

# **WILLAMETTE RIVER BASIN TOTAL MAXIMUM DAILY LOAD PROJECT**

## **A Basin-Specific Aquatic Food Web Biomagnification Model for Estimation of Mercury Target Levels**

FINAL

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## SUMMARY

In Oregon's Willamette River Basin (WRB), health advisories currently limit consumption of fish that have accumulated methylmercury to levels posing a potential health risk for humans. Under the Clean Water Act, these advisories create the requirement for a Total Maximum Daily Load (TMDL) for mercury in the WRB. A TMDL is a calculation of the maximum amount of a pollutant that a waterbody can receive and still meet water quality standards. Because methylmercury is known to biomagnify in aquatic food webs, a biomagnification factor can be used, given a protective fish tissue criterion, to estimate total mercury concentrations in surface waters required to lower advisory mercury concentrations currently in fish in the WRB. This paper presents a basin-specific aquatic food web biomagnification model that simulates inorganic ( $\text{Hg}[\text{II}]$ ) and methylmercury accumulation in fish tissue and estimates WRB-specific biomagnification factors for resident fish species of concern to stakeholders. It was calibrated with WRB-specific fish tissue and surface water data. Probabilistic (two-dimensional Monte Carlo) techniques propagate stochastic variability and uncertainty throughout the model, providing decision makers with credible range information and increased flexibility in establishing a specific mercury target level. The model predicts the probability of tissue mercury concentrations in eight fish species within the range of concentrations actually measured in these species during 25+ years of water quality monitoring. Estimated mean biomagnification factor values range from  $1.12 \times 10^6$  to  $7.66 \times 10^6$  and are within the range of such values estimated by U.S. EPA on a national basis. Several WRB-specific mercury target levels are generated, which vary by their probability of affording human health protection relative to the U.S. EPA methylmercury tissue criterion of 0.30 mg/kg. Establishing a specific numeric target level is, however, a public policy decision, and one that will require further discussions among the various WRB stakeholders.

## INTRODUCTION

A Total Maximum Daily Load (TMDL) is a calculation of the maximum amount of a pollutant that a waterbody can receive and still meet water quality standards, and an allocation of that amount to the pollutant's sources. Water quality standards, which are set by States, Territories, or Tribes, identify the beneficial uses for each waterbody (e.g., drinking water supply, contact recreation (swimming), aquatic life support (fishing)) and the scientific criteria to support those uses. A TMDL is the sum of the allowable loads of a single pollutant from all contributing point and nonpoint sources, including natural background. The calculation must include a margin of safety to ensure that the waterbody can be used for its designated purposes. The Clean Water Act mandates the establishment of water quality standards and defines the requirements of the TMDL program [1].

The Willamette River Basin (WRB) occupies an area of approximately 32,000 km<sup>2</sup> in northwestern Oregon, USA. The Willamette River is the 13<sup>th</sup> largest river in the coterminous United States in terms of streamflow and produces more runoff per square mile than any of the larger rivers. Oregon's three largest urban areas, the cities of Portland, Salem, and Eugene, border the river. In the WRB, consumption of fish that have accumulated levels of mercury, particularly methylmercury (MeHg), is a significant mercury health risk for humans. A mercury advisory warning of health risks from consumption of fish has been in effect at Cottage Grove Reservoir (located on the Coast Fork Willamette River in the southern WRB) since 1979 [2,3]. In February 1997, the Oregon Department of Human Services issued a mercury advisory for consumption of largemouth bass, smallmouth bass, and northern pikeminnow for the entire mainstem Willamette River, including the Coast Fork to Cottage Grove Reservoir; a separate advisory was issued for Dorena Reservoir, also located on the Coast Fork [3]. The Oregon Department of Human Services issued a consolidated (all species) fish consumption advisory for the entire WRB in 2001. These advisories create, per Clean Water Act Section 303d, the legal requirement for a mercury TMDL for the WRB.

One of the goals within the overall WRB TMDL process is establishment of a target level for total mercury in surface water that is linked to the protection of specified beneficial uses (e.g., sport and subsistence fishing) [2,3]. Two pieces of information are needed to establish such a target level: (1) an acceptable methylmercury concentration in fish tissue (tissue criterion) and (2) a defined relationship between total mercury concentrations in surface water and total mercury concentrations in fish tissue. This target analysis uses the fish tissue MeHg criterion

value of 0.30 mg/kg (wet weight) developed by the U.S. Environmental Protection Agency for protection of human health [4,5]. Although the Oregon Department of Human Services uses a level of 0.35 mg/kg to trigger fish consumption advisories, utilization of U.S. EPA's 0.30 mg/kg provides an additional margin of safety. Some fish tissue concentrations protective of piscivorous wildlife (0.028 to 0.68 mg/kg) fall below both criteria, suggesting that neither may be protective of all such wildlife [6]. However, a target analysis incorporating wildlife receptors was not performed at this time as the legal basis for this TMDL rests solely on fish consumption advisories for human health.

Aerobic surface waters without known sources of mercury contamination generally contain <5 ng/L of total mercury [7]. Approximately 7.8 percent of total dissolved mercury in the epilimnion (36 percent in the hypolimnion) is in the form of dissolved MeHg, the species predominantly accumulated by aquatic organisms [4,8]. MeHg concentrations in surface water may be better predictors of MeHg levels in fish than are MeHg levels in sediment [9]. Biomagnification of MeHg through diet, rather than gill uptake from water, is considered the dominant basis for elevated concentrations in fish [10,11,12,13,14]. Methylmercury accumulation via either surface water or food sources may be substantial, but the relative contribution of each pathway may vary with fish species [15,16,17,18,19]. The transfer efficiency of mercury through the food web is affected by the form of mercury. Although inorganic mercury is the dominant form in the environment and easily accumulated, it is also depurated quickly. The digestive wall is much more permeable for MeHg than for inorganic mercury, allowing MeHg to be readily transferred to other tissues [12]. Methylmercury accumulates quickly, is depurated very slowly, and therefore has a greater potential to biomagnify in higher-trophic-level species. The half-life of total mercury in fish is approximately 5 days to 5 months but 1 to >3 years for MeHg [4]. Due to its preferential uptake, ability to be transferred among tissues, and slow depuration, most (~99%) of the mercury in fish muscle tissue is MeHg [4].

Estimating a target mercury water concentration requires a biomagnification factor (BMF) and a fish tissue criterion protective of human health. A target concentration in the ng/L range could be set simply through use of a default BMF of approximately  $10^6$  to  $10^7$  [4]. It is understood, however, that such factors are often influenced by local conditions and that default factors derived from nation-wide averages may be potentially unresponsive to, or inappropriate for, a given regional ecosystem. U.S. EPA suggests that, for a particular area of

concern, BMFs derived from data collected within the area are preferred to default values [4]. They also suggest inclusion of site-specific considerations when calculating a surface water target level [5]. In addition, the WRB TMDL stakeholders expressed a desire for information on the behavior and levels of mercury in fish species of special interest to them. A default approach cannot adequately address this specific request. Thus, for the WRB, a food web biomagnification model, focusing on resident fish species of concern to stakeholders, and calibrated with basin-specific tissue and water data, was used to bring regional specificity to estimates of BMF values. A fundamental assumption of this approach, which is of particular importance to its use in the TMDL process, is that the BMF between MeHg in water and total mercury in fish will remain constant under new mercury loading regimes.

This model simulates mercury (as Hg[II] and MeHg) accumulation in fish through a basin-specific food web in response to chemical exposure, based upon chemical mass balances for aquatic biota. It equates rates of change in chemical concentration within a fish (and other aquatic organisms) to the sum of chemical fluxes into and out of the organism. These fluxes include direct uptake of the chemical from water, uptake through feeding, loss of the chemical due to elimination (desorption and excretion), and dilution due to growth. It addresses the potential for bioconcentration (concentration from water), bioaccumulation (concentration from diet as well as water), and biomagnification (systematic concentration as chemicals are passed to higher trophic levels) of Hg[II] and MeHg. To predict tissue levels in fish destined for human consumption, the model is repeatedly applied to organisms at each trophic level to simulate Hg[II] and MeHg transfer from primary and secondary producers, through a variety of intermediate invertebrate and fish species, to top predators (humans).

Due to their differing physicochemical properties, Hg(II) and MeHg are handled in separate sub-models, whose outputs are then combined to yield BMF and total mercury surface water concentration estimates that equate to the protective criterion. These estimates are provided for each fish species in the model. Probabilistic (two-dimensional Monte Carlo) techniques are used to propagate both stochastic variability and uncertainty (lack of knowledge) throughout the model. This approach provides decision makers with information as to the credible range of target levels, as well as the probability of any given target level, to give them increased flexibility in establishing a specific mercury target level [4,20]. At present, there is only one food web model, with one human health endpoint, for the entire WRB. Because the WRB can be divided into four reaches on the basis of aquatic ecosystems and fish species assemblages,

reach-specific sub-models may eventually be needed to better assess local conditions [21]. Adding a sensitive ecological endpoint (e.g., avian eggs) would extend protection to encompass both human and wildlife receptors. Food web models of this type have been developed for various hydrophobic organic chemicals [22] and metals [23], including mercury [24,25,26].

## **METHODS**

### **Model Compartments**

The model food web consists of 17 compartments selected to represent important components of the WRB aquatic ecosystem: 3 source media, 1 secondary carbon source (detritus), 3 primary producers, 6 primary consumers, 2 secondary consumers, 1 tertiary consumer, and 1 top (human) consumer (Figure 1). Primary (1°) producers are organisms that convert CO<sub>2</sub> to biomass. The term usually refers to photosynthesizers, but also includes chemosynthetic bacteria that use chemical instead of light energy for CO<sub>2</sub> fixation. Primary (1°) consumers are organisms that must eat other organisms for their energy metabolism, since they cannot produce new organic matter by photosynthesis or chemosynthesis (as can producers). Primary consumers, according to the ecological pyramid concept, are herbivorous grazers. Secondary (2°) consumers include herbivorous fish, whose diet is confined to plant items, as well as invertivorous and omnivorous fish whose diet can include both plant and animal items. As the invertebrate food source decreases in abundance and diversity due to habitat degradation (e.g., anthropogenic stressors), there is typically a shift from insectivorous to omnivorous fish species. Tertiary (3°) consumers include only larger piscivorous fish at the fourth trophic level. Although humans could be included at this level, they are kept separate to indicate their status as assessment endpoints for this target analysis.

To identify major fish species and their potential food items within the WRB, information was assembled from the general literature, previous regional studies, and data collected during water quality monitoring events. Biota compartments were defined based on organism anatomy and morphology, primary exposure medium, dietary (feeding) preferences, general life history, and local abundance. Aquatic species representative of each trophic level are listed in Table 1. Aquatic species selected for inclusion in this model, including eight of the eighteen fish species sampled during water quality monitoring events, and their food preferences, are listed in Table 2. Criteria for selecting the representative fish species were: (1) species consumed by humans, (2) abundant or common in their respective communities,

(3) prominent species at each trophic level in regional food webs, (4) representative of a specific functional group, and (5) species for which mercury tissue data are currently available. Juvenile (young-of-year (YOY),  $\leq 1$  year in age) and adult fish are modeled separately because their feeding strategies often differ and juveniles are often prey items for adult omnivorous and piscivorous fish.

Compartments within the food web are linked by one or more discrete food chains or paths, each of which leads from surface water through varying intermediate biotic compartments, to the human (top level consumer) compartment. Pathways analysis is used to define feeding relationships represented in the model. This approach analyzes the flux of mercury along each of these individual paths, then combines individual path results to form an estimate of mercury concentrations in fish species available to, and consumed by, humans [27]. As summarized in Table 2, these paths are defined largely by the feeding preferences of representative species above the 1<sup>o</sup> producer level; feeding preferences are quantified with the dietary fraction variable.

### **Model Variables**

With pathway analysis, mercury fluxes can be modeled with a limited number of variables. Some of these variables, such as bioconcentration factors, are readily obtainable for different species, whereas others, such as elimination rate constants and assimilation efficiencies, are less readily available and must be derived or estimated from data available for similar species. Each food web compartment is defined by eight variables:

- (1) *Bioconcentration Factor* (BCF, L/kg) For a given biotic compartment, the BCF represents simple partitioning between the compartment and MeHg and Hg[II] in surface water. BCFs for all aquatic invertebrate and fish compartments are relative to surface water concentrations.
- (2) *Chemical Dietary Assimilation Efficiency* (AE,  $\mu\text{g}$  chemical absorbed/ $\mu\text{g}$  chemical ingested) This is the fraction of ingested chemical that is absorbed across the gut lining of an organism [28].
- (3) *Chemical Elimination Rate* ( $k_2$ ,  $\text{d}^{-1}$ ) Chemicals that have been fully absorbed are assumed to deposit in storage sites determined by the selective preference of the chemical (proteins for mercury); depletion from these sites is a function of the chemical-specific elimination rate. Loss is assumed to be first-order for each model compartment and chemical.

- (4) *Normalized Food Intake Rate* (NIR, g intake/g body weight/d) The amount of food (wet weight) ingested by an organism per day, expressed as a percentage of its body weight.
- (5) *Body weight* (BW, g) This is used as an independent variable in the determination of food intake rates for several compartments.
- (6) *Dietary Fraction* ( $DF_{ij}$ , unitless) For each predator (i) and prey (j) combination in the model, this term represents the fraction of predator (i) diet consisting of prey (j). For each predator, the sum of dietary fractions equals 1.
- (7) *Predator-Prey Size Relationship Factor* ( $\Phi$ , unitless) The ratio of prey length to predator length for partially or exclusively piscivorous model compartments. This term is included to prevent implausible model behavior such as a predator consuming prey that are near its own size.
- (8) *Water Temperature* (T, °C) This is used as an independent variable in the determination of food intake rates for several compartments as well as in the calculation of compartment-specific MeHg elimination rates.

### Model Algorithms

Bioaccumulation factors are calculated for each pathway (i.e., food chain) leading from surface water through intermediate compartments, to humans [27]. Given that each step along this pathway represents a trophic level in the food chain, then:

Level 1	$BCF_1 = C_B/C_W$	{1}
Level 2	$BAF_2 = BCF_2 + f_2 BCF_1$	{2}
Level 3	$BAF_3 = BCF_3 + f_3 BCF_2 + f_3 f_2 BCF_1$	{3}
Level 4	$BAF_4 = BCF_4 + f_4 BCF_3 + f_4 f_3 BCF_2 + f_4 f_3 f_2 BCF_1$	{4}

where:

$C_B$	Concentration of chemical in biota (mg/kg)
$C_W$	Concentration of chemical in surface water (mg/L)
$BCF_k$	Bioconcentration factor for the $k^{\text{th}}$ trophic level (L/kg)
$BAF_k$	Bioaccumulation factor for the $k^{\text{th}}$ trophic level (unitless)
$f_k$	Food term for the $k^{\text{th}}$ trophic level (unitless)

The food term ( $f_k$ ) is dependent on the trophic level in question and has been adapted to

describe bioaccumulation in an entire food web, as opposed to a single food chain, by use of a dietary fraction variable [27]. To avoid the  $DF_{i,j}$  sum among all prey for a given predator from exceeding 1 during model simulations, individual  $DF_{i,j}$  values are normalized relative to their sum to result in a total of 1. The food term and normalized DF were calculated as,

$$f_k = \left( \frac{AE \cdot NIR \cdot NDF_{i,j}}{k_2} \right) \quad \{5\}$$

$$NDF_{i,j} = \left( \frac{DF_{i,j} \cdot S}{\sum (DF_{i,j} \cdot S)} \right) \quad \{6\}$$

where:

$f_k$	Food term for the $k^{\text{th}}$ trophic level (unitless)
AE	Toxicant assimilation efficiency ( $\mu\text{g}$ toxicant absorbed/ $\mu\text{g}$ toxicant ingested)
NIR	Weight-normalized food intake rate (intake (g)/body weight (g)/d)
$DF_{i,j}$	Dietary fraction of $j^{\text{th}}$ prey item in $i^{\text{th}}$ predator diet (unitless)
$NDF_{i,j}$	Dietary fraction normalized over all preferred food items (unitless)
$k_2$	Toxicant elimination rate ( $\text{d}^{-1}$ ); Equation {16} or {17}
S	Size switch (unitless); Equation {19}

Organisms at Level 1 are assumed to be autotrophic, such that all mercury accumulation results from uptake from surface water per Equation {1}. Small aquatic organisms with varied feeding habits (microinvertebrates, macroinvertebrates, zooplankton) are also included at Level 1, on the assumption that uptake from water (or surface adsorption) would outweigh uptake from diet to the point where uptake from diet was insignificant [27].

