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An overview of mercury concentrations in freshwater fish species: a national fish mercury dataset for Canada

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Abstract: Fish mercury (Hg) concentrations have been measured over the last 30–40 years in all regions of Canada as part of various monitoring and research programs. Despite this large amount of data, only regional assessments of fish Hg trends and patterns have previously been attempted. The objective of this study was to assemble available freshwater fish Hg concentration data from all provinces and territories and identify national patterns. The Canadian Fish Mercury Database includes over 330 000 records representing 104 species of freshwater fish collected from over 5000 locations across Canada between 1967 and 2010. Analysis of the 28 most frequently occurring species (>1000 records) showed that the majority of variation in Hg concentrations (when normalized to a standard size) was accounted for by geographic location. Median Hg concentrations increased with trophic level ($r = 0.40$, $p < 0.05$), with the highest Hg concentrations found in piscivorous species such as walleye (*Sander vitreus*), northern pike (*Esox lucius*), and lake trout (*Salvelinus namaycush*). The Canadian Fish Mercury Database provides the most comprehensive summary of fish Hg measurements in Canada, and the results indicate that several regionally observed trends in fish Hg concentrations (e.g., Hg biomagnification and geographic variation) are observed at a national scale. Implications for the effective assessment of changes in fish Hg concentrations in relation to changes in Hg emission regulations are discussed.

Résumé : Les concentrations de mercure (Hg) dans le poisson sont mesurées depuis 30 ou 40 ans dans toutes les régions du Canada, dans le cadre de divers programmes de surveillance et de recherche. Malgré cette vaste quantité de données, seules des évaluations régionales des tendances et distributions du Hg dans le poisson ont été effectuées par le passé. L'objectif de l'étude consistait à regrouper les données disponibles sur les concentrations de Hg dans les poissons dulcicoles pour toutes les provinces et tous les territoires et à cerner les motifs de distribution à l'échelle nationale. La base de données canadienne sur le mercure dans le poisson (« Canadian Fish Mercury Database ») comprend plus de 330 000 entrées représentant 104 espèces de poissons dulcicoles échantillonnés dans plus de 5000 localités à la grandeur du pays, de 1967 à 2010. L'analyse des 28 espèces les plus fréquentes (>1000 entrées) a démontré que la situation géographique expliquait la majeure partie de la variabilité des concentrations de Hg (normalisées en fonction d'une taille standard). Les concentrations médianes de Hg augmentaient parallèlement au niveau trophique ($r = 0{,}40$, $p < 0{,}05$), les poissons piscivores comme le doré (*Sander vitreus*), le grand brochet (*Esox lucius*) et le touladi (*Salvelinus namaycush*) présentant les plus fortes concentrations. La base de données canadienne sur le mercure dans le poisson constitue la synthèse la plus exhaustive des mesures de Hg dans le poisson au Canada, et les résultats indiquent que plusieurs tendances observées à l'échelle régionale des concentrations de Hg dans le poisson (p. ex. bioamplification et variations géographiques du Hg) sont également observées à l'échelle nationale. Les conséquences de ces résultats en ce qui concerne l'évaluation efficace de l'évolution des concentrations de Hg dans le poisson par rapport aux modifications de la réglementation en matière d'émissions de Hg sont abordées. [Traduit par la Rédaction]

Introduction

Anthropogenic activities have contributed to the increase in the global atmospheric mercury (Hg) pool during the 20th century (Pacyna et al. 2010). In Canada, with the exception of known point-source industrial Hg pollution, most aquatic ecosystems receive the bulk of their Hg loadings from atmospheric deposition of inorganic Hg directly to the water surface and to their catchments (Lockhart et al. 1998). Natural processes within lakes and their watersheds convert a portion of atmospherically deposited inorganic Hg to the more toxic methylmercury (MeHg), which is strongly biomagnified in aquatic food webs and consistently results in high concentrations in predatory

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fish and other top aquatic predators worldwide (Wiener et al. 2003).

The biogeochemistry of Hg in aquatic ecosystems and their catchments is sufficiently complex that relationships between the quantity of deposited inorganic Hg and the extent of bioaccumulation in fish are not always straightforward, but have been statistically and experimentally verified (Hammerschmidt and Fitzgerald 2006; Harris et al. 2007; Orihel et al. 2007). Environmental factors at the landscape level (e.g., watershed and lake size, wetland area and connectivity, type of vegetative cover), and water body level such as physico-chemical (e.g., pH, sulphur cycling, dissolved organic matter, nutrients, and temperature) and biological properties (e.g., productivity, food web structure) are known to influence the amount of inorganic Hg available for methylation, rate of methylating microorganisms, and ultimately uptake and transfer through the aquatic food web into fish (Winfrey and Rudd 1990; Bodaly et al. 1993; Cabana et al. 1994; St. Louis et al. 1994; Driscoll et al. 1995; Kidd et al. 1995; Benoit et al. 1999; Hrabik and Watras 2002; Grigal 2002; Pickhardt et al. 2002; Graydon et al. 2008). Consequently, Hg concentrations measured in the same species of fish may vary considerably from one water body to another, despite geographic proximity (Kamman et al. 2005; Lockhart et al. 2005).

For the past three to four decades, as part of monitoring programs and research projects, a large number of freshwater fishes from across Canada have been analyzed to determine Hg concentrations in various tissues. Extensive sampling has been carried out by federal and provincial government agencies for the issuance of fish consumption advisories (MWS 2007; AHW 2010; BCMWLAP 2011; EY 2011; MDDEPQ 2011; NSDFA 2011; PEIDEEF 2011; OME 2011), assessment of fishery yields and commercial fish quality (Stewart et al. 2003), and monitoring of Hg accumulation in fish from newly impounded hydroelectric reservoirs (Bodaly et al. 1984; Verdon et al. 1991; Schetagne and Verdon 1999). In addition, many targeted studies have been carried out usually with the objective of describing the levels of Hg in specific fish species from a particular location or region in relation to wildlife health risks (Scheuhammer and Blancher 1994; Burgess and Meyer 2008), point sources of contamination (Parks et al. 1991; Weech et al. 2004), bioaccumulation patterns as a result of food web alterations (Johnston et al. 2003), geographical patterns in relation to various environmental parameters known to affect Hg availability (Evans et al. 2005), and baseline and operational environmental monitoring for industrial projects. Collectively, these independent efforts across Canada have resulted in a substantial volume of fish Hg data that span variable temporal and spatial scales. Some of these provincial and regional datasets are periodically analyzed and published in the primary scientific literature (e.g., Verdon et al. 1991; Bodaly et al. 2007; Bhavsar et al. 2010), but many remain unpublished. The magnitude and variability of Hg concentration in freshwater fish species on a national scale across Canada has not yet been comprehensively evaluated.

The Clean Air Regulatory Agenda (CARA) was implemented by Environment Canada in 2007 to develop a nationally coordinated strategy for assessing and managing atmospheric pollutants within Canada (Morrison 2011). To date, several region-specific Hg science programs have been developed and successfully implemented in parts of North America. These include the Great Lakes Basin (Evers et al. 2011), Arctic Canada (CACAR 2003), northeastern North America (Evers and Clair 2005), and the Florida Everglades (Krabbenhoft et al. 2000). Previous efforts to compile and analyze these regional databases (i.e., northeastern North America (Kamman et al. 2005); northern Canada (Lockhart et al. 2005)) have been limited in spatial scope or focussed on particular species of interest, but have nonetheless served to advance knowledge about the state and extent of Hg concentrations in freshwater fishes on a scale critical for understanding atmospheric Hg contamination of aquatic ecosystems. However, a comparable strategy and program at a national scale in Canada has thus far remained elusive (Smith and Trip 2005; Morrison 2011).

As part of the first phase of the CARA Hg science program (Morrison 2011), we sought to compile available data on fish Hg across Canada to (*i*) establish a baseline for assessing the effectiveness of any future management actions aimed at reducing atmospheric Hg pollution, (*ii*) facilitate a national-scale ecological risk assessment to identify regions where piscivorous fish and wildlife may face an elevated risk owing to Hg exposure, and (*iii*) provide national data for Hg-related ecosystem modelling activities, as outlined by Morrison (2011). This paper serves to describe the process of data amalgamation and provide a point of reference for forthcoming papers detailing subsequent CARA mercury studies (e.g., Depew et al. 2013).

Methods

Source datasets

We collected, sorted, and formatted datasets of Hg measurements in freshwater fish from a total of 256 distinct sources (Supplementary Table S1[1]) into a Microsoft Access database, hereafter referred to as the Canadian Fish Mercury Database (CFMD). Contaminant monitoring programs at the provincial (Ontario, Manitoba, Saskatchewan, Quebec) and federal level (Fisheries and Oceans Canada Inspection Branch) provided the largest datasets. Datasets from intensive monitoring of hydroelectric reservoirs by Fisheries and Oceans Canada and hydroelectric industrial partners (Hydro Québec, BC Hydro) were also substantial. The remaining datasets were derived from contributed data collected from ad hoc surveys, nonroutine sampling programs, wildlife and food web studies, and environmental impact assessments for industrial projects (Supplementary Table S1[1]). Most source datasets were provided electronically in delimited text or spreadsheet form, but we also extracted data from scientific technical reports (microfiche and hard copy) from academic and government libraries, as well as those publicly available from online environmental assessment registries (Canadian Environmental Assessment Act registry, http://www.ceaa.gc.ca/050/index_e.cfm; Nunavut Impact Review Board, http://www.nirb.ca/PublicRegistry.html; Mackenzie Valley Review Board, http://reviewboard.ca/registry/index.php; British Columbia Environmental Project Information Centre, http://a100.gov.bc.ca/appsdata/epic/html/deploy/epic_home.html).

Georeferencing, site information, and classification criteria

Data quality varied considerably across all contributed datasets in terms of completeness and accuracy. Data collected prior to 1990 was generally not precisely georeferenced owing to the limited availability of global positioning system technology. One of the objectives during the assembly of the CFMD was to ensure that geographical locations (e.g., latitude and longitude) were correctly identified so that future studies could be placed in the appropriate spatial context. Geographic location information provided by data partners was used to locate sampled water bodies using geographic information systems (GIS). Data sources included the National Hydrological Network layer (NHN; http://geobase.ca/geobase/en/data/nhn/description.html), the Canadian Geographic Names Database (CGNDB; http://www.nrcan.gc.ca/earth-sciences/search/search_e.php). These sources were used to cross-reference named features (primarily lakes), and the CGNDB identifier was

[1]Supplementary data are available with the article through the journal Web site at http://nrcresearchpress.com/doi/suppl/10.1139/cjfas-2012-0338.

adopted for integration of features into the database. For unnamed features, the supplied or local name was used.

