*	White sucker	White sucker	Warmer, shallow lakes or warm, shallow bays, and tributary rivers of larger lakes. Generally found at depths <30 feet.	Yes	Insectivore/molluscovore	Freshwater benthic				Scott and Crossman, 1973	
2	*Erimyzon sucetta*	Lake chubsucker	White sucker	Small, shallow, warm, weedy ponds.	No, northern extreme of range is Lake Erie and Lake St. Clair	Insectivore	Freshwater benthic				Scott and Crossman, 1973	
2	*Cyprinus carpio*	Common carp	White sucker	Warm, turbid waters.	Yes	Herbivore/Insectivore/ Mulloscovore	Freshwater benthic				Scott and Crossman, 1973	
3	*Coregonus artedii*	Cisco	Forage fish	Deeper waters of lakes.	Yes	Omnivore	Freshwater pelagic				Scott and Crossman, 1973	
3	*Couesius plumbeus*	Northern lake chub	Forage fish	Deeper waters of lakes and large rivers.	Yes	Omnivore	Freshwater pelagic				Scott and Crossman, 1973	
3	*Amia calva*	Bowfin	Walleye	Swampy, vegetated bays of warm lakes and rivers.	Yes	Piscivore	Freshwater benthic				Scott and Crossman, 1973	
3	*Catastomous catastomous*	Longnose sucker	White sucker	Lakes/ponds/rivers (almost everywhere in clear, cold water)	Yes	Invertebrates	Freshwater benthic				Scott and Crossman, 1973	
3	*Cottus cognatus*	Slimy sculpin	White sucker	Deeper waters of lakes and cooler streams on rocky or gravelly substrate	Yes	Insectivore	Freshwater benthic				Scott and Crossman, 1973	
3	*Prosopium cylindraceum*	Round whitefish	White sucker	Lakes at depths less than 150 feet	Yes	Omnivore	Freshwater benthic				Scott and Crossman, 1973	
2 and 3	*Lepomis macrochirus*	Bluegill	Forage fish	Shallow, weedy, warm water of large and small lakes, ponds, and heavily vegetated, slowly flowing areas of small rivers and large creeks. Shallow water, < 20 feet deep.	Yes	Insectivore/omnivore	Freshwater benthic				Scott and Crossman, 1973	
2 and 3	*Coregonus clupeaformis*	Lake whitefish	Forage fish	Cool water of lakes, spawns in shallower water. Depth range of 60 to 174 feet.	Yes	Omnivore	Freshwater benthic				Scott and Crossman, 1973	
2 and 3	*Ictalurus punctatus*	Channel catfish	Walleye/white sucker	Cool, clear, deeper waters of large lakes and rivers	Yes	Omnivore	Freshwater benthic				Scott and Crossman, 1973	
2 and 3	*Perca flavescens*	Yellow perch	Yellow perch	Warm to cool water habitats of all types. Prefer clear water and abundant vegetation. Shallow water <30 feet deep.	Yes	Omnivore	Freshwater pelagic and benthic				Scott and Crossman, 1973	
2 and 3	*Pomoxis nigromaculatus*	Black crappie	Yellow perch	Clear, quiet, warm water of large ponds, small lakes, bays and shallower areas of larger lakes, and areas of low flow of larger rivers.	Yes	Omnivore	Freshwater benthic				Scott and Crossman, 1973	
2 and 3	*Ictalurus nebulosus*	Brown bullhead	Yellow perch/white sucker	Shallow, warm-water areas of ponds/lakes/rivers. Depths of <40 feet.	Yes	Omnivore	Freshwater benthic				Scott and Crossman, 1973	
3 and 4	*Lutra canadensis*	River otter	American mink	Lakes/ponds/rivers	Yes	Piscivore	Freshwater pelagic and benthic		0.10-0.17	>30 cm	Sample and Suter, 1999; CCME, 1999; USEPA, 1997	100% of diet is fish
3 and 4	*Mustela vison*	American mink	American mink	Lakes/ponds/rivers	Yes	Omnivore	Freshwater pelagic and benthic		0.14-0.24	0-20 cm	Sample and Suter, 1999; CCME, 1999; USEPA, 1997	33-90% of diet is fish or aquatic prey (mean=55%)
3 and 4	*Gavia immer*	Common loon	Great blue heron	Lakes/ponds/rivers (primary habitat)	Yes	Piscivore	Freshwater pelagic	Diver	0.18		CWS, 2002; CCME, 1999	
3 and 4	*Pandion haliaetus*	Osprey	Great blue heron	Lakes/ponds/rivers (teritary habitat)	Yes	Piscivore	Freshwater pelagic	Foot plunger	0.2	0-40 cm	CWS, 2002; CCME, 1999; Sample and Suter, 1999	
3 and 4	*Ardea herodias*	Great blue heron	Great blue heron	Lakes/ponds/rivers (teritary habitat)	Yes	Piscivore	Freshwater pelagic	Ambusher	0.21	0-30 cm	CWS, 2002; CCME, 1999; Sample and Suter, 1999	
3 and 4	*Stizostedion vitreum*	Walleye	Walleye	Shallow, turbid lakes; large streams or rivers	Yes	Piscivore	Freshwater pelagic and benthic				Scott and Crossman, 1973	
3 and 4	*Esox lucius*	Northern pike	Walleye	Heavily vegetated slow-moving rivers or weedy bays of lakes	Yes	Piscivore/Omnivore	Freshwater pelagic and benthic				Scott and Crossman, 1973	
3 and 4	*Micropterus salmoides*	Largemouth bass	Walleye	Shallow bays of larger lakes, more rarely large, slow-moving rivers	Yes	Omnivore	Freshwater pelagic and benthic				Scott and Crossman, 1973	Adult diet is 50-90% small fishes
3 and 4	*Esox niger*	Chain pickerel	Walleye	Sluggish streams and heavily vegetated lakes and ponds; water < 10 feet deep	Yes	Piscivore	Freshwater pelagic and benthic				Scott and Crossman, 1973	
3 and 4	*Esox americanus americanus*	Redfin pickerel	Walleye	Sluggish, heavily vegetated acidic streams; less frequently in ponds and weedy backwaters/quiet bays of larger lakes/rivers	Yes	Piscivore	Freshwater pelagic and benthic				Scott and Crossman, 1973	
3 and 4	*Salvelinus namaycush*	Lake trout	Walleye	Deep lakes; less frequently in northern half of range in shallow lakes and in rivers	Yes	Omnivore	Freshwater pelagic				Scott and Crossman, 1973	
3 and 4	*Lota lota* (Linnaeus)	Burbot	Walleye	In central/southern Canada, the deep waters of lakes and rivers. Restricted to below hypolimnion in summer.	Yes	Omnivore	Freshwater pelagic and benthic				Scott and Crossman, 1973	

