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Assessing potential health risks to fish and humans using mercury concentrations in inland fish from across western Canada and the United States

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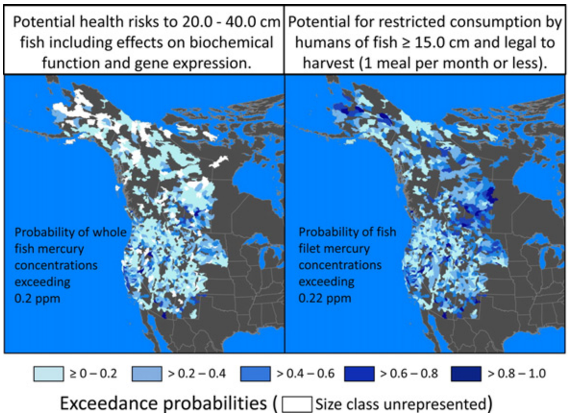
HIGHLIGHTS

- Fish and human health risks from Hg exist in western Canada and the United States.
- We used a hierarchical statistical model characterizing Hg risks and uncertainty.
- Potential health risk was heterogeneous across the region, and higher in some areas.
- Targeted monitoring could improve understanding and mitigation of Hg contamination.

GRAPHICAL ABSTRACT



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ABSTRACT

Fish represent high quality protein and nutrient sources, but Hg contamination is ubiquitous in aquatic ecosystems and can pose health risks to fish and their consumers. Potential health risks posed to fish and humans by Hg contamination in fish were assessed in western Canada and the United States. A large compilation of inland fish Hg concentrations was evaluated in terms of potential health risk to the fish themselves, health risk to predatory fish that consume Hg contaminated fish, and to humans that consume Hg contaminated fish. The

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probability that a fish collected from a given location would exceed a Hg concentration benchmark relevant to a health risk was calculated. These exceedance probabilities and their associated uncertainties were characterized for fish of multiple size classes at multiple health-relevant benchmarks. The approach was novel and allowed for the assessment of the potential for deleterious health effects in fish and humans associated with Hg contamination in fish across this broad study area. Exceedance probabilities were relatively common at low Hg concentration benchmarks, particularly for fish in larger size classes. Specifically, median exceedances for the largest size classes of fish evaluated at the lowest Hg concentration benchmarks were 0.73 (potential health risks to fish themselves), 0.90 (potential health risk to predatory fish that consume Hg contaminated fish), and 0.97 (potential for restricted fish consumption by humans), but diminished to essentially zero at the highest benchmarks and smallest fish size classes. Exceedances of benchmarks are likely to have deleterious health effects on fish and limit recommended amounts of fish humans consume in western Canada and the United States. Results presented here are not intended to subvert or replace local fish Hg data or consumption advice, but provide a basis for identifying areas of potential health risk and developing more focused future research and monitoring efforts.

1. Introduction

Mercury (Hg) is an important contaminant due to its widespread distribution and tendency to bioaccumulate in organisms to levels that impact the health of humans, wildlife, and ecosystems worldwide (Driscoll et al., 2007; Mergler et al., 2007). Hg enters the landscape through a variety of natural and anthropogenic pathways including volcanoes, forest fires, erosion, fossil fuel burning, waste incineration, mining operations, and cement production (Pirrone and Mason, 2009). Inorganic Hg can be converted into methylmercury, the form of Hg that can be taken up in living tissues as the contaminant moves through food webs (Harris et al., 2007; Power et al., 2002; [USEPA] United States Environmental Protection Agency, 2001). Methylmercury is a neurotoxin that can adversely affect fish, humans, and wildlife in a variety of ways, potentially impacting behavior, cognition, growth, reproduction, and survival (Depew et al., 2012; Mergler et al., 2007; Scheuhammer et al., 2007). Despite health concerns related to Hg contamination in fish, the benefits of fish consumption are well documented and have been postulated by some to outweigh the health risks from Hg exposure under certain conditions (Institute of Medicine of the National Academies et al., 2007; Knuth et al., 2003; Mergler et al., 2007).

A variety of Hg exposure benchmarks associated with potential fish and human health effects have been established, but these benchmarks can range widely due to a variety of factors. In fish, factors like species, gender, and age have been considered when determining Hg exposure benchmarks that may have health impacts on individuals and at the population level (see reviews in Depew et al., 2012; Sandheinrich and Wiener, 2011). Further, many different endpoints have been selected to evaluate effects from Hg exposure on fish including, but not limited to, effects on behavior, gene expression, growth, metabolism, gonadal somatic indices, and plasma and blood characteristics (see reviews in Depew et al., 2012; Sandheinrich and Wiener, 2011). In humans, similar to fish, gender and age can impact how Hg exposure affects the health of individuals consuming Hg contaminated fish, primarily affecting children during development and consequently a concern for women who are pregnant or intend to become pregnant (Mergler et al., 2007; [WHO] World Health Organization, 1990). There are also many different endpoints selected to evaluate effects from Hg contamination in humans. Perhaps some of the most important endpoints are those related to intelligence, because fish consumption can positively and negatively affect cognition, meaning that fish consumption is a balance between the advantages and disadvantages it has during human development (Institute of Medicine of the National Academies et al., 2007; Mergler et al., 2007; Oken et al., 2005). An additional complication for evaluating potential health risks from Hg contaminated fish is that their Hg concentrations are inherently variable across species, sizes and locations (e.g., Depew et al., 2013; Evers et al., 2007, 2011). This makes using mean fish Hg concentrations (that often have a lognormal distribution; e.g., Eagles-Smith et al., 2016-in this issue) to assess potential health risks challenging because fish themselves are rarely “average” and piscivorous fish and humans rarely consume “average” fish. Thus, these factors can confound how Hg exposure benchmarks associated with potential fish and human health effects are characterized and how potential health risks are assessed.

Despite these challenges, syntheses of fish Hg datasets in North America have been conducted at a variety of large scales including, but not limited to, the Northeast region (e.g., Evers et al., 2007), the Great Lakes region (e.g., Evers et al., 2011) and the continental United States (e.g., Stahl et al., 2009; Xue et al., 2015). These efforts can be highly useful and provide a basis for presenting and comparing results across broad geographical regions. Data compiled by the Western North America Hg Synthesis Group were used to characterize fish Hg concentrations relevant to fish and human health (see Eagles-Smith et al., 2016-in this issue for further detail about data compilation). The expansive study area includes a variety of different habitats with disparate precipitation regimes across the largest range of elevations found in North America. These characteristics influence the distribution of fish species throughout the area, and ultimately determine the potential for health risks associated with Hg contaminated fish. The overall goal was to identify and characterize potential health risks posed to fish and humans by Hg contamination across the western United States and Canada to inform future monitoring and advisory development efforts. Specific objectives were to develop health risk and fish consumption advice maps for inland fish species; 1) characterizing health risks posed to contaminated fish due to their own Hg concentrations, 2) characterizing health risks posed to fish consuming Hg contaminated prey fish, and 3) characterizing fish consumption advice for humans. These risks were assessed across a range of fish size classes and at various fish Hg concentrations associated with deleterious health effects (fish) and recommended levels of fish consumption (humans; developing children and women who are or intend to become pregnant). This approach was novel in that it characterized potential health risks across a range of fish size classes and at multiple health-relevant Hg concentration benchmarks so results were applicable and interpretable across a variety of fish species and their consumers with differing foraging habits. This method was also particularly relevant to health risk assessment because fish Hg concentration data were presented in terms of probabilities (and importantly their corresponding uncertainties) derived in part from empirical data obtained directly from the fish species and sizes of interest for each sampling location.

2. Materials and methods

2.1. Data description

Empirical total Hg concentration data were used from approximately 150 fish species sampled from 891 hydrologic units (8-digit hydrologic units in the United States and equivalent hydrologic units in Canada) and over 3000 unique sampling locations. Eight-digit hydrologic unit codes were selected because they are delineated by watershed boundaries which are relevant to Hg contamination as water is an important transporter of Hg and can support conditions conducive to Hg methylation and bioaccumulation. In addition, 8-digit hydrologic units were used because they combine data from relatively standardized areas of approximately 1800 km^2, and equivalents of these delineations exist in both Canada and the United States for use across countries. Only samples with valid lengths and sampling locations were used in these analyses ($N = 84.082$). It was assumed that measurements of total Hg in fish were representative of methylmercury, the primary form of Hg that bioaccumulates. Nearly all Hg in whole fish and muscle tissue is in the methylated form (Bloom, 1992; Drysdale et al., 2005; Greenfield and Jahn, 2010). Briefly, this dataset was assembled from information provided by a variety of institutions and agencies across the western United States and Canada. Data were available from 1969 to 2014 from a combination of individual and composite fish samples within the study area. For all analyses, wet Hg concentrations were used and are presented in parts per million (ppm or milligrams per kilogram) wet weight. Composite samples were treated the same as individual samples. Detection limits ranged from 0.001 to 0.1 ppm, but values from <100 samples were equal to or less than reported detection limits, so they were included without adjustment. Measurements of whole-body fish Hg concentrations were used for determining potential health risk posed to contaminated fish due to their own Hg concentrations and risk posed to fish consuming Hg contaminated prey fish, while measurements of fish axial muscle Hg concentrations were used for determining risk posed to humans consuming contaminated fish. In all cases, fish lengths were assessed using total length (cm) with the exception of sturgeons (Acipenseridae family) for which fork length was used when determining whether fish exceeded minimum legal harvest length when applicable (analysis of potential health risks to humans from fish consumption, described in Section 2.5). Fork length is the most common metric used to assess sturgeon size (minimum legal length limit) in the provinces, states and territories sampled, while total length is the most common metric used in the United States and Canada for all other species within the dataset based on respective contemporary (2015) province, state and territory general angling regulations.