For locations that could be georeferenced with confidence, we created a water body specific identifier (WATERBODY_ID) that consisted of a unique georeferenced datum corresponding to either the centroid of the sampled water body (for small features; <300–500 km²) or the specified sample location for very large water bodies, such as the Laurentian Great Lakes, Lake Winnipeg, Great Slave Lake, Great Bear Lake, and some large reservoirs. Spatial reference information for rivers and watercourses (stream, creek, brook) was generally specific enough (either from the source dataset or supporting literature) to ascertain the geographic location to within a few hundred metres. These unique identifiers and geographic coordinates served to both allow for amalgamation of records taken from approximately the same location or water body collected by different groups in the same year and allow for indexing within the database to facilitate searches for duplicate records. Original location coordinates were retained in separate data fields (if present). Locations that could not be adequately georeferenced using the above approaches were compared with printed topographical maps and (or) source document descriptions of the sampling sites. If this did not improve confidence in the identification of the sampling location, such data were excluded from the database.

Sampled locations were classified following the convention in the supplied datasets, taking into consideration the general guidelines set forth by the Geographic Names Board of Canada (GNBC 2001). A full set of classification and decision rules can be found in Supplementary Table S2[1]. For some sampling locations, specific classifiers related to designation as a lake or reservoir (pre- or post-impoundment), relative position to impounded reservoirs (upstream or downstream), or status as a Hg-contaminated water body were employed. Although we recognize that other types of point sources of Hg (e.g., municipal solid waste incinerators, coal-fired power plants) are distributed throughout much of the study region, in the context of this study we restricted the use of "Hg-contaminated" water-bodies to those that were impacted by direct-water discharges of Hg (i.e., chlor-alkali plant discharge) or contamination from mine tailings or processing waste only. There are two principle reasons for this decision. First, Hg emitted to the atmosphere from point sources such as coal-fired power plants and incinerators contributes to Hg contamination in both the near- and far-field, and the relative contribution of these sources to Hg contamination in aquatic ecosystems has been difficult to quantify (Cheng et al. 2012). Second, while recently impounded reservoirs and direct Hg discharge impacted water bodies are undoubtedly influenced by atmospheric Hg deposition, the magnitude of Hg input from industrial sources in Hg-contaminated water bodies, the generation of MeHg from the decomposition of terrestrial vegetation, and the inundation of soils in flooded reservoirs generally far exceeds the amount of MeHg generated from atmospheric deposition in the absence of these inputs (Kelly et al. 1997; Lockhart et al. 2000). Methylation of Hg in these affected aquatic systems universally leads to a rapid increase to high levels of Hg in fish that may persist for a substantial period of time (>15 years) even after the cessation of point-source inputs or exhaustion of the Hg pool in flooded soils and vegetation (Parks et al. 1991; Verdon et al. 1991; Anderson et al. 1995; Bodaly et al. 2007; Munthe et al. 2007; Anderson 2011). Consequently, recently impounded reservoirs and direct Hg-contaminated water bodies are unlikely to reflect patterns and (or) trends in atmospheric Hg deposition.

For verification of reservoirs and impounded water bodies, unless indicated in the source dataset, we used information from published literature (e.g., Bodaly et al. 2007) and hydroelectric and water regulatory agencies (BC Hydro, Nalcor Energy, Manitoba Hydro, Saskatchewan Watershed Authority, Ontario Power Generation) to ascertain the year of reservoir impoundment. Records denoting fish collected within 30 years of impoundment were classed as "reservoir". Hg concentrations in fish residing in downstream water bodies can also be impacted through the export of Hg in water, sediments, and plankton (Schetagne et al. 2000) and (or) a change in feeding guild due to the increased availability of dead or moribund fish macerated by passage through turbines (Brouard et al. 1994). Locations classed as "downstream of reservoir" were used only if indicated in the source dataset because the degree of influence of upstream reservoirs varies from region to region and may not be universally consistent (Bodaly et al. 2007).

Locations with a documented history of known direct-water discharge Hg contamination were classified as "contaminated" based on available information. This included instances of Hg-laden mine tailings from cinnabar mines (e.g., Pinchi Lake, British Columbia; Reid and Morley 1975) or tailings from gold mines where Hg was used in ore amalgamation and recovery processes (e.g., Giauque Lake, North West Territories; Moore and Sutherland 1980), water bodies affected by the direct discharge of Hg from chlor-alkali or pulp mill complexes (e.g., South Saskatchewan River, Saskatchewan, Wobeser et al. 1970; Clay Lake, Ontario, Armstrong and Scott 1979). At least five chlor-alkali plants in the vicinity of the Laurentian Great Lakes and St. Lawrence River released over 700 t of Hg into the lakes between 1935 and 1995 (Tripp and Thorleifson 1998), and the use of phenyl mercuric acetate slimicides by the pulp and paper industry is also thought to have contributed to Hg pollution at some locations (Fimreite 1970). Many of these areas are designated as areas of concern owing to persistent pollution problems, of which Hg is often included (Weis 2004). We classified records from these locations as "contaminated" if the sampling coordinates fell within the designated area of concern boundaries as delineated by Environment Canada (see http://www.ec.gc.ca/raps-pas/default.asp?lang%20=%20En&n%20=%20A290294A-1).

Description of dataset structure and variables of interest

Every effort was made to ensure that information on detection limits, analytical methods, identification of species, and sample type were included in the CFMD and that Hg concentrations were reported on a wet mass basis. For data reported on a dry mass basis, we used the supplied moisture content (or 78% if unavailable; Baker et al. 2004) to convert dry mass concentrations to wet mass. In cases where analytical and sample characteristic information was unavailable, we used source documents or published literature to obtain the missing information. In general, the provided datasets clearly identified the species sampled and the portion of the fish (e.g., whole fish, fillet, skin-free muscle) that was analyzed, although in some cases identification to the species level was not done. A list of common and scientific names for fish species contained in the CFMD is available in Supplementary Table S3[1].

Mercury concentrations generally increase with the age of the fish, and age can be assessed using scales, otoliths, or other structures (Scott and Armstrong 1972). However, proxies for age (length, mass) are simpler and more cost effective to measure, and as a result the majority of provided datasets included information on fish length or mass rather than age. Length data were typically reported as fork or total length, either measured for a single fish or as the mean length of a composite number of fish. When the length measurement was not specified (fork or total) in the source dataset or was not apparent from related literature, we assumed that the measurement of length was equivalent to total length. While most data records provided a measure of length (usually to the nearest 10 mm) for a single fish, several datasets used slot sizes for composite samples for analysis. If the mean length of composited fish was not reported, we used the mid-point of the slot range as an approximation of the mean size in the composite sample. For records reporting a measure of fork length, we used fork length to total length ratios for each species as reported in Fish-

Base (Froese and Pauly 2011) to convert to a total length equivalent. For species that did not have a fork length to total length ratio, we used a ratio from a species with similar morphology. For composite fish samples, we retained the number of fish composited, while single fish or composite samples of unknown number were assigned a value of one.

Analytical methods

The majority of records in the database (~96%) were analyzed using a variation of the cold vapour atomic absorption spectroscopy method and report Hg concentrations as total Hg (Uthe et al. 1970; Hendzel and Jamieson 1976; Loescher and Neary 1981; Scheuhammer and Bond 1991; USEPA 2001*a*; Centre d'expertise en analyse environmentale du Québec 2003; OME 2006). In more recent years, investigators have shifted to higher precision or throughput methods such as cold vapour atomic fluorescence spectrophotometry (Method 1631e USEPA 2002) and thermal decomposition followed by atomic absorption spectrophotometry (USEPA 1998). Other less frequently used instrumental methods include inductively coupled plasma atomic emission spectroscopy (USEPA 2001*b*) and inductively coupled plasma mass spectrometry (USEPA 1994). Detection limits varied across datasets but were generally low (<0.05 $\mu g \cdot (g \text{ wet mass})^{-1}$), and samples that were at or below the detection limit were usually appropriately indicated. For summary purposes, samples at or below the detection limit were assigned a value equivalent to half the reported detection limit. Although the utility of detection limit substitution has been questioned (Helsel 2005), based on the small number of records at or below detection limits (2.8% of total records; 6461), large sample sizes for the majority of species, and low relative detection limit compared with measured Hg levels, we suggest that any bias resulting from our substitution approach will be small unless the species sample sizes and measured Hg levels are small. For species and sample types (see below) that had more than 10% of records at or below the detection limit, we have indicated these in Table 1 so that the summary statistics can be interpreted accordingly.

Sample characteristics

The most commonly sampled portion in the dataset is skin-free dorsal muscle tissue with no bones. This is generally considered the portion most commonly consumed by humans. Other commonly analyzed sample types include whole fish (used primarily for wildlife or ecosystem status studies) and fillets with the skin on. Nonlethal methods to sample dorsal muscle with biopsy needles have become more popular, since they permit live release of fish and provide Hg concentrations equivalent to those measured in larger muscle samples that require lethal sampling (Baker et al. 2004). For classification purposes, these are treated as equivalent to skinless fillets. Other sampled portions consisted of organs (liver, gonads, kidney), eggs, unspecified fish composites, and eviscerated fish composites. In some cases it is not clear whether the composite types are of muscle only, muscle with skin, or whole fish, but these other tissue types represented <1% of the records. For brevity, summary statistics for these rare tissue types are not presented in this paper.

Quality control and quality assurance

In addition to verification checks on location information (described above), a series of action queries were designed in Microsoft Access to locate duplicate records based on fields describing location, year of sampling, species, length (fork and (or) total), mass, and Hg concentration. Identification of anomalous length and Hg values, usually resulting from misplaced decimal points and improper units, was accomplished using Q–Q plots, length–mass plots, and cumulative distribution function plots. Records identified as possible outliers or anomalies were then examined and corrected from source or purged from the CFMD.

Statistical analysis

Here we aim to summarize the scope and breadth of the CFMD and to discuss the merits of fish Hg monitoring initiatives as they relate to the objectives of the CARA Hg science program. This overview of ongoing efforts will be a valuable contribution towards a better understanding of the state and extent of Hg contamination of freshwater fishes in Canada and provide a baseline for development of future monitoring frameworks for fish Hg assessment. However, the CFMD is complex and there is insufficient space for detailed spatial or temporal analysis in this paper. Comparable efforts on smaller spatial and temporal scales have investigated broad-scale correlations of standard-size fish Hg with physico-chemical parameters known to influence Hg availability (e.g., pH, dissolved organic carbon; Evans et al. 2005; Kamman et al. 2005) and have generally provided insightful patterns over large geographic regions. However, we presently lack an adequate representation or resolution of such ancillary data relative to the scale of the CFMD. We have therefore chosen to focus on more descriptive and broad-scale analysis of available data to set the groundwork for additional detailed analyses in future papers.