APPENDIX B. *Mercury in sediment and biota and biological effects from previous surveys*

Table B1. Total and methyl mercury concentrations in sediments collected from St. Lawrence River (Cornwall) Area of Concern from 1985 to 2000. Concentrations are in top 10 cm unless otherwise noted. Biological effects from *BEAST* analysis.

	Total Hg in Sediment (ng/g)							Methyl Hg Sediment (ng/g)	Total Hg Biota (ng/g)	Biological Effects[g]	
Location/Site	1985[a]	1991[b]	1992[c]	1993[c]	1994[d]	1997[e]	2000[f]	1991[b]	1991[b]	Community	Toxicity
Lake St. Lawrence											
083 (top 3 cm)	-	90	-	-	-	-	-	1.0	11.9	-	-
082 (top 3 cm)	-	120	-	-	-	-	-	1.3	26.5	-	-
Zone 1											
166	-	-	-	-	-	790	-	-	-	-	-
167	-	-	-	-	-	1190	-	-	-	Unstressed	Poss. toxic
168	-	-	-	-	-	1710	-	-	-	-	-
Zone 2											
5	-	-	-	-	13820	1670	8400	-	-	Unstressed	Poss. toxic
9	-	-	-	-	7740	4830	12000	-	-	Unstressed	Non-toxic
16	-	-	-	-	10810	-	-	-	-	-	-
17	-	-	-	-	1770	2000	-	-	-	Unstressed	Poss. toxic
19	-	-	-	-	7300	-	-	-	-	-	-
27	-	-	-	-	890	4320	-	-	-	Poss. Stressed	Poss. toxic
31	-	-	-	-	12250	19500	3700	-	-	Unstressed	Non-toxic
46	-	-	-	-	6340	-	-	-	-	-	-
54	-	-	-	-	750	-	-	-	-	-	-
64	-	-	-	-	5300	3090	12000	-	-	Poss. Stressed	Non-toxic
7 sites (top 3 cm)	40 – 4400	160 –3130	-	-	-	-	-	3.9 – 10.3	6.4– 68.0	-	-
Zone 3											
404 (top 3 cm)	-	560	-	-	-	-	-	7.0	-	-	-
17 sites (top 2 cm)	-	-	130 – 500	-	-	-	-	-	-	-	-
16 sites (top 6 cm)	-	-	-	11 – 1000	-	-	-	-	-	-	-
Zone 4											
173	-	-	-	-	-	130	-	-	-	-	-
175	-	-	-	-	-	80	-	-	-	Poss. stressed	Non-toxic
176	-	-	-	-	-	150	-	-	-	-	-
177	-	-	-	-	-	120	-	-	-	-	-
179	-	-	-	-	-	140	-	-	-	Poss. stressed	Non-toxic
181	-	-	-	-	-	130	-	-	-	-	-
182	-	-	-	-	-	120	-	-	-	-	-
Farfield											
171	-	-	-	-	-	440	-	-	-	-	-
172	-	-	-	-	-	620	-	-	-	-	-
376, 373 (top 3 cm)	770	60 - 1180	-	-	-	-	-	0.95 – 2.5	4.7 – 14.0	-	-

[a] Anderson 1990
[b] Richman 1994
[c] Metcalfe-Smith et al. 1995
[d] Richman 1996
[e] Richman 1999
[f] Richman 20
[g] Reynoldson et al. 1997

APPENDIX C. *Conversion of total and methyl mercury levels in biota to wet weights*

Table C1. Total mercury in biota (***converted to ng/g wet weight***) collected from the St. Lawrence River (Cornwall) Area of Concern.

Area/Zone	Site	BIOTA – Total Hg		
		Chironomid	Snail	Amphipod
Ref. Upstream	1319	3.9	-[a]	-
	1320	3.8	23.9	-
	1321	8.3	18.2	8.5
	1322	6.0	11.7	7.6
	1323	5.2	-	-
	1325	8.2(6.1)[b]	-	-
	1326	1.8	-	-
	1331	3.0	-	-
	1332	11.5	16.6	-
Ref. Downstream	1327	8.5	6.4	-
	1328	10.0	5.7	-
	A1	11.3	18.0	-
Zone 1	167	14.1	30.1	-
	168	17.3(16.0)[b]	22.0	-
	183	109.9	67.3(77.8)[b]	-
	184	41.8	39.4(44.9)[b]	29.2
Zone 3	101	46.3	51.0	-
	108	135.4	120.7	32.9
Zone 2	5	16.6	9.9	44.9
	9	69.8	29.7	-
	16	12.6	34.1	28.4
	17(10 cm)	3.3	21.0	30.8
	17(5 cm)	8.0	23.5	-
	19	87.2	31.7	-
	27	44.9	37.0	-
	31	241.6	72.8(82.7)[b]	76.9
	46	17.7(15.6)[b]	13.7	-
	54	12.8	17.8	-
	64	45.8	28.8	30.6
Zone 4	175	10.0	17.1	-
	179	7.0(7.7)[b]	75.6	21.7(25.6)[b]
	173	11.2	24.8	-
	176	6.4	22.5	-
	182	6.9	8.4	-
Downstream	171	9.1	18.1	-

[a] no data
[b] laboratory duplicate

Table C2. Methyl mercury in biota (***converted to ng/g wet weight***) collected from the St. Lawrence River (Cornwall) Area of Concern.