2.2. Analytical approach

A statistical upscaling model was developed that provided inference at the level of 8-digit hydrologic units (and equivalent hydrologic units in Canada) developed in the United States by the U.S. Geological Survey (see Eagles-Smith et al., 2016-in this issue for more details). Upscaling is a statistical term for the rigorous statistical process of formally aggregating inference at a scale larger than the scale at which the data were originally collected. To obtain the correct inference at larger scales using a single estimation framework, smaller-scale uncertainty must be propagated during aggregation. Note that "scale" need not be spatial, but could be ecological (e.g., food web structure), or temporal for example. Hierarchical approaches are particularly useful as statistical upscaling methods that rely on fewer assumptions and result in more dependable statistical inference. Interested readers are referred to Gelman and Hill (2007), and Hobbs and Hooten (2015) for detailed mechanics behind hierarchical models and uncertainty propagation. In an effort to characterize these data in the most beneficial and applicable way to represent potential fish and human health risks, the probability was calculated that a fish of a given size collected from a given location (hydrologic unit) within the study area would exceed Hg concentration benchmarks (described in Sections 2.3, 2.4 and 2.5 and represented in Table 1) relevant to a variety of health risks (fish) or consumption advice (humans). Exceedance probabilities were presented at multiple health-relevant benchmarks and across several fish size classes in each analysis. However, exceedance probabilities were only presented for a given size class of fish in a particular hydrologic unit if Hg concentration data were available for at least one fish within that size class in that hydrologic unit. For example, if a sampling area only contained Hg concentration data from fish <40.0 cm, exceedance probabilities were not extrapolated and provided for size classes >40.0 cm that were not evaluated in that area.

Fish Hg concentration exceedance probabilities were calculated by letting $y_{i,j}$ represent the jth ($j = 1, \ldots, m_i$) Hg concentration measured in hydrologic unit i (for $i = 1, \ldots, n$). Because of differences in protocol across the data collected within this compilation, individual sites within hydrologic units were not necessarily sampled with the same amount of effort. Thus, a resampling approach was used to represent individual sites within a hydrologic unit evenly with respect to sample size. The resampling approach ensured that results were not biased toward oversampling or limited sampling at any particular site; a known contaminated site for instance. Then, for each measurement, auxiliary information $x_{i,j}$ was observed. Using $x_{i,j}$ as predictor variables, y_i was predicted, an unobserved future Hg concentration in region i. To estimate uncertainty associated with y_i, the predictive distribution of y_i given all available data $\mathbf{y} \equiv \{y_{i,j}$, for all $i, j\}$ was considered

$$[y_i|\mathbf{y}] = \int \cdots \int [y_i|\mathbf{y}, \boldsymbol{\beta}_i, \sigma^2]\,[\{\boldsymbol{\beta}_i, \forall i\}, \sigma^2, \boldsymbol{\mu}, \boldsymbol{\Sigma}|\mathbf{y}]\,d\boldsymbol{\beta}_1 \cdots d\boldsymbol{\beta}_n d\sigma^2 d\boldsymbol{\mu} d\boldsymbol{\Sigma} \quad (1)$$

$$= \int \cdots \int [y_i|\boldsymbol{\beta}_i, \sigma^2]\,[\{\boldsymbol{\beta}_i, \forall i\}, \sigma^2, \boldsymbol{\mu}, \boldsymbol{\Sigma}|\mathbf{y}]\,d\boldsymbol{\beta}_1 \cdots d\boldsymbol{\beta}_n d\sigma^2 d\boldsymbol{\mu} d\boldsymbol{\Sigma} \quad (2)$$

where the last equality holds if the y_i (for $i = 1, \ldots, n$) are conditionally independent given the model parameters β_i and σ^2. The predictive distribution $[y_i|\mathbf{y}]$ was obtained by integration via Eq. (2) (Hobbs and Hooten, 2015). The hierarchical model was formulated as

$$y_{i,j} \sim \mathrm{N}\left(\mathbf{x}'_{i,j}\boldsymbol{\beta}_i, \sigma^2\right) \quad (3)$$

$$\boldsymbol{\beta}_i \sim \mathrm{N}\left(\boldsymbol{\mu}_\beta, \boldsymbol{\Sigma}_\beta\right) \quad (4)$$

$$\sigma^2 \sim \mathrm{IG}(q, r) \quad (5)$$

$$\boldsymbol{\mu}_\beta \sim \mathrm{N}(\boldsymbol{\mu}_0, \boldsymbol{\Sigma}_0) \quad (6)$$

$$\boldsymbol{\Sigma}_\beta^{-1} \sim \mathrm{Wish}\left((\mathbf{S}\nu)^{-1}, \nu\right). \quad (7)$$

Table 1
Fish Hg concentration benchmarks relevant to fish and human health used to categorize the available data. Hg concentrations are presented as whole body (effects within fish and effects on fish consuming fish) and axial muscle (advice for sensitive humans) values. Parenthetical information are associated health risks (effects within fish and effects on fish consuming fish) and consumption advice (advice for sensitive humans).
Benchmarks were established based on information available from Beckvar et al. (2005) and Sandheinrich and Wiener (2011) (effects within fish), Depew et al. (2012) (effects on fish consuming fish), and the Great Lakes Fish Advisory Workgroup (2007) (advice for sensitive humans).

Effects within fish	Effects on fish consuming fish	Advice for sensitive humans
>0.2 ppm (biochemical/gene expression)	>0.05 ppm (reproduction/biochemical)	>0.05–0.11 ppm (2 fish meals/week)
>0.3 ppm (behavior/reproduction/histology)	>0.5 ppm (behavioral)	>0.11–0.22 ppm (1 fish meal/week)
>1.0 ppm (growth)	>1.44 ppm (growth)	>0.22–0.95 ppm (1 fish meal/month)
		>0.95 ppm (no fish consumption)

This linear mixed model has random effects $\boldsymbol{\beta}_i$ that allow for variation at the hydrologic unit level (indicative of the species present) in the relationship between fish length and Hg concentration. It was expected that there was some general coherence in the fish length–Hg concentration relationship among the hydrologic units, but with variability controlled by Σ_β. Thus, the latent parameters $\boldsymbol{\mu}_\beta$ provide insight about the fish length–Hg concentration relationship at the scale of the entire dataset, and the coefficients $\boldsymbol{\beta}_i$ provide insight about the relationship at the scale of individual hydrologic units. Accounting for the variability in the fish length-Hg concentration relationships was critical for obtaining appropriate inference about important derived quantities related to the predictions y_i (e.g., exceedance probabilities) for each hydrologic unit. Variable descriptions can be found in Table 2.

Ultimately the posterior exceedance probabilities for each hydrologic unit i were sought. Thus, given a health-relevant fish Hg concentration benchmark y^*, the posterior exceedance probability was of interest

$$P(y_i > y^* | \mathbf{y}). \tag{8}$$

By fitting the model using a Markov Chain Monte Carlo algorithm, the posterior exceedance probabilities were obtained by calculating the empirical cumulative distribution function based on the Markov Chain Monte Carlo samples using

$$P(y_i > y^* | \mathbf{y}) \approx \frac{\sum_{k=1}^{K} I_{\left\{y_i^{(k)} > y^*\right\}}}{K}, \tag{9}$$

where, $y_i^{(k)}$ are the Markov Chain Monte Carlo samples (10,000 iterations) for y_i on each iteration ($k = 1, \ldots, K$) and $I_{\{y_i^{(k)} > y^*\}}$ are indicator variables equal to one if the condition in the subscript is true and zero otherwise. The posterior exceedance probability appropriately incorporated the uncertainty (standard deviation; maximum of 0.5) associated with the extent of knowledge of the study area given the data available. By formally connecting multiple levels of the hierarchy, uncertainty at the data level can be separated from the process level, ensuring that uncertainty propagates through the levels of the model space appropriately, providing more accurate inference. The maximum standard deviation of 0.5 is derived because binary variables, such and exceedance probabilities, arise as random variables from Bernoulli distributions. Bernoulli random variables are designated as either zero or one; with a mean probability p and a variance $p(1-p)$. Thus, if the probability of exceedance is either zero or one, the variance of exceedance is zero because there is no uncertainty in the estimate (e.g., one knows the exact probability of exceedance). However, if the probability of exceedance is 0.5, the standard deviation of the estimate is at a maximum (0.5), because the lowest certainty occurs when the odds are balanced between the exceedance probability being either zero or one.

This approach precluded having to select a size and species (or group of species) of fish for normalization purposes and allowed for empirical data from the fish species and sizes present within a hydrologic unit to be incorporated in the characterization of exceedance probabilities and their uncertainties. It was also not necessary to select a single health-relevant benchmark of fish Hg concentrations to evaluate. This approach was used because it was challenging to objectively select a single fish species or group of species, a standard size of fish, and a fish Hg concentration benchmark that possessed the set of qualities of interest to adequately characterize the potential health risks evaluated here. For example, we were interested in; 1) potential health risks associated with Hg concentrations within all fish species sampled, including rare species that may be threatened or endangered, and juvenile and adult fish of various lengths with species-specific sensitivities to Hg contamination, 2) potential health risks to predatory fish consuming Hg contaminated fish of a variety of potential prey species across a range of sizes, and 3) providing relevant information to sensitive demographics of fish consumers across ethnic groups with differing fish harvest and consumption habits. In addition, a variety of potential sub-lethal health effects posed by Hg contamination in the environment were of interest. Thus, selecting one representative fish Hg concentration benchmark to represent all potential health risks would have been difficult. This approach was also selected to represent empirical fish Hg concentration data across the study area with as little modification as possible to represent "realized" health risks based on empirical data from fish species and sizes represented within a particular hydrologic unit of interest. In other words, health risks were considered to be those associated with Hg contaminated fish of the species and sizes physically collected within a hydrologic unit because they represent the fish that could be at risk, or pose potential health risks to organisms that consume them. This is an important distinction because it does not represent relativized Hg exposure within fish communities, but instead potential health risks to fish and humans based on unadjusted empirical fish Hg concentrations. When fish Hg concentrations are relativized there is control for spatial, temporal, and species-based variation across the landscape that can result in substantially different portrayals of data relative to unadjusted raw data (Eagles-Smith et al., 2016-in this issue).