During assembly of the database, it was clear that sampling effort was strongly biased toward locations subjected to direct point-source Hg discharges (i.e., chlor-alkali facilities) and newly impounded reservoirs. This is not surprising given the requirements of various federal and provincial agencies and hydroelectric utilities to establish and monitor Hg levels in fish from these systems, to provide consumption advisories for recreational and subsistence fishers. However, this sample collection bias may skew summary statistics if other less impacted locations are not sampled with the same frequency or intensity. We therefore elected to exclude sites classified as "contaminated", "reservoirs", or "downstream of reservoir" (Supplementary Fig. S1[1]) from the summary statistics and analyses presented here to better characterize Hg levels in fish across Canada where Hg loads are primarily derived from atmospheric deposition. For these data, we generated descriptive statistics (minimum, median, and maximum) for all species represented for total Hg and total length equivalents for the three most commonly measured portion types: skinless fillets, skin on fillets, and whole fish (Table 1).

To assess the breadth of sampling effort in the context of available water body entities, we used the fraction of water surface area (FWS) of Canada (1 km^2 grid raster; Canada Centre for Remote Sensing, Natural Resources Canada, Ottawa, Ont.; Supplementary Fig. S2[1]) as a proxy for surface water body availability. Water-covered areas in the FWS dataset are defined as all water body entities (lakes, rivers, wetlands, fens, etc.) reported in the Canadian National Topographic Database. Source maps for the FWS dataset were collected after snow-melt periods and should be relatively unbiased, although some uncertainty remains owing to the considerable temporal variation encompassed in the Canadian National Topographic Database data collection (Fernandes et al. 2001). Although the FWS dataset does not distinguish the number of water body entities or fishless water bodies, for the purposes of the coarse analysis presented here, the FWS data are the best available data source for the scale of interest. Zonal summary statistics for the FWS data and the CFMD (mean fraction water surface; number of sampled sites, events, and fish collected) were computed on the basis of NHN unit limits using ArcGIS 9.3.1 (ESRI 2009). Associations among these variables were computed using Spearman rank correlation coefficients in the statistical software R version 2.11.1 (R Development Core Team 2010).

The summary statistics in Table 1 provide some information regarding differences in Hg concentrations measured in various species. However, it is important to note that different species may be collected from very different regions. Moreover, if there are strong geographic differences in Hg availability, species with a limited distribution will only provide representative information for a small region and possibly confound interpretation. We de-

Table 1. Median total mercury (Hg) concentrations (minimum–maximum in parentheses) by sample type for 104 freshwater fish species in the Canadian Fish Mercury Database (1967–2010).

	Skinless fillet			Skin-on fillet			Whole fish			
Species	Count	Length (cm)	Total Hg (μg·g−1)	Count	Length (cm)	Total Hg (μg·g−1)	Count	Length (cm)	Total Hg (μg·g−1)	RelC*
Lake sturgeon	1 010	95.5 (14.2–173.7)	0.23 (<DL–2.90)	16	102.7 (58.5–124.2)	0.31 (0.07–0.57)	6	52.0 (35.5–92.0)	0.03 (0.01–0.06)	11.9
Atlantic sturgeon	4	131.0 (72.1–166.8)	0.04 (0.02–0.09)							ND
Sturgeon (unspecified)	25	95.7 (41.8–135.9)	0.15 (<DL–3.60)							ND
White sturgeon	79	52.2 (28.3–94.1)	0.17 (0.05–1.28)							<0.1
Bowfin	115	51.5 (35.2–73.6)	0.27 (0.06–1.60)							0.4
American eel	410	73.0 (28.0–112.6)	0.32 (<DL–2.08)	6	42.8 (33.4–59.0)	0.67 (0.39–2.54)	6	38.9 (32.4–55.4)	0.35 (0.14–0.77)	2.5
Longnose sucker	3 104	38.6 (7.0–74.3)	0.14 (<DL–2.55)	751	38.3 (15.2–59.1)	0.16 (0.02–1.33)	119	42.2 (5.6–54.1)	0.06 (<DL–0.46)	56.3
White sucker	12 717	41.3 (5.4–73.0)	0.12 (<DL–4.39)	843	40.2 (18.0–60.6)	0.09 (<DL–0.90)	1132	36.5 (5.6–68.5)	0.13 (<DL–1.40)	35.8
Largescale sucker	210	44.5 (21.8–59.7)	0.17 (<DL–0.82)	6	43.2 (38.3–48.4)	0.14 (0.10–0.27)				0.4
Northern hog sucker	1	29.0	0.28							ND
Bigmouth buffalo	63	44.6 (22.6–76.7)	0.19 (0.01–0.78)							0.2
Silver redhorse	63	35.5 (8.6–54.5)	0.07 (0.01–0.55)							5.5
River redhorse	2	57.2 (54.0–60.4)	0.38 (0.17–0.59)							ND
Shorthead redhorse	609	40.2 (19.9–59.3)	0.23 (0.02–1.21)							15.6
Redhorse (unspecified)	321	42.7 (20.5–74.8)	0.24 (<DL–3.10)							9.8
Greater redhorse	1	44.1	0.38							ND
Rock bass	2 621	17.9 (5.5–29.6)	0.19 (<DL–1.20)				52	13.3 (7.0–18.0)	**0.08 (<DL–0.71)**	7.7
Sunfish family	121	37.7 (12.0–58.2)	0.22 (0.07–1.52)							ND
Bluegill	801	17.7 (9.9–24.1)	**0.08 (<DL–0.69)**	10	15.3 (14.4–17.2)	0.03 (<DL–0.05)	11	11.1 (11.1–13.9)	**0.07 (<DL–0.14)**	0.5
Pumpkinseed	1 643	16.2 (3.3–24.0)	**0.09 (<DL–1.10)**	10	15.6 (13.8–16.2)	0.04 (<DL–0.11)	62	11.2 (6.4–17.5)	**0.06 (<DL–0.30)**	2.5
Smallmouth bass	10 436	31.8 (6.5–59.4)	0.33 (<DL–5.00)	48	28.8 (9.0–43.0)	0.38 (0.07–0.86)	81	17.0 (3.3–36.6)	0.20 (<DL–0.87)	12.4
Largemouth bass	3 453	31.6 (11.2–60.3)	0.29 (<DL–3.40)	5	21.0 (20.0–22.3)	0.19 (0.16–0.20)	15	17.5 (13.5–21.6)	0.09 (0.04–0.42)	1.8
Bass (unspecified)	56	32.5 (27.5–35.0)	0.36 (0.11–2.87)							0.4
White crappie	150	22.3 (4.8–36.6)	0.08 (<DL–1.34)							<0.1
Black crappie	1 480	23.4 (11.0–40.1)	**0.11 (<DL–1.60)**							3.6
Alewife	38	16.9 (10.0–21.4)	0.07 (0.02–0.40)				45	15.0 (11.0–21.0)	**0.07 (<DL–0.19)**	0.6
Gizzard shad	115	37.8 (8.5–47.0)	**0.04 (<DL–0.28)**							0.2
Prickly sculpin	41	15.2 (7.0–26.8)	0.32 (0.01–2.19)				3	6.9 (4.8–9.5)	0.13 (0.11–0.21)	ND
Slimy sculpin	4	6.8 (5.3–9.6)	0.03 (0.03–0.07)				224	6.1 (3.8–12.2)	0.03 (<DL–0.75)	5.5
Sculpin	3	8.0 (6.4–8.3)	**0.06 (<DL–0.07)**							ND
Staghorn sculpin	36	19.2 (15.1–30.1)	0.04 (0.02–0.67)							ND
Fourhorn sculpin	3	21.2 (19.2–22.0)	**0.06 (<DL–0.08)**							ND
Arctic sculpin	1	24.3	0.15							ND
Shorthorn sculpin	3	27.7 (22.7–31.2)	0.17 (0.16–0.32)							ND
Mottled sculpin	1	7	0.05							ND
Chiselmouth				3	27.2 (23.7–32.4)	0.08 (0.08–1.13)				ND
Goldfish	9	27.0 (23.5–59.2)	**0.09 (<DL–0.33)**							ND
Quillback	58	42.9 (22.6–57.5)	0.21 (<DL–1.04)							ND
Lake chub	144	12.4 (6.5–31.0)	0.15 (<DL–0.78)				22	13.4 (5.5–28.5)	0.22 (0.06–0.83)	28.5
Carp	3 573	59.5 (15–104.5)	0.17 (<DL–1.23)	4	50.3 (43.2–58.5)	0.09 (0.07–0.11)	10	11.2 (3.2–24.5)	**0.02 (<DL–0.08)**	11.6
Brassy minnow							13	6.8 (3.0–9.8)	0.08 (0.03–0.14)	ND
Pearl dace	99	9.0 (6.4–13.0)	0.14 (0.03–0.83)				22	9.2 (5.8–15.0)	0.19 (0.06–0.32)	15.9
Peamouth chub	115	25.3 (14.9–34.7)	0.23 (0.07–0.69)	3	25.2 (25.0–26.9)	0.21 (0.18–0.22)				1.8
Golden shiner	32	9.0 (8.0–15.1)	0.19 (<DL–0.43)				91	9.9 (6.8–19.5)	0.16 (0.01–0.58)	2.9
Emerald shiner							29	5.9 (4.5–11.0)	**0.01 (<DL–0.11)**	ND
Common shiner	29	9.7 (7.4–13.0)	0.32 (0.13–0.64)				64	6.9 (4.4–16.0)	**0.05 (<DL–0.38)**	1.6

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Table 1 (*continued*).