Area/Zone	Site	BIOTA – Methyl Hg		
		Chironomid	Snail	Amphipod
Ref. Upstream	1319	*0.5*	-[a]	-
	1320	*1.0*	*4.1*	-
	1321	*1.0*	*3.4*	*1.6*
	1322	*1.3(1.1)[b](1.5)[c]*	*2.0*	*2.1*
	1323	*1.3*	-	-
	1325	*0.4*	-	-
	1326	*0.3*	-	-
	1331	*0.2*	-	-
	1332	*0.4*	*1.3*	-
Ref. Downstream	1327	*1.3*	*4.5*	-
	1328	*1.5*	*3.8*	-
	A1	*1.0(1.1)[c]*	*5.7*	-
Zone 1	167	*0.6*	*4.7*	-
	168	*0.6*	*4.6(5.5)[c]*	-
	183	*1.1*	*13.4*	-
	184	*0.5*	*4.8(4.8)[b]*	*2.2*
Zone 3	101	*3.1*	*14.7*	-
	108	*2.9*	*17.7*	*10.3*
Zone 2	5	*0.8*	*2.7*	*1.9*
	9	*0.9*	*1.4*	-
	16	*1.3*	*3.1*	*8.6*
	17(10 cm)	*0.6*	*4.9*	*3.8*
	17(5 cm)	*1.2*	*5.2*	-
	19	*1.4*	*6.4*	-
	27	*1.4*	*6.7*	-
	31	*4.1*	*13.8*	*7.8*
	46	*0.5*	*3.7(3.1)[b]*	-
	54	*0.6*	*4.7*	-
	64	*0.7*	*3.8(3.8)[c]*	*6.2*
Zone 4	175	*1.7*	*2.3*	-
	179	*1.4*	*13.6*	*4.2(4.9)[c]*
	173	*1.1*	*3.5*	-
	176	*1.4*	*3.7*	-
	182	*0.8*	*2.0*	-
Downstream	171	*1.1(0.8)[b](0.8)[c]*	*5.1*	-

[a] no data
[b] repeat analysis
[c] laboratory duplicate

APPENDIX D. *Quality assurance/Quality control results*

Table D1. Sediment nutrient concentrations and particle size fractions, and overlying water alkalinity and nutrient concentrations for field replicate samples.

Station		TOC	TP	TN	% silt	% sand	% clay	Alk	NO_3NO_2	NH3-N	TKN	TP(W)
1332	Mean	4.383	1496.67	4683.33	64.79	0.99	34.21	88.63	0.204	0.014	0.211	0.014
	SD	0.006	11.55	213.85	1.52	0.44	1.29	0.12	0.002	0.0006	0.010	0.001
	CV	0.1	0.8	5	2	44	4	0.1	0.8	4	5	8
184	Mean	20.670	980.67	3600	32.21	46.14	21.65	89.47	0.188	0.017	0.208	0.014
	SD	1.914	94.24	70	7.75	8.12	1.04	0.21	0.003	0.0006	0.006	0.0002
	CV	9	10	2	24	18	5	0.2	1	3	3	2
5	Mean	4.2	1096.67	4178	62.04	14.61	23.35	88.27	0.221	0.013	0.215	0.0235
	SD	0.184	45.09	238.60	2.57	1.61	0.98	0.64	0.001	0.001	0.016	0.004
	CV	4	4	6	4	11	4	0.7	0.5	8	8	15
171	Mean	3.367	1093.33	2340	40.03	34.49	25.48	88.50	0.212	0.012	0.239	0.017
	SD	0.064	60.28	303.15	1.82	0.66	1.18	0.61	0.002	0.0006	0.007	0.002
	CV	2	6	13	5	2	5	0.7	0.7	5	3	12

Table D2. Metal concentrations in sediment for field replicate samples. (Mercury data are from Flett Research.)

Station		Total Hg	Methyl Hg	Al	Cr	Cu	Fe	Mn	Ni	Pb	Zn
1332	Mean	137.67	0.665	1.88	48.22	56.22	2.67	501.76	43.91	50.10	168.15
	SD	31.47	0.209	0.07	1.33	1.60	0.07	6.90	1.05	1.84	3.93
	CV	23	32	4	3	3	3	1	2	4	2
184	Mean	783.33	1.803	0.50	19.64	46.23	0.97	100.36	23.05	34.89	113.34
	SD	373.54	1.164	0.02	0.88	8.95	0.04	1.93	4.05	5.06	9.56
	CV	48	65	3	4	19	4	2	18	15	8
5	Mean	1875.67	3.84	0.69	29.94	61.55	1.33	217.00	27.92	91.59	642.27
	SD	141.51	0.063	0.05	1.76	4.13	0.06	5.19	1.60	10.59	54.33
	CV	8	2	7	6	7	5	2	6	12	8
171	Mean	253.00	0.898	0.80	22.29	34.68	1.37	247.66	26.11	28.45	117.12
	SD	83.37	0.282	0.04	1.01	4.94	0.05	5.25	0.83	1.06	7.15
	CV	32.95	31	5	5	14	4	2	3	4	6

Table D3. Laboratory quality assurance/quality control data from Flett Research Ltd.