Total residue accumulation from all pathways to a given compartment at level 2 or higher is termed the biomagnification factor (BMF) to indicate that residue accumulation in the entire food web to that compartment is being addressed. For food chains of aquatic organisms, BAF values are not directly additive at Level 2 and higher because their bioconcentration would then be counted more than once [27]. To sum residue accumulation in multiple food chains containing several trophic levels of aquatic organisms, variations of the following equations are used:

$$\text{Level 2} \quad BMF_2 = (BCF_2 + \sum f_2 BCF_1) \cdot f_E \quad \{7\}$$

$$\text{Level 3} \quad BMF_3 = (BCF_3 + \sum f_3 BMF_2) \cdot f_E \quad \{8\}$$

$$\text{Level 4} \quad BMF_4 = (BCF_4 + \sum f_4 BMF_3) \cdot f_E \quad \{9\}$$

$$f_E = 1 - \exp(-k_2 \cdot (t \cdot 365)) \quad \{10\}$$

where:

- BMF<sub>1-4</sub> Biomagnification factor at trophic levels 1 through 4 (L/kg)
- BCF<sub>1-4</sub> Bioconcentration factor at trophic levels 1 through 4 (L/kg)
- f<sub>1-4</sub> Food term at trophic levels 1 through 4 (unitless)
- f<sub>E</sub> Fraction of equilibrium attained at time of consumption (unitless)
- k<sub>2</sub> Elimination rate (d<sup>-1</sup>); Equation {17} or {18}
- t Average age at time of consumption (years); Equation {15}

These equations are used to calculate the BMF for aquatic organisms at Level 2 and higher because they account for total residue accumulation in all pathways in lower trophic levels. The fraction of contaminant equilibrium attained ( $f_E$ ) is dependent on an organism's age (c.f., Equation {15}) at time of consumption; it applies only to the invertebrate and fish compartments.

Given estimates of inorganic mercury and MeHg BMF values, the total mercury concentration in fish tissue is estimated as,

$$C_{Tn} = \left[ \frac{(C_{IN} \cdot BMF_{IN}) + (C_{MEHg} \cdot BMF_{MEHg})}{CF} \right] \quad \{11\}$$

and the target level for total mercury in surface water as,

$$TL_n = \left[ \frac{TC}{BMF_{MEHg} \cdot \Omega} \right] \cdot CF \quad \{12\}$$

where:

- C<sub>Tn</sub> Total mercury concentration in the n<sup>th</sup> fish species (mg/kg)
- C<sub>IN</sub> Inorganic mercury concentration in surface water (ng/L)
- C<sub>ME</sub> Methylmercury concentration in surface water (ng/L)

$BMF_{INn}$	Inorganic mercury biomagnification factor for the $n^{\text{th}}$ fish species (L/kg)
$BMF_{ME n}$	MeHg biomagnification factor for the $n^{\text{th}}$ fish species (L/kg)
$TL_n$	Total mercury target level for the $n^{\text{th}}$ fish species (ng/L)
TC	U.S. EPA fish tissue criterion for MeHg (0.30 mg/kg)
$\Omega$	Ratio of dissolved MeHg to total mercury in surface water (unitless)
CF	Conversion factor ( $1 \times 10^6$ ng/mg)

### Model Input Variables

As in most probabilistic modeling analyses, selection of values and definition of distributions is based in part on site-specific data, in part on literature values, and in part on best professional judgment (i.e., informed assumptions). Although their use can obscure significant physiological and ecological differences between fish species, several generalized relationships (e.g., Equations {17}, {18}, {19}) are used to address a lack of species-specific information. The procedures used to derive distributions for the probabilistic analysis generally follow those of MacIntosh et al. [25,26]. All input variables, any distributions defining them, parameter values for these distributions, and references to information which formed the basis for selecting these parameters are summarized in Table 3. Selected variables are discussed further below.

To maintain understood relationships between length, age, and weight, and because substantial length data were available for all eight fish species, body length was used to derive values for both body weight and age. A continuous distribution was fit to measured length data for each species; separate distributions were formed for adult and juvenile fish. Adult fish were assumed to be those 1 year or older. The adult length distribution for a given species was truncated low at a length equivalent to age 1 and truncated high at the mean asymptotic length (length at an infinitely high age,  $L_{\infty}$ ). Adult length at age 1 was determined by the von Bertalanffy growth function,

$$L_t = L_{\infty} (1 - \exp(-K \cdot (t - t_0))) \quad \{13\}$$

where:

$L_t$	Predicted length of fish at age $t$ (cm)
$L_{\infty}$	Asymptotic length [length at an infinitely high age] (cm)
K	Time factor ( $\text{year}^{-1}$ )

t	Age (years)
t <sub>0</sub>	Theoretical age at length 0 (years)

Values for L<sub>∞</sub> and K were obtained from the literature [29]. For consistency, a default t<sub>0</sub> value for each species was estimated from L<sub>∞</sub> and K with the empirical relationship [29],

$$\log(-t_0) = -0.3922 - 0.2752 \cdot \log(L_\infty) - 1.038 \cdot \log(K) \quad \{14\}$$

Growth parameter values are summarized in Table 4. All juvenile length distributions were assumed to be uniform, with a lower bound of 1 cm and an upper bound equal to length at age 1, estimated with the von Bertalanffy growth function (Equation {13}) [29]. Parameters for these distributions are given in Table 3. Comparisons of measured length data and resulting fitted distributions for adults are provided in Appendix A.

The average age at time of consumption of individuals comprising the invertebrate and fish compartments was used only to determine the fraction of contaminant equilibrium attained ( $f_E$ ; Equation {10}) in those compartments. Lifespan in all other compartments is assumed to be great enough for practical equilibrium (≈90% of theoretical) to be reached. For juvenile fish, age was estimated simply as the ratio of a value drawn from a uniform juvenile length distribution to a point estimate of length at end of year 1. For adult fish, age was estimated from length data through back-calculation of the von Bertalanffy growth function [29],

$$t = t_0 - \ln(1 - L/L_\infty) / K \quad \{15\}$$

where:

t	Age (years)
L <sub>t</sub>	Length of fish at age <i>t</i> (cm)
L <sub>∞</sub>	Asymptotic length [length at an infinitely high age] (cm)
K	Time factor (year <sup>-1</sup> )
t <sub>0</sub>	Theoretical age at length 0 (years)

For all fish species, body weight was estimated from length as,

$$BW = a \cdot L^b \quad \{16\}$$

where:

- BW Fish body weight (g)
- a Species-specific coefficient (unitless)
- b Species-specific coefficient (unitless)

Initial estimates of “a” and “b” values were obtained from the literature [29], then adjusted using length and weight measurements of fish collected in the WRB. Adjusted values for “a” and “b” are given in Table 3. Comparisons of modeled and measured length-weight relationships are provided in Appendix B. For invertebrates at trophic level 1, body weight and age were assigned a positive 1:1 correlation.

The methylmercury elimination rate in all fish species was estimated as a function of fish body weight and water temperature [30],

$$\ln k_{2(ME)} = c \cdot T - d \cdot \ln BW + e - f \quad \{17\}$$

where:

- $k_{2(ME)}$  MeHg elimination rate ( $d^{-1}$ )
- c Temperature coefficient (unitless); literature value 0.066 (0.019, standard error (SE)); model values in Table 3
- d Body weight coefficient (unitless); literature value 0.20 (0.06 SE); model values in Table 3
- e Acute / chronic exposure value (unitless); literature value 0.73 (0.24 SE) for chronic, 0 for acute; model values in Table 3
- f Constant (unitless); literature value 6.56 (0.45 SE); model values in Table 3
- BW Body weight of fish (g)
- T Surface water temperature ( $^{\circ}C$ )

The elimination rate for Hg[II] in all fish species was estimated as a function of fish weight [25]:

$$k_2 = 0.111 \cdot BW^{0.46} \quad \{18\}$$

where:

$k_2$  Hg(II) elimination rate ( $d^{-1}$ )  
BW Body weight of fish (g)

Food assimilation efficiency has been shown to be directly related to temperature and inversely to fish body size for grass carp [1], and similar relationships may exist for the assimilation efficiency of dietary MeHg. However, because no published information was available to support derivation of such a relationship specifically for MeHg, its assimilation was treated as independent of metabolic rate. Reported point estimates for assimilation efficiency of dietary MeHg range from <0.20 to >0.80, with 0.80 a typical default value [16,30]. However, MeHg bioavailability estimates obtained for channel catfish (*Ictalurus punctatus*) with pharmacological methods were lower (non-compartmental average 0.33, range 0.14 - 0.55; compartmental average 0.29, range 0.12 - 0.42) than those obtained with mass balance methods, suggesting that mass balance methods overestimate the bioavailability of toxicants in fish [32]. The initial, pre-calibration distribution of dietary MeHg assimilation efficiency values for all fish species spanned <0.20 to >0.80, with an assumed median value of 0.50, so that  $AE_{MeHg} \sim \text{Triangular}(0.05, 0.50, 0.95)$ . This distribution was subsequently customized for each species during model calibration.

The daily ingestion rate for all fish was estimated as a function of water temperature and fish body weight using a bioenergetics-based model [22],

$$IR = (0.022 \cdot BW^{0.85}) (\exp(0.06 \cdot T)) \quad \{19\}$$

where:

IR Ingestion rate (kg food / day)  
BW Body weight of fish (kg)  
T Water temperature ( $^{\circ}\text{C}$ )

Food preferences of invertebrates and fish are summarized in Table 2; a matrix of predator-prey interactions included in the model is shown in Table 5 [25,29]. Precise quantification of the dietary fraction ( $DF_{ij}$ ) for each predator-prey interaction was not attempted. For non-preferred food items,  $DF_{ij} = 0$ . For a preferred food item, variability in its actual consumption was expressed by defining  $DF_{ij}$  as a uniform distribution, with a minimum of 0.001 and a maximum of 1.0 (c.f., Table 5). Dietary fractions were normalized to 1 with respect to all preferred food

items ( $NDF_{ij}$ ) before being used for food term ( $f_k$ , Equation {5}) estimation.

Prey size was a factor only for the fish compartments. A predator-prey size relationship factor ( $\Phi$ ) expresses prey size as a function of predator length. For largemouth bass a mean value for  $\Phi$  of 0.34, with a standard deviation of 0.028, has been reported [33]. The ratio of predator-prey sizes is approximately 4:1 (geometric mean ratio  $\approx 3.5:1$ ) among fishes of different species, when sizes are expressed as body lengths [29]. Distributions for  $\Phi$  for each species are given in Table 3. The size switch (S) was computed as:

$$S = \begin{cases} 0 & \text{if } L_{PREY} > (L_{PRED} \times \Phi) \\ 1 & \text{if } L_{PREY} \leq (L_{PRED} \times \Phi) \end{cases} \quad \{20\}$$

where:

S	Size switch (unitless)
$\Phi$	Predator-prey size ratio (unitless)
$L_{PREY}$	Length of prey (cm)
$L_{PRED}$	Length of predator (cm)

## Model Operation

In a quantitative probabilistic exposure model, variability in the result may be due to stochastic variability (heterogeneity), uncertainty (lack of knowledge), or some combination of the two. If both stochastic variability and uncertainty are negligible, the outcome is purely deterministic - a rare occurrence. When uncertainty is negligible, variation in the result is described by a single cumulative density function (CDF) representing the expected statistical variation in, for example, tissue concentration or target level. If neither uncertainty or stochastic variability are negligible, there are multiple CDFs representing variability because, due to lack of knowledge, the exact position of the one CDF that correctly represents variability cannot be known [34]. For uncertainty analysis, considerable judgment is involved in distinguishing which input variables should be modeled as only stochastic, which as only imperfectly known (uncertain), and which as influenced by both sources of variation [35]. However, making this distinction allows establishing two confidence intervals: one due to stochastic variability that is impractical to reduce and another due to uncertainty (lack of knowledge) that may be reducible with judicious collection of additional data. For one-dimensional (1-D) Monte Carlo (MC) analyses all variables were considered to be purely stochastic. In a two-dimensional (2-D) MC analysis of tissue concentration eleven variables (adult body length in eight species, surface

water total dissolved mercury concentration, surface water dissolved methylmercury concentration, and water temperature) were identified as uncertain; for target level analysis ten variables (adult body length in eight species, water temperature, and the dissolved MeHg:total mercury ratio) were identified as uncertain. A variable was identified as uncertain primarily on the basis of the practicality of obtaining further knowledge about it through measurement (within the constraints typically imposed by budget, schedule, and logistics). No variables were treated as second-order random variables [36].

The model was constructed in an MS Excel® (Microsoft Corporation, Redmond, Washington) spreadsheet environment. Circular cell references must be permitted to allow for simulation of food web feedback loops (e.g., adult-juvenile predation within the same species). Probabilistic analyses were performed with an MS Excel® compatible software capable of performing 1-D and 2-D MC analyses (Crystal Ball®, Decisioneering, Inc., Denver, Colorado), using Latin hypercube sampling, with a fixed random number seed. One-dimensional results are based on 10,000 model iterations; two-dimensional results derive from 50 outer loop (uncertainty) iterations of 100 inner loop (variability) iterations.

## **RESULTS AND DISCUSSION**

### **Model Calibration**

Prior to model calibration, empirical density functions (EDFs) were formed from measured tissue concentrations of dissolved total mercury and MeHg water column concentration distributions were estimated from measured data. Cumulative density functions (CDFs) of estimated tissue concentrations were then generated using literature values for all variables, with the exception of the empirically-derived surface water concentrations, and 1-D MC techniques. Pre-calibration results for tissue concentration are shown in Figures 5 to 12 and for tissue-body length relationships in Appendix C. A strong positive correlation between fish length and mercury tissue concentration has been reported for large (>120 mm) fish in Oregon state-wide [37]. Measured data from the WRB show a negligible to moderate positive relationship between tissue concentration and body length depending on species (c.f., Appendix C). Calibrating the model to WRB conditions involved minimizing the differences, particularly at the median, between model-generated CDFs and measured EDFs of mercury tissue concentration and replicating, to the extent practicable, observed mercury tissue concentration - body length relationships. During calibration the number of variables adjusted was kept to a minimum and all adjustments were maintained within limits imposed by measured and literature

data. Following calibration, estimated tissue concentration CDFs were generated with 1-D and 2-D MC techniques. Two-dimensional MC analyses assessed the effect of uncertainty in dissolved total mercury and MeHg concentrations in surface water, water temperature, the dissolved MeHg:total mercury ratio, and adult fish body length) on tissue concentration and target level estimates.

The Weibull plotting position  $\{\text{rank}/(n+1)\}$  method was used to generate EDFs for each fish species, using data collected between 1969 and 1997 for basin-wide water quality monitoring (Figure 2). These EDFs indicate the probability of observing a specific tissue concentration for a given species on a basin-wide, long-term average basis. Measured tissue total mercury concentrations are highest in adult piscivorous northern pikeminnow (median  $\approx 0.57$  mg/kg, wet weight) and lowest in adult, largely invertivorous, cutthroat trout (median  $\approx 0.11$  mg/kg, wet weight) (Figure 2). Similarly, the probability of a northern pikeminnow exceeding the U.S. EPA tissue criterion is  $\approx 80\%$ . Mercury concentrations obtained through water quality monitoring are higher than those for large ( $>120$  mm) invertivores and piscivores probability sampled throughout western Oregon (c.f., Table 7) [37]. But average mercury concentrations for western Oregon are higher than the national average of 0.10 mg/kg [7]. In certain WRB reservoirs, elevated tissue mercury concentrations ( $>1$  mg/kg, wet weight) are thought to be associated with mercury releases from legacy mining activities [38]. This suggests that some monitoring events occurred, intentionally or unintentionally, at locations with anthropogenically elevated mercury levels.

Of interest is whether observed mercury tissue concentrations could pose a risk to fish themselves, as well as to those preying on these fish. Lowest lowest-observed-effect-[tissue] concentrations for several toxicological endpoints (behavior, biochemical, cellular, developmental, growth, mortality, physiological) range from 0.25 to 2.4 mg/kg (wet weight) in rainbow trout, chum salmon, and walleye [39]. As some mercury tissue concentrations measured in the WRB are within this range (Figure 3), the potential exists for observed concentrations to cause adverse health effects in the fish themselves. This potential may be mitigated by acclimatization to naturally elevated ambient mercury levels, as well as other factors not considered in the laboratory tests forming the basis for these residue effect levels.