Table 2
Variable description table. Hydrologic unit codes are referred to as HUC.

Variable description	
$y_{i,j}$	Log Hg measurement from sample *j* in HUC *i*
y_i	Predicted log Hg measurement for HUC *i*
$\mathbf{x}_{\mathrm{i,j}}$	Covariates for sample *j* in HUC *i* (i.e., intercept and log fish length)
$\boldsymbol{\beta}_i$	HUC specific regression coefficients relating log length to log Hg
σ^2	Variance component for errors associated with length-mercury relationship at the sample-level
$\boldsymbol{\mu}_\beta$	Central tendency of HUC regression coefficients ($\boldsymbol{\beta}_i$)
$\boldsymbol{\Sigma}_\beta$	Covariance of HUC regression coefficients ($\boldsymbol{\beta}_i$)
q	Shape hyperparameter for the variance component distribution
r	Scale hyperparameter for the variance component distribution
S^{-1}	Mean matrix of the inverse-covariance distribution
v	Degrees of freedom for the inverse-covariance distribution

2.3. Analysis 1: characterizing health risks posed to contaminated fish due to their own Hg concentrations

To characterize potential health risks to fish from elevated Hg concentrations within their own bodies, fish Hg concentrations were evaluated using benchmarks associated with observed health effects summarized across studies on multiple fish species. The benchmarks considered were whole body Hg concentrations >0.2 ppm (potential for impacts on biochemical function and gene expression; little or no health effects were observed below 0.2 ppm, Beckvar et al., 2005), >0.3 ppm (potential for impacts on behavior, reproduction and histology), and >1.0 ppm (potential for impacts on growth and other deleterious effects) (Sandheinrich and Wiener, 2011; see Table 1). These benchmarks corresponded well to an analysis using lethality-equivalent test endpoints as continuous variables to characterize health effects associated with Hg concentrations in fish (Dillon et al., 2010). Benchmarks were evaluated for fish within size classes of 0 to 20, >20 to 40, >40 to 60, >60 to 80, and >80 cm total length to best represent the data, bearing in mind the size distribution of the samples available. Exceedance probabilities and their uncertainties were calculated in terms of these fish size classes to determine the likelihood that a newly sampled fish from a given hydrologic unit would exceed a given Hg concentration benchmark and potentially experience deleterious health effects. This approach resulted in 15 exceedance probability and uncertainty distribution maps across the study area.

2.4. Analysis 2: characterizing health risks posed to fish from consuming Hg contaminated prey fish

To characterize potential health risks to piscivorous fish from consuming contaminated fish, Hg concentrations of small fish that could be potential prey for larger fish were evaluated using benchmarks associated with observed health effects resulting from dietary Hg exposure summarized across studies on multiple fish species. The benchmarks considered were whole body Hg concentrations >0.05 ppm (potential for impacts on biochemical function and reproduction; little or no health effects were observed below 0.05 ppm), >0.5 ppm (potential for impacts on behavior), and >1.44 ppm (potential for impacts on growth and other deleterious effects) (Depew et al., 2012; see Table 1). These benchmarks were evaluated for fish within size classes of 0 to 10, >10 to 20, >20 to 30, >30 to 40, and >40 to 50 cm total length to represent size ranges relevant to fish predators of a variety of different sizes represented in the dataset. For reference, it has been shown that freshwater fish species can consume prey fish up to approximately one third to one half of their own body length (Yule and Luecke, 1993; Mittlebach and Persson, 1998; Ruzycki et al., 2003). Exceedance probabilities and their uncertainties were calculated in terms of these fish size classes to determine the likelihood that a newly sampled fish from a given hydrologic unit would exceed a given Hg concentration benchmark and potentially cause deleterious health effects in fish that might consume it. This approach resulted in 15 exceedance probability and uncertainty distribution maps across the study area.

2.5. Analysis 3: characterizing recommended amounts of fish consumption for humans

To characterize potential health risks to humans sensitive to Hg exposure, including developing children and women who are or intend to become pregnant, from fish consumption, fish Hg concentrations were evaluated using fish consumption advice recommended for this most sensitive demographic (Great Lakes Fish Advisory Workgroup, 2007). Benchmarks for advice about dietary Hg exposure from consuming fish muscle tissues were <0.05 ppm (no recommended restrictions on fish consumption), >0.05 to 0.11 ppm (two fish meals per week), >0.11 to 0.22 ppm (one fish meal per week), and >0.22 to 0.95 ppm (one fish meal per month; no fish consumption was recommended above 0.95 ppm) (Great Lakes Fish Advisory Workgroup, 2007; see Table 1). These benchmarks were evaluated for fish within groups relevant to harvest and consumption habits of humans. The first group was unfiltered, including all fish available within the dataset. The second group included fish that were ≥15.0 cm and were also of legal minimum harvestable length and of a species legal to possess by anglers based on respective contemporary (2015) province, state and territory-wide general angling regulations. Anglers rarely harvest fish below 15.0 cm and this length category has been applied previously for similar purposes (Cook and Younk, 1998; Wiener et al., 2012). The third group included fish legal to harvest and possess that were also ≥30.48 (12 in.). This length threshold represents a common minimum length limit applied to black basses which were a large proportion of the data set ($N = 6432$). The fourth group included fish legal to harvest and possess that were also ≥45.72 cm (18 in.). This length threshold represents a large yet common minimum length limit applied to fish in general province, state and territory-wide angling regulations often applied to walleye (*Sander vitreus*) which is sought after for harvest and consumption, and represented the species with the highest sample size within the full dataset ($N = 17.171$). Exceedance probabilities and their uncertainties were calculated in terms of these fish size classes to determine the likelihood that a newly sampled fish from a given hydrologic unit would exceed a given Hg concentration benchmark and would have some recommended restriction associated with the frequency it should be consumed. This approach resulted in 16 exceedance probability and uncertainty distribution maps across the study area. It is important to note that fish consumption advice is based on ranges of Hg concentrations. Benchmarks were evaluated as upper limits of fish Hg concentration, above which lower fish consumption rates are recommended. Thus, if exceedance is low for a particular fish size class and Hg concentration benchmark in a given hydrologic unit, one can evaluate whether a higher consumption frequency might be appropriate within that area by examining the figure panel associated with the next lowest benchmark and so on (e.g., from the 0.95 to 0.22 to 0.11 ppm Hg concentration benchmarks) until exceedances are probable. Also note that this analysis is intended as a large-scale assessment to identify potential areas of concern, and is not a substitute for local fish consumption advice.

2.6. Mapping Hg contamination exceedance probabilities

To create the paneled maps for the figures within this manuscript, a contiguous North American hydrologic unit dataset was created by merging the 8-digit hydrologic unit codes from the Watershed Boundary Dataset ([USDA]. United States Department of Agriculture-Natural Resources Conservation Service, U.S. Geological Survey, U.S. Environmental Protection Agency) with Work Unit boundaries from the National Hydro Network Index dataset ([NRC]. Natural Resources Canada). In areas along the Canadian and United States border, where polygons overlapped, linework from the Watershed Boundary Dataset was preferentially used. The resulting dataset was processed to resolve topological issues and exceedance probabilities and their associated uncertainties were assigned to each polygon when data were available.

3. Results and discussion

3.1. Relationships between exceedance probabilities, fish size and Hg concentration benchmarks

In general across the study area, larger size classes of fish had higher probabilities of exceeding a given health-relevant Hg concentration benchmark within a given hydrologic unit relative to smaller fish size classes (Table 3). This was expected given the nature of fish length–Hg concentration relationships (Sackett et al., 2013). Similarly, low health-relevant fish Hg concentration benchmarks were exceeded more often than high benchmarks. Importantly, the results from all three analyses evaluated different fish size classes as well as different health-relevant fish Hg concentration benchmarks and the analyses were also conducted with different objectives. Thus, comparisons made across the results from the three analyses should be made keeping these factors and others mentioned in Materials and Methods in mind.

3.2. Analysis 1: health risks posed to contaminated fish due to their own Hg concentrations

Of the 891 hydrologic units represented in this dataset, exceedance probabilities and corresponding uncertainty estimates were provided for 533, 764, 677, 408, and 233 hydrologic units in the 0 to 20, >20 to 40, >40 to 60, >60 to 80, and >80 cm total length size classes, respectively for this analysis. There were fewer data on Hg concentrations for larger fish size classes relative to smaller ones, and this was the primary cause of the decline in the number of hydrologic units represented by fish sampled within the >60 to 80, and >80 cm total length size classes. Based on the data available, it was evident that across the study area, exceedance probabilities were relatively low at the smallest fish size classes (>0 to 20.0 cm, and >20.0 to 40.0 cm) and at the highest (1.0 ppm) health relevant fish Hg concentration benchmark (Fig. 1). However, there were some localized areas where the exceedance probabilities of the two smallest fish size classes were elevated relative to other areas when evaluated at the 0.2 and 0.3 ppm fish Hg benchmarks (Fig. 1) and even at the 1.0 ppm benchmark. For example, when considering fish Hg concentrations from the two smallest size classes (>0 to 20.0 cm and >20.0 to 40.0 cm) at the 1.0 ppm benchmark, three subregions had multiple hydrologic units with predicted exceedance probabilities larger than 0.15. These included the Klamath-Northern California Coastal Subregion with the highest exceedance probabilities being 0.17 and 0.18, the Middle Gila Subregion within the Santa Cruz River Basin with the highest exceedance probabilities being 0.21 and

Table 3
Median exceedance probabilities (and median standard deviations) for all hydrologic units with available data for analyses 1–3 at applicable size class and health-relevant fish Hg concentration benchmarks. Analyses are separated by gray rows; size classes increase from left to right; and fish Hg concentration benchmarks increase from top to bottom within each analysis. Parenthetically below each size class is the number of fish that fell within that particular size class.