SEDIMENT: Total Mercury

Sample Recovery

Site	comments	Recovery
1319		96.5
1320		96.5
1321		96.5
1322		96.5
1322	dup	96.5
1323		102.2
1325		102.2
1326		102.2
1331		102.2
133200		102.2
133200	dup	102.2
133201		102.2
133202		102.2
1327		102.2
1328		102.2
A1		99.6
167		99.6
168		99.6
183		99.6
18400		99.6
18401		97.5
18401	dup	97.5
18402		99.6
101		96.5
108		96.5
5-00		99.6
5-01		99.6
5-02		96.5
9		96.5
16		96.5
17		96.5
19		96.5
27		96.5
31		96.5
46		96.5
54		96.5
54	dup	96.5
64		96.5
175		99.6
179		99.6
173		97.5
173	dup	97.5
176		99.6
173		97.5
173	dup	97.5
182		99.6
17100		99.6
17101		99.6
17102		99.6
	mean	**98.73**
	range	**96.5-102.2**

Matrix Spike recovery

Site	comments	Recovery
1322		100
54		93.1
133200		110.7
6669		93.7
6662		91
667802		92.7
6690		80.5
6696		96.8
173		99.4
18401		95.7
	mean	**95.36**

Reference Sediment

Marine sediment standard (MESS - 2: 92 ng/g)

Run	THg	Mean % Recovery
1	96.0	
2	87.2	99.6
3	88.8	
4	90.2	97.3
5	90.6	
6	86.9	96.5
7	81.9	
8	84.6	90.5
9	89.7	
10	87.9	96.5
11	89.7	
12	87.9	96.5
mean	**88.45**	**96.15**

range 90.5-99.6

SEDIMENT: Methyl mercury

Sample Recovery

Site	comments	Recovery
1319		84.7
1320		84.7
1321		81.1
1322		84.7
1322	dup	84.7
1323		82.4
1325		82.4
1326		82.4
1331		103.4
1331	dup	103.4
133200		91.5
133201		91.5
133202		96.2
1327		96.2
1328		96.2
A1		76.7
167		105.1
168		105.1
183		76.7
18400		76.7
18401		76.7
18402		105.1
101		81.1
108		84.7
5-00		76.7
5-01		76.7
5-01	dup	76.7
5-02		81.1
9		81.1
16		85.7
17		85.7
19		85.7
27		103.4
27	dup	103.4
31		84.7
46		81.1
54		103.4
54	repeat	103.4
64		81.1
175		76.7
179		85.7
173		105.1
173	dup	105.1
176		85.7
176	dup	85.7
182		85.7
17100		105.1
17101		105.1
17102		105.1
	mean	**89.56**
	range	**76.7-105.1**

Matrix Spike Recovery

Site	comments	Recovery
18402	spike	107.7
17101	spike	102.6
19	spike	86.6
182	spike	84.9
5-00	spike	79.1
18401	spike	74.4
5-02	spike	91.2
101	spike	67.5
101	spike-repeat	74.7
1319	spike	84.7
133201	spike	79.9
1325	spike	80
1327	spike	91.3
1331	spike	88.1
54	spike	117.4
54	spike-repeat	119.8
	mean	**89.36875**
	range	**74.4-119.8**

Table D3. Laboratory quality assurance/quality data from Flett Research Ltd. (cont.)

BIOTA: Total Mercury

Reference Material (DORM-2, MQAP fish samples)

Run	Standard	THg	Expected THg	% Recovery	Mean
1	DFO Bag 296	500	449	111	109
	DFO Bag 296	478	449	107	
	DFO Bag 297	221	205	108	108
	DFO Bag 297	219	205	107	
	NRC (Dorm2)	4613	4640	99	97
	NRC (Dorm2)	4424	4640	95	
2	DFO Bag 296	458	449	102	101
	DFO Bag 296	448	449	100	
	DFO Bag 297	224	205	109	105
	DFO Bag 297	208	205	101	
	NRC (Dorm2)	4659	4640	100	99
	NRC (Dorm2)	4536	4640	98	
3	DFO Bag 296	464	449	103	104
	DFO Bag 296	470	449	105	
	DFO Bag 297	236	205	115	109
	DFO Bag 297	211	205	103	
	NRC (Dorm2)	4372	4640	94	96
	NRC (Dorm2)	4554	4640	98	
4	DFO Bag 296	456	449	102	101
	DFO Bag 296	453	449	101	
	DFO Bag 297	218	205	106	104
	DFO Bag 297	206	205	101	
	NRC (Dorm2)	4449	4640	96	95
	NRC (Dorm2)	4330	4640	93	
5	DFO Bag 296	470	449	105	103
	DFO Bag 296	456	449	102	
	DFO Bag 297	211	205	103	103
	DFO Bag 297	212	205	104	
	NRC (Dorm2)	4406	4640	95	96
	NRC (Dorm2)	4485	4640	97	
6	DFO Bag 296	463	449	103	104
	DFO Bag 296	472	449	105	
	DFO Bag 297	224	205	109	106
	DFO Bag 297	212	205	103	
	NRC (Dorm2)	4564	4640	98	97
	NRC (Dorm2)	4465	4640	96	
7	DFO Bag 296	470	449	105	105
	DFO Bag 296	469	449	104	
	DFO Bag 297	225	205	110	107
	DFO Bag 297	215	205	105	
	NRC (Dorm2)	4578	4640	99	98
	NRC (Dorm2)	4529	4640	96	
8	DFO Bag 296	470	449	105	104
	DFO Bag 296	469	449	104	
	DFO Bag 297	225	205	110	107
	DFO Bag 297	214	205	105	
	NRC (Dorm2)	4577	4640	99	98
	NRC (Dorm2)	4528	4640	98	
				overall mean	**102.3**