Model estimation of tissue mercury concentrations requires knowledge of dissolved total (inorganic and MeHg concentrations combined) and MeHg concentrations in surface water.

Such data are sparse for the WRB, as are synoptic surface water-fish tissue data, owing largely to the lack, until very recently, of adequately sensitive analytical techniques for differentiating inorganic mercury and MeHg in surface water. Early (pre-1995) studies have reported inorganic mercury concentrations in surface water possibly influenced by anthropogenic mercury sources (e.g., legacy mining activities) [40,41]. Samples collected in 1995 from the Row River and Coast Fork Willamette River (southern WRB) had total mercury concentrations of 2.60 and 5.00 ng/L (unfiltered), respectively and 1.47 and 2.70 ng/L (filtered), respectively. MeHg concentrations in unfiltered samples were 0.1 and 0.3 ng/L, respectively, while those in filtered samples were 0.05 and 0.2, respectively [42]. Samples collected in June 1998 near Cottage Grove Reservoir (southern WRB) showed dissolved MeHg concentrations ranging from 0.022 to 0.255 ng/L and total MeHg concentrations ranging from 0.022 to 0.142 ng/L [42]. Samples collected in 2001 in the Willamette River (including Portland Harbor) showed total mercury concentrations (unfiltered) from <0.05 ng/L (detection limit, EPA Method 1631) to 4.95 ng/L (highest maximum) [43]. Total mercury values in unfiltered samples from the Tualatin River, a tributary of the Willamette, ranged from <1.0 ng/L to 1.9 ng/L [43]. Total mercury concentrations in filtered samples ranged from <0.05 ng/L to 1.65 ng/L (highest maximum) in the Willamette River (including Portland Harbor) [43]. A two-year study to gather mercury data specifically in support of the WRB TMDL was initiated by the Oregon Department of Environmental Quality in 2002. Water and sediment samples were collected throughout the WRB in 2002 and analyzed for both total mercury and MeHg (using EPA Method 1631) [44]. First round results show mean total mercury concentrations in filtered and unfiltered samples of 0.58 ng/L and 1.03 ng/L, respectively, with mean MeHg concentrations in filtered and unfiltered samples of 0.08 ng/L and 0.10 ng/L, respectively [44]. Distributions for dissolved (filtered) total mercury and MeHg concentrations given in Table 3 are derived from these data, as they have met strict quality control / quality assurance criteria and are assumed to best reflect basin-wide concentrations and are consistent with previous post-1995 results for specific locations in the WRB.

Sensitivity analysis shows (Table 6) that contributions to variance in tissue estimates come primarily from variables associated with the dietary exposure pathway: MeHg elimination rate coefficients, MeHg assimilation efficiency, adult body length, and, for some species, MeHg bioconcentration factors for their food items. Pre-calibration tissue concentration distributions were within an order of magnitude (Figures 4 to 11) and tissue concentration-length relationships generally within the measured 90 percent confidence interval (Appendix C). The pre-calibration model could have been used, although with greater uncertainty, to estimate

target levels. Calibration served to reduce uncertainty in the model and make it clearly WRB-specific. Calibration thus focused on changing (1) MeHg assimilation rate distributions for four species (CAR, LSS, CTT, SMB), (2) the MeHg elimination rate body weight coefficient (“d” in Equation {17}) for four species (NPM, LMB, LSS, CAR), and (3) the MeHg bioconcentration factor for BLU. The MeHg elimination rate acute/chronic variable (“e” in Equation {17}) was set at zero for all species. Although the MeHg elimination rate temperature coefficient (“c” in Equation {17}) did not require adjustment, its contribution to variance (Table 6) suggests that multiple sub-basin models may be needed to more accurately represent the water temperature differences across the WRB system. Changes necessary for calibration indicate the challenges posed when generic uptake or loss relationships are applied to a given species in a given environment, as well as the need to better understand MeHg uptake and loss kinetics in all fish species, but particularly trophic level 2 fish species such as carp and largescale sucker.

### **Fish Tissue Concentrations**

Measured values for total and MeHg mercury concentrations, Equation {11}, and 1-D MC methods (variability and uncertainty combined) were used to generate tissue total mercury concentrations CDFs for comparison with EDFs of measured tissue concentrations. Measured tissue concentration EDFs ( $\square$ ) and 1-D MC modeled tissue concentration CDFs ( $\times$ ) are in general agreement and estimated and measured median and mean values are within one standard deviation (Figures 4 to 11 and Table 7). A 2-D MC analysis produces multiple CDFs representing variability because, due to uncertainty, the exact position of the one “correct” CDF is unknown. There is a 90 percent probability that the correct CDF will fall within the 90 percent confidence bounds on the variability in the tissue concentration estimate [34]. Measured tissue concentration EDFs ( $\square$ ) fall within this confidence range for all species (Figures 4 to 11). The northern pikeminnow, for example, has measured and estimated mean mercury tissue concentrations of 0.60 and 0.83 mg/kg, respectively, and measured and estimated median values of 0.57 and 0.55 mg/kg, respectively (Table 7). The 90 percent confidence range of the model estimate was 0.05 to 5.12 mg/kg, fully bracketing the measured EDF (Figure 4).

### **Biomagnification Factors**

Model estimates of biomagnification factors for each of the eight species are summarized in Table 8. Modeled BMF values are highest for trophic level 4 piscivorous species (northern pikeminnow, large and smallmouth bass), somewhat lower for trophic level 3 omnivorous species, and within range of “direct estimate” mercury BMF values reported by U.S. EPA for

trophic level 3 and 4 fish species [4]. This model does not, therefore, represent a sharp departure from prior mercury BMF estimates derived with other methods. Nationally, U.S. EPA's tissue criterion of 0.30 mg/kg (wet weight) has been equated to an average MeHg water concentration of 0.058 ng/L for an age-3 largemouth bass [9]. For modeled largemouth bass BMF values in Table 8, equivalent MeHg water concentrations range from 0.01 ng/L to 0.25 ng/L, overlapping the national estimate. Using U.S. EPA's direct estimated BMF values, this range is 0.02 ng/L to 0.92 ng/L. This suggests that applying a national default BMF, rather than the model-derived BMF, to fish species in the WRB could result in a mercury target level less than that necessary for protection of human health.

### **Surface Water Mercury Target Level**

A fish tissue criterion of 0.30 mg/kg, a distribution for the fraction of total mercury that is dissolved MeHg ( $\Omega$ ), a model estimated value for  $BMF_{ME}$ , and Equation {12} were used to estimate total mercury surface water target levels for each fish species. The distribution of ratios of dissolved MeHg to total dissolved inorganic mercury ratios measured in the WRB (mean 0.20, range 0.072 - 0.35,  $n = 20$ ) falls between U.S. EPA values for the epilimnion (mean 0.078) and the hypolimnion (mean 0.36) (Figure 12). However, water quality-based pollution control activities traditionally rely on the total concentration of the inorganic metal form, not the dissolved organic form, making  $\Omega$  the ratio of dissolved MeHg to total mercury. For the WRB, the mean value of  $\Omega$  for dissolved MeHg to total mercury is 0.13 (range 0.01 to 0.27,  $n = 20$ ) [44]. The 2-D analysis of target levels treated  $\Omega$  as an uncertain variable. Target level calculation results are summarized in Table 9. For the northern pikeminnow, for example, when the total mercury water concentration exceeds 2.62 ng/L (1-D MC estimate), there is a 95 percent probability that the tissue concentration in an individual fish will exceed the criterion. This probability falls to 50 percent when the water concentration is  $\approx 0.4$  ng/L, and to 5 percent at 0.05 ng/L. When uncertainty is separated from stochastic variability (2-D MC estimate), there is a 95 percent probability of a tissue criterion exceedance in 95 percent of the population when the target level exceeds 9.20 ng/L. Conversely, at a target level of 0.03 ng/L, there is only a 5 percent probability of a criterion exceedance in 5 percent of the population.

Selection of an actual mercury TMDL target level for the WRB is a matter of public policy, will require further discussions among the Agency and WRB stakeholders, and will likely not depend on this model alone. The model does, however, provide decision makers with several choices, with differing degrees of uncertainty, for a total mercury surface water target level. For

example, a conservative choice would be the upper 95<sup>th</sup> percentile for the northern pikeminnow or  $\approx 0.03$  ng/L (Table 9). At this level, it is expected that 95 percent of the northern pikeminnow population in the WRB would achieve U.S. EPA's tissue criterion. The Oregon Department of Human Services initiates a fish consumption advisory when the average tissue concentration exceeds its tissue criterion of 0.35 mg/kg. Thus the mean, 0.77 ng/L (1-D MC) could be chosen (Table 9), with the expectation that U.S. EPA's tissue criterion would not be exceeded, on average, for an individual northern pikeminnow. A much less conservative choice would be the lower 5<sup>th</sup> percentile for the northern pikeminnow, or  $\approx 9.20$  ng/L. At this level, it is expected that only 5 percent of the northern pikeminnow population in the WRB would achieve the protective tissue criterion. The California Regional Water Quality Control Board has proposed a mercury target for the San Francisco Bay estuary of 0.05 ng/L (the lower of human (0.1 ng/L) and wildlife (0.05 ng/L) targets) [6]. Their 0.1 ng/L human health target was based on the Food and Drug Administration action level of 1 mg/kg MeHg in fish tissue and a default bioaccumulation factor of  $10^7$ . Their 0.05 ng/L value is U.S. EPA's 1997 criterion value for wildlife [4,6]. In the southeastern U.S., U.S. EPA Region 6 has proposed MeHg targets ranging from 0.2 to 0.4 ng/L for the Ouachita River Basin and Bayou Bartholomew in Arkansas and Louisiana, using a BAF of  $6.8 \times 10^6$  L/kg and an MeHg:total Hg ratio of 0.2 [45].

When selecting any TMDL target level it should be noted that detection limits for cold vapor atomic absorption (CVAA) are approximately 40 to 500 ng/L and for U.S. EPA Method 1631 (low level, clean sampling) approximately 0.05 to 2 ng/L. For any target level  $\leq 2.0$  ng/L, the more logistically demanding and costly Method 1631 will be required for compliance verification, even though false negatives would still occur. These increased demands and costs may have an impact on the nature and extent of water quality and compliance monitoring activities. In addition, U.S. EPA's fish tissue criterion for mercury simply reflects a level of mercury that could be consumed by humans without inducing adverse health effects. It was developed without reference to specific conditions in regions to which it might be applied. In some regions, natural levels (i.e., those not associated with anthropogenic sources of mercury contamination), that may be difficult, if not impossible, to mitigate or eliminate, may be sufficient to generate tissue concentrations equal to or greater than this criterion. Such ambient levels should be a factor when selecting a target level.

## **Conclusion**

At its current level of development, this model appears able to reasonably approximate the

behavior of Hg(II) and MeHg in WRB aquatic food webs. For selected species in the WRB, it is capable of (a) estimating the probability of a specific fish tissue mercury concentration within the range of such probabilities actually measured in these species and (b) closely approximating observed fish tissue concentration-body length relationships. It can thus be used to estimate a surface water mercury concentration linked to acceptable tissue levels in WRB fish populations. Confidence range widths suggest that further quantification of up to eleven variables categorized as uncertain (but measurable) would enhance the WRB-specificity of the model and its usefulness for establishing a target level. Developing and manipulating this model has been a useful exercise. Aside from supporting target level selection, it has highlighted both data gaps and the assumptions made to bridge them, suggested where such gaps and assumptions could be filled or tested with further research and data collection, and provided a guide for asking more informed questions about MeHg behavior in aquatic systems, and in the WRB in particular.

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**Table 1. Compartments and representative species for differing trophic levels in the Willamette River food web model**

COMPARTMENT	TROPHIC LEVEL	REPRESENTATIVE SPECIES
Surface water	Level 0 [source media]	Open water overlying sediment.
Detritus	Level 1 [2° carbon source]	Dead or decaying algae and weeds; technically called organic detritus to distinguish it from the mineral detritus classified by geologists. Serves as a secondary organic carbon source
Aquatic macrophytes	Level 1 [1° producer]	Higher aquatic plants; in the sense of "higher" evolutionarily than algae and having roots and differentiated tissues; may be emergent (cattails, bulrushes, reeds, wild rice), submergent (water milfoil, bladderwort) or floating (duckweed, lily pads).
Phytoplankton	Level 1 [1° producer]	Microscopic floating plants, mainly algae, that live suspended in water and that drift about because they cannot move by themselves or because they are too small or too weak to swim effectively against a current. A basic food source in many aquatic ecosystems.
Periphyton	Level 1 [1° producer]	A complex matrix of algae and heterotrophic microbes attached to submerged substrata in almost all aquatic ecosystems. An important food source for invertebrates and some fish.
Zooplankton	Level 2 [1° consumer]	Animal portion of the living particles in water that freely float in open water, eat bacteria, algae, detritus and sometimes other zooplankton and are in turn eaten by planktivorous fish. May include planktonic caldocerans, copepods, ostracods, mites
Aquatic insect larvae	Level 2 [1° consumer]	Primarily benthic, including chironomid, trichopteran, ephemeropteran, and dipteran larvae
Aquatic crustaceans	Level 2 [1° consumer]	Crayfish
Aquatic insects	Level 2 [1° consumer]	Primarily pelagic, including a variety of diving insects and those that skim the water surface such as water boatmen, pond skimmers, etc.
Aquatic mollusks	Level 2 [1° consumer]	Mussels, snails, clams, etc.
Aquatic worms	Level 2 [1° consumer]	Oligochaetes

**Table 1. Compartments and representative species for differing trophic levels in the Willamette River food web model**

COMPARTMENT	TROPIC LEVEL	REPRESENTATIVE SPECIES
Omnivorous fish	Level 3 [2° consumer]	Northern pikeminnow (juvenile) Largemouth bass (juvenile) Largescale sucker (juvenile, adult) Common carp (juvenile, adult) Rainbow trout (juvenile, adult) Cutthroat trout (juvenile, adult) Smallmouth bass (juvenile) Bluegill (juvenile, adult)
Piscivorous fish	Level 4 [3° consumer]	Northern pikeminnow (adult) Largemouth bass (adult) Smallmouth bass (adult)
Top piscivorous predator	Level 5	Humans

**Table 2. Food item preferences of representative species for different trophic levels in the Willamette River food web model**

REPRESENTATIVE SPECIES	FOOD PREFERENCE	
	JUVENILE	ADULT
Zooplankton		Phytoplankton Detritus
Aquatic insect larvae	Periphyton Aquatic macrophytes Detritus Zooplankton	
Aquatic crustaceans		Aquatic macrophytes Detritus
Aquatic insects		Aquatic insect larvae Aquatic macrophytes Aquatic worms Zooplankton
Aquatic mollusks		Phytoplankton Zooplankton
Aquatic worms		{Sediment} * Detritus Periphyton
Northern pikeminnow [46,47] <i>Ptychocheilus oregonensis</i> piscivore, edible	Fork Length (FL) < 30cm Zooplankton Aquatic insect larvae Aquatic insects	Fish (salmonids) Aquatic crustaceans Aquatic insects Aquatic mollusks
Largemouth bass [46] <i>Micropterus salmoides</i> piscivore, edible	Zooplankton Aquatic insect larvae Aquatic crustaceans Aquatic worms Small fish (FL < 2-5 cm)	Aquatic insect larvae Aquatic crustaceans Aquatic worms Fish (fishminnows, juvenile carp)
Smallmouth bass [47,48] <i>Micropterus dolomieu</i> piscivore, edible	Zooplankton Aquatic insect larvae Aquatic insects + small fish bulk of diet at FL > 4 cm	Aquatic crustaceans Small fish
Largescale sucker [46,48] <i>Catostomus macrocheilus</i> benthivorous omnivore, edible	Detritus Zooplankton Aquatic insect larvae	Phytoplankton Periphyton Aquatic insects Aquatic mollusks

**Table 2. Food item preferences of representative species for different trophic levels in the Willamette River food web model**

REPRESENTATIVE SPECIES	FOOD PREFERENCE	
	JUVENILE	ADULT
Common carp [49] <i>Cyprinus carpio</i> benthivorous omnivore, edible	Phytoplankton Periphyton Zooplankton Aquatic insect larvae	Aquatic macrophytes Periphyton Aquatic crustaceans Aquatic insects Aquatic worms Aquatic mollusks
Rainbow trout [50] <i>Oncorhynchus mykiss</i> pelagic invertivore, edible	Zooplankton	Aquatic insect larvae Aquatic crustaceans Aquatic insects Aquatic mollusks Small fish
Cutthroat trout [51] <i>Oncorhynchus clarkii</i> pelagic invertivore, edible	Zooplankton	Aquatic insect larvae Aquatic crustaceans Aquatic insects Small fish
Bluegill [46] <i>Lepomis macrochirus</i> pelagic omnivore, edible	Aquatic macrophytes Detritus Phytoplankton Zooplankton Aquatic crustaceans Aquatic insect larvae Aquatic worms	{Sediment} * Aquatic macrophytes Phytoplankton Zooplankton Aquatic insects larvae Aquatic crustaceans Small fish

\* Incidental ingestion of sediment was not included as an exposure pathway.