	Skinless fillet			Skin-on fillet			Whole fish			
Species	Count	Length (cm)	Total Hg (μg·g^{-1})	Count	Length (cm)	Total Hg (μg·g^{-1})	Count	Length (cm)	Total Hg (μg·g^{-1})	RelC*
Blacknose shiner							6	6.4 (5.2–6.6)	0.08 (0.07–0.19)	ND
Spottail shiner	106	7.3 (6.7–12.5)	0.08 (0.02–0.35)	7	7.4 (4.2–8.2)	0.08 (0.05–0.15)	24	5.4 (4.9–7.3)	**0.03 (<DL–0.06)**	16.8
Northern redbelly dace	26	8.0 (4.6–10.0)	0.09 (0.01–0.43)				21	5.1 (4.0–7.8)	0.16 (0.07–0.24)	7.1
Finescale dace	11	8.3 (6.3–11.3)	0.13 (0.04–0.40)				1	5.0	0.17	ND
Bluntnosed minnow							50	5.9 (5.0–7.8)	**0.05 (<DL–0.09)**	ND
Fathead minnow	20	7.9 (7.4–8.6)	0.06 (0.03–0.12)				8	5.7 (4.3–7.5)	0.01 (<DL–0.21)	ND
Flathead chub	10	27.0 (20.5–31.2)	0.11 (0.05–0.18)							ND
Northern pikeminnow	190	31.0 (10.7–54.4)	0.33 (0.02–1.99)	8	45.5 (24.3–57.7)	0.49 (0.13–1.79)				2.1
Blacknose dace	258	6.0 (2.7–8.2)	0.26 (0.05–1.05)				5	3.7 (3.5–5.1)	**0.01 (<DL–0.02)**	0.3
Longnose dace	11	11.2 (5.7–25.9)	0.19 (0.06–0.64)				35	7.5 (4.5–10.9)	0.13 (0.06–0.43)	8.9
Creek chub	145	10.9 (5.2–29.0)	0.13 (0.03–0.82)				81	11.3 (4.2–18.0)	0.11 (0.03–0.32)	2.1
Fallfish	126	12.5 (7.6–59.9)	0.15 (0.03–1.63)				25	15.1 (8.9–31.9)	0.08 (0.02–0.79)	2.4
Northern pike	52 881	60.7 (9.1–137.8)	0.38 (<DL–10.9)	584	67.4 (23.6–122.1)	0.56 (0.04–3.40)	182	49.8 (8.0–91.1)	0.18 (0.02–1.62)	41.4
Muskellunge	132	77.3 (11.9–206.2)	0.46 (0.04–3.20)				3	44.6 (8.6–50.5)	0.11 (0.02–0.13)	12.8
Chain pickerel	82	42.7 (18.6–71.3)	0.57 (0.13–1.58)	8	38.2 (26.4–56.0)	0.38 (0.15–0.54)				0.3
Banded killifish							57	8.1 (5.9–9.9)	0.18 (0.04–0.60)	<0.1
Burbot	2 524	53.6 (12.4–131.0)	0.30 (<DL–6.40)	72	54.0 (29.5–78.5)	0.30 (0.05–1.71)	12	62.5 (14.5–75.0)	0.12 (0.03–0.22)	64.6
Brook stickleback							2	4.3 (4.0–4.5)	0.13 (0.12–0.14)	ND
Threespined stickleback	2	5.1 (5–5.2)	0.10 (0.09–0.11)				4	4.2 (3.4–5.6)	0.15 (0.11–0.21)	ND
Ninespined stickleback							8	4.9 (4.2–5.7)	0.17 (0.02–0.34)	ND
Goldeye	1 765	35.7 (14.3–51.8)	0.31 (<DL–3.50)				11	27.6 (25.4–33.1)	0.15 (0.09–0.21)	24.0
Mooneye	612	31.5 (13.4–51.6)	0.22 (0.01–1.30)	9	36.1 (34.4–49.0)	0.46 (0.22–0.68)	16	27.0 (21.5–37.1)	0.09 (0.06–0.20)	12.6
Yellow bullhead	3	25.5 (20.4–26.2)	0.27 (0.25–2.74)							ND
Brown bullhead	3 890	27.0 (11.0–43.0)	**0.10 (<DL–1.10)**	33	26.3 (14.0–36.3)	0.04 (0.01–0.39)	154	24.5 (11.1–39.0)	**0.14 (<DL–0.87)**	6.3
Channel catfish	1 741	46.5 (16.5–87.0)	0.20 (<DL–2.50)							10.4
Catfish (unspecified)	10	24.6 (16.2–31.7)	0.15 (<DL–0.21)							ND
Stonecat	3	19.1 (19.1–19.8)	0.47 (0.21–0.47)							ND
Longnose gar	9	79.0 (53.3–93.0)	0.58 (0.06–1.80)							ND
White perch	931	21.7 (10.2–32.1)	0.10 (<DL–1.37)	95	24.3 (11.7–36.7)	0.86 (0.08–2.30)	4	21.8 (9.0–25.3)	0.55 (0.33–0.87)	1.8
White bass	2 304	30.4 (9.7–44.3)	0.16 (<DL–2.20)				7	18.0 (12.0–23.0)	**0.11 (<DL–0.39)**	3.0
Striped bass	10	41.3 (34.3–72.8)	0.34 (0.11–0.89)	2	36.4 (35.4–37.3)	0.36 (0.28–0.44)				ND
Rainbow smelt	295	18.5 (9.2–34.0)	0.19 (<DL–1.41)				662	15.0 (5.0–35.1)	0.08 (<DL–0.84)	12.1
Yellow perch	12 557	20.4 (4.0–50.6)	0.14 (<DL–2.70)	44	17.3 (4.6–28.1)	0.27 (0.02–2.67)	2058	10.3 (3.3–32.1)	0.11 <DL– 1.60)	21.6
Perch (unspecified)	335	24.6 (12.4–34.0)	0.15 (<DL–0.62)							1.1
Sauger	3 881	32.3 (11.5–65.0)	0.43 (<DL–4.30)	60	26.3 (16.6–34.7)	0.21 (0.08–0.60)	18	27.4 (20.4–33.0)	0.35 (0.22–1.03)	18.5
Walleye	64 898	46.8 (6.7–101.0)	0.41 (<DL–10.4)	165	46.3 (17.3–66.4)	0.41 (0.03–1.88)	90	43.8 (17.3–69.2)	0.36 (<DL–1.64)	28.9
Trout perch	13	5.8 (5.0–12.4)	0.14 (0.03–0.29)				4	5.7 (5.4–9.3)	0.07 (0.06–0.11)	ND
Cisco (lake herring)	4 576	29.0 (8.1–63.3)	0.14 (<DL–1.63)	26	26.9 (13.3–41.4)	0.10 (0.05–0.41)	88	21.0 (8.0–48.2)	0.11 (<DL–0.58)	26.7
Arctic cisco	3	44.5 (43.9–45.5)	0.08 (0.06–0.10)							ND
Lake whitefish	16 779	43.1 (5.2–99.7)	0.09 (<DL–3.73)	1573	40.3 (15.1–60.1)	0.17 (<DL–2.40)	215	34.5 (11.5–50.2)	0.07 (0.01–0.92)	65.2
Dwarf lake whitefish	203	18.0 (12.3–56.8)	0.21 (0.11–1.36)							ND
Bloater	581	27.3 (7.8–41.6)	0.08 (<DL–0.72)							0.7
Deepwater cisco	67	28.0 (14.9–35.6)	0.18 (0.07–0.46)							0.6
Broad whitefish	221	52.2 (38.3–63.9)	0.05 (<DL–0.22)							2.8
Least cisco	24	23.0 (14.4–27.2)	0.04 (0.03–0.12)							ND

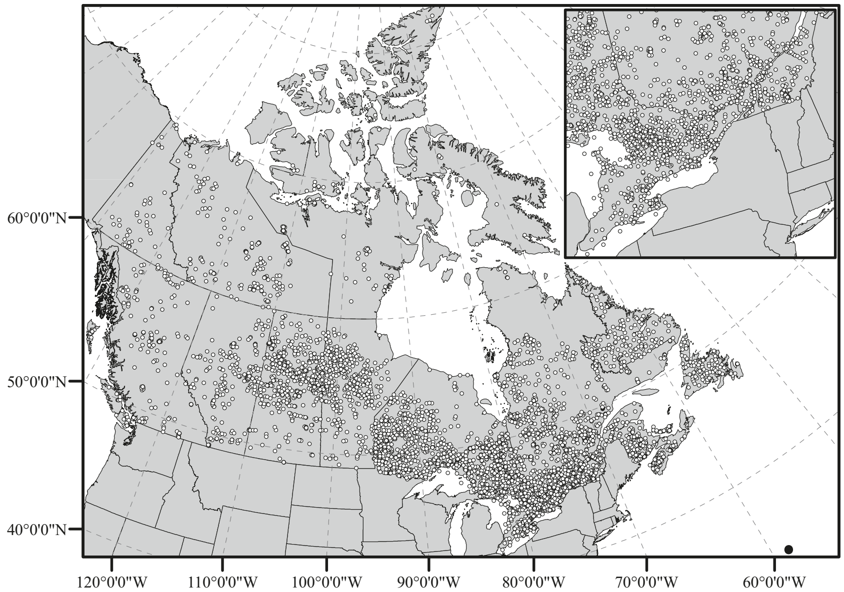
Table 1 (*concluded*).