Analysis 1: direct risk to fish from Hg contamination					
Size class/benchmark	>0–20.0 cm (15,116)	>20.0–40.0 cm (25,462)	>40.0–60.0 cm (30,345)	>60.0–80.0 cm (11,243)	>80.0 cm (1916)
>0.2 ppm	0.05 (0.23)	0.18 (0.38)	0.40 (0.46)	0.53 (0.44)	0.73 (0.42)
>0.3 ppm	0.02 (0.14)	0.08 (0.27)	0.24 (0.42)	0.31 (0.43)	0.53 (0.46)
>1.0 ppm	0.00 (0.04)	0.00 (0.05)	0.02 (0.13)	0.02 (0.13)	0.06 (0.24)
Analysis 2: risk to fish from consuming Hg contaminated prey					
Size class/benchmark	>0–10.0 cm (9270)	>10.0–20.0 cm (5846)	>20.0–30.0 cm (10,729)	>30.0–40.0 cm (14,733)	>40.0–50.0 cm (17,820)
>0.05 pm	0.54 (0.43)	0.57 (0.46)	0.71 (0.44)	0.83 (0.36)	0.90 (0.29)
>0.5 ppm	0.00 (0.04)	0.00 (0.05)	0.01 (0.11)	0.03 (0.17)	0.08 (0.28)
>1.44 ppm	0.00 (0.00)	0.00 (0.00)	0.00 (0.01)	0.00 (0.03)	0.00 (0.07)
Analysis 3: fish meal frequency for humans					
Size class/benchmark	All fish (84,082)	Legal and >15.0 cm (71,648)	Legal and >30.48 cm (57,511)	Legal and >45.72 cm (33,213)	
>0.05 ppm	0.86 (0.35)	0.89 (0.32)	0.95 (0.22)	0.97 (0.16)	
>0.11 ppm	0.60 (0.46)	0.63 (0.46)	0.76 (0.42)	0.85 (0.35)	
>0.22 ppm	0.31 (0.45)	0.34 (0.45)	0.46 (0.46)	0.56 (0.46)	
>0.95 ppm	0.02 (0.13)	0.02 (0.15)	0.04 (0.19)	0.06 (0.23)	

0.25, and the Central Lahontan Subregion within the Carson River Basin near the Carson River Hg Superfund Site with the highest exceedance probabilities being 0.17, 0.33 and 0.50. Similar locations were identified as potential health risks to avian species using a disparate (avian) dataset by Ackerman et al. (2016-in this issue). Larger fish size classes had higher exceedance probabilities, especially when evaluated at the

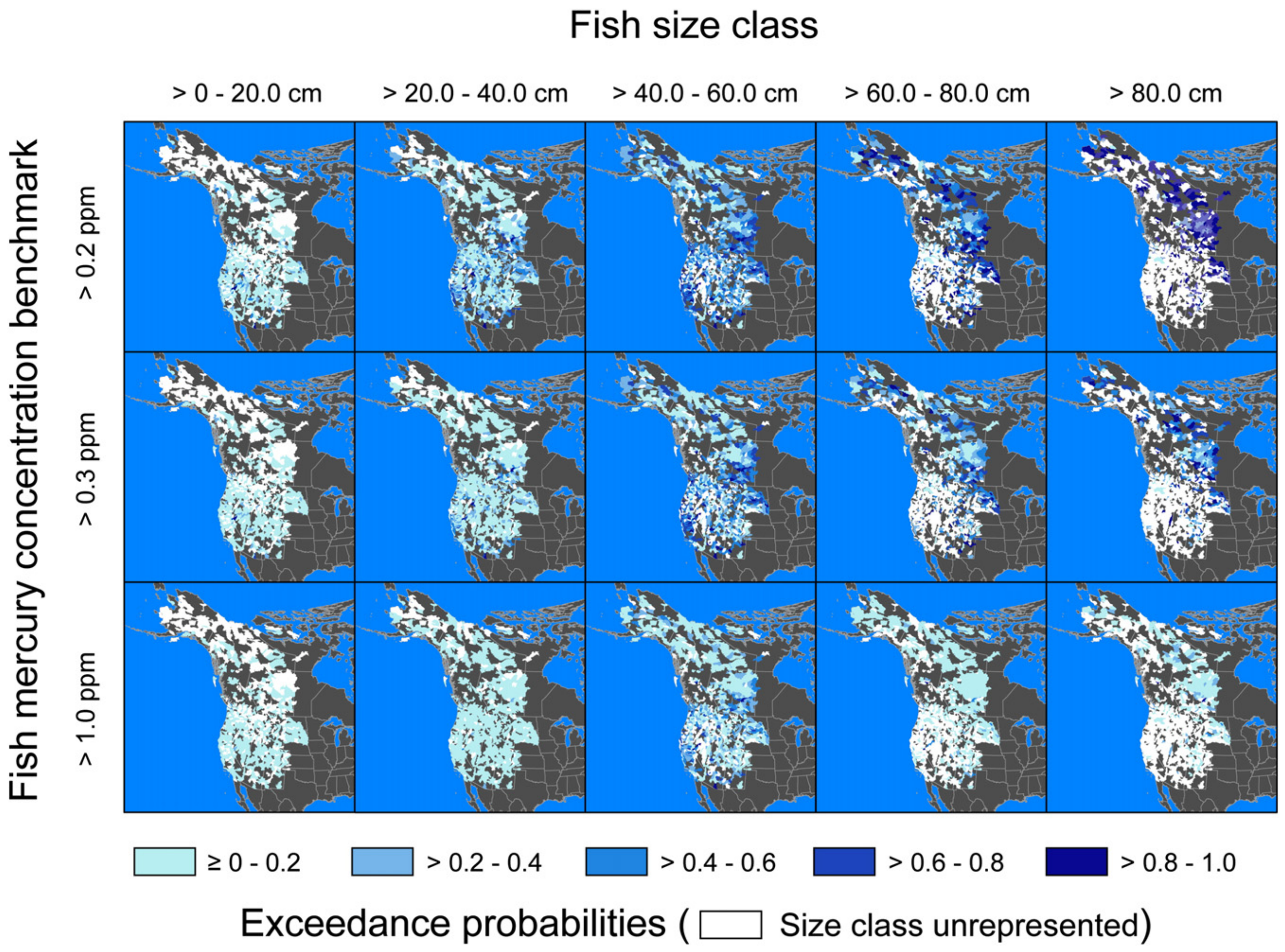

Fig. 1. Hydrologic unit-specific exceedance probabilities related to health risks posed to contaminated fish due to their concentrations of bioaccumulated Hg throughout the study area. Fish size classes (0 to 20, >20 to 40, >40 to 60, >60 to 80, and >80 cm total length) increase from left to right. Health-relevant fish Hg concentration benchmarks increase from top to bottom and are >0.2 to 0.3 ppm (potential for impacts on biochemical function and gene expression), >0.3 to 1.0 (potential for impacts on behavior, reproduction and histology), and >1.0 ppm (potential for impacts on growth and other deleterious effects). Hydrologic units where fish were not sampled in a given size class but other data were collected are shown in white.

0.2 and 0.3 ppm benchmarks. Thus, results indicated that, within the study area, there were potential health risks posed to smaller size classes of fish in some areas, and that those risks increased with increasing fish size. Fish in larger size classes (especially those in the >60.0–80.0 cm and >80.0 cm size classes) commonly had elevated probabilities of exceeding health-relevant Hg concentration benchmarks despite relatively few areas having significantly elevated exceedance probabilities at the highest Hg concentration benchmark (1.0 ppm). This suggests that some fish are potentially impacted by Hg contamination throughout the study area and could be experiencing deleterious sublethal health effects impacting biochemical function, gene expression, behavior, reproduction, and histological function associated with their own tissues exceeding the 0.2 and 0.3 ppm fish Hg concentration benchmarks (Sandheinrich and Wiener, 2011). Importantly, Hg contamination in the environment which might negatively affect fish reproduction represents potential population-level effects on multiple fish species and populations. Thus, given the arguably disproportionately high ecological significance of rare, threatened, or endangered fish species of concern, identifying and characterizing potential population-level effects using data collected from these species in areas where they persist is important, and imperative for the protection of these valuable species.

Although exceedance probabilities were predicted at various health-relevant benchmarks across the landscape, the uncertainty associated with those predictions was relatively high. Uncertainty was generally high for the three largest fish size classes evaluated at the 0.2 and 0.3 ppm benchmarks with the exception of some of the northern most hydrologic units sampled where uncertainty was lower for the largest fish size classes. This was because of higher certainty that exceedance probabilities evaluated at the 0.2 ppm benchmark in those areas were approaching 1 (Fig. 2). At the 1.0 ppm Hg concentration benchmark, there was a higher level of certainty that fish in the smaller size classes would not exceed that benchmark. However, uncertainty did generally increase for the larger size classes of fish evaluated at the 1.0 ppm benchmark. The combination of elevated exceedance probabilities coupled with prediction uncertainty suggests that more fine-scale evaluations may be required to accurately characterize potential health risks to fish in many locations within the study area.