**Table 3. Input variables and parameter values used in the Monte Carlo simulations of total mercury concentrations in fish tissue from the Willamette River Basin.**

VARIABLE	UNITS	DISTRIBUTION	PARAMETERS	COMMENTS / REFERENCES
<b>ENVIRONMENTAL VARIABLES</b>				
Total dissolved mercury concentration in surface water	ng/L	Lognormal	0.58, 0.49	Best-fit to field data, $n = 20$ . Based on recently (2002) measured total dissolved mercury levels in WRB surface water [44].
Dissolved MeHg concentration in surface water	ng/L	Lognormal	0.08, 0.02	Best-fit to field data, $n = 20$ . Based on recently (2002) measured dissolved MeHg levels in WRB surface water [44].
Dissolved MeHg-total mercury ratio ( $\Omega$ )	unitless	Gamma	Loc = -0.05, scale = 0.03, shape = 7.117, lower bound = 0; upper bound = 1	Best fit to recently (2002) measured ratio of total to MeHg levels in WRB surface water [44]. Kolmogorov-Smirnov (K-S) $p = 0.075$ .
Water temperature	°C	Triangular	6.0, 12.5, 22.0	Between 1969 and 1992, median temperatures in the Willamette River temperatures at Portland ranged from a low of 5.6 °C to a high of 21.8 °C, with a most likely value of $\approx 12.5$ °C [41]. This range was assumed to encompass all temperature regimes within the WRB.
<b>MeHg BIOCONCENTRATION FACTOR</b>				
Detritus (DET)	L/kg	Logtriangular	2.50, 3.00, 3.50	Assumed to be the same as that for Hg[III] (see below).
Aquatic macrophytes (AQP)	L/kg	Logtriangular	0.80, 2.15, 3.50	Values for submergent and emergent portions of six species of aquatic vascular plants from 34 to 3500 and 6 to 32, respectively [52]. Values for duckweed ( <i>Lemna minor</i> ) (480, 2950); reed grass ( <i>Phragmites communis</i> ) (850, 25, 530, 74, 139); bulrush ( <i>Scirpus lucustris</i> ) (90, 790, 8, 1250, 39, 190); yellow iris ( <i>Iris pseudacorus</i> ) (20, 40, 18, 34, 31, 90) [53]. Distribution defined by minimum, geometric mean, and maximum of these data.
Phytoplankton (PHY)	L/kg	Triangular	3.50, 4.50, 5.50	Values for <i>Scenedesmus obliquus</i> and <i>Microcystis incerta</i> from 761 to 2100 and from 461 to 990, respectively [25,54]. Values of 1200 and 2610 for filamentous algae <i>Oedogonium</i> sp. [53]. Values of 3400, 38400, 107000, and 133000 have been calculated for phytoplankton [4]. Distribution spans these calculated values.
Periphyton (PER)	L/kg	Triangular	3.50, 4.50, 5.50	Assumed same as PHY compartment
Zooplankton (ZOO)	L/kg	Logtriangular	2.45, 3.90, 5.40	Value for <i>Gammarus</i> sp. of approximately 8000 [50]. A value of 249000 for the marine copepod <i>Acartia clausi</i> [55]. Values of 3570 and 286 for water fleas ( <i>Daphnia</i> sp.) and caldocerans ( <i>Eurycerus</i> ), respectively [53]. Distribution defined by minimum, geometric mean, and maximum of these data.

**Table 3. Input variables and parameter values used in the Monte Carlo simulations of total mercury concentrations in fish tissue from the Willamette River Basin.**

VARIABLE	UNITS	DISTRIBUTION	PARAMETERS	COMMENTS / REFERENCES
Aquatic insect larvae (AQL)	L/kg	Logtriangular	2.80, 3.40, 4.10	Values for <i>Chironomus riparius</i> from 3000 to 5000. Values for bloodworms (Chironomidae) (988, 3070, 12700); mayfly (Ephemeroidea) naiads (900, 3290, 690); caddisfly ( <i>Tricoptera</i> sp.) larve (710), dragonfly ( <i>Odonata</i> sp.) nymphs (1296), damselfly ( <i>Odonata</i> sp.) nymphs (1186), crane fly ( <i>Tipula</i> sp.) larvae (625), alderfly ( <i>Sialis lutaria</i> ) larvae (1270), great diving beetle ( <i>Dytiscus marginalis</i> ) larvae and imago (3134, 800) [53]. Distribution defined by minimum, geometric mean, and maximum of these data.
Aquatic crustaceans (AQC)	L/kg	Loguniform	2.45, 5.40	Specific BCF data not available for this compartment. Uncertainty bounds span to those for AQL and ZOO compartments.
Aquatic insects (AQI)	L/kg	Logtriangular	2.80, 3.15, 3.50	Values for adult lesser water boatman ( <i>Corixa</i> sp.) (4200, 8470, 740), water boatman ( <i>Notonecta glauca</i> ) (2460, 674), pond skater ( <i>Gerris najas</i> ) (754), aquatic saw bug ( <i>Asellus aquaticus</i> ) (954), and water spiders (Hydracnidae) (624) [53]. Distribution defined by minimum, geometric mean, and maximum of these data.
Aquatic mollusks (AQM)	L/kg	Logtriangular	3.00, 4.20, 5.40	Values for pond snails ( <i>Planorbis</i> sp.) (1280, 3570, 1970), giant pond snails ( <i>Lymnaea stagnalis</i> ) (1800, 3480, 1178), and the snail <i>Physa fontinalis</i> (4266) [53]. Value of 249000 calculated from uptake and depuration data for marine bivalve <i>Crassostrea virginica</i> exposed to (CH <sub>3</sub> COO) <sub>2</sub> for 45 days [56]. Distribution defined by minimum, geometric mean, and maximum of these data.
Aquatic worms (AQW)	L/kg	Logtriangular	2.00, 2.65, 3.30	Values for annelids <i>Haemopsis sanguisuga</i> (2030, 450, 1148) and <i>Glossosiphonia complanata</i> (110, 640), as well as Oligochaeta (1780, 690) [53]. Distribution defined by minimum, geometric mean, and maximum of these data.
NPM, LMB, LSS, CAR, RBT, CTT, SMB, BCF	L/kg	Logtriangular	3.00, 4.50, 6.00	Values for brook trout (10000, 12000, 23000) and one for rainbow trout (11000) [54]. Values for juvenile rainbow trout ( <i>Salmo gairdneri</i> ) (4525, 6628, 8033), bluegill sunfish ( <i>Lepomis macrochirus</i> ) (1138, 2454), and pike ( <i>Esox lucius</i> ) organs (7673, 7230, 2002, 2198) [57,58]. Values for brook trout exposed to varying concentrations of MeHg for 28 to 38 weeks, were 127000 [59] and 69000 to 630000 [60]. Distribution spans these data.
BLU <sup>†</sup>	L/kg	Logtriangular	3.00, 6.50, 7.00	Calibration adjustment.
<b>MeHg ASSIMILATION EFFICIENCY</b>				
ZOO	unitless	Triangular	0.20, 0.5, 0.8	[25]
AQL	unitless	Triangular	0.50, 0.76, 0.95	[25]

**Table 3. Input variables and parameter values used in the Monte Carlo simulations of total mercury concentrations in fish tissue from the Willamette River Basin.**

VARIABLE	UNITS	DISTRIBUTION	PARAMETERS	COMMENTS / REFERENCES
AQC	unitless	Uniform	0.50, 0.95	Efficiency of 72% and 76% reported for blue crabs and pink shrimp, respectively [61]. Mode is mean of these values, minimum and maximum are $\pm 30\%$ of this mean.
AQI	unitless	Uniform	0.5, 0.95	Based on [25]; but a high minimum is conservatively assumed.
AQM	unitless	Triangular	0.5, 0.75, 0.95	Efficiency of 72% reported for marine bivalve, <i>Mytilus edulis</i> , exposed to MeHg for 80 days [62]. Distribution bounds reflect high uncertainty owing to lack of freshwater mollusk data.
AQW	unitless	Uniform	0.50, 0.95	Specific assimilation efficiency data were not available for this compartment. Maximum approaches maximum possible; a high minimum is conservatively assumed.
BLU, RBT	unitless	Triangular	0.05, 0.50, 0.95	Distribution based on assimilation efficiencies of 0.94, 0.815, and 0.15 for yellow perch, mosquito fish, predacious fish, respectively [16,25,63].
NPM <sup>†</sup>	unitless	Triangular	0.40, 0.45, 0.50	Distribution established during calibration.
LMB <sup>†</sup>	unitless	Triangular	0.50, 0.55, 0.60	Distribution established during calibration.
CAR <sup>†</sup>	unitless	Triangular	0.05, 0.05, 0.20	Distribution established during calibration.
LSS <sup>†</sup>	unitless	Triangular	0.05, 0.15, 0.20	Distribution established during calibration.
CTT <sup>†</sup>	unitless	Triangular	0.05, 0.05, 0.50	Distribution established during calibration.
SMB <sup>†</sup>	unitless	Triangular	0.05, 0.10, 0.95	Distribution established during calibration.
<b>MeHg ELIMINATION RATE</b>				
ZOO	day <sup>-1</sup>	Logtriangular	-1.17, -0.69, -0.22	[25]
AQL	day <sup>-1</sup>	Logtriangular	-1.48, -1.00, -0.52	[25]
AQC	day <sup>-1</sup>	Logtriangular	-1.37, -1.06, -0.76	[25]
AQI	day <sup>-1</sup>	Loguniform	-1.48, -1.00, -0.52	Assumed same as AQL compartment.
AQM	day <sup>-1</sup>	Loguniform	-3.00, -0.22	Elimination rates for two marine bivalves, <i>Mytilus edulis</i> and <i>Crassostrea virginica</i> , are 0.0003 and 0.001, respectively [24,64]. Minimum is highest of these data; maximum is highest invertebrate value.
AQW	day <sup>-1</sup>	Loguniform	-2.00, -0.22	Estimated from AQM data.
SMB, CTT, RBT, BLU	day <sup>-1</sup>	Normal	c(0.066, 0.019); d(0.20, 0.06); e(0) f(6.56, 0.45)	Estimated on basis of body weight and temperature with Equation {17}. Coefficient values as given in the literature [30].

**Table 3. Input variables and parameter values used in the Monte Carlo simulations of total mercury concentrations in fish tissue from the Willamette River Basin.**

VARIABLE	UNITS	DISTRIBUTION	PARAMETERS	COMMENTS / REFERENCES
NPM <sup>†</sup>	day <sup>-1</sup>	Normal	c(0.066, 0.019); d(0.28, 0.06); e(0) f(6.56, 0.45)	Estimated on basis of body weight and temperature with Equation {17}. Calibration resulted in body weight coefficient (d) higher than literature value [30].
LMB <sup>†</sup>	day <sup>-1</sup>	Normal	c(0.066, 0.019); d(0.18, 0.06); e(0) f(6.56, 0.45)	Estimated on basis of body weight and temperature with Equation {17}. Calibration resulted in body weight coefficient (d) within 1 SE of literature value [30].
LSS <sup>†</sup>	day <sup>-1</sup>	Normal	c(0.066, 0.019); d(0.54, 0.06); e(0) f(6.56, 0.45)	Estimated on basis of body weight and temperature with Equation {17}. Calibration resulted in body weight coefficient (d) higher than literature value [30].
CAR <sup>†</sup>	day <sup>-1</sup>	Normal	c(0.066, 0.019); d(0.55, 0.06); e(0) f(6.56, 0.45)	Estimated on basis of body weight and temperature with Equation {17}. Calibration resulted in body weight coefficient (d) higher than literature value [30].
<b>Hg[II] BIOCONCENTRATION FACTOR</b>				
DET	L/kg	Logtriangular	2.50, 3.00, 3.50	Value is approximately 1100 [65]. Distribution as given in [25].
AQP	L/kg	Logtriangular	0.50, 1.65, 2.80	Values for six species of aquatic vascular plants reported 3 to 77 and 4 to 264, respectively [52]. Values for duckweed ( <i>Lemna minor</i> ) (70); reed grass ( <i>Phragmites communis</i> ) (56, 149); bulrush ( <i>Scirpus lucustris</i> ) (77, 70); yellow iris ( <i>Iris pseudacorus</i> ) (18, 23) [53], as well as water hyacinth ( <i>Eichhornia crassipes</i> ) (580) [66]. Distribution defined by minimum, geometric mean, and maximum of these data.
PHY	L/kg	Logtriangular	2.90, 3.45, 4.00	Values for four algae types reported from 853 to 10920 [25,54]. Values of 8537 and 871 reported for <i>Crocomonas salina</i> and <i>Oedogonium</i> sp., respectively [53,67]. Distribution defined by minimum, geometric mean, and maximum of these data.
PER	L/kg	Logtriangular	2.90, 3.45, 4.00	Assumed same as PHY compartment.
ZOO	L/kg	Logtriangular	3.40, 3.65, 3.90	Value for <i>Gammarus</i> sp. of 2500. Value of 7600 for the copepod <i>Acartia clausi</i> [54,55]. Distribution defined by minimum, geometric mean, and maximum of these data.
AQL	L/kg	Logtriangular	2.10, 3.20, 4.30	Values for caddisfly ( <i>Tricoptera</i> sp.) larve (513), damselfly ( <i>Odonata</i> sp.) larvae (655), cranefly ( <i>Tipula</i> sp.) larvae (840), and great diving beetle ( <i>Dytiscus marginalis</i> ) larvae and imago (603, 862) [53]. Values for larva and pupa life stages of <i>Chironomus riparius</i> of 19600 and 15600, respectively [68]. Value of 138 for mayfly (Ephemeroidea) larvae [53]. Distribution defined by minimum, geometric mean, and maximum of these data.