Sample Recovery

Site	Chir	Snail	Amph
1319	103		
1320	100		
1321	103	101.6	104.6
1322	100		104.6
1323	104.6		
1325	100		
1326	103		
1331	100		
1332	100		
1327	103	101.6	
1328	101.6	101.6	
A1	100	104.6	
167	100	103	
168	103	101.6	
183	100		
184	103	103.3	104.6
101	103	103	
108	100	104.6	104.6
5	103	103	104.6
9	100		
16	100	104.6	101.6
17	100	101.6	
19	100	101.6	
27	100	101.6	
31	103	103	101.6
46	101.6	104.6	
54	103	104.6	
64	103	104.6	104.6
175	101.6	104.6	
179	100	104.6	100
173	101.6	103	
176	100	103	
182	100	101.6	
171	101.6	103	
mean	**101.34**	**103.08**	**103.42**
range	**100-104.6**	**101.6-104.6**	**100-104.6**

Matrix Spike Recovery

sample	Recovery
3008	99.8
3047	100.6
3108	97.2
3285	88.4
3430	97.9
3325	99.5
3419	97.0
3002	100.5
3430	100.7
3325	103.1
3349	94.5
mean	**98.1**

BIOTA: Methyl Mercury

Sample Recovery

Site	Snail	Chir	Amph
1319		92.5	
1320	91.2	95.9	
1321	96.1	99.6	91.8
1322	91.2	95.9	91.8
1323		96.6	
1325		95.9	
1326		99.6	
1331		95.9	
1332	91.2	88.3	
1327	96.1	92.5	
1328	96.1	88.3	
A1	92.2	99.6	
167	92.2	92.5	
168	96.1	99.6	
183	92.2	95.9	
184	96.1	99.6	91.8
101	92.5	99.6	
108	92.2	99.6	96.6
5	92.2	99.6	91.8
9	88.3	95.9	
16	99.6	99.6	91.8
17	96.1	95.9	91.8
19	96.1	92.5	
27	96.1	92.5	
31	92.5	92.5	91.8
46	92.2	91.2	
54	96.6	92.5	
64	92.2	92.5	91.8
175	96.6	88.3	
179	96.6	99.6	91.8
173	92.2	88.3	
176	92.2	95.9	
182	96.1	95.9	
171	92.2	88.3	
mean	**93.90**	**94.95**	**92.20**
range	**88.3-99.6**	**88.3-99.6**	**91.8-96.6**

Matrix Spike Recovery

Site	Snail	Chir	Amph
5			107.4
64			96
17	101.5		
19	92.1		
46	102.3	96.5	
171	98.1		
17		103	
31		105	
176		94.5	
16		96.6	
184		100.7	
mean	**98.5**	**99.4**	**101.7**

Reference Material (DORM-2: 4470 +/- 340 ng/g)

Run	MeHg	Mean % Recovery
1	4544	
2	4088	96.6
3	3937	
4	4272	91.8
5	4194	
6	4393	96.1
7	4045	
8	4202	92.2
9	3999	
10	4271	92.5
11	4259	
12	4315	95.9
13	4369	
14	4535	99.6
15	3793	
16	4103	88.3
17	4368	
18	4440	98.5
19	4309	
20	4559	99.2
21	3898	
22	4291	91.6
23	4179	
24	3977	91.2
25	3988	
26	3910	88.3
27	4113	
28	4233	93.4
29	3785	
30	4071	87.9
mean	**4181.33**	**93.5**
SD	**214.71**	

Table D4 Laboratory duplicate analysis from Caduceon Laboratory

			Laboratory Duplicate (Site 1325)			Laboratory Duplicate (Site 175)		
Analyte	**Units**	**Det Limit**	**Concn 1**	**Concn 2**	**CV**	**Concn 1**	**Concn 2**	**CV**
K	pct	0.05	0.25	0.23	0.059	0.08	0.08	0.000
Li	µg/g	1	20	19	0.050	12	11	0.061
Mg	pct	0.01	0.94	0.88	0.047	1.46	1.45	0.005
Mn	µg/g	1	641	614	0.030	315	309	0.014
Mo	µg/g	1	1	<1		2	2	0.000
Na	pct	0.01	0.04	0.04	0.000	0.05	0.04	0.157
Nb	µg/g	5	<5	<5		<5	<5	
Ni	µg/g	1	42	39	0.045	28	26	0.052
Pb	µg/g	1	33	34	0.021	23	23	0.000
Sb	µg/g	5	<5	<5		<5	<5	
Sn	µg/g	20	<20	<20		<20	<20	
Sr	µg/g	1	43	41	0.029	44	43	0.016
Ti	µg/g	1	1107	950	0.108	559	519	0.052
V	µg/g	25	45	41	0.071	23	23	0.000
W	µg/g	20	<20	<20		<20	<20	
Y	µg/g	1	20	19	0.033	11	11	0.000
Zn	µg/g	1	137	133	0.021	106	91	0.108
Al2O3	pct	0.01	12.89	12.9	0.001	11.01	11.05	0.003
BaO	pct	0.001	0.071	0.071	0.005	0.064	0.065	0.011
CaO	pct	0.01	2.77	2.82	0.013	5.45	5.38	0.009
Cr2O3	pct	0.01	0.01	0.01	0.079	0.01	0.01	0.000
Fe2O3	pct	0.01	5.76	5.72	0.005	3.84	4.03	0.034
K2O	pct	0.01	2.90	3	0.024	2.74	2.74	0.000
MgO	pct	0.01	2.23	2.24	0.002	2.82	2.79	0.008
MnO	pct	0.01	0.10	0.1	0.006	0.06	0.07	0.109
Na2O	pct	0.01	1.75	1.61	0.059	2.14	1.96	0.062
P2O5	pct	0.03	0.42	0.39	0.052	0.33	0.34	0.021
SiO2	pct	0.01	55.43	55.38	0.001	59.1	59.3	0.002
TiO2	pct	0.01	0.73	0.73	0.003	0.59	0.58	0.012
LIO	pct	0.05	13.34	13.47	0.007	11.67	11.8	0.008
				mean CV	**0.031**		**mean CV**	**0.029**

Table D5. Reference material data from Caduceon Laboratory.