3.3. Analysis 2: health risks posed to fish from consuming Hg contaminated prey fish

Of the 891 hydrologic units represented in this dataset, exceedance probabilities and corresponding uncertainty estimates were provided for 143, 521, 677, 672, and 640 hydrologic units in the 0 to 10, >10 to 20, >20 to 30, >30 to 40, and >40 to 50 cm total length size classes, respectively for this analysis. Fish Hg concentration data were relatively sparse within the smallest size class evaluated with the exception of 2 hydrologic units which comprised approximately 75% of all of the fish Hg concentration data within the >0 to 10.0 cm size class. Thus, the majority of hydrologic units were not represented in the health risk map pertaining to the smallest fish size class. Based on the data available, it was evident that many hydrologic units had exceedance probabilities that were elevated for all size classes of fish considered in analysis 2 when evaluated at the 0.05 ppm fish Hg concentration benchmark (Fig. 3). This fish Hg concentration

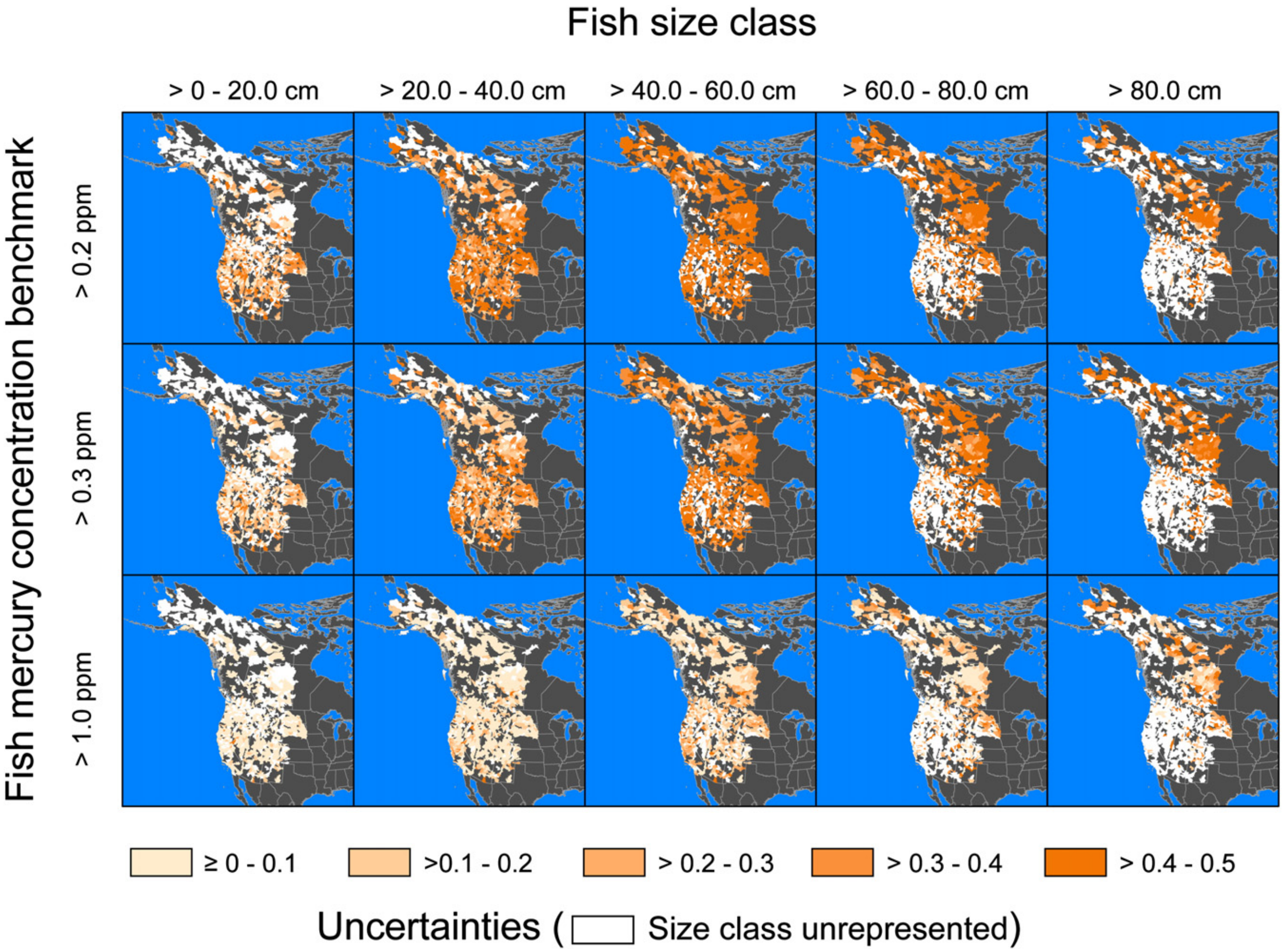

Fig. 2. Hydrologic unit-specific uncertainties associated with exceedance probabilities related to health risks posed to contaminated fish due to their own Hg concentrations throughout the study area. Hydrologic units where fish were not sampled in a given size class but other data were collected are shown in white. See Fig. 1 caption for further detail.

benchmark was relatively low when compared to the lowest benchmark of 0.2 ppm evaluated in analysis 1, so exceedance probabilities were higher. The relatively high exceedance probabilities at the 0.05 ppm benchmark across fish size classes suggested that in many locations within the study area, there was potential for deleterious health effects in fish consuming the prey fish represented by these data. These effects include alterations of important biochemical functions and reproductive mechanisms from consuming Hg contaminated prey (Depew et al., 2012). It is important to note that the potential health risks evaluated in analysis 2 apply to piscivorous fish rather than other species that may be omnivorous or planktivorous for example. However, these types of health impacts could have population-level consequences, and if piscivorous fish have the potential to consume these contaminated prey, their populations may be experiencing deleterious health effects in some areas.

Uncertainty associated with exceedance probabilities evaluated at the 0.5 and 1.44 ppm fish Hg concentration benchmarks was relatively low compared to uncertainty at the 0.05 ppm benchmark (Fig. 4). Uncertainty generally increased with fish size class, with the exception of some decreasing uncertainties associated with elevated exceedance probabilities in hydrologic units in the eastern portion of the study area near the Canadian and United States border as well as some of the eastern and western portions when evaluated at the 0.05 ppm benchmark. Similar to analysis 1, this decrease in uncertainty was due to exceedance probabilities approaching 1 in larger fish size classes such that there was little uncertainty associated with their high probability of exceeding the relatively low fish Hg concentration benchmark of 0.05 ppm (Fig. 4).

3.4. Analysis 3: characterizing recommended amounts of fish consumption for humans

Of the 891 hydrologic units represented in this dataset, exceedance probabilities and corresponding uncertainty estimates were provided for 891, 873, 777, and 611 hydrologic units for the size classes including; all fish, fish legal to harvest and possess that were also ≥15.0 cm, fish legal to harvest and possess that were also ≥30.48 cm (12 in.), and fish legal to harvest and possess that were also ≥45.72 cm (18 in.). Exceedance probabilities were relatively high at the lowest fish Hg concentration benchmark of 0.05 ppm that was evaluated (Fig. 5). Exceedance probabilities were lower at the benchmark of 0.11 ppm and still lower at 0.22 ppm, but still present across the study area. At the fish Hg concentration benchmark of 0.95 ppm, relatively few exceedances were observed (Fig. 5). Although exceedance probabilities changed as fish size class increased, exceedance probabilities appeared to be driven primarily by the fish Hg concentration benchmark that was being evaluated (Fig. 5). These results indicate that in many areas across all the size classes of fish evaluated here, some form of restricted fish consumption would be recommended based on the advice presented by the Great Lakes Fish Advisory Workgroup (2007).