**Table 3. Input variables and parameter values used in the Monte Carlo simulations of total mercury concentrations in fish tissue from the Willamette River Basin.**

VARIABLE	UNITS	DISTRIBUTION	PARAMETERS	COMMENTS / REFERENCES
AQC	L/kg	Logtriangular	2.00, 2.25, 2.50	Three values reported for crayfish ( <i>Procambarus clarkii</i> ): 121, 158, 216 [69]. Value of 333 grass shrimp ( <i>Palaemonetes pugio</i> ) [70]. Distribution defined by minimum, geometric mean, and maximum of these data.
AQI	L/kg	Logtriangular	2.60, 3.25, 3.90	Value of 7500 for the adult life stage of <i>Chironomus riparius</i> [68]; 414 and 483 for adult lesser water boatman ( <i>Corixa</i> sp.), water boatman ( <i>Notonecta glauca</i> ), and pond skater ( <i>Gerris najas</i> ) (431) [53]. Distribution defined by minimum, geometric mean, and maximum of these data.
AQM	L/kg	Logtriangular	2.30, 2.60, 2.90	Values for mussels ( <i>Mytilus edulis</i> ) (664, 236), short-necked clams ( <i>Venerupis philiooinarum</i> ) (190), pond snails ( <i>Planorbis</i> sp.) (795), giant pond snail ( <i>Lymnaea stagnalis</i> ) (297), and the snail <i>Physa fontinalis</i> (637) [53]. Distribution defined by minimum, geometric mean, and maximum of these data.
AQW	L/kg	Logtriangular	2.30, 2.78, 3.25	Values reported for annelid <i>Haemopis sanguisuga</i> (670) and Oligochaeta (517) [53]. Distribution defined by minimum, geometric mean, and maximum of these data.
NPM, SMB, LMB, LSS, CAR, RBT, CTT, BLU	L/kg	Logtriangular	0.70, 2.20, 3.70	1800 and 4994 for rainbow trout and fathead minnow, respectively [25]. 97 and 2560 for <i>Serranus cabrilla</i> (marine species) and <i>Gambusia affinis</i> , respectively [60,71]. Juvenile rainbow trout ( <i>Salmo gairdneri</i> ) values of 5, 12, 26 [72]. Distribution defined by minimum, geometric mean, maximum of these data.
<b>Hg[II] ASSIMILATION EFFICIENCY</b>				
ZOO	unitless	Triangular	0.5, 0.6, 0.9	[25]
AQL	unitless	Triangular	0.5, 0.6, 0.9	[25]
AQC	unitless	Triangular	0.5, 0.6, 0.9	[25]
AQI	unitless	Triangular	0.5, 0.6, 0.9	[25]
AQM	unitless	Triangular	0.013, 0.04, 0.12	An efficiency of 4% was reported for the marine bivalve, <i>Mytilus edulis</i> , exposed to Hg[II] for 80 days [62]. Distribution bounds are $\pm 3 \times$ this value.
AQW	unitless	Triangular	0.5, 0.6, 0.9	[25]
NPM, SMB, LMB, LSS, CAR, RBT, CTT, BLU	unitless	Triangular	0.112, 0.172, 0.264	[25]
<b>Hg[II] ELIMINATION RATE</b>				
All invertebrate compartments	day <sup>-1</sup>	Logtriangular	0.0126, 0.126, 1.26	[25]

**Table 3. Input variables and parameter values used in the Monte Carlo simulations of total mercury concentrations in fish tissue from the Willamette River Basin.**

VARIABLE	UNITS	DISTRIBUTION	PARAMETERS	COMMENTS / REFERENCES
NPM, SMB, LMB, LSS, CAR, RBT, CTT, BLU	day <sup>-1</sup>	---	---	Estimated on basis of body weight with Equation {18}.
<b>BODY WEIGHT</b>				
ZOO	g	Triangular	$1.4 \times 10^{-5}$ , $3.3 \times 10^{-5}$ , $7.6 \times 10^{-5}$	[25]
AQL	g	Triangular	$4 \times 10^{-4}$ , $6.25 \times 10^{-4}$ , $9.8 \times 10^{-4}$	[25]
AQC	g	Loguniform	0.1, 4	[25]
AQI	g	Triangular	$4 \times 10^{-4}$ , $6.25 \times 10^{-4}$ , $9.8 \times 10^{-4}$	Assumed same as AQL compartment.
AQM	g	---	n/a	Estimate of mollusk body weight not required, as an estimated body-weight normalized food intake rate value was available (see below)
AQW	g	Loguniform	0.0023, 0.019	Range is based on reported fresh weights for <i>Tubifex tubifex</i> [73]
BLU	g	---	0.05 L <sup>2.8702</sup>	Best-fit to field data, Spearman rank correlation ( $r_s$ ) = 0.736, $p < 0.001$ .
NPM	g	---	0.006 L <sup>3.1079</sup>	Initial values from [74], then best-fit to field data, $r_s = 0.962$ , $p < 0.001$ .
LMB	g	---	0.0185 L <sup>2.292</sup>	Initial values from [29], then best-fit to field data, $r_s = 0.979$ , $p < 0.001$ .
LSS	g	---	0.0175 L <sup>2.8687</sup>	Initial values from [75], then best-fit to field data, $r_s = 0.964$ , $p < 0.001$ .
CAR	g	---	0.028 L <sup>2.8289</sup>	Best-fit to field data, $r_s = 0.933$ , $p < 0.001$ .
RBT	g	---	0.0146 L <sup>2.9748</sup>	Best-fit to field data, $r_s = 0.964$ , $p < 0.001$ .
CTT	g	---	0.009 L <sup>3.0044</sup>	Initial values from [75,76], then best-fit to field data, $r_s = 0.848$ , $p < 0.001$ .
SMB	g	---	0.012 L <sup>3.057</sup>	Best-fit to field data, $r_s = 0.729$ , $p < 0.001$ .
<b>FOOD INTAKE RATE</b>				
ZOO	g/day	Logtriangular	-1.04, -0.56, -0.09	[25,77]
AQL	g/day	Loguniform	-1.00, -0.39	[25]
AQC	g/day	Loguniform	-1.00, -0.39	Assumed same as AQL compartment.
AQI	g/day	Loguniform	-1.00, -0.39	Assumed same as AQL compartment.
AQM	g/day	Logtriangular	-1.65, -1.525, -1.40	Intake rate of 0.025 g(dry)/g(dry)/d estimated for marine bivalve <i>Mytilus edulis</i> on basis of bivalve respiration, growth rate, and food assimilation efficiency [59]. Distribution bounds are $\pm 3 \times$ this estimated value.
AQW	g/day	Loguniform	-1.00, -0.39	Assumed same as AQL compartment.

**Table 3. Input variables and parameter values used in the Monte Carlo simulations of total mercury concentrations in fish tissue from the Willamette River Basin.**

VARIABLE	UNITS	DISTRIBUTION	PARAMETERS	COMMENTS / REFERENCES
All juvenile and adult fish	g/day	---	---	Estimated on basis of body weight and water temperature using Equation {19}.
<b>FISH LENGTH</b>				
BLU (juv)	cm	Uniform	1.0, 10.3	Length range equivalent to age $\leq 1$ year; assumes juvenile populations dominated by younger, smaller individuals.
NPM (juv)	cm	Uniform	1.0, 12.0	See BLU.
LMB (juv)	cm	Uniform	1.0, 17.2	See BLU.
LSS (juv)	cm	Uniform	1.0, 22.3	See BLU.
CAR (juv)	cm	Uniform	1.0, 18.8	See BLU.
RBT (juv)	cm	Uniform	1.0, 21.5	See BLU.
CTT (juv)	cm	Uniform	1.0, 21.5	See BLU.
SMB (juv)	cm	Uniform	1.0, 16.2	See BLU.
BLU (adult)	cm	Weibull	Location = 90.93, Scale = 76.80, Shape = 1.5869	Best-fit to field data. $n = 25$ . Kolmogorov-Smirnov (K-S) $p = 0.098$ . Lower and upper bounds provided in Table 4.
NPM (adult)	cm	Logistic	Mean = 383.00, Scale = 38.00	Best-fit to field data. $n = 62$ . K-S $p = 0.077$ . Lower and upper bounds provided in Table 4.
LMB (adult)	cm	Beta	$\alpha = 6.68$ , $\beta = 6.65$ , Scale = 678.60	Best-fit to field data. $n = 192$ . K-S $p = 0.066$ . Lower and upper bounds provided in Table 4.
LSS (adult)	cm	Logistic	Mean = 458.04, Scale = 30.10	Best-fit to field data. $n = 90$ . K-S $p = 0.094$ . Lower and upper bounds provided in Table 4.
CAR (adult)	cm	Logistic	Mean = 554.41, Scale = 45.20	Best-fit to field data. $n = 43$ . K-S $p = 0.061$ . Lower and upper bounds provided in Table 4.
RBT (adult)	cm	Pareto	Location = 213.94, Shape = 5.61	Best-fit to field data. $n = 36$ . K-S $p = 0.083$ . Lower and upper bounds provided in Table 4.
CTT (adult)	cm	Beta	$\alpha = 16.26$ , $\beta = 4.65$ , Scale = 356.40	Best-fit to field data. $n = 25$ . K-S $p = 0.069$ . Lower and upper bounds provided in Table 4.
SMB (adult) <sup>†</sup>	cm	Uniform	Minimum = 190, Maximum = 410	Best-fit to field data. $n = 10$ . K-S $p = 0.129$ . Lower and upper bounds from field data.
<b>PREDATOR - PREY SIZE RATIO</b>				
NPM, SMB, LSS, CAR, RBT, CTT, BLU	unitless	Triangular	0.225, 0.25, 0.275	Generic species value of 0.25 [from 29] $\pm 10\%$ .
LMB	unitless	Normal	0.34, 0.028	Value for largemouth bass from [33].
<b>LIFESPAN</b>				
ZOO	days	Uniform	10, 20	[25]
AQL	days	Loguniform	1.47, 2.55	[25]

**Table 3. Input variables and parameter values used in the Monte Carlo simulations of total mercury concentrations in fish tissue from the Willamette River Basin.**

VARIABLE	UNITS	DISTRIBUTION	PARAMETERS	COMMENTS / REFERENCES
AQC	days	Loguniform	1.47, 2.55	[25]
AQI	days	Loguniform	1.47, 2.55	[25]
AQM	days	Loguniform	1.47, 2.55	[25]
AQW	days	Loguniform	1.47, 2.55	[25]

† Initial, literature-based, value altered during model calibration.

**Table 4. Parameters for the von Bertalanffy growth function (Equation {12}) used to estimate age of adult fish from measured length data.**

Species	$L_{\infty}$ <sup>a</sup>	K <sup>b</sup>	$t_0$ <sup>c</sup>	$L_0$ <sup>d</sup>	$L_1$ <sup>e</sup>	$L_{max}$ <sup>f</sup>	R- $L_{max}$ <sup>g</sup>	Ref
LMB	65.1	0.170	-0.808	1.0	17.2	97	58	[29,79]
BLU	31.4	0.231	-0.718	1.0	10.3	41	27	[29]
SMB	49.0 <sup>h</sup>	0.210	-0.682	1.0	19.0 <sup>h</sup>	69	39	[29,79]
CAR	74.8	0.157	-0.845	1.0	18.8	120	72	[29]
LSS	61.0	0.300	-0.456	1.0	21.5	61	59	[29,80]
NPM	54.9	0.100	-1.469	1.0	12.0	63	54	[29,78]
CTT	51.8	0.397	-0.356	1.0	21.5	99	33	[29]
RBT	51.8	0.397	-0.356	1.0	21.5	120	38	[29]

- a) Asymptotic length [length at an infinitely high age] (cm). Upper bound of adult length distribution.
- b) Time factor (year<sup>-1</sup>)
- c) Theoretical age at length 0 (years). Estimated with Equation {14}.
- d) Assumed length (cm) at age 0. Lower bound of juvenile length distribution.
- e) Length (cm) at age 1 from Equation {13}. Upper bound of juvenile length distribution; Lower bound of adult length distribution.
- f) Maximum length (cm) reported in the literature [29].
- g) Maximum length (cm) measured in fish collected from the Willamette River Basin.
- h) Values based on best-fit to limited existing data, theoretical values are  $L_{\infty} = 54.5$  and  $L_1 = 16.2$ .

**Table 5. Matrix of predator-prey interactions included in the model.**

pred →											NPM		LMB		SMB		LSS		CAR		RBT		CTT		BLU		
prey ↓		DET	AQP	PHY	PER	ZOO	AQL	AQC	AQI	AQM	AQW	J	A	J	A	J	A	J	A	J	A	J	A	J	A	J	A
DET		■				●	●	●	●		●							●	●		●					●	●
AQP			■				●	●	●									●	●		●					●	●
PHY				■		●				●								●	●	●	●				●	●	●
PER					■		●	●			●							●	●	●	●					●	●
ZOO						■	●	●	●	●		●		●		●		●	●	●	●	●		●	●	●	●
AQL							■		●			●	●	●	●	●	●	●	●	●	●	●	●	●	●	●	●
AQC								■					●	●	●	●	●	●	●	●	●	●	●	●	●	●	●
AQI									■			●	●	●	●	●	●	●	●	●	●	●	●	●	●	●	●
AQM										■	●							●	●	●	●	●	●	●	●	●	●
AQW											■			●		●		●	●	●	●	●	●	●	●	●	●
NPM	J											■	●	●	●	●	●					●		●		●	
	A												■		●		●										
LMB	J											●	●	■	●	●	●					●		●		●	
	A												●		■		●										
SMB	J											●	●	●	●	■	●					●		●		●	
	A												●		●	■											
LSS	J											●	●	●	●	●	●	■				●		●		●	
	A												●		●		●	■									
CAR	J											●	●	●	●	●	●		■			●		●		●	
	A												●		●		●		■								
RBT	J											●	●	●	●	●	●					■	●		●		●
	A												●		●		●						■				
CTT	J											●	●	●	●	●	●					●	■	●		●	
	A												●		●		●							■			
BLU	J											●	●	●	●	●	●					●		●	■	●	
	A												●		●		●									■	

●:  $DF_U \sim \text{Uniform}(0.001, 1.00)$ ; otherwise  $DF_U = 0$ .

**Table 6. Results of a 1-D MC sensitivity analysis of the WRB food web model. Sensitivity is expressed as percentage contribution to variance of the tissue concentration estimate. Only values  $\geq \pm 1.5\%$  are shown. [Numbers in parentheses indicate a negative contribution to variance.]**

Variable	% Contribution to Variance							
	NPM	SMB	LMB	BLU	CAR	LSS	CTT	RBT
Water (filtered) concentration, Hg[II]	8.0	3.7	8.5	6.1	5.7	5.3	6.6	4.4
Water (filtered) concentration, MeHg	8.1	3.8	8.6	6.1	5.8	5.3	6.6	4.4
Water temperature	2.7	1.6	3.8				4.2	4.6
MeHg elimination rate (body weight coefficient)	5.0	4.5	7.5	1.6	13.0	8.3	5.1	5.0
MeHg elimination rate (temperature coefficient)	(6.7)	(2.6)	(4.8)		(2.0)	(2.2)		(1.5)
MeHg elimination rate (constant)	1.8	6.3			8.7	5.7	5.7	3.1
MeHg assimilation efficiency	1.5	29.1		6.3	11.4	6.9	16.7	15.2
Adult body length	9.1	8.8	7.6	4.7	5.1	6.1	2.6	5.8
Dietary fraction, BLU juveniles in diet	2.9	1.8	2.9					1.6
Ingestion rate, AQL	1.5							
Juvenile body length, BLU		2.7		(2.2)			(4.9)	(4.1)
Juvenile body length, CAR					(5.8)			
Juvenile body length, CTT							(2.4)	
Juvenile body length, LSS						(15.0)		
Juvenile body length, RBT								(4.1)
MeHg bioconcentration factor, AQC						2.0		
MeHg bioconcentration factor, BLU	12.3	4.9	11.3	32.1			3.9	3.6
MeHg bioconcentration factor, PER	8.5	4.7	8.3	3.6	9.8	10.5	9.6	10.4
MeHg bioconcentration factor, PHY	3.9		2.6		5.9	6.4		
MeHg bioconcentration factor, ZOO	4.2	2.7	4.2		2.9	2.9	1.5	2.5
MeHg elimination rate, AQW	(2.8)		2.3		(2.6)	(2.1)	(3.5)	(3.5)

**Table 7. Comparison of measured and model estimated (post-calibration) tissue mercury concentrations for eight species of adult fish in the Willamette River Basin.**

	1-D MC Model Estimate (mg/kg) <sup>a</sup>			2-D MC Model Estimate (mg/kg) <sup>b</sup>		
<b>Trophic level 4 species</b>						
Fish Species	5 <sup>th</sup> -%tile	50 <sup>th</sup> -%tile	Mean <sup>d</sup>	95 <sup>th</sup> -%tile	lower bound <sup>e</sup>	upper bound <sup>f</sup>
Northern pikeminnow	0.12 [0.13] <sup>c</sup>	0.55 [0.57]	0.83 ± 1.00 {0.60 ± 0.31} n = 95	2.50 [1.33]	0.05	5.12
Largemouth bass	0.08 [0.12]	0.39 [0.43]	0.62 ± 0.80 {0.52 ± 0.32} n = 192	1.90 [1.14]	0.03	4.28
Smallmouth bass	0.02 [0.09]	0.18 [0.16]	0.41 ± 0.73 {0.27 ± 0.20} n = 10	1.47 [0.70]	0.01	3.40
Large piscivores	---	---	0.225 <sup>g</sup> (0.161 - 0.315)	---	---	---
<b>Trophic level 3 species</b>						
Rainbow trout	0.02 [0.02]	0.12 [0.16]	0.25 ± 0.418 {0.22 ± 0.25} n = 36	0.86 [0.60]	0.02	1.54
Cutthroat trout	0.01 [0.02]	0.07 [0.06]	0.13 ± 0.22 {0.13 ± 0.14} n = 25	0.44 [0.46]	0.01	0.59
Carp	0.04 [0.10]	0.19 [0.22]	0.34 ± 0.57 {0.24 ± 0.17} n = 64	1.06 [0.50]	0.02	1.33
Largescale sucker	0.03 [0.05]	0.16 [0.18]	0.29 ± 0.46 {0.22 ± 0.15} n = 135	0.89 [0.62]	0.02	1.61
Bluegill	0.03 [0.01]	0.15 [0.25]	0.23 ± 0.27 {0.35 ± 0.31} n = 27	0.70 [1.00]	0.02	0.82
Large invertivores	---	---	0.042 <sup>g</sup> (0.035 - 0.049)	---	---	---

- a) Calculated using Equation {11} and 1-D MC analysis (stochastic variability and uncertainty combined).
- b) 90 percent confidence range of tissue concentration estimates, calculated using Equation {11}, and 2-D MC analysis, with eleven (adult body length (8), dissolved total mercury concentration, dissolved MeHg concentration, and water temperature) treated as uncertain.
- c) Values in [brackets] are from empirical distribution functions formed from measured tissue concentration data for adult fish.
- d) Model estimated arithmetic mean tissue concentration  $\pm$  one standard deviation. Values in {braces} are the mean and standard deviation of the measured tissue concentration data for all sampling locations within the WRB combined ( $n$  = number of tissue samples for a given species).
- e) Lower bound - 5<sup>th</sup> percentile of the distribution of 5<sup>th</sup> percentile tissue concentration estimates.
- f) Upper bound - 95<sup>th</sup> percentile of the distribution of 95<sup>th</sup> percentile tissue concentration estimates.
- g) Least-squares mean (with 95% confidence interval) mercury concentration in large (>120 mm) fish sampled in the Western aggregate ecoregion of Oregon, as reported in [37].