		WH89-1			STSD-3			SO-2			STSD-1		
Analyte	**Units**	**measured**	**reference**	**recovery**	**measured**	**reference**	**recovery**	**measured**	**reference**	**recovery**	**measured**	**reference**	**recovery**
K	pct												
Li	µg/g				19	23	0.826						
Mg	pct												
Mn	µg/g				2340	2630	0.890				3480	3740	0.930
Mo	µg/g				6	7	0.857				2	2	1.000
Na	pct												
Nb	µg/g												
Ni	µg/g				28	25	1.120				23	18	1.278
Pb	µg/g				42	39	1.077				37	34	1.088
Sb	µg/g												
Sn	µg/g												
Sr	µg/g												
Ti	µg/g												
V	µg/g				50	61	0.820				35	47	0.745
W	µg/g												
Y	µg/g												
Zn	µg/g				175	192	0.911				145	165	0.879
Al2O3	pct	12.2	12.1	1.008				14.51	15.1	0.961			
BaO	pct	0.288	0.29	0.993				0.101	0.111	0.910			
CaO	pct	5.91	5.9	1.002				2.72	2.77	0.982			
Cr2O3	pct	0.04	0.04	1.000				<0.01	<0.01				
Fe2O3	pct	6.97	6.9	1.010				7.76	7.89	0.984			
K2O	pct	2.25	2.21	1.018				3.06	2.94	1.041			
MgO	pct	3.5	3.5	1.000				0.87	0.89	0.978			
MnO	pct	0.09	0.09	1.000				0.09	0.07	1.286			
Na2O	pct	1.25	1.34	0.933				2.41	2.48	0.972			
P2O5	pct	0.2	0.19	1.053				0.66	0.69	0.957			
SiO2	pct	60.96	60.5	1.008				54	53.42	1.011			
TiO2	pct	0.9	0.9	1.000				1.31	1.43	0.916			
LIO	pct	4.75	5	0.950									
			mean recovery	**0.998**		**mean recovery**	**0.929**		**mean recovery**	**1.000**		**mean recovery**	**0.987**

Inter-Laboratory Comparison of Analyses of Total Hg in Sediment from St. Lawrence River (Cornwall) Area of Concern

Analyses for concentrations of total mercury (THg) in sediment were performed by 2 laboratories: Flett research Ltd., which was selected to measure THg and methyl mercury in sediment and biological samples, and Caduceon Environmental Laboratory, which conducted physico-chemical analyses on sediment that included THg determination. Each lab received a sediment subsample from the same homogenized sample collected at each site. Those submitted to Flett were sent frozen, and those submitted to Caduceon were first freeze-dried. Figure D1 shows how the site measurements compare graphically.

Overall agreement between labs for the determinations of THg in sediment is indicated by the slope of a regression involving the two variables. As recommended by McArdle (1988) and Draper and Smith (1998), the regression was estimated by the geometric mean (GM, aka reduced major axis) method instead of the ordinary least squares (OLS) method. The OLS method assumes negligible error in the *X* variable, and can result in biased slope estimates when applied to data in which both *X* and *Y* variables are subject to errors of the same magnitude, a situation which clearly applies here. Rather than minimizing the sum of the squares of the deviations of observed *Y* values from the regression line, as in the OLS method, the GM method minimizes the sum of the areas of the triangles formed by the data point, the point on the line corresponding to the *X* value, and the point on the line corresponding to the *Y* value. Geometric Mean slope, b_{GM}, was estimated by

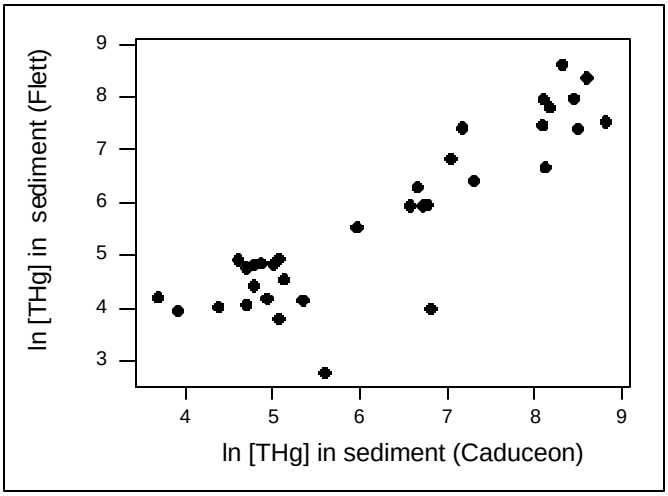


Figure D1. Comparison of total Hg measurements between labs.

$$b_{GM} = s_y / s_x \qquad \text{(Legendre and Legendre 1998)}$$

where s_y = standard deviation of *Y* - values, and s_x = standard deviation of *X* - values. The b_{GM} estimate is also the geometric mean of the OLS slope of *Y* on *X* and the reciprocal of the slope of *X* on *Y*. (Note that when the purpose of the analysis is not to estimate functional parameters such as the slope, but only to predict values of *Y* for given *X*'s, OLS regression is suitable (Legendre and Legendre 1998). For this reason, the GM method was not used for the invertebrate Hg – sediment Hg regressions.)