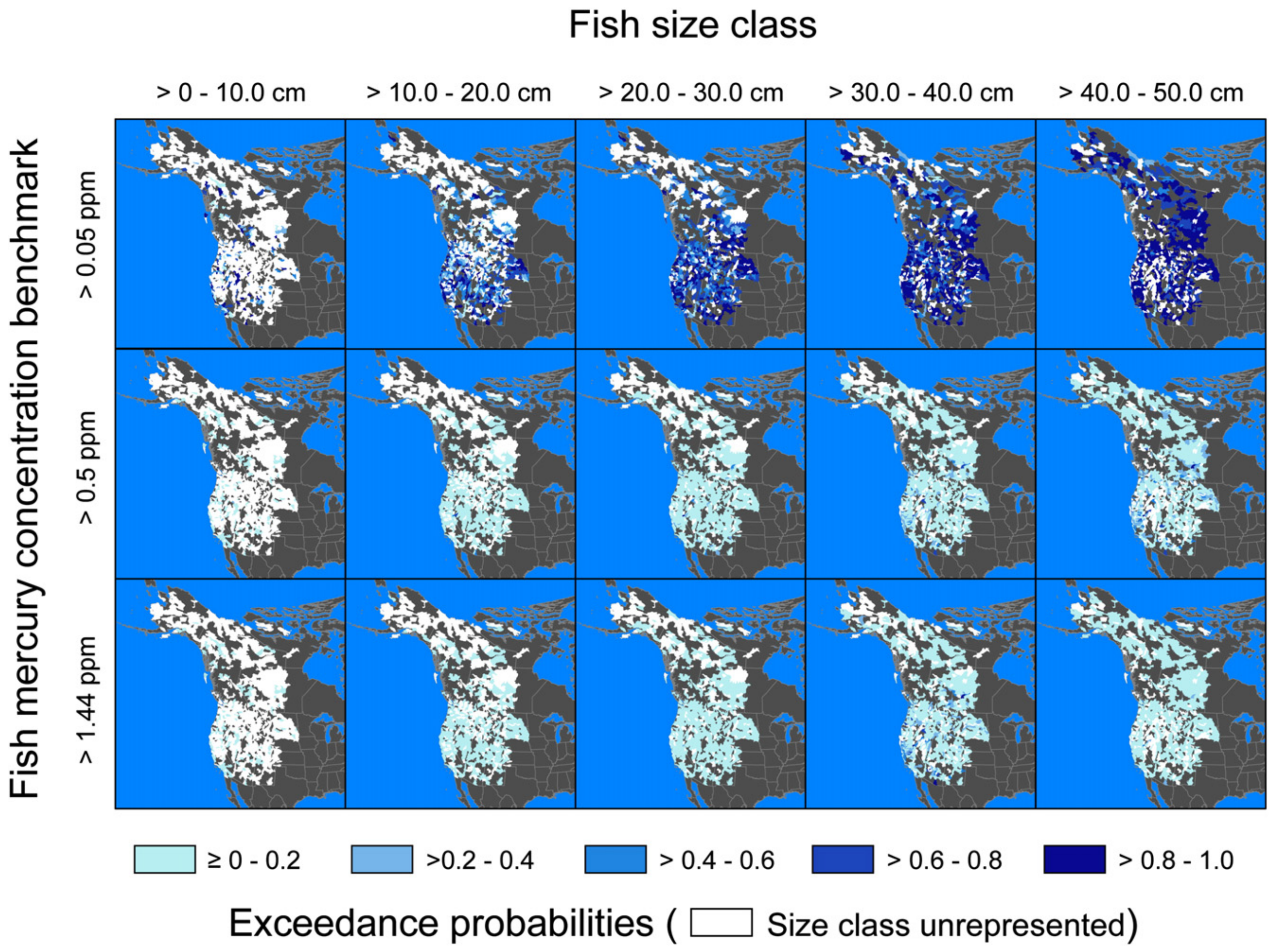

Fig. 3. Hydrologic unit-specific exceedance probabilities related to health risks posed to piscivorous fish from consuming Hg contaminated prey fish throughout the study area. Fish size classes (0 to 10, >10 to 20, >20 to 30, >30 to 40, and >40 to 50 cm total length) increase from left to right. Health-relevant fish Hg concentration benchmarks increase from top to bottom and are >0.05 to 0.5 ppm (potential for impacts on biochemical function and reproduction), >0.5 to 1.44 (potential for impacts on growth and other deleterious effects), and >1.44 ppm (potential for impacts on growth and other deleterious effects). Hydrologic units where fish were not sampled in a given size class but other data were collected are shown in white.

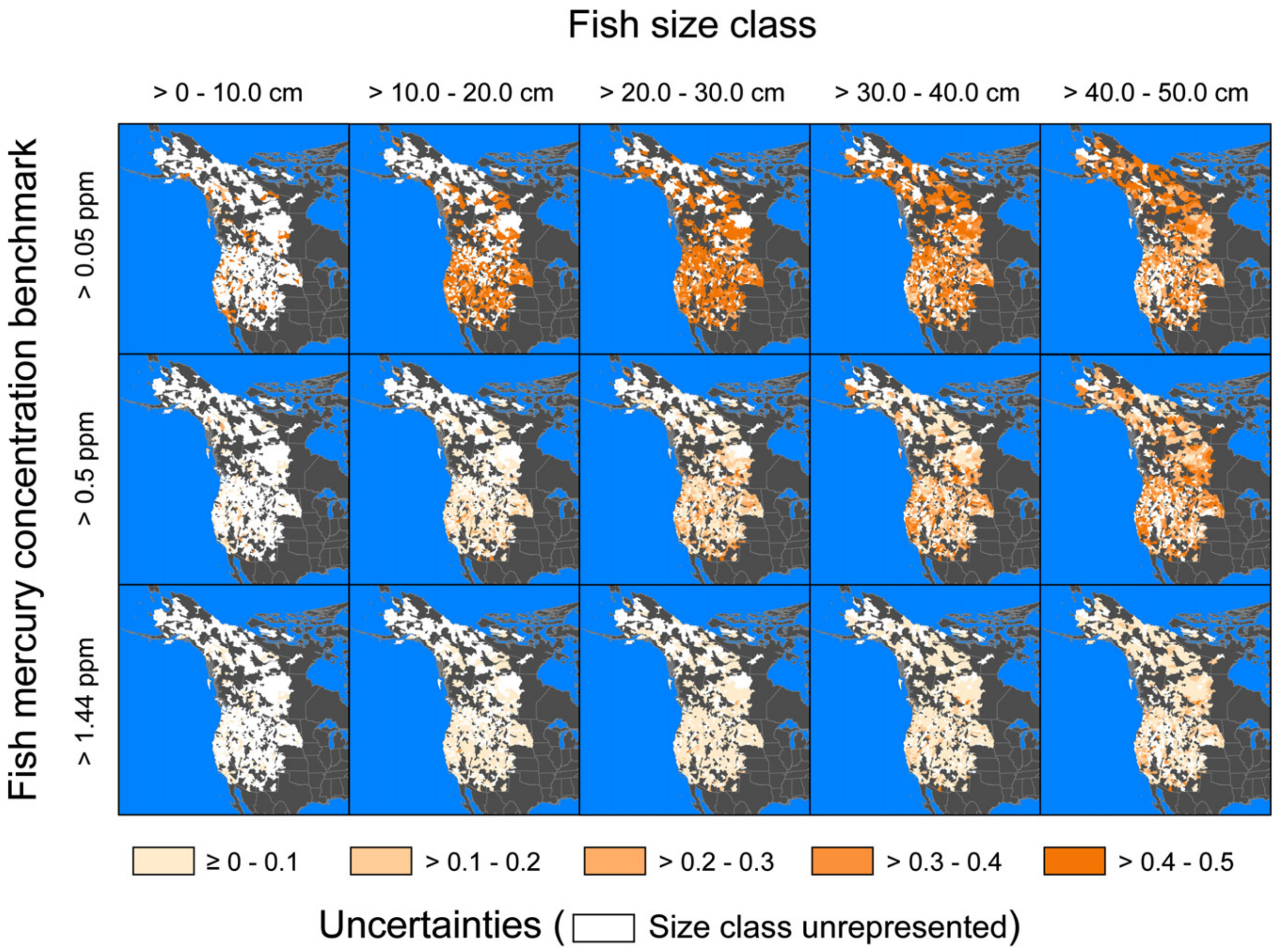

Fig. 4. Hydrologic unit-specific uncertainties associated with exceedance probabilities related to health risks posed to piscivorous fish from consuming Hg contaminated prey fish throughout the study area. Hydrologic units where fish were not sampled in a given size class but other data were collected are shown in white. See Fig. 3 caption for further detail.

Similar to the exceedance probabilities, their associated uncertainties appeared to be driven primarily by which health-relevant fish Hg concentration benchmark was being evaluated (Fig. 6). A relatively large fraction of the hydrologic units in the northern most portion of the study area as well as some of the eastern, western, and central portions appeared to have elevated exceedance probabilities when evaluated at the 0.05 ppm fish Hg concentration benchmark. The uncertainties associated with those areas are relatively low because fish are indeed likely to exceed that benchmark in many cases (Fig. 6). Thus, fish consumption advice can be developed in those situations with some level of confidence when evaluated at a finer scale. However, high uncertainty was common across the landscape, especially when evaluated at the 0.11 ppm and 0.22 ppm fish Hg concentration benchmarks (Fig. 6). The relatively high exceedance probabilities in the area combined with the relatively high uncertainties associated with them is challenging to address when developing fish consumption advisories. This is because health risks can be increased when uncertainty about fish Hg concentrations leads to consumption of fish that are believed to be less contaminated than they actually are. Perhaps a more robust or complimentary approach to providing some metric of average fish Hg concentrations in an area is providing probabilities that fish may exceed a given Hg concentration threshold as described here. Since fish Hg concentrations are inherently variable, including uncertainty around estimates and characterizations of potential health risks is also imperative for appropriate development of fish consumption advisories, especially in cases where consumers may believe they are being exposed to less Hg than they actually are. These situations may warrant more targeted monitoring and research as well as a review of historical and contemporary data to better characterize potential health risk and appropriately inform fish consumption advisory development.

Importantly, individuals consume different types and amounts of fish, some harvesting many fish and focusing on top predators, some harvesting few fish of smaller sizes or lower trophic levels, and others harvesting species that are the most available in a given area. This has direct health risk implications, as some fish species (like highly piscivorous fish) are more prone to Hg bioaccumulation (Eagles-Smith et al., 2016-in this issue), and therefore represent an elevated health risk to humans relative to other species at lower trophic levels. More than half ($N = 42.114$ of 84,082) of the dataset used here to characterize health risks from Hg contamination consisted of four fish species that are largely piscivorous (Scott and Crossman, 1973) including lake trout (*Salvelinus namaycush*; $N = 4.362$), largemouth bass (*Micropterus salmoides*; $N = 4.892$), northern pike (*Esox lucius*; $N = 15.689$), and walleye ($N = 17.171$). Thus, in this respect, the potential health risks to humans presented here from Hg contaminated fish consumption are somewhat conservative, and fish with lower trophic levels harvested by some anglers are likely to have lower exceedance probabilities. For example, species of salmon were largely excluded from these analyses with only those captured in inland waters being considered (<1% of the data), and salmon captured in the ocean (a conventional source of fish protein for many) were excluded entirely. However, these species generally contain relatively high ratios of omega-3 fatty acids relative to their Hg content, making them beneficial for consumption relative to other fish species that are more contaminated by Hg like piscivorous apex predators (Eagles-Smith et al., 2016-in this issue; Great Lakes Fish Advisory Workgroup, 2007). The exceedance