**Table 8. Comparison of model estimated biomagnification factors for eight species of Willamette River fish and U.S. EPA national bioaccumulation factors for mercury.**

Fish Species	1-D MC Model Estimates (L/kg) <sup>a</sup>			
	5 <sup>th</sup> -%tile	50 <sup>th</sup> -%tile	Mean	95 <sup>th</sup> -%tile
<b>TROPHIC LEVEL 4 SPECIES</b>				
Northern pikeminnow	$1.56 \times 10^6$	$7.03 \times 10^6$	$1.04 \times 10^7$	$2.93 \times 10^7$
Largemouth bass	$1.17 \times 10^6$	$4.99 \times 10^6$	$7.81 \times 10^6$	$2.38 \times 10^7$
Smallmouth bass	$3.09 \times 10^5$	$2.29 \times 10^6$	$5.09 \times 10^6$	$1.77 \times 10^7$
U.S. EPA direct estimate bioaccumulation factor for trophic level 4 species [4, Tables D-8 and -19]	$3.26 \times 10^5$	$6.81 \times 10^6$	$1.11 \times 10^7$	$1.42 \times 10^7$
<b>TROPHIC LEVEL 3 SPECIES</b>				
Rainbow trout	$2.78 \times 10^5$	$1.59 \times 10^6$	$3.17 \times 10^6$	$1.03 \times 10^7$
Carp	$4.77 \times 10^5$	$2.38 \times 10^6$	$4.23 \times 10^6$	$1.35 \times 10^7$
Largescale sucker	$4.45 \times 10^5$	$2.06 \times 10^6$	$3.58 \times 10^6$	$1.09 \times 10^7$
Bluegill	$3.65 \times 10^5$	$1.91 \times 10^6$	$2.84 \times 10^6$	$8.32 \times 10^6$
Cutthroat trout	$1.92 \times 10^5$	$9.51 \times 10^5$	$1.67 \times 10^6$	$5.46 \times 10^6$
U.S. EPA direct estimate bioaccumulation factor for trophic level 3 species [4, Tables D-7 and -18]	$4.61 \times 10^5$	$1.58 \times 10^6$	$2.09 \times 10^6$	$5.41 \times 10^6$

a) Biomagnification factor estimates based on a 1-D MC (stochastic variability and uncertainty combined) analysis.

**Table 9. Potential species-specific surface water target levels for total mercury in the Willamette River Basin, based on a post-calibration model.**

Fish Species	1-D MC Model Estimate (ng/L) <sup>a</sup>				2-D MC Model Estimate (ng/L) <sup>b</sup>	
	5 <sup>th</sup> -%tile <sup>c</sup>	50 <sup>th</sup> -%tile <sup>d</sup>	Mean	95 <sup>th</sup> -%tile <sup>e</sup>	lower bound <sup>f</sup>	upper bound <sup>g</sup>
Northern pikeminnow	2.62	0.39	0.77	0.05	9.20	0.03
Largemouth bass	3.55	0.57	1.19	0.08	6.78	0.04
Smallmouth bass	13.50	1.19	3.16	0.12	19.64	0.08
Rainbow trout	4.31	1.67	4.31	0.22	60.11	0.11
Bluegill	3.40	1.44	3.40	0.22	18.75	0.10
Largescale sucker	9.70	1.37	3.24	0.18	11.60	0.08
Carp	7.43	1.10	2.32	0.19	17.46	0.08
Cutthroat trout	22.20	3.14	6.59	0.31	43.83	0.13

- a) Calculated using Equation {12} and 1-D MC methods, with biomagnification factor and  $\Omega$  as distributions.
- b) Calculated using Equation {12} and 2-D MC methods, with biomagnification factor and  $\Omega$  as distributions and ten variables (adult body lengths (8), water temperature, and  $\Omega$ ) treated as uncertain.
- c) Total mercury concentration that would achieve the U.S. EPA tissue criterion in 5 percent of individuals.
- d) Total mercury concentration that would achieve the U.S. EPA tissue criterion in 50 percent of individuals.
- e) Total mercury concentration that would achieve the U.S. EPA tissue criterion in 95 percent of individuals.
- f) Lower bound - total mercury concentration that would achieve the U.S. EPA tissue criterion in 5 percent of the population.
- g) Upper bound - total mercury concentration that would achieve the U.S. EPA tissue criterion in 95 percent of the population.

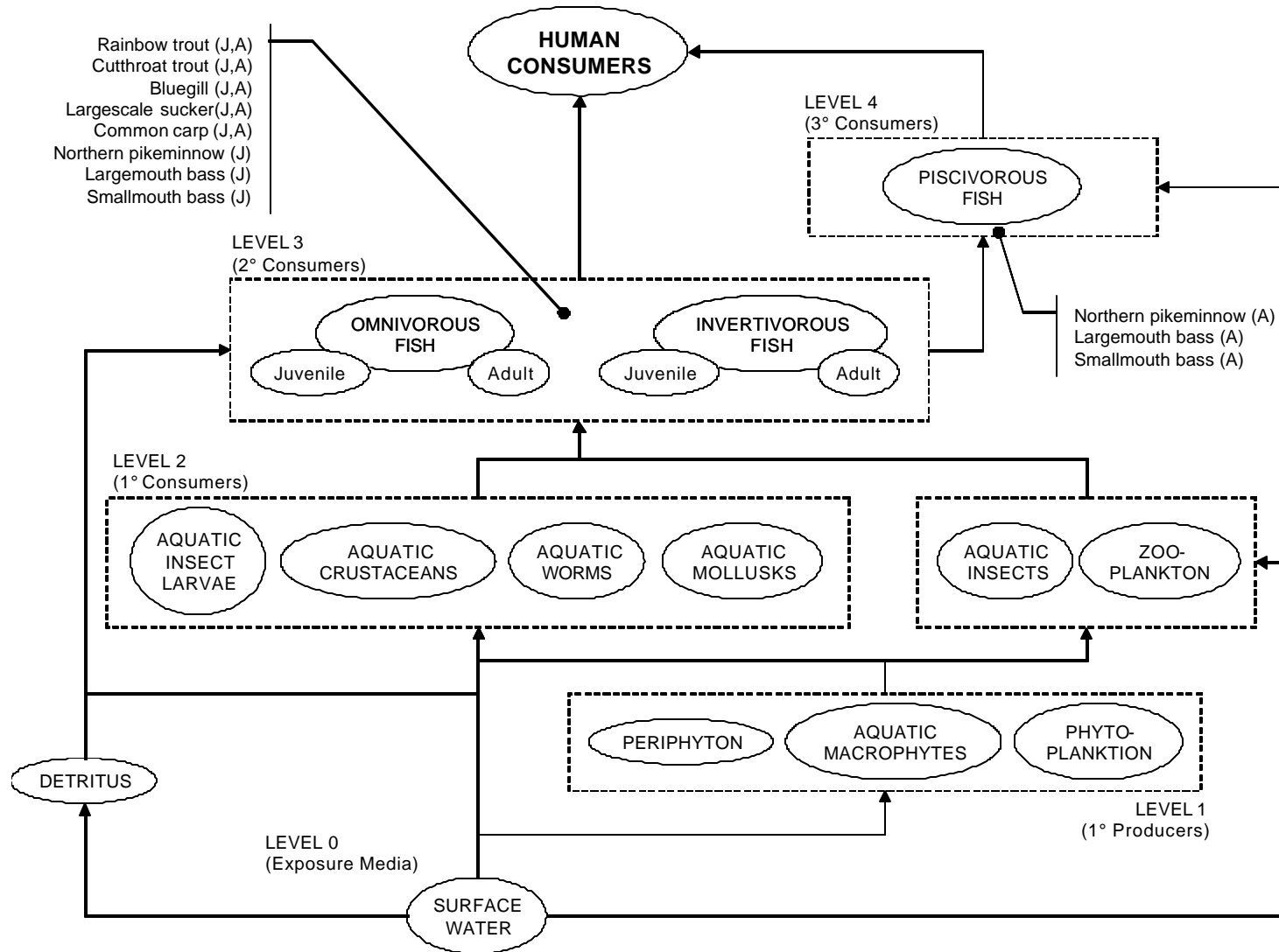


Figure 1. Overview of representative aquatic species and feeding relationships included in the Willamette River Basin food web model. (J = juvenile fish; A = adult fish),

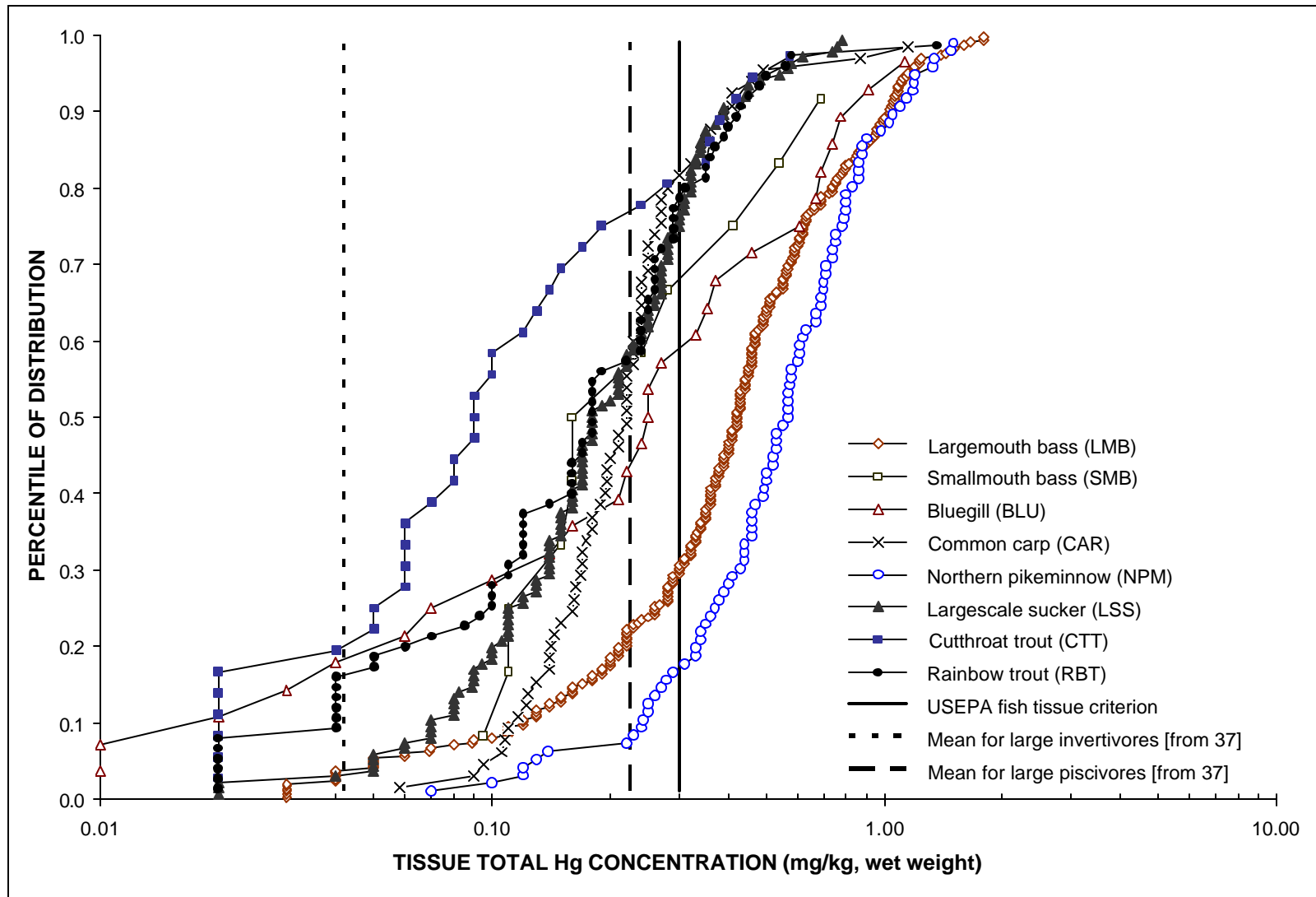


Figure 2. Empirical density functions of tissue total mercury concentrations in eight species of Willamette River Basin fish. U.S. EPA human health fish tissue criterion (0.3 mg/kg) and results of recent survey of mercury in Oregon fish [37] provided for comparison.

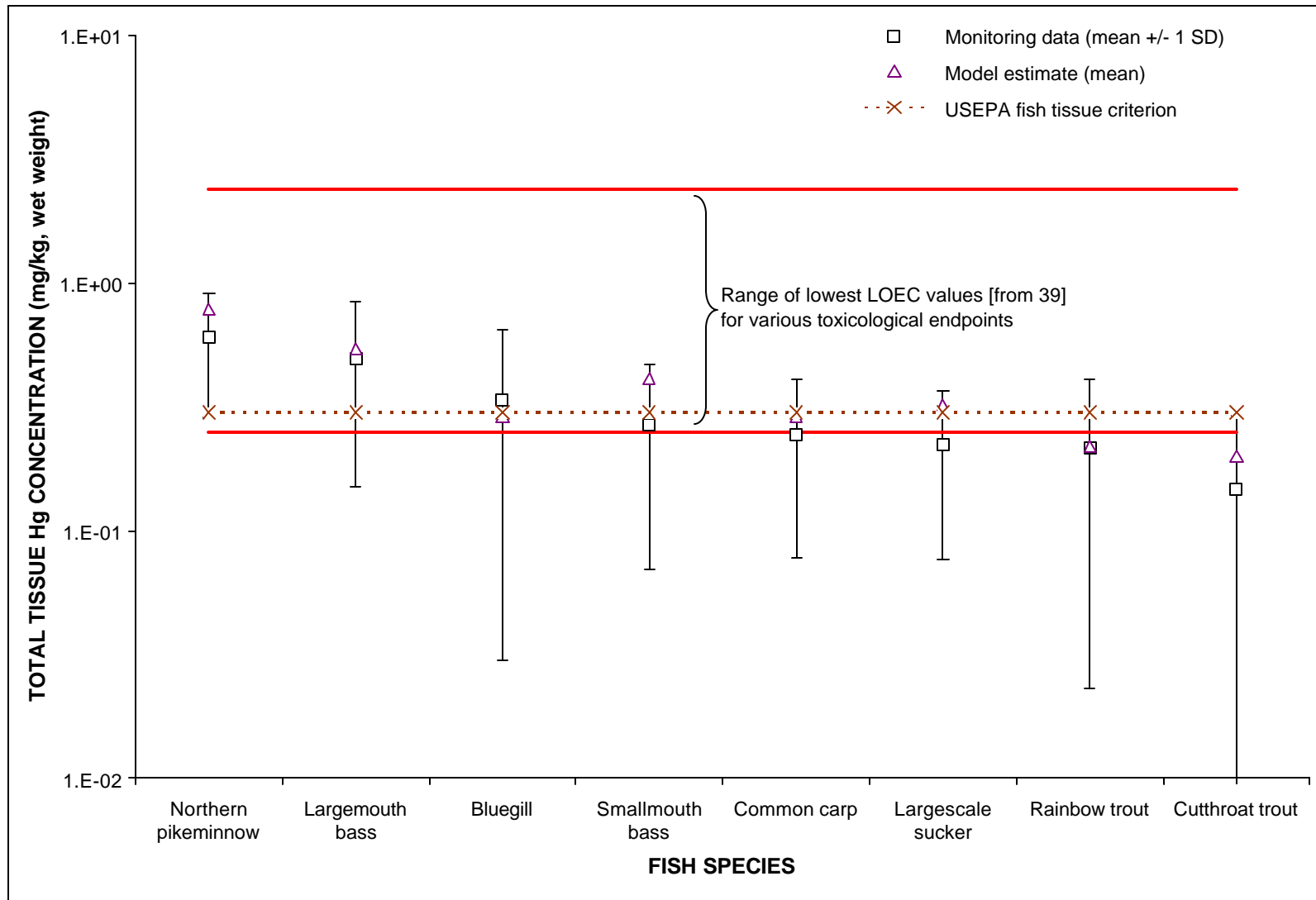


Figure 3. Modeled and measured mean total mercury concentrations in eight species of Willamette River Basin fish in comparison to lowest-observed-effect [tissue] concentrations reported for similar freshwater species [39].