Geometric mean regression slope for $\ln[THg]_{Flett}$ vs $\ln[THg]_{Caduc}$:

Standard deviation of $\ln[THg]_{Flett} = 1.5992 = s_y$
Standard deviation of $\ln[THg]_{Caduc} = 1.5737 = s_x$

$b_{GM} = s_y / s_x = 1.5992/1.5737 =$ **1.0162**

OLS regression of *Y* vs *X*: $\ln[THg]_{Flett} = 0.0375 + 0.90197 \ln[THg]_{Caduc}$
OLS regression of *X* vs *Y*: $\ln[THg]_{Caduc} = 1.2955 + 0.87341 \ln[THg]_{Flett}$

For both regressions $P<0.001$ and $r^2 = 78.8\%$.

As a check, using the alternate slope estimation method: $b_{GM} = (0.90197 \times [1 / 0.87341])^{½} = 1.0162$

The overall agreement in measurements of THg in sediment is therefore very good because the slope estimate is close to 1. This suggests that either (a) the analyses of the labs are accurate or (b) analyses are biased in identical ways. The unexplained 21.2% of the variation of the regression should be attributed to laboratory measurement error.

APPENDIX E. *Supplementary physico-chemical environmental data for the St. Lawrence River (Cornwall) Area of Concern sites*

Table E1. Grain size and nutrient concentrations in sediment.

Area	Site	Sand	Silt	Clay	Gravel	TOC	Total N	Total P
		%	%	%	%	%	µg/g	µg/g
Reference	1319	1.54	59.39	39.07	0	3.09	3200	1140
	1320	0.24	48.03	51.73	0	2.96	3480	1360
	1321	69.65	16.46	13.89	0	1.46	1100	650
	1322	60.96	24.56	14.48	0	0.85	969	882
	1323	14.86	66.86	18.27	0	2.11	1860	977
	1325	2.25	58.64	39.11	0	4.57	4100	1380
	1326	1.79	49.76	48.45	0	4.92	4990	1450
	1331	5.09	69.15	25.76	0	3.08	2880	1000
	1332[a]	0.99	64.79	34.21	0	4.38	4683	1497
	1327	48.79	36.01	15.21	0	2.51	2270	934
	1328	39.52	36.24	17.70	0	1.00	816	689
	A1	8.81	54.47	34.72	0	3.75	4200	1070
Zone 1	167	15.10	57.53	27.37	0	4.16	3600	1060
	168	21.88	49.55	28.57	0	3.84	2600	983
	183	69.19	16.02	14.79	0	21.16	3150	1030
	184[a]	46.14	32.21	21.65	0	20.67	3600	981
Zone 3	101	79.88	10.42	9.70	0	6.21	1620	766
	108	68.54	19.56	11.90	0	5.06	1340	727
Zone 2	5[a]	14.61	62.04	23.35	0	4.20	4178	1097
	9	23.41	54.52	22.07	0	3.84	3350	1050
	16	50.01	31.25	15.10	3.64	2.72	2080	814
	17 (5 cm)	27.01	48.51	24.48	0	3.86	3440	1100
	17 (10 cm)	59.82	23.70	15.03	1.45	3.25	1770	806
	19	44.33	36.52	16.50	2.65	3.04	2280	1020
	27	59.83	24.52	12.91	2.74	3.11	1310	729
	31	25.72	54.22	19.79	0.27	2.62	1500	1000
	46	19.30	53.15	27.55	0	3.87	3870	1120
	54	54.78	26.63	18.03	0.56	3.16	1760	844
	64	21.87	54.76	23.37	0	5.02	4030	970
Zone 4	173	11.71	55.43	32.86	0	3.50	3690	1110
	175	23.04	49.11	27.85	0	2.96	2480	1070
	176	6.90	58.49	34.61	0	3.91	3670	1190
	179	19.55	51.43	29.03	0	3.55	3470	1160
	182	29.33	43.63	27.05	0	2.75	2510	1100
D/S	171[a]	34.49	25.48	40.03	0	3.37	2340	1093

[a] QA/QC site. Values represent the mean of three field replicates.

Table E2. Metal concentrations in sediment.

Area	Site	Al	As	Cd	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Zn
	Units	pct	µg/g	µg/g	µg/g	µg/g	pct	µg/g	µg/g	µg/g	µg/g	µg/g
	Detection Limit	0.01	5	1	1	1	0.01	0.005	1	1	1	1
Reference	1319	1.9	<5	<1	38	31	2.7	0.21	639	34	24	101
	1320	2.7	<5	<1	46	33	3.7	0.27	980	40	20	124
	1321	0.5	<5	<1	12	10	0.9	0.16	158	13	18	45
	1322	0.4	<5	<1	13	10	0.9	0.05	209	17	25	51
	1323	0.8	<5	1	23	26	1.4	0.11	293	27	34	114
	1325	2.2	<5	1	44	43	2.8	0.12	641	42	33	137
	1326	2.2	<5	<1	43	42	3.0	0.11	800	40	36	125
	1331	1.2	<5	1	32	40	1.8	0.12	289	32	38	117
	1332[a]	1.9	<5	1.7	48	56	2.7	0.16	502	44	50	168
	1327	0.9	<5	1	22	24	1.6	0.08	379	26	29	90
	1328	0.7	<5	<1	17	16	1.4	0.04	405	19	32	52
	A1	1.2	<5	1	28	41	2.0	0.10	378	32	22	134
Zone 1	167	0.8	<5	1	23	43	1.4	0.87	174	26	31	117
	168	0.7	<5	1	22	37	1.3	0.72	167	26	29	106
	183	0.4	<5	1	57	55	0.9	4.88*	79	30	30	110
	184[a]	0.5	<5	1	20	46	1.0	3.36*	100	23	35	113
Zone 3	101	0.3	<5	<1	12	27	0.8	1.49	118	16	21	54
	108	0.4	<5	<1	12	21	0.8	3.31*	154	16	20	65
Zone 2	5[a]	0.7	<5	1	30	62	1.3	6.71*	217	28	92	642
	9	0.9	<5	1	42	86	1.6	3.54*	209	32	302*	1162*
	16	0.6	<5	<1	21	40	1.2	1.14	275	22	46	204
	17 (5 cm)	0.7	<5	1	24	42	1.3	1.31	235	24	45	219
	17 (10 cm)	0.5	<5	1	33	35	1.0	1.31	201	22	51	425
	19	0.6	<5	1	24	65	1.1	4.10*	232	22	45	206
	27	0.6	<5	<1	16	24	1.1	3.26*	249	21	29	113
	31	0.6	<5	1	19	28	1.3	5.42*	273	22	40	171
	46	1.0	<5	1	27	43	1.6	0.78	242	29	38	181
	54	0.5	<5	<1	15	23	1.0	0.83	180	19	24	149
	64	0.9	<5	2	36	123*	1.6	4.70*	231	30	477*	2300*
Zone 4	173	1.1	<5	1	29	45	1.8	0.15	355	32	32	123
	175	1.0	<5	1	25	34	1.7	0.13	315	28	23	106
	176	1.2	<5	<1	31	46	2.1	0.17	410	34	25	137
	179	1.2	<5	2	29	38	1.9	0.91	309	31	23	116
	182	0.9	<5	1	21	28	1.5	0.14	365	25	20	87
D/S	171[a]	0.8	<5	1	22	35	1.4	0.39	248	26	28	117
	LEL	-	6	0.6	26	16	2%	0.20	460	16	31	120
	SEL	-	33	10	110	110	4%	2.00	1100	75	250	820