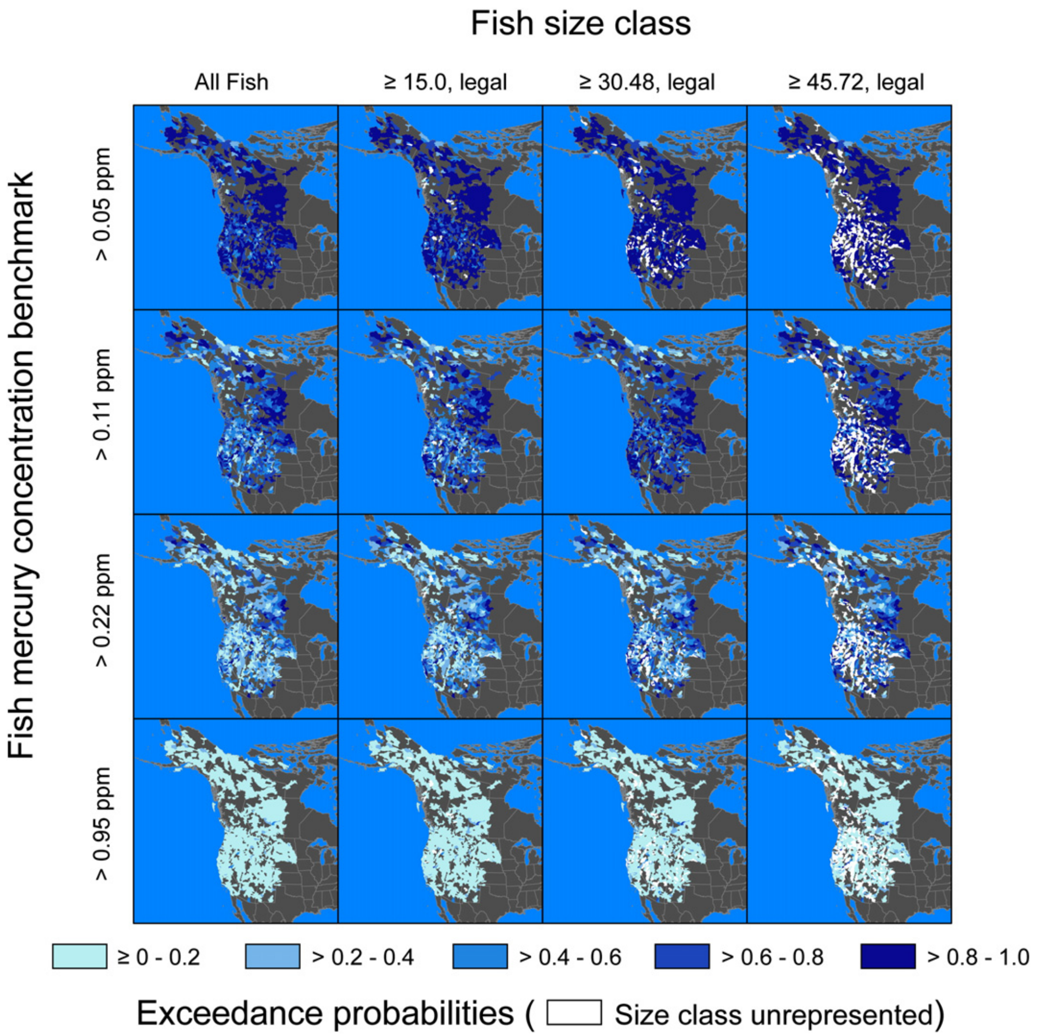

Fig. 5. Hydrologic unit-specific exceedance probabilities related to consumption advice for developing children and women who are or intend to become pregnant throughout the study area. Fish size classes increase from left to right and include all fish (all fish), fish of legal minimum harvestable length and of a species legal to possess by anglers based on respective contemporary (2015) province, state and territory-wide general angling regulations that were also ≥15.0 cm (≥15.0 cm, legal), fish legal to harvest and possess that were also ≥30.48 cm or 12 in. (≥30.48 cm, legal), and fish legal to harvest and possess that were also ≥45.72 cm or 18 in. (≥45.72 cm, legal). Benchmarks for advice about dietary Hg exposure from consuming fish muscle tissues increase from top to bottom and were ≤0.05 ppm (no recommended restrictions on fish consumption), ≤0.11 ppm (two fish meals per week), ≤0.22 ppm (one fish meal per week), and ≤0.95 ppm (one fish meal per month; no fish consumption was recommended above 0.95 ppm) (Great Lakes Fish Advisory Workgroup, 2007). Hydrologic units where fish were not sampled in a given size class but other data were collected are shown in white.

benchmarks evaluated here are also conservative in that they are based on the demographics (developing children and women who are of childbearing age) most sensitive to Hg exposure through fish consumption (Great Lakes Fish Advisory Workgroup, 2007). However, importantly, some ethnic groups within the United States (tribal populations, for example) consume more fish than other groups, and this can result in Hg exposure that is three to ten times higher than the general population and represents health risks (Xue et al., 2015). Thus, health risks associated with fish consumption for these groups are disproportionately higher because of higher fish consumption when compared to the general population. These factors, and others like them, must be taken into consideration when interpreting the results presented here to evaluate and balance potential health benefits and risks posed to humans by the consumption of fish throughout the study area. It is also imperative when evaluating potential human health risks from consuming Hg contaminated fish that advisories for specific fish species and water bodies be consulted if available.

3.5. Context relative to other synthesis efforts in North America

Because approach used in this manuscript was novel and presents potential health risks in terms of exceedance probabilities and uncertainty, it was difficult to make direct comparisons to other studies. However, to put these results in a larger context, Eagles-Smith et al. (2016-in this issue) found that geometric mean Hg concentrations of whole, inland fish collected in western Canada and the United States exceeded 0.2 ppm wet weight at 20% of the sites analyzed, and exceeded 0.3 ppm at 17% of the sites analyzed. In a similar large-scale synthesis effort, Sandheinrich et al. (2011) found that sexually mature female largemouth bass, northern pike, smallmouth bass (*Micropterus dolomieu*), and walleye collected from the states bordering the Great Lakes had whole-body Hg concentrations exceeding 0.2 ppm wet weight at 8% to 43% of sites, and exceeding 0.3 ppm wet weight at 3% to 18% of sites depending on the species being considered. A synthesis effort in northeastern North America found that standard-length brook trout (*Salvelinus fontinalis*) and yellow perch (*Perca flavescens*) fillets had Hg concentrations exceeding 0.3 ppm wet weight in 14% and 42% of sites respectively (Kamman et al., 2005). Although these studies are not directly comparable due to different methodologies, the synthesis in the Great Lakes region had similar results (ranges of sites where exceedances occurred) and species composition (piscivorous sport fish) relative to the results of Eagles-Smith et al. (2016-in this issue). Northeastern North America in particular is influenced by elevated Hg deposition and has characteristics known to promote bioaccumulation

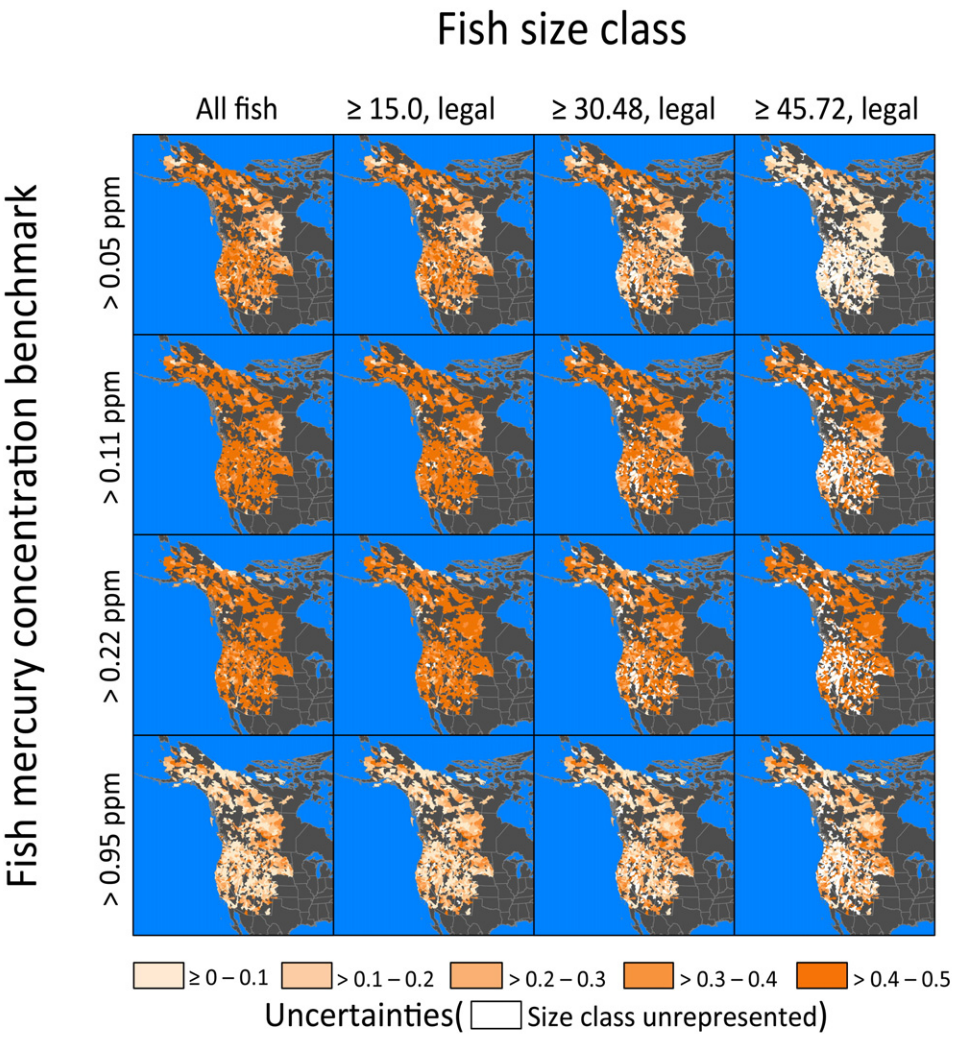

Fig. 6. Hydrologic unit-specific uncertainties associated with exceedance probabilities related to consumption advice for developing children and women who are or intend to become pregnant throughout the study area. Hydrologic units where fish were not sampled in a given size class but other data were collected are shown in white. See Fig. 5 caption for further detail.