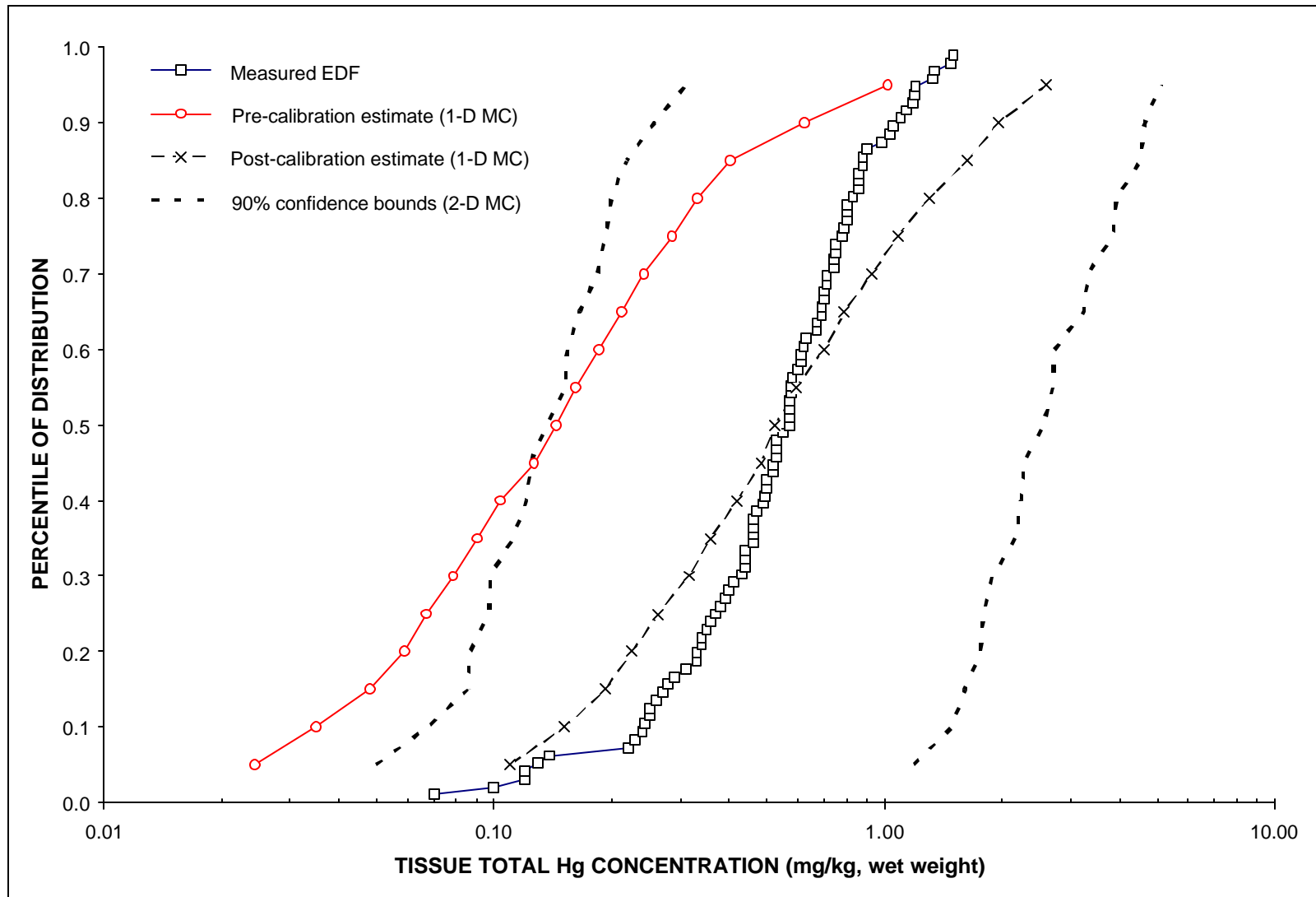


Figure 4. Comparison of (a) measured and modeled estimated (1-D MC) tissue total mercury concentrations, (b) pre- and post calibration model estimates, and (c) the 90 percent confidence bounds (2-D MC) on the post-calibration model estimate for northern pikeminnow (NPM).

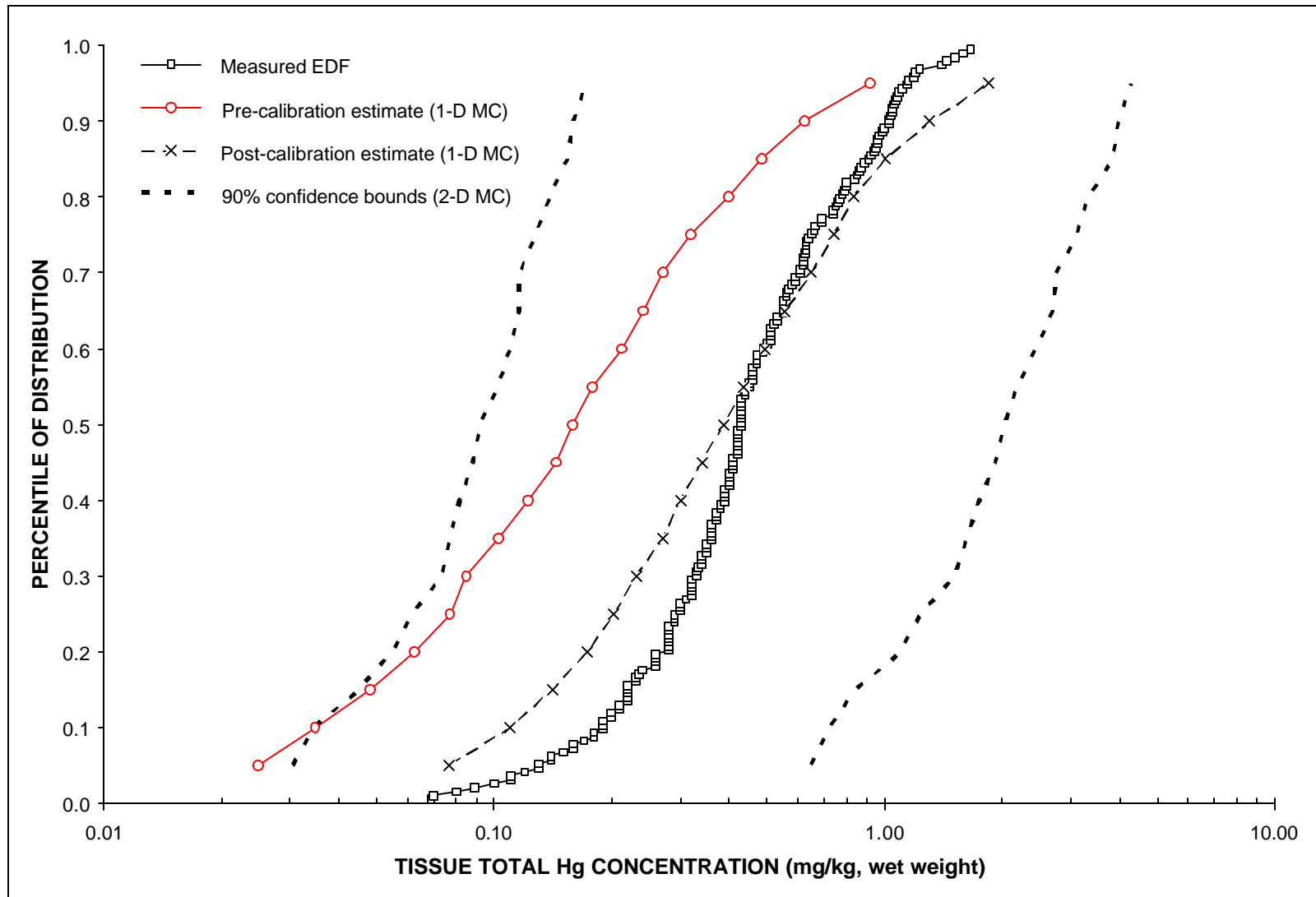


Figure 5. Comparison of (a) measured and modeled estimated (1-D MC) tissue total mercury concentrations, (b) pre- and post calibration model estimates, and (c) the 90 percent confidence bounds (2-D MC) on the post-calibration model estimate for largemouth bass (LMB).

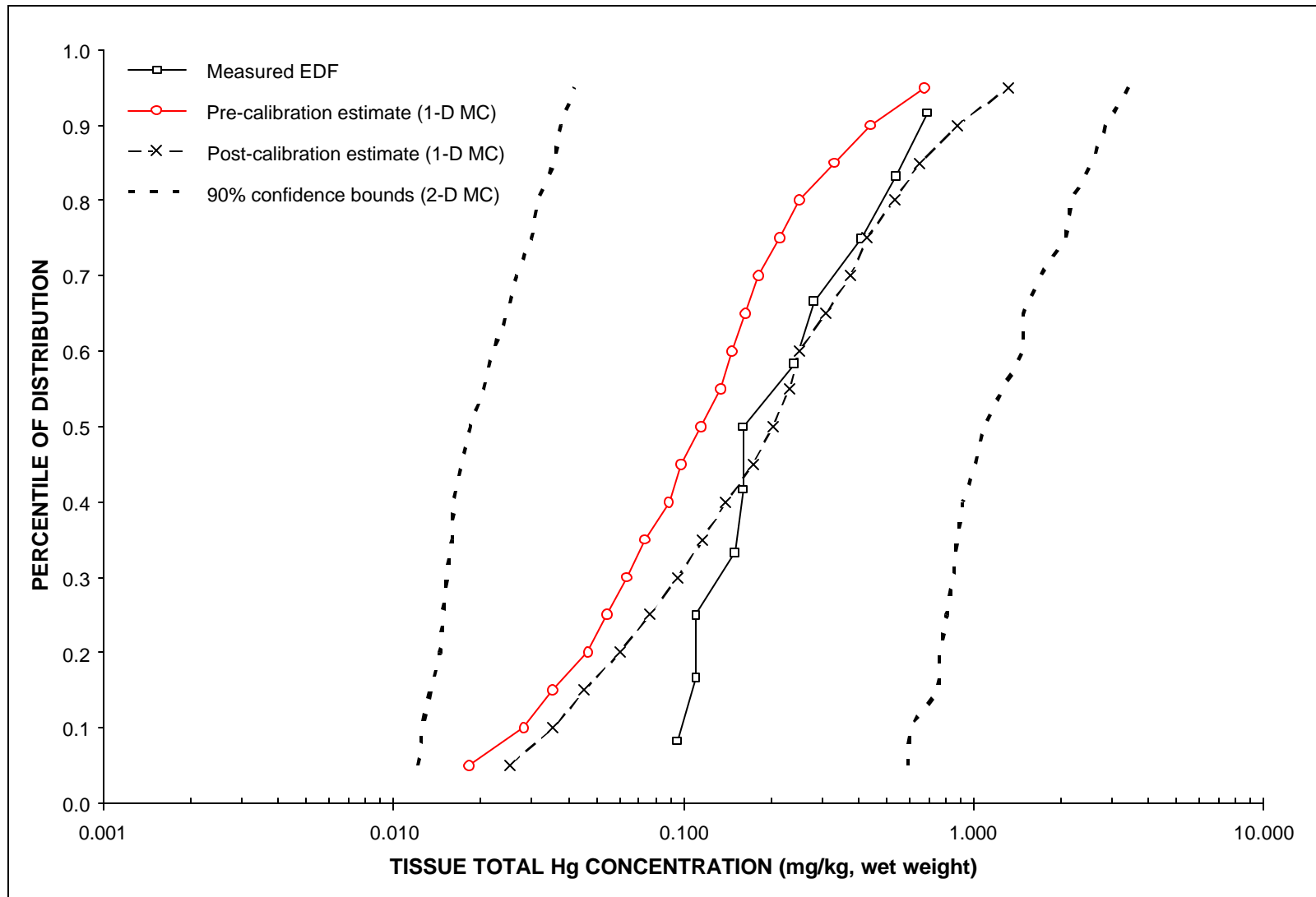


Figure 6. Comparison of (a) measured and modeled estimated (1-D MC) tissue total mercury concentrations, (b) pre- and post calibration model estimates, and (c) the 90 percent confidence bounds (2-D MC) on the post-calibration model estimate for smallmouth bass (SMB).

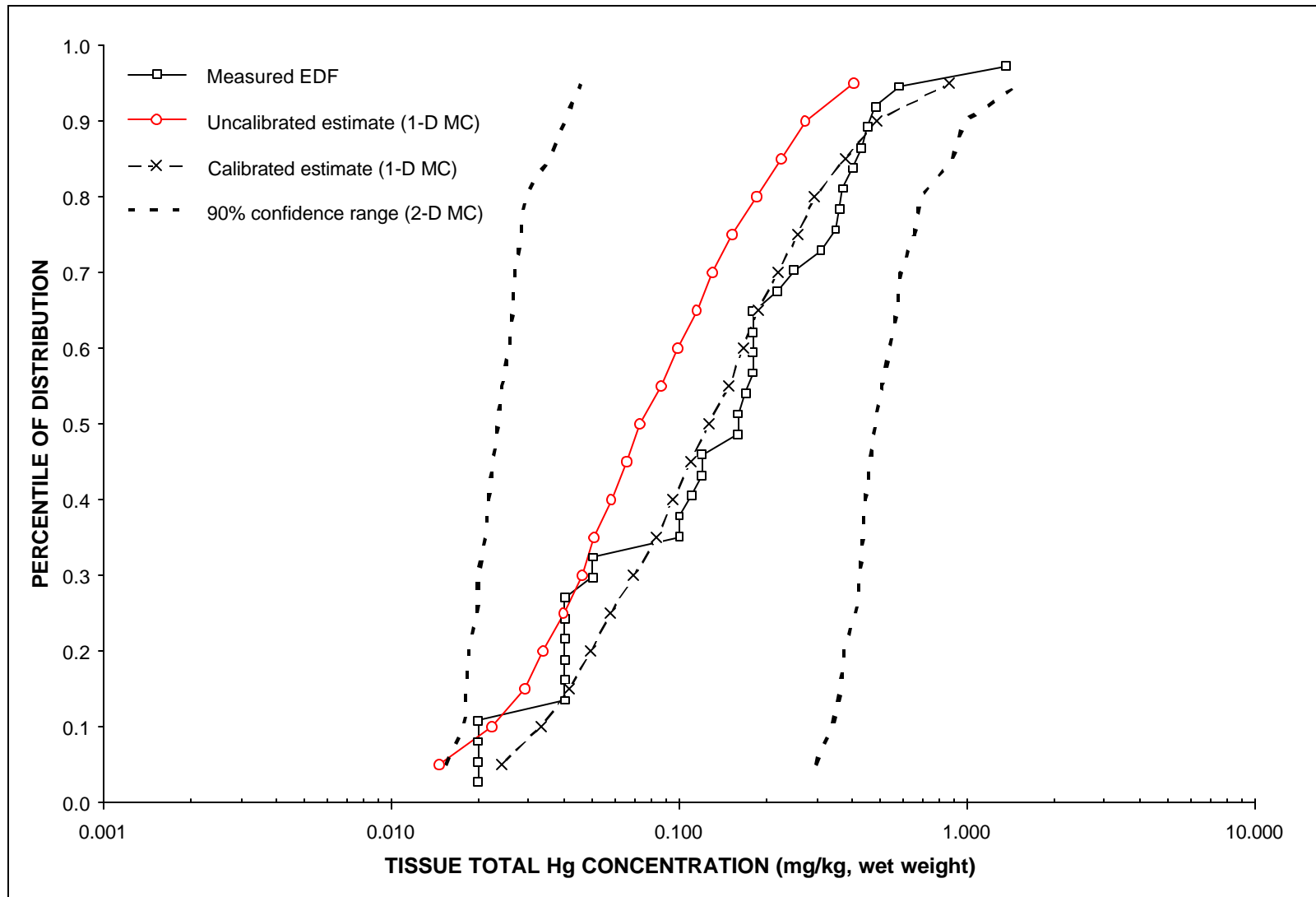


Figure 7. Comparison of (a) measured and modeled estimated (1-D MC) tissue total mercury concentrations, (b) pre- and post calibration model estimates, and (c) the 90 percent confidence bounds (2-D MC) on the post-calibration model estimate for rainbow trout (RBT).