	Skinless fillet			Skin-on fillet			Whole fish			
Species	Count	Length (cm)	Total Hg (μg·g⁻¹)	Count	Length (cm)	Total Hg (μg·g⁻¹)	Count	Length (cm)	Total Hg (μg·g⁻¹)	RelC*
Whitefish (unspecified)	170	42.3 (16.0–89.4)	0.08 (<DL–0.56)							37.4
Splake	357	43.7 (13.5–79.1)	0.17 (<DL–2.60)							2.8
Tiger trout	1	10.9	0.03							ND
Cutthroat trout	155	30.7 (8.1–57.8)	0.20 (0.01–1.30)				26	20.9 (14.0–28.4)	0.17 (0.03–0.32)	4.4
Pink salmon	269	43.0 (31.2–59.7)	**0.07 (<DL–0.92)**							0.7
Chum salmon	4	64.6 (61.0–67.1)	0.06 (0.05–0.07)							ND
Coho salmon	1 479	63.0 (4.4–102.0)	0.14 (<DL–1.72)	55	68.0 (29.8–85.0)	0.17 (<DL–0.29)				14.2
Rainbow trout	3 014	54.0 (6.8–100.5)	0.11 (<DL–0.94)	172	36.0 (10.0–101.9)	0.10 (<DL–1.95)	57	27.1 (7.6–29.0)	**0.01 (<DL–0.34)**	28.4
Rainbow trout (anadromous)	30	42.5 (25.0–60.3)	0.11 (0.03–1.00)							0.2
Kokanee	40	52.0 (19.6–74.8)	0.03 (<DL–0.16)	24	23.9 (13.7–48.2)	0.06 (0.01–0.15)				0.7
Sockeye salmon	38	60.3 (12.5–75.6)	0.05 (<DL–0.07)							0.7
Chinook salmon	1 857	78.4 (36.3–123.3)	0.22 (<DL–0.97)	125	88.0 (36.3–108.4)	0.23 (0.06–0.44)				8.2
Round whitefish	853	37.3 (9.7–58.3)	**0.06 (<DL–1.17)**	92	27.6 (19.5–44.1)	0.07 (0.03–0.36)	41	24.5 (14.7–29.8)	0.05 (0.02–0.08)	41.2
Mountain whitefish	524	32.3 (5.1–51.7)	0.06 (<DL–0.33)	62	39.7 (19.2–47.9)	**0.07 (<DL–0.17)**	12	41.9 (33.4–47.6)	0.07 (0.02–0.40)	9.8
Atlantic salmon	122	35.7 (9.8–58.4)	0.19 (0.03–0.81)	16	31.9 (22.6–50.3)	0.53 (0.04–2.17)	9	38.9 (21.6–45.1)	**0.21 (<DL–0.47)**	5.5
Atlantic salmon (landlocked)	505	32.7 (11.5–66.0)	0.29 (<DL–2.53)	382	26.7 (10.8–58.6)	0.24 (<DL–2.30)				8.5
Brown trout	1 315	53.0 (8.9–80.0)	0.17 (<DL–1.40)	21	58.8 (50.8–74.5)	0.17 (0.09–0.27)				25.1
Arctic char (anadromous)	384	68.5 (19.0–95.2)	0.05 (<DL–1.00)	12	25.2 (18.6–46.9)	0.04 (<DL–0.08)	15	23.5 (19.0–28.5)	0.18 (<DL–0.52)	2.6
Arctic char (landlocked)	643	37.1 (10.8–85.3)	0.16 (<DL–3.40)	255	47.8 (17.1–75.3)	0.11 (<DL–1.75)	25	15.9 (10.2–32.6)	0.22 (0.09–1.93)	30.0
Arctic char (dwarf)	16	19.6 (14.5–27.0)	0.23 (0.09–0.52)							ND
Bull trout	66	30.4 (8.3–83.4)	0.07 (0.01–0.84)							3.9
Brook trout	3 889	26.0 (4.7–89.9)	0.14 (<DL–3.33)	1052	24.3 (8.6–61.2)	0.10 (<DL–1.60)	404	17.2 (7.5–40.7)	0.10 (<DL–0.80)	20.9
Dolly Varden	191	15.0 (5.6–60.6)	0.02 (<DL–0.56)	10	19.8 (8.1–31.1)	**0.04 (<DL–0.09)**				6.6
Lake trout	21 324	54.1 (4.5–121.7)	0.28 (<DL–10.0)	673	56.0 (12.4–106.2)	0.42 (<DL–2.72)	95	41.8 (7.5–70.1)	0.11 (<DL–0.81)	65.2
Lake trout (humper)	44	58.4 (24.4–82.6)	0.45 (0.06–1.40)							ND
Siscowet	69	58.3 (22.3–91.4)	0.49 (0.10–1.60)							ND
Inconnu	171	76.9 (38.4–107.9)	0.14 (<DL–0.59)	1	73.9	0.97				6.4
Arctic grayling	201	35.9 (7.5–54.1)	0.05 (<DL–0.47)	22	36.1 (21.2–44.5)	0.08 (0.02–0.22)	27	35.0 (7.6–44.3)	**0.11 (<DL–0.19)**	25.7
Freshwater drum	1 387	33.6 (11.0–72.3)	0.17 (<DL–2.00)							6.1

Note: Median and range (minimum–maximum) of lengths presented as total length equivalents (cm). Median and range (minimum–maximum) of total Hg concentrations (μg·(g wet mass)⁻¹) are presented. RelC is a relative measure of spatial coverage represented by the 95% MCP expressed as a percentage of Canada for species sampled at a minimum of 10 or more locations. Note that data from contaminated sites, reservoirs, and identified downstream water bodies are not included here. Bolded median and ranges of total Hg indicate species–sample types where >10% of records were reported to be at or below the detection limit (DL).

*ND refers to species for which insufficient numbers of locations are present for computation of minimum convex polygons.

Fig. 1. Distribution of freshwater locations across Canada where fish have been sampled for Hg analysis between 1967 and 2010 (n = 5202). Note that reservoirs and designated downstream water bodies, locations impacted by Hg point-source pollution, and estuarine or marine locations are not included here. Inset panel shows central and southern Ontario and southwestern Quebec in greater detail.



rived an estimate of spatial scale for each species (where possible) by calculating the minimum convex polygon area (MCP; Mohr 1947) bounding georeferenced locations where each species has been recorded in the CFMD. Both the 100% (all locations) and 95% (95% of locations) MCP were computed using the home range tools extension in ArcGIS (Rodgers et al. 2007). The floating mean algorithm was used to derive the 95% MCP to reduce the bias from the inclusion of rare events (Horne and Garton 2006). Areas for each MCP were subsequently calculated using a polygon mask defined by the Canadian land mass and are expressed as a relative percentage of coverage based on the area of the masking polygon (10 051 388 km^2). Although less precise and statistically robust compared with kernel density estimation (Hemson et al. 2005), the purpose of the derived MCP here is to provide a relative index of spatial coverage and variation among species listed in Table 1, not to calculate exact range limits for each species. All calculations were conducted after projection of geographic data to a modified Canada Albers Equal-Area Conic projection (latitude of origin: 49°N; reference latitudes: 49°N and 77°N; central meridian: 96°W; units: km).

To examine the influence of spatial scale on Hg concentrations in fish, we extracted the variance components from linear mixed-effects models (Pinhiero and Bates 2000) fit to each species with more than 1000 records in the database (see Supplementary Table S4[1]). Briefly, mixed-effects models were fit in R (R Development Core Team 2010) using the "lme4" package (Bates and Maechler 2010) and restricted maximum likelihood methods. WATERBODY_ID (i.e., location) and year of collection (rescaled as SAMPLE_YEAR minus minimum of SAMPLE_YEAR) were treated as random effects, while portion type, water body type, and length (centered on species mean length) were treated as fixed effects. Variation due to measurement, analytical, sample processing, or data treatment error (e.g., converting from dry to wet mass concentrations) we termed "sampling error" and incorporated into the residual error. Formulated in this manner, this model allowed us to estimate both the among-site and among-year variability in Hg concentrations in a standard-size fish (based on the species-specific mean; see Supplementary Table S4[1]) while controlling for variation in Hg concentration due to fish size, water body type and sample portion type. Direct assessments of variation due to sample portion type and water body type were not evaluated because of the highly unbalanced dataset (see Table 1) and the difficulty in estimating precise variance components for random effects with few levels (<5–6; Crawley 2002, p. 670). Interannual variation within sites (e.g., a site × year interaction) was also not evaluated directly because the majority of sites in the CFMD are sampled only once over the period of record.

Results and discussion

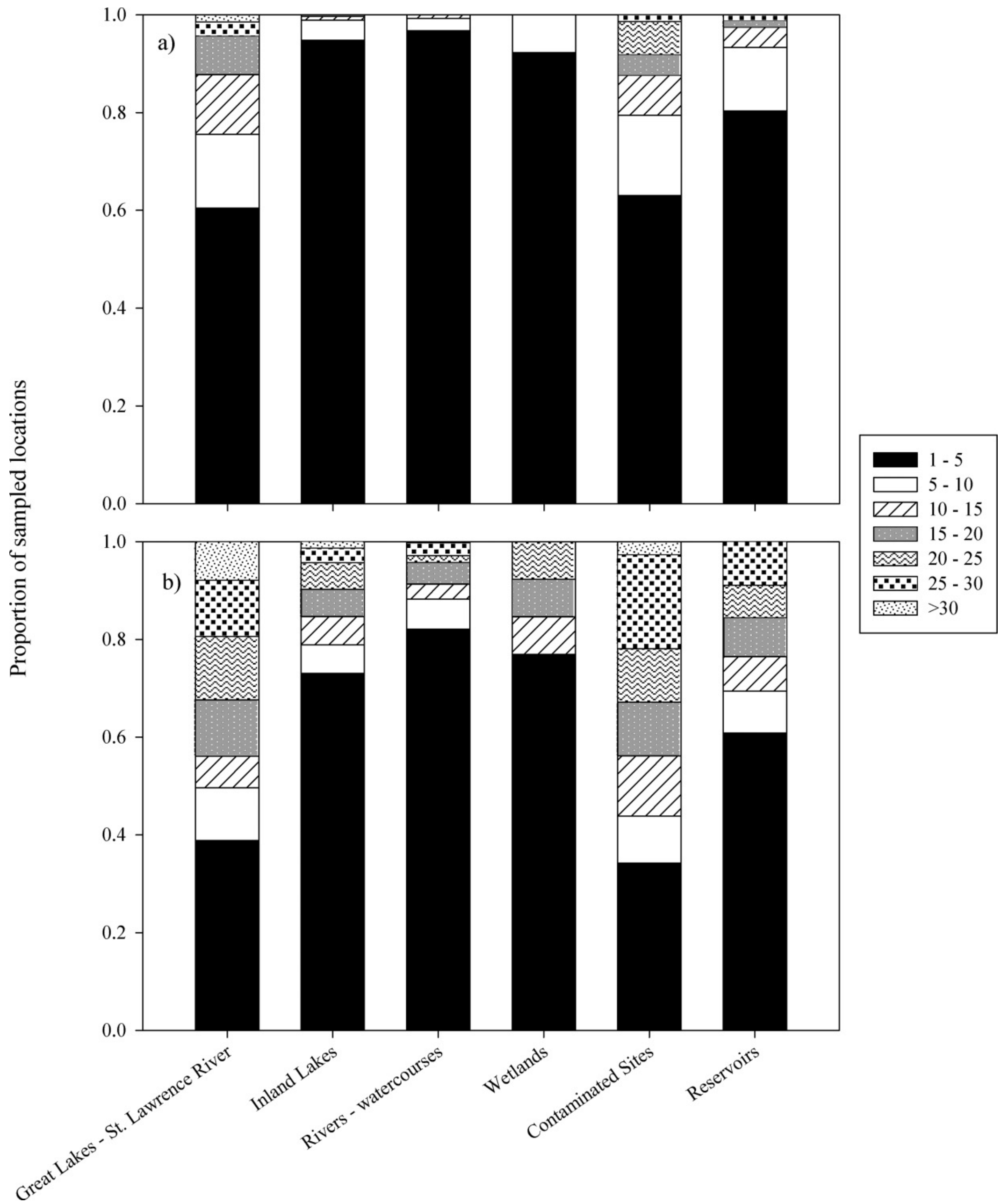
In total, 406 542 records were initially made available, but only 387 872 records were retained in the CFMD after quality assurance checks (see Methods). Of the nearly 18 000 records excluded, approximately 13 000 records were duplicates that resulted primarily from interagency data sharing. The remaining excluded records could not be adequately georeferenced, did not contain suitable taxonomic information, or did not have associated measures of total Hg, length, or mass or had uncertainty regarding reporting of units (wet or dry mass).