(Driscoll et al., 2007). Perhaps not surprisingly, smaller fish species in northeastern North America generally considered to have lower trophic positions (i.e., brook trout and yellow perch) had similar or even higher exceedances of Hg concentration benchmarks (Kamman et al., 2005) relative to the larger piscivores evaluated by Sandheinrich et al. (2011) in the Great Lakes and many of the species evaluated during the Western North American Hg Synthesis described in part here. Thus, although there were differences in study design and analyses, these data suggest that potential health risks to fish and humans from Hg contamination were more similar in western Canada and the United States to the Great Lakes region. By contrast, some areas of northeastern North America may have the potential for higher health risks to fish and humans from Hg contamination, but note that this is an oversimplification when making comparisons across these large spatial scales.

4. Conclusions

This manuscript provides a basis for identifying areas of concern for further investigation, and informing the design of comprehensive monitoring and additional research efforts to assess potential health risks to fish and humans from Hg contamination. A relatively unique approach was used to characterize potential health risks associated with Hg contamination by calculating probabilities that fish from locations across western Canada and the United States would exceed Hg concentration benchmarks relevant to fish and human health. Importantly, the uncertainty associated with these probabilities was characterized, which is often overlooked but crucial for assessing health risks and informing fish consumption advisory development. Exceedance probabilities and their uncertainties were evaluated for a range of fish size classes and Hg concentration benchmarks to ensure that results were applicable and interpretable across a variety of fish species and sizes. This was also done to ensure that these results were relevant to the broad range of fish consumers (focusing here on other fish and humans) with a variety of different fish consumption habits. In fact, these results were comparable to efforts evaluating wildlife health in western Canada and the United States using analyses based on avian Hg concentration data (Ackerman et al., 2016-in this issue) and an effort using the same dataset to identify biological Hg "hotspots" that represent potential health risks to avian piscivores (Jackson et al., 2016-in this issue).

Often, data syntheses use some form of normalization approach, selecting a common fish species or group of interest and a standard size of fish for data characterization (e.g., Depew et al., 2013; Evers et al., 2007, 2011; Eagles-Smith et al., 2016-in this issue), subsequently transforming other data sources in terms of those species and sizes. These approaches are valuable, especially when one is considering relative amounts of Hg in systems to address mechanistic questions (e.g., which regions, systems or system types are more likely to produce fish with elevated Hg concentrations) under ceteris paribus (all else being equal) conditions (e.g., Eagles-Smith et al., 2016-in this issue). However, the approach used here was designed to allow the data from different species to influence the representation of potential health risks. In some ways this limits the interpretation of the data such that comparisons can be made across the landscape, but these should not be made with the goal of identifying mechanistic differences in Hg bioaccumulation spatially (e.g., geographical differences in Hg deposition or methylation; see Eagles-Smith et al., 2016-in this issue), but rather to compare relative health risks across the landscape that are representative of the species collected and analyzed within a hydrologic unit. The objective here was to characterize "realized" potential health risks posed to fish and their consumers from Hg contamination. Thus, it was difficult to select a fish species or group of species and a particular size that was distributed in waterbodies throughout western Canada and the United States, and would be simultaneously relevant to the health of multiple species and sizes of Hg contaminated fish, multiple fish species and sizes that consume them, and humans that consume a variety of different fish species and sizes. In addition, the Hg concentration benchmarks used to evaluate potential health risks in fish were based on studies involving many species and multiple life stages and sizes of fish (Depew et al., 2012; Sandheinrich and Wiener, 2011). This made it more difficult to select a particular species and size of fish indicative of the health of a variety of others. Instead, all fish species were included in these analyses to represent exceedance probabilities across a range of size classes of fish and health-relevant Hg concentration benchmarks, while simultaneously incorporating location-specific information (hydrologic-unit specific regression coefficients relating log fish length to log fish Hg concentration) to increase the applicability of the results across the landscape.

The approach described here was beneficial in many ways, but there were limitations as well. Fish Hg concentrations were not collected mechanistically across studies, or necessarily with the intention of a large-scale compilation. Thus, there was sampling bias associated with the data available. This bias was accounted for by evenly representing data from multiple sites within a hydrologic unit to correct for oversampling or limited sampling at any one site (a known contaminated site for instance). However, there was still bias in that sampling locations were not all selected at random, and may not be representative of all the available sites, fish species or fish sizes in a particular hydrologic unit. Without intensive and comprehensive sampling (which can be difficult, not feasible, or impossible) sampling bias is inherent in these types of data compilations. The scale at which these analyses were structured was both a benefit and limitation. Data characterizations were designed to be representative of the large spatial scale of this study, but simultaneously relevant across a variety of fish size classes and health-relevant benchmarks of fish Hg concentrations. Balancing these factors and presenting the results in a meaningful way was challenging, and readers interested in finer details of the data presented in Figs. 1–6 are directed to the supplemental information for this manuscript. Further, uncertainty was found to be high in many cases across these analyses despite using all available data that fit predetermined criteria to increase sample size. Although this was somewhat of a limitation and could be improved with the collection of more data, variability in fish Hg concentrations is ubiquitous. Arguably, one of the largest health risks posed to humans from Hg contamination in fish is when one is relatively confident that they are consuming fish of a given Hg content when it is actually higher. This situation could result in an overexposure of Hg caused by unrecognized uncertainty. Thus, it is important to identify areas where fish Hg concentrations are variable, and accurately quantify uncertainty, as well as identify areas that have fish with consistently elevated Hg concentrations.

The data characterization presented here indicated that there were many locations within the study area where sampling took place, but fish in certain size classes were not collected and analyzed for Hg concentration (areas indicated in white). It should be emphasized that these areas (and those that were not sampled at all) may still hold potential health risks, and this should be considered when interpreting results presented here. It may be beneficial to focus additional effort on areas where little information is available to determine whether potential health risks (or benefits) exist. It should also be noted that Hg concentrations in fish are not static, having the potential to change (sometimes rapidly) through time (e.g., Eagles-Smith et al., 2008; Johnson et al., 2015; Lepak et al., 2012b) so routine, targeted monitoring is imperative to adequately characterize potential health risks posed to fish and humans from Hg contamination in the environment. Thus, one should always refer to available, current data and health risk assessments specific to the system and fish species and size of interest to make fish consumption decisions.

Temporal trends in fish Hg concentrations (Eagles-Smith et al., 2016-in this issue) and Hg deposition (Weiss-Penzias et al., 2016) have been evaluated in western North America. Eagles-Smith et al. (2016-in this issue) observed a decrease in fish Hg concentrations from 1969 to 1977, no discernable trends from 1978 to 2012, and a decline in 2013 to 2014 (but with very little support and representative of only a few sites in the North American Desert and Northwest Forested Mountain ecoregions; <1% of the data available). Interestingly, Weiss-Penzias et al. (2016) observed more sites with significant increasing trends in Hg deposition relative to sites with decreasing trends based on data available from western and central Canada and the United States from 2008 to 2013, while many sites showed no significant changes through time. Since the observed temporal trends in fish Hg concentrations and Hg deposition were inconsistent, all available fish Hg concentration data for analyses presented here. Over 90% of the data available were from 1978 to 2012 when no significant trends in fish Hg concentrations were observed by Eagles-Smith et al. (2016-in this issue). Though a decrease in fish Hg concentrations was observed from 1969 to 1977, including data from these years resulted in a more conservative perspective with regard to potential health risks. This seemed appropriate, especially when considering more recent increasing trends observed in Hg deposition by Weiss-Penzias et al. (2016), suggesting that one might expect potential health risks from Hg contamination to increase as a result in the future. Although incorporating a temporal component would have been informative (see Eagles-Smith et al., 2016-in this issue), the data available could only support a finite number of hierarchical levels to be appropriately and simultaneously considered at the broad spatial scale represented here. Since the objective was to identify potential health risks and their associated uncertainty, it was decided to focus on estimating parameters that informed these attributes most directly. Because a temporal component was not considered, it is noted that the information presented here is intended as a large-scale risk assessment and not intended to replace or subvert local fish consumption advisories, and current data are the most important for understanding the potential for contemporary health risks at the local scale.

Fish, humans, and wildlife (e.g., Ackerman et al., 2016-in this issue; Jackson et al., 2016-in this issue) are faced with potential health risks from Hg contamination in western Canada and the United States. To reduce potential health risks posed by Hg contamination in the short-term to humans, fish consumption advice, and fisheries and system management techniques (e.g., Johnson et al., 2015; Lepak et al., 2012a, b; Mailman et al., 2006) can be used. Humans benefit from having the ability to use judgment based on available data and consumption advice to make decisions about which fish to eat (i.e., the potential to eat fish species with relatively elevated omega-3 fatty acids relative to Hg concentrations such as salmon), when, and how often. By developing comprehensive monitoring and fish consumption advisory programs, one can balance the health benefits of fish consumption versus the potential risks. However, fish and wildlife are unable to assess health risks in this manner, lacking the ability to select alternate prey species based on Hg concentrations. Therefore, to be protective of ecosystems and the organisms that live within them (including humans) in the long-term, it would be beneficial to continue research efforts to help mitigate Hg contamination and ultimately reduce the amount of Hg cycling within the environment.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.03.031.

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