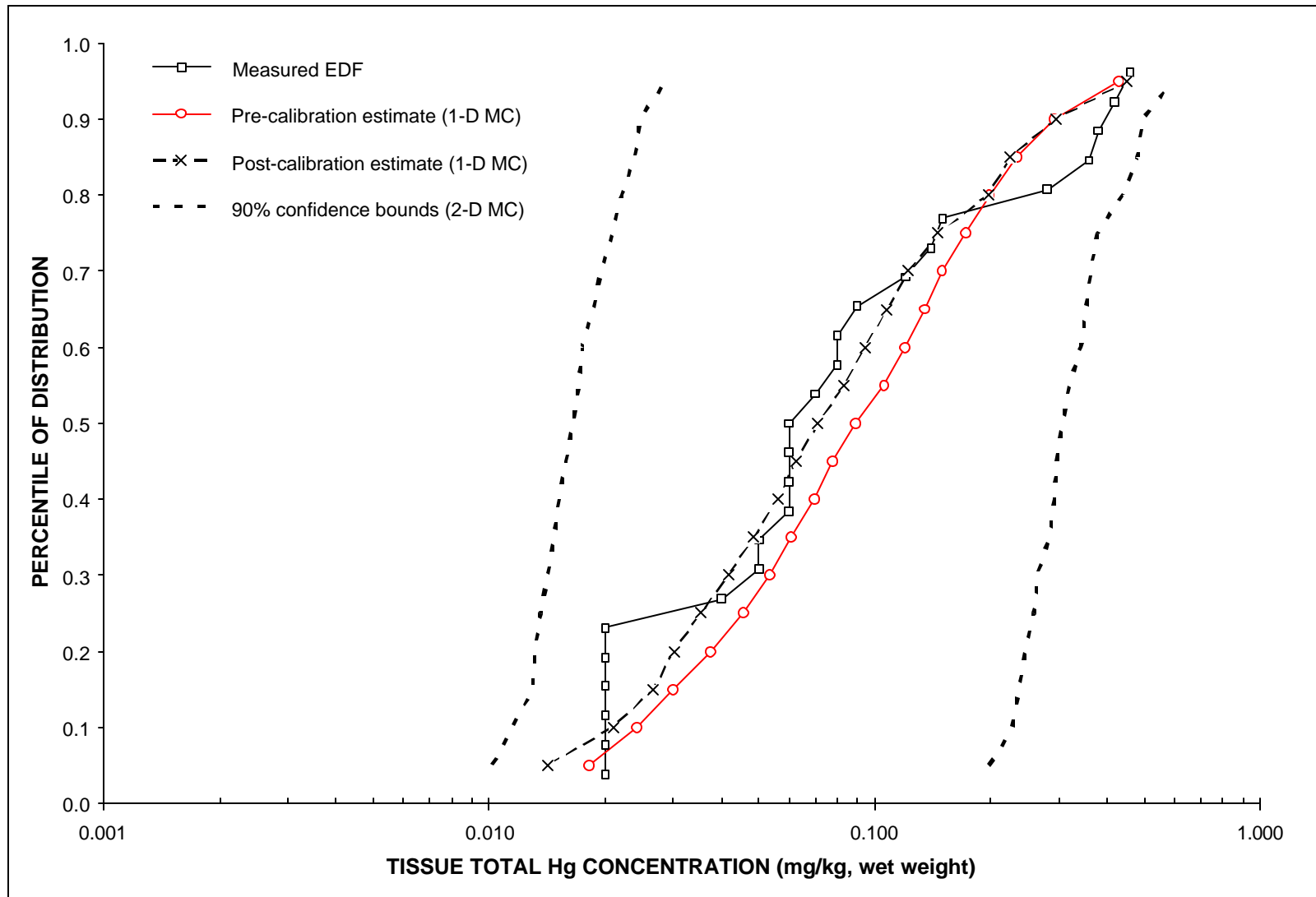


Figure 8. Comparison of (a) measured and modeled estimated (1-D MC) tissue total mercury concentrations, (b) pre- and post calibration model estimates, and (c) the 90 percent confidence bounds (2-D MC) on the post-calibration model estimate for cutthroat trout (CTT).

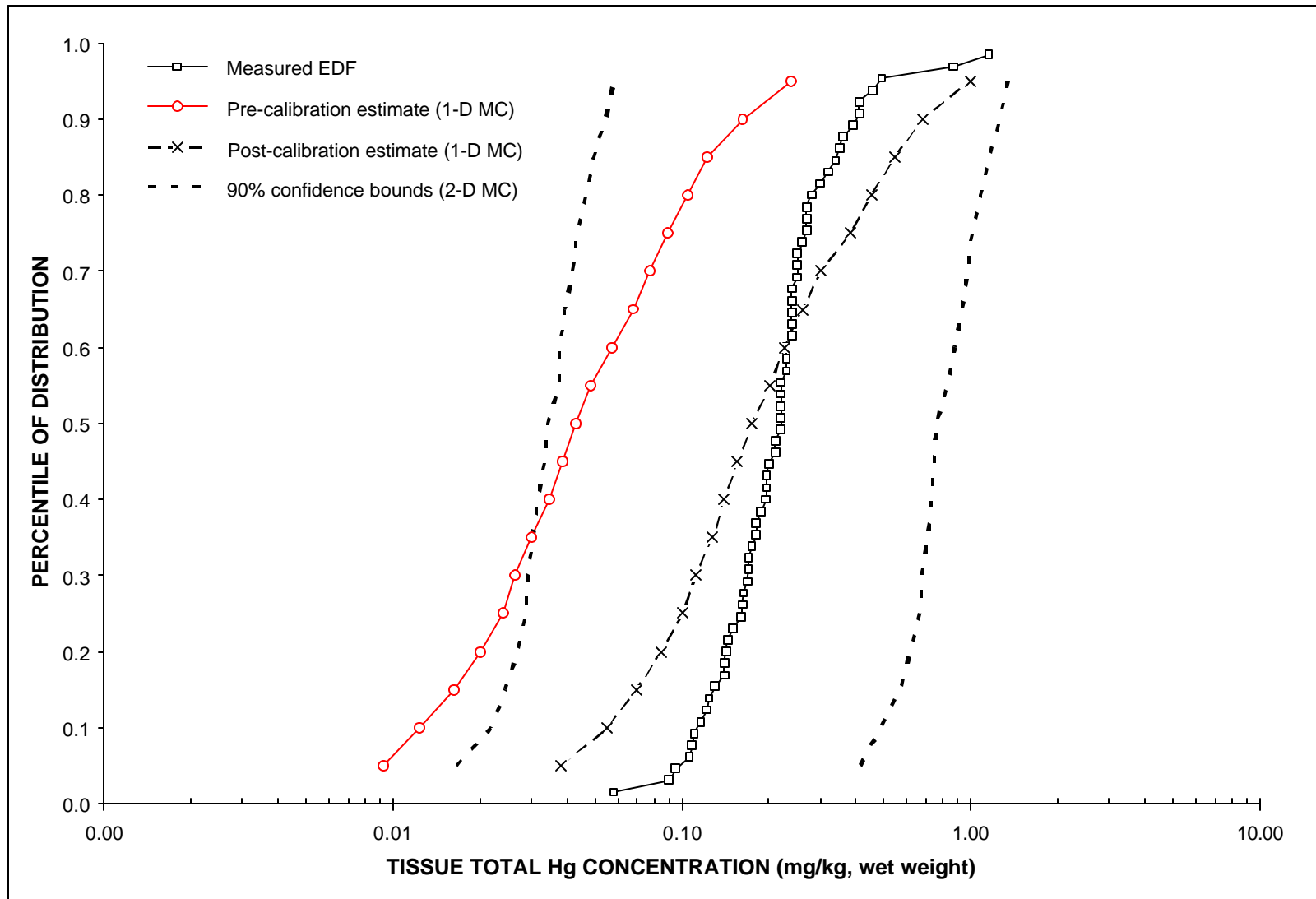


Figure 9. Comparison of (a) measured and modeled estimated (1-D MC) tissue total mercury concentrations, (b) pre- and post calibration model estimates, and (c) the 90 percent confidence bounds (2-D MC) on the post-calibration model estimate for common carp (CAR).

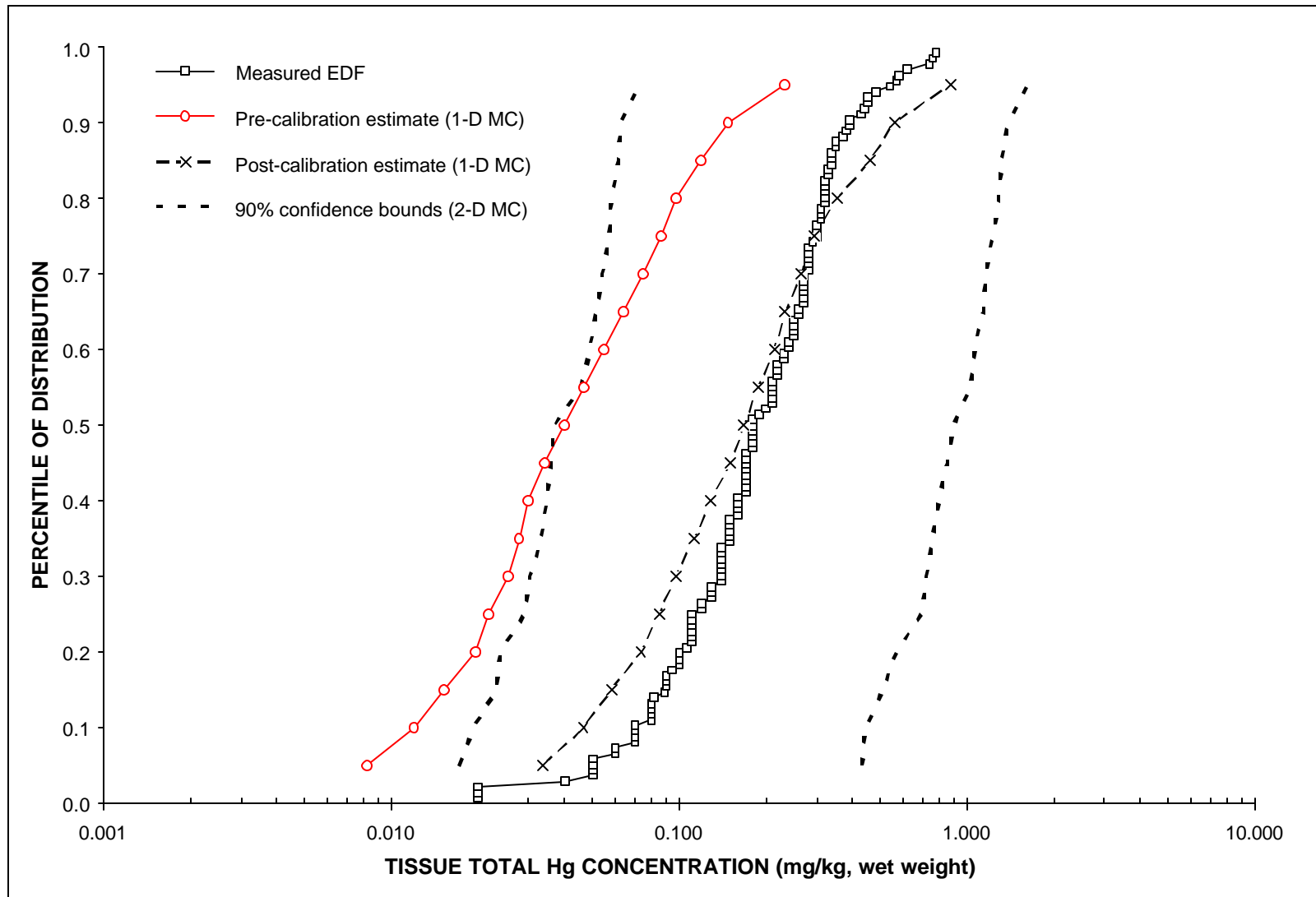


Figure 10. Comparison of (a) measured and modeled estimated (1-D MC) tissue total mercury concentrations, (b) pre- and post calibration model estimates, and (c) the 90 percent confidence bounds (2-D MC) on the post-calibration model estimate for largescale sucker (LSS).

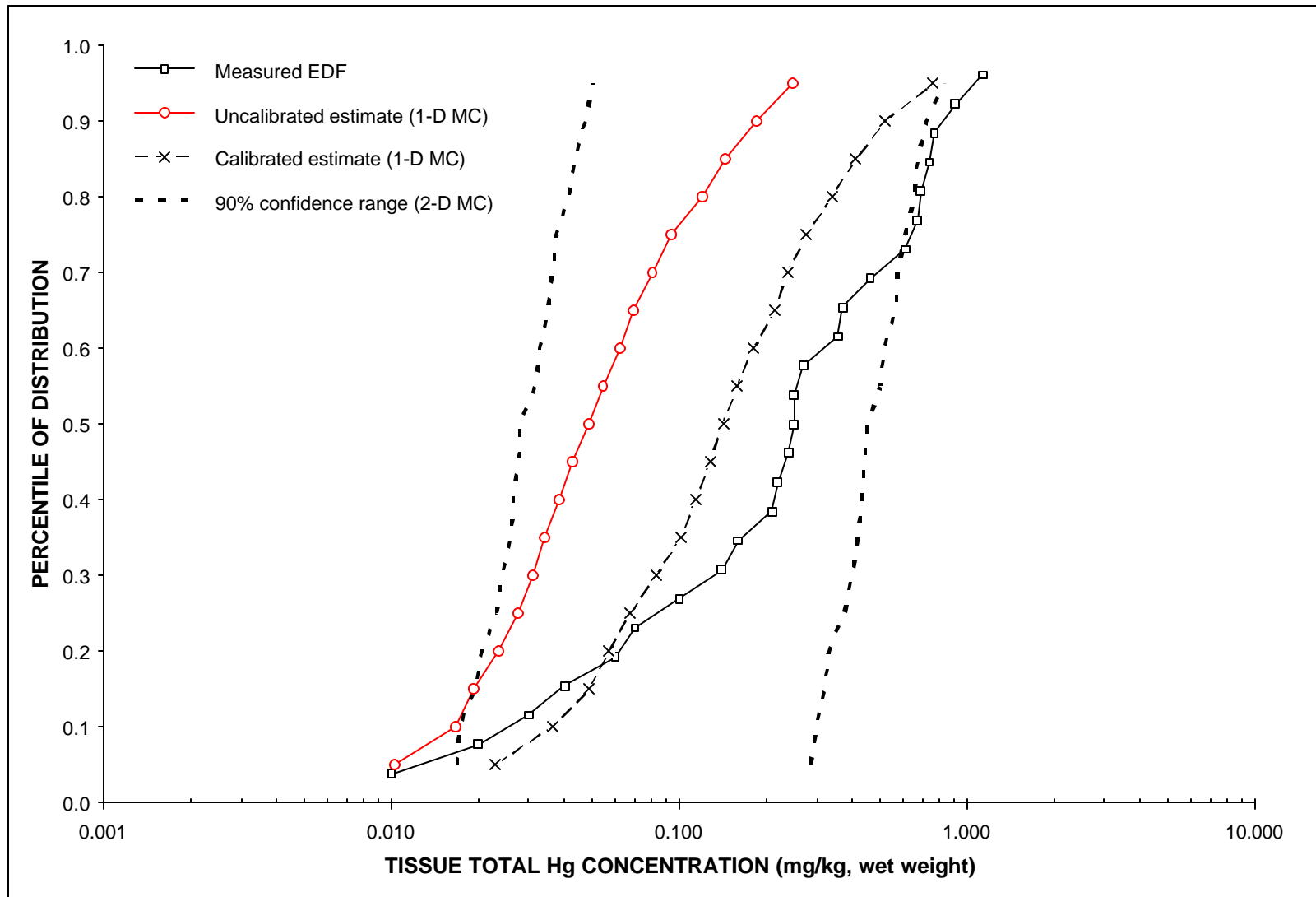


Figure 11. Comparison of (a) measured and modeled estimated (1-D MC) tissue total mercury concentrations, (b) pre- and post calibration model estimates, and (c) the 90 percent confidence bounds (2-D MC) on the post-calibration model estimate forfor bluegill (BLU).