Nearly one-third (29%; 110 597) of the included records originated from locations known to be affected by point-source Hg inputs or newly impounded reservoirs and associated water bodies (see Methods). However, this large volume of records represented only about 6% of the total locations sampled, and given the elevated Hg concentrations typically observed in these environments, the potential for bias in the summary statistics was considerable. After exclusion of records from reservoirs and contaminated sites (see Methods), 269 622 records remained, providing Hg data that covered 43 years (1967–2010) and spanned the entire country (Fig. 1). These records represent measurements of Hg concentrations from 104 species (including landlocked and anadromous types, species identified to genus only, and two hybridized species; Table 1) and eight different portion types collected from 5202 unique locations across Canada (Fig. 1).

Spatial and temporal scope of fish Hg sampling in Canada

While almost 9% of Canada is covered by water, surface water is irregularly distributed across Canada (Supplementary Fig. S2[1]). The geographical distribution of locations represented in the CFMD follows a similar pattern, as the mean fraction of surface water was positively correlated with the number of sampled locations at the hydrological work unit level (Spearman ρ = 0.27, p < 0.001, n = 1157; Supplementary Fig. S3[1]). Some inland regions

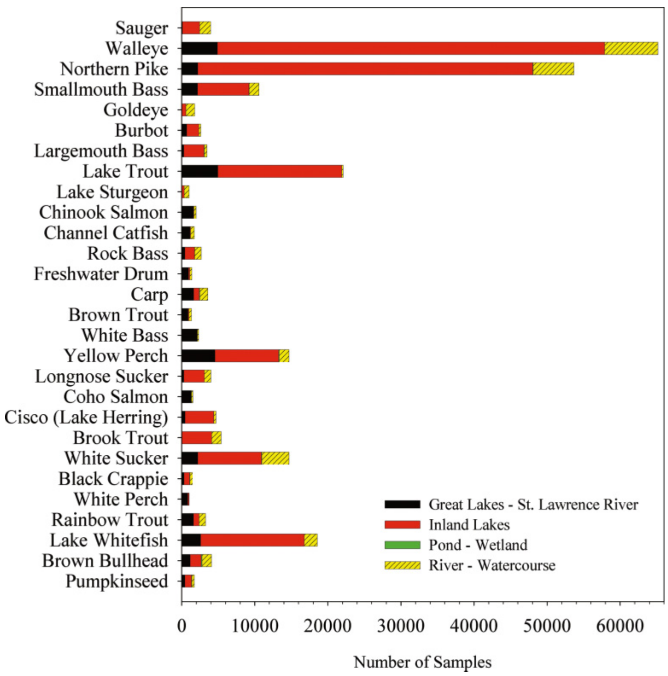
Fig. 2. Proportion of sampled locations displaying (*a*) resampling count (number of years sampled) and (*b*) the maximum length of temporal record (in years) for freshwater locations in the database. Sample locations are grouped by major class of water body. Note that contaminated sites, newly impounded reservoirs, and downstream water bodies have been resampled more intensively and have longer temporal records, but are excluded from the data summary and analysis in this paper (see Methods).



are clearly undersampled relative to surface water availability (Northwest Territories, Nunavut, northern Ontario and Quebec, and the interior of British Columbia), but the relatively similar geographical distribution between the number of sampled locations and available surface water indicates that the locations in the database provide a reasonable geographic representation at a national scale.

Sampling effort, represented here by the number of sampling events (an event is defined by a location–year combination) and the number of records (proxy for number of fish sampled), varied considerably both spatially and temporally, with hundreds to thousands of fish collected in some regions, while in others a much smaller number of records exist or no sampling has occurred (Supplementary Figs. S4 and S5[1]). For inland locations, most regions have received a similar level of sampling intensity, although for many locations (>90%) this represents limited temporal sampling (1–5 times over 43 years; Fig. 2). In contrast, sampling in the Great Lakes and St. Lawrence River, large inland lakes such as Lake Winnipeg, Great Slave Lake, and Lake Athabasca, and larger lakes in northern Saskatchewan and Manitoba has been comparatively more intense (Supplementary Figs. S4 and S5[1]). Consequently, both the number of sampling events and the total number of records sampled were positively correlated to the fraction of surface water available within each NHN work unit (Spearman ρ = 0.27 and 0.28, p < 0.001, n = 1157 respectively; Supplementary Figs. S4 and S5[1]), likely reflecting the importance of these regions for both commercial and subsistence fishing (DFO 2011).

Fig. 3. Count of records (aggregated by major water body types) for species within the database that contributed >1000 records between the years 1967 and 2010 from freshwater locations across Canada. Species are arranged from top to bottom in order of decreasing median total Hg concentration. Records from contaminated water bodies, reservoirs, and downstream locations are not included here.



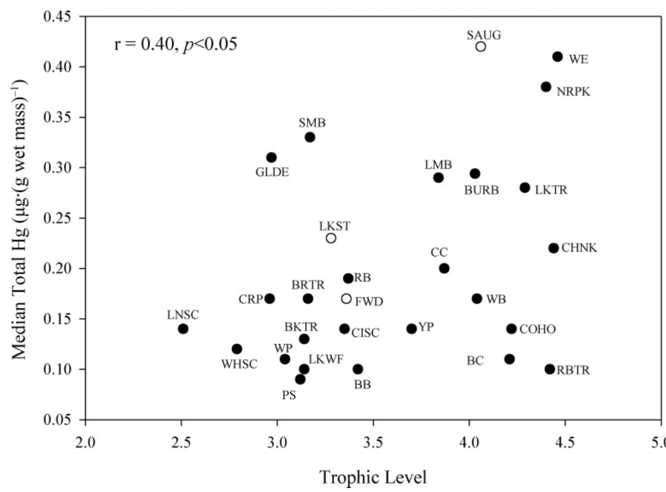
Hg concentrations in fish collected across Canada

Despite data being available for 104 species (Table 1), 28 species alone each contributed more than 1000 records. The representation of these species is heavily skewed towards eight key species (Fig. 3). These key species include top piscivores such as walleye (*Sander vitreus*), northern pike (*Esox lucius*), lake trout (*Salvelinus namaycush*), and smallmouth bass (*Micropterus dolomieu*), representing 24.1%, 19.6%, 7.9%, and 3.9%, respectively, and lower trophic level species such as lake whitefish (*Coregonus clupeaformis*), white sucker (*Catostomus commersonii*), and yellow perch (*Perca flavescens*), representing 6.2%, 4.7%, and 4.7%, respectively. The relative abundance of these species in the dataset is perhaps not entirely unexpected given the status of these species as biomonitors for contaminants, prized sport fish, and species of significance for indigenous and commercial fisheries.

Of the remaining species with ≥1000 total records, only longnose sucker (*Catostomus catostomus*), brook trout (*Salvelinus fontinalis*), brown bullhead (*Ictalurus nebulosus*), cisco (*Coregonus artedi*), sauger (*Sander canadensis*), largemouth bass (*Micropterus salmoides*), carp (*Cyprinus carpio*), rainbow trout (*Oncorhynchus mykiss*), and burbot (*Lota lota*) accounted for more than 1% of total records (Table 1; Fig. 3).

Total Hg levels across all fish species ranged from below detection (highest reported detection limit is 0.11 μg·(g wet mass)$^{-1}$) to 10.9 μg·(g wet mass)$^{-1}$ (Table 1). For the 28 species examined in detail, despite substantial variability in numbers and sizes of fish collected and the geographic scale of species occurrences, median Hg concentrations increased significantly with trophic level (Pearson r = 0.40, p < 0.05, n = 28; Fig. 4), with the highest Hg concentrations consistently found in predatory species (Fig. 5). The species with the highest median concentrations included piscivorous and omnivorous species such as sauger (*Sander canadensis*; 0.42 μg·g^{-1}), walleye (0.41 μg·g^{-1}), northern pike (0.38 μg·g^{-1}), smallmouth bass (0.33 μg·g^{-1}), goldeye (*Hiodon alosoides*; 0.31 μg·g^{-1}), and burbot (*Lota lota*; 0.30 μg·g^{-1}), while the species with the lowest median concentrations included yellow perch (0.14 μg·g^{-1}), longnose sucker (0.14 μg·g^{-1}), brook trout (*Salvelinus fontinalis*; 0.13 μg·g^{-1}), white sucker (0.12 μg·g^{-1}), rainbow trout (*Oncorhynchus mykiss*; 0.11 μg·g^{-1}), lake whitefish (0.10 μg·g^{-1}), and brown bullhead (*Ictalurus nebulosus*; 0.10 μg·g^{-1}) (Fig. 5).

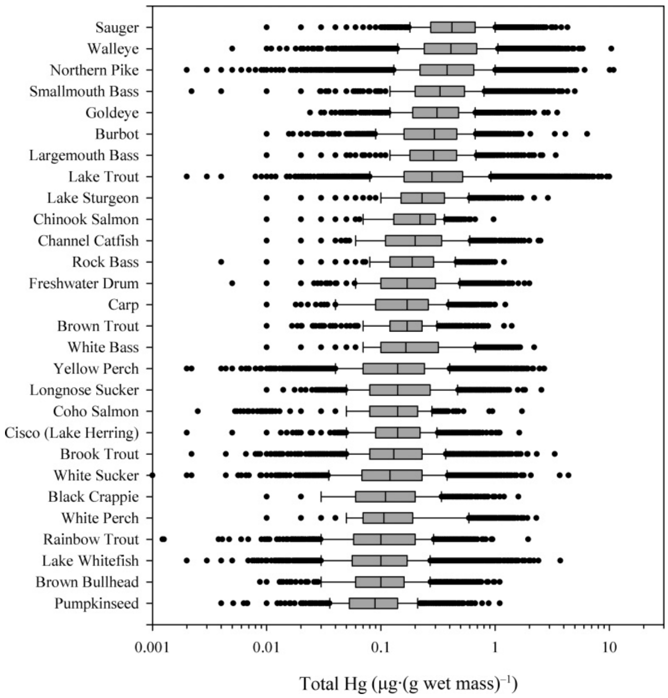
Fig. 4. Scatterplot of species-specific median total Hg concentration (μg·(g wet mass)$^{-1}$) against trophic level for species with >1000 total records. Records from contaminated water bodies, reservoirs, and downstream locations are not included here. Trophic level is as reported in FishBase (Froese and Pauly 2011) based on dietary studies (closed circles) except for sauger (SAUG), freshwater drum (FWD), and lake sturgeon (LKST) (open circles), whose values are derived based on a Monte Carlo procedure (see Froese and Pauly 2011 for details). Other species codes indicated on the figure are as follows: RB (rock bass), LNSC (longnose sucker), WHSC (white sucker), CISC (cisco), LKWF (lake whitefish), CRP (carp), NRPK (northern pike), GLDE (goldeye), BB (brown bullhead), CC (channel catfish), PS (pumpkinseed), BURB (burbot), SMB (smallmouth bass), LMB (largemouth bass), WB (white bass), WP (white perch), YP (yellow perch), BC (black crappie), RBTR (rainbow trout), CHNK (Chinook salmon), COHO (coho salmon), BKTR (brook trout), LKTR (lake trout), BRTR (brown trout), WE (walleye). Species scientific names can be found in Supplementary Table S3[1].