[a] QA/QC site. Values represent the mean of three field replicates.

* exceeding the Severe Effect Level

Table E3. Physico-chemical conditions of overlying water.

Area	Site	Alkalinity	NO_3NO_2	NH_3-N	TKN	TP	pH	Conductivity	Temp	DO	Site depth
	Units	mg/L	mg/L	mg/L	mg/L	mg/L		µS/cm	°C	mg/L	m
Reference	1319	85.6	0.105	0.013	0.221	0.0148	8.56	285	13.2	10.7	2.6
	1320	85.9	0.103	0.027	0.313	0.0241	8.52	287	13.0	10.8	6.4
	1321	87.8	0.203	0.014	0.214	0.0136	8.61	292	15.3	9.8	5.4
	1322	87.1	0.193	0.016	0.221	0.0154	8.41	292	15.4	9.6	6.0
	1323	87.7	0.192	0.015	0.202	0.0131	8.46	290	15.3	9.6	6.8
	1325	87.3	0.127	0.012	0.242	0.0135	8.65	289	14.8	10.6	5.8
	1326	86.3	0.121	0.013	0.241	0.0125	8.56	287	14.3	10.0	8.4
	1331	89.0	0.207	0.022	0.218	0.0144	8.55	293	15.2	9.8	8.0
	1332[a]	88.6	0.204	0.014	0.211	0.0137	8.57	293	15.1	9.5	10.6
	1327	87.2	0.209	0.014	0.207	0.0134	8.23	293	15.1	9.3	5.2
	1328	88.7	0.209	0.013	0.226	0.0120	8.20	293	15.0	9.4	8.6
	A1	86.4	0.215	0.017	0.264	0.0432	8.51	291	15.6	9.8	5.8
Zone 1	167	89.4	0.203	0.016	0.225	0.0151	8.43	299	15.6	9.4	7.1
	168	89.0	0.209	0.017	0.214	0.0148	8.44	299	15.6	10.3	7.9
	183	88.1	0.178	0.015	0.222	0.0138	8.35	295	15.8	9.9	1.9
	184[a]	89.5	0.188	0.017	0.208	0.0137	8.37	299	15.6	9.7	4.0
Zone 3	101	89.2	0.215	0.014	0.204	0.0168	8.26	297	15.0	9.6	7.1
	108	89.2	0.215	0.013	0.221	0.0163	8.26	297	15.0	9.7	6.3
Zone 2	5[a]	88.3	0.221	0.013	0.215	0.0235	8.33	299	15.6	9.4	6.1
	9	89.1	0.217	0.015	0.207	0.0172	8.34	298	15.6	9.5	6.5
	16	88.1	0.218	0.012	0.200	0.0166	8.33	301	15.6	10.8	9.2
	17	88.5	0.220	0.014	0.201	0.0248	8.31	296	15.6	10.2	8.9
	19	88.7	0.219	0.022	0.217	0.0202	8.28	297	15.5	9.9	9.0
	27	88.9	0.224	0.018	0.221	0.0618	8.27	296	15.7	10.3	10.7
	31	88.6	0.223	0.017	0.187	0.0166	8.25	301	15.4	10.5	8.7
	46	89.1	0.218	0.012	0.206	0.0143	8.52	296	15.6	9.8	10.1
	54	88.7	0.212	0.011	0.197	0.0139	8.52	296	15.7	9.9	8.1
	64	88.9	0.216	0.012	0.217	0.0155	8.30	303	15.6	10.6	3.1
Zone 4	173	87.1	0.201	0.020	0.187	0.0138	8.48	292	15.6	9.7	10.5
	175	88.6	0.210	0.011	0.190	0.0168	8.47	293	15.6	9.7	13.4
	176	89.2	0.207	0.021	0.265	0.0155	8.46	291	15.6	9.7	14.5
	179	88.6	0.210	0.014	0.204	0.0152	8.47	291	15.7	9.9	15.0
	182	87.5	0.210	0.014	0.198	0.0135	8.46	292	15.6	10.2	11.0
D/S	171[a]	88.5	0.212	0.012	0.239	0.0166	8.58	296	15.7	10.0	10.0

[a] QA/QC site. Values represent the mean of three field replicates for alkalinity, NO_3NO_2, NH_3-N, TKN and TP only.

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