Considerable range in Hg concentration is apparent both among and within species (Table 1; Fig. 5). Yet compared with the high concentrations measured in fish from grossly polluted systems (>5 μg·g^{-1}; Fimreite et al. 1971; Scott 1974) or newly impounded reservoirs (>3 μg·g^{-1}; Verdon et al. 1991; Bodaly et al. 2007), Hg concentrations from fish collected in unimpacted aquatic systems rarely exceeded these levels (Fig. 3). Some of these comparably high Hg concentrations are from fish collected at locations deemed to be separate but hydrologically connected (i.e., upstream) from reservoirs or contaminated sites. These few very high Hg concentrations are clearly anomalous from other individual fishes collected at the same location and likely reflect immigration of fish from more impacted locations (data not shown).

There are also several locations within the CFMD where elevated Hg concentrations in fish are thought to be driven by the enhanced availability of MeHg derived from the weathering or degassing of geogenic Hg sources such as tetrahedrite, cinnabar, or sulphide-rich Proterozoic black shales (Shilts and Coker 1995; Rasmussen et al. 1998). However, in the absence of anthropogenic disturbance (i.e., mineral extraction and processing), Hg concentrations in fish from these locations rarely exceeded the mean levels found in contaminated sites or newly flooded reservoirs (Shilts and Coker 1995; Weech et al. 2004; Ethier et al. 2008, but see MacCrimmon et al. 1983).

Fig. 5. Boxplot of total Hg concentration (μg·(g wet mass)$^{-1}$) for species with >1000 records between the years 1967 and 2010 from freshwater locations (excluding contaminated water bodies, reservoirs, and downstream water bodies) across Canada. Species are sorted from top to bottom by decreasing median total Hg concentration. Lower and upper limits of the box represent the 25th and 75th percentiles. The solid line represents the median concentration, while lower and upper ends of the whiskers represent the 10th and 90th percentiles, respectively.



Influence of spatial scale on variation in fish Hg concentration

The geographic coverage represented by different species in the CFMD varied considerably (Fig. 6; Table 1). Thirty of 104 species had a 95% MCP that represented at least 10% in terms of relative coverage, but only seven species (lake whitefish, lake trout, burbot, longnose sucker, northern pike, walleye, and yellow perch) had geographic coverage or representation that could be considered reasonably close to a national scale (Table 1). Thus, for the majority of species summarized in Table 1, Hg concentrations and ranges need to be cautiously interpreted, as they only represent a small regional area.

Previous examinations of regional datasets have indicated that the greatest variation in standard-size fish Hg concentrations (within species) generally occurs at the water body (or location) level (Kamman et al. 2005; Lockhart et al. 2005). For key species with >1000 records in the CFMD, this pattern generally holds true; as the geographic range of sampled locations increases, so too does the among-site percentage of total variance (Pearson $r = 0.58$, $p < 0.001$; Fig. 7*a*). Among-site variance appears to reach a maximum between 70% and 75% of total variance (Fig. 7*a*; Table S4[1]). Although the among-year and residual variance increases as the among-site variance declines, no clear pattern emerges in among-year variation, and it tends to remain low relative to among-site variation except for a few species and rarely exceeds the residual variance (Fig. 7*b*; Table S4[1]). For some species, residual geographic variation may be apportioned into among-year variance if exclusive sampling of a particular species occurred in very different regions in different years. Additional residual geographic variability may also be unaccounted for if different portion types are restricted to specific geographic areas or water body types (Supplementary Fig. S6[1]). The remaining variation in Hg concentrations is presumably due to water body type, the type of sample portion analyzed, and the factors that affect the residual error (see Methods) that we are unable to assess using this approach.

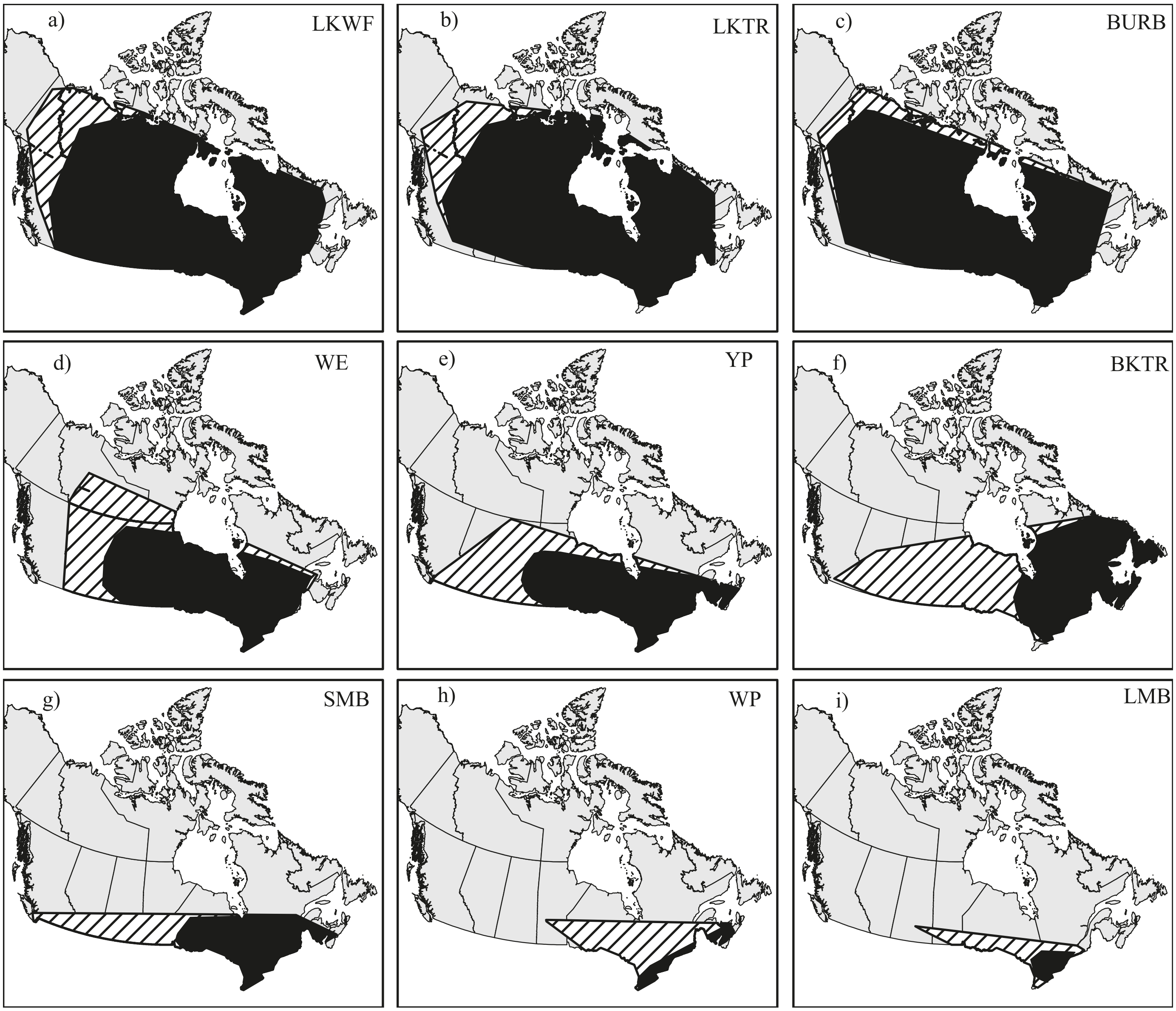
It is possible that this observed pattern is simply an artefact of including several species that are primarily sampled from a small geographic region where Hg concentrations do not vary significantly. For example, Chinook salmon (*Oncorhynchus tshawtyshca*), coho salmon (*Oncorhynchus kisutch*), freshwater drum (*Aplodinotus grunniens*), channel catfish (*Ictalurus punctatus*), brown trout (*Salmo trutta*), white bass (*Morone chrysops*), and white perch (*Morone americana*) have been sampled primarily from the Laurentian Great Lakes and have a relatively low proportion of total variation associated with among-site variability (Figs. 3 and 7*a*; Table 1). Hg concentrations in these species have been consistently low since the 1980s as evidenced by their general exemption from Hg-related consumption advisories (Bhavsar et al. 2011). However, we note that for other species sampled from relatively small geographic areas (i.e., small 95% MCP) not dominated by sites in the Laurentian Great Lakes (e.g., lake sturgeon (*Acipenser fulvescens*), goldeye (*Hiodon alosoides*), rock bass (*Ambloplites rupestris*), black crappie (*Pomoxis nigromaculatus*), and pumpkinseed (*Lepomis gibbosus*)), the among-site variance is still generally low and the same general pattern of increasing among-site variance with increased geographic coverage appears to hold (Fig. 7*a*). The increase in among-site variability with geographic scope has been observed at subcontinental scales (Kamman et al. 2005) and in this compilation of data appears to hold across different species and trophic levels. This is suggestive of relatively broad gradients of MeHg bioavailability, presumably reflecting an increase in the variability of atmospheric Hg deposition (Engle et al. 2010), landscape sensitivity to Hg transport and methylation (Driscoll et al. 2007), and other factors that affect the magnitude and extent of MeHg bioaccumulation in aquatic food webs as the geographic range of investigation increases (Evers et al. 2007).

Gaps, limitations, and implications for effective assessment of Hg regulations

Although there are remaining uncertainties and data gaps in both local and global estimates of Hg emission inventories (Pirrone et al. 2010) and uncertainties related to quantifying net Hg deposition over large spatial scales (Prestbo and Gay 2009), reductions in atmospheric Hg loads are expected to lead to reductions in net Hg deposition and ultimately a reduction in MeHg generation and subsequent bioaccumulation of MeHg in fish (Munthe et al. 2007; Harris et al. 2007). However, the ability to comprehensively assess such changes at a national level within Canada remains a challenging task.

The detection of changes in fish Hg in response to changes in atmospheric Hg deposition on a national scale is difficult for a number of reasons. While there is a paucity of Hg deposition monitoring stations in Canada (Miller et al. 2005), the general consensus from assessments of long-term records of Hg deposition, whether measured directly or inferred from sediment cores, suggests that both year-to-year and long-term variability in atmospheric Hg deposition is relatively small, although this depends on the location and duration of the record in question (~±2% per year; Swain et al. 1992; Prestbo and Gay 2009). Recent manipulative experiments at the ecosystem level have demonstrated that fish Hg concentrations can respond quickly (on the order of years) to changes in atmospheric Hg deposited directly to the water body surface (Hrabik and Watras 2002; Harris et al. 2007; Orihel et al. 2007), but the magnitude and duration of the response will ultimately depend both on the extant characteristics of the system in question and other factors that may affect the more gradual export of Hg stored in the upland and lowland regions of the catchments, the availability of Hg compounds for methylation, and the

Fig. 6. Spatial plots of 100% and 95% minimum convex polygons (MCP) for (*a*) lake whitefish (LKWF), (*b*) lake trout (LKTR), (*c*) burbot (BURB), (*d*) walleye (WE), (*e*) yellow perch (YP), (*f*) brook trout (BKTR), (*g*) smallmouth bass (SMB), (*h*) white perch (WP), and (*i*) largemouth bass (LMB) sorted in order from largest 95% MCP to smallest 95% MCP. Black regions in each panel represent the area bounded by the 95% MCP, while the hatched area represents the area bounded by the 100% MCP for each species. Grey area represents regions where selected species do not have a recorded occurrence in the database, but may nonetheless be present. Locations representing contaminated water bodies, reservoirs, and downstream water bodies are not included here.

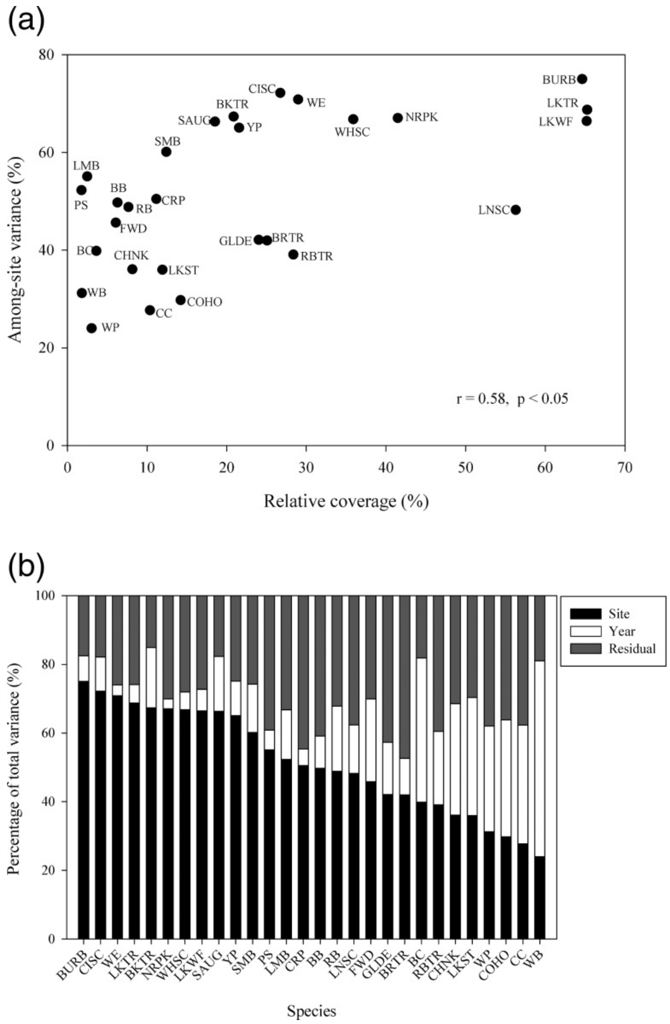


subsequent bioaccumulation of MeHg up the food chain (Harris et al. 2007).

Further confounding the ability to adequately detect changes in fish Hg concentrations in the context of gradual changes in atmospheric Hg deposition is the interaction of a number of ecosystem factors that can strongly influence the extent and magnitude of Hg bioaccumulation in fish. Changes in land use or disturbance within catchments such as forest fires (Garcia and Carignan 2005; Kelly et al. 2006), alteration of food web structure by exotic species (Southward Hogan et al. 2007; Lepak et al. 2009), eutrophication (Pickhardt et al. 2002; Essington and Houser 2003), and, under some circumstances, intensive fishing (Verta 1990), can alter fish Hg concentrations on the time scale of years. Other changes, such as increases or decreases in acidic deposition (Hrabik and Watras 2002; Drevnick et al. 2007) or temperature increases (Carrie et al. 2010), may alter fish Hg concentrations over longer periods of time (decades). Consequently, attributing changes in fish Hg across both gradients in time (short versus long) and space (single site versus synoptic surveys) in relation to changes in atmospheric Hg inputs will remain a challenging task, particularly in the absence of corroborative evidence to rule out other potential factor(s).

A second important issue is that, in Canada, the most intensively sampled water bodies and therefore those with the most frequent and longest time series of fish Hg measurements are those that have historically been impacted by either point-source Hg pollution such as the English-Wabigoon River system in northwestern Ontario (Kinghorn et al. 2007) and the Great Lakes areas of concern (Weis 2004) or the creation of large hydroelectric reservoirs in northern Manitoba, Quebec, and Labrador (Verdon et al.

Fig. 7. (*a*) Scatterplot showing the percentage of total variation in Hg concentration in a standard-length fish attributed to among-site variation for species with >1000 total records as a function of relative coverage (95% minimum convex polygon area expressed as a percentage of Canadian land mass area). Species codes as in Fig. 4. (*b*) Percentage of total variation in standard-length fish Hg concentrations attributable to among-site, among-year, and residual components for each of the 28 species in the CFMD with >1000 records. Species codes as in Fig. 4.



1991; Bodaly et al. 2007; Anderson 2011) (Fig. 2). These sites have been extensively monitored both for the issuance of fish consumption advisories and to further elucidate mechanisms responsible for increases in fish Hg. While appropriate and useful for assessing the safety of fish consumption for recreational and subsistence fish consumers, these datasets are likely to be less useful for deciphering changes in fish Hg in relation to changes in atmospheric Hg deposition, because the initial load of Hg from point-source discharges and generation of MeHg during the inundation of terrestrial vegetation and soils likely far exceeds the input of Hg from the atmosphere in the absence of these disturbances (Kelly et al. 1997; Lockhart et al. 2000; Snodgrass et al. 2000). Moreover, fish Hg concentrations in these affected systems require significant temporal lags (>15–30 years) to return to concentrations comparable to initial conditions or unimpacted systems (Verdon et al. 1991; Bodaly et al. 2007; Kinghorn et al. 2007; Munthe et al. 2007; Anderson 2011).

Lastly, while we observed some degree of correspondence between the sampling effort documented to date and the inferred availability of surface water across Canada, and despite the substantial volume of data represented in the CFMD, the number of locations sampled represent only a fraction (approximately 0.5%) of available lakes with a surface area >0.1 km² estimated to be in Canada, one of the most water-rich countries in the world (~910 400 lakes; Minns et al. 2008). A synoptic estimate of rivers or watercourses on a national scale is not available, but we surmise that the degree of representation in the CFMD is probably quite similar (i.e., <1%). Consequently, it is difficult to ascertain to what degree the CFMD truly represents the range and distribution of fish Hg concentrations across the country. We recognize that most fish monitoring surveys (including those used in the construction of the CFMD) are rarely designed in a probabilistic framework that would allow for an unbiased assessment of regional or national patterns and trends (sensu Stahl et al. 2009). Thus the ability to extrapolate inferences from further analysis of the CFMD to regional or national scales is complicated by site selection and other intrinsic biases.

Despite these challenges, we believe that the CFMD can contribute to a greater understanding of fish Hg concentrations across Canada and provide a baseline for future monitoring and assessment with regards to potential effects of regulating Hg emissions. For example, the data within the CFMD likely subsume many of the factors known to affect Hg concentrations in fish on both short and long time scales (see above) and provide a means to incorporate such variability into regional analysis and interpretation of changes in fish Hg concentrations over time, as demonstrated by several recent efforts at subcontinental scales (Rasmussen et al. 2007; Monson 2009; Monson et al. 2011). Additionally, the CFMD may also be of use for designing and implementing an expanded national monitoring program. At present, the Clean Air Regulatory Agenda Freshwater Inventory and Surveillance of Mercury (CARA FISHg; M. Sekela, C.S. Eckley, A. Armellin, J. Syrgiannis, M. Keir, S. Backus, J. Pomeroy, B. McNaughton, M. Gledhill, D. Donald, and M. Neilson, unpublished data) is the only fish Hg monitoring program with national coverage in Canada, but it lacks the temporal history (initiated in 2008) and spatial extent covered by the CFMD. Regardless of the approach taken, a thorough assessment of program design and feasibility is required before a more comprehensive program can be initiated.

Synopsis and conclusions

To the best of our knowledge, the CFMD provides the most comprehensive summary of freshwater fish Hg measurements in Canada published to date. Although marine and (or) farmed fish species were outside the scope of this study, it may nonetheless be valuable to conduct a similar assessment given the widespread nature of Hg contamination. Regardless, it is clear that a wealth of information on representative predator (walleye, northern pike, lake trout, and smallmouth bass) and nonpredator (lake whitefish, white sucker, and longnose sucker) species presently exists within Canada. Initial assessment of major patterns and trends in the CFMD confirm the findings of many previous studies that reflect both consistent and widespread bioaccumulation and biomagnification of Hg in predatory species of fish, but also the importance of variability in landscape and aquatic ecosystem characteristics that contribute to variation in the extent and magnitude of Hg accumulation in fish.

The global nature of Hg contamination requires a broader scale of analysis and assessment to effectively assess the ability of regulatory activities to manage Hg pollution from the atmosphere. Through merging regional databases we are able to develop a comprehensive dataset with expansive coverage that will contribute to further assessment of Hg contamination of freshwater fishes across Canada. An ability to conduct a national assessment of Hg in freshwater fish in Canada is within reach, but careful consideration of sampling approaches to maximize efficiency and reduce uncertainties is required to establish a rigorous framework. Continued collaboration between different levels of government, academia, and industry is also encouraged. As national assessment capability increases, so too should the ability to evaluate the effectiveness of Hg regulatory activities in relation to Hg accumulation in fish on a national scale.

Disclaimer

The data for this initiative were provided under a limited research data-sharing agreement. Consequently, the entity "Canadian Fish Mercury Database" (CFMD) refers to a temporary amalgamation of shared and compiled datasets from willing contributors and various publicly available sources. For data not already in the public domain, ownership of provided data remains with and is the responsibility of the contributing individual, organization, or agency. The corresponding author(s) are not authorized to release data provided by all contributors, and queries regarding such data must be directed to the contributing individual, organization, or agency. A full list of data sources is provided in the online supplementary information (Table S1[1]).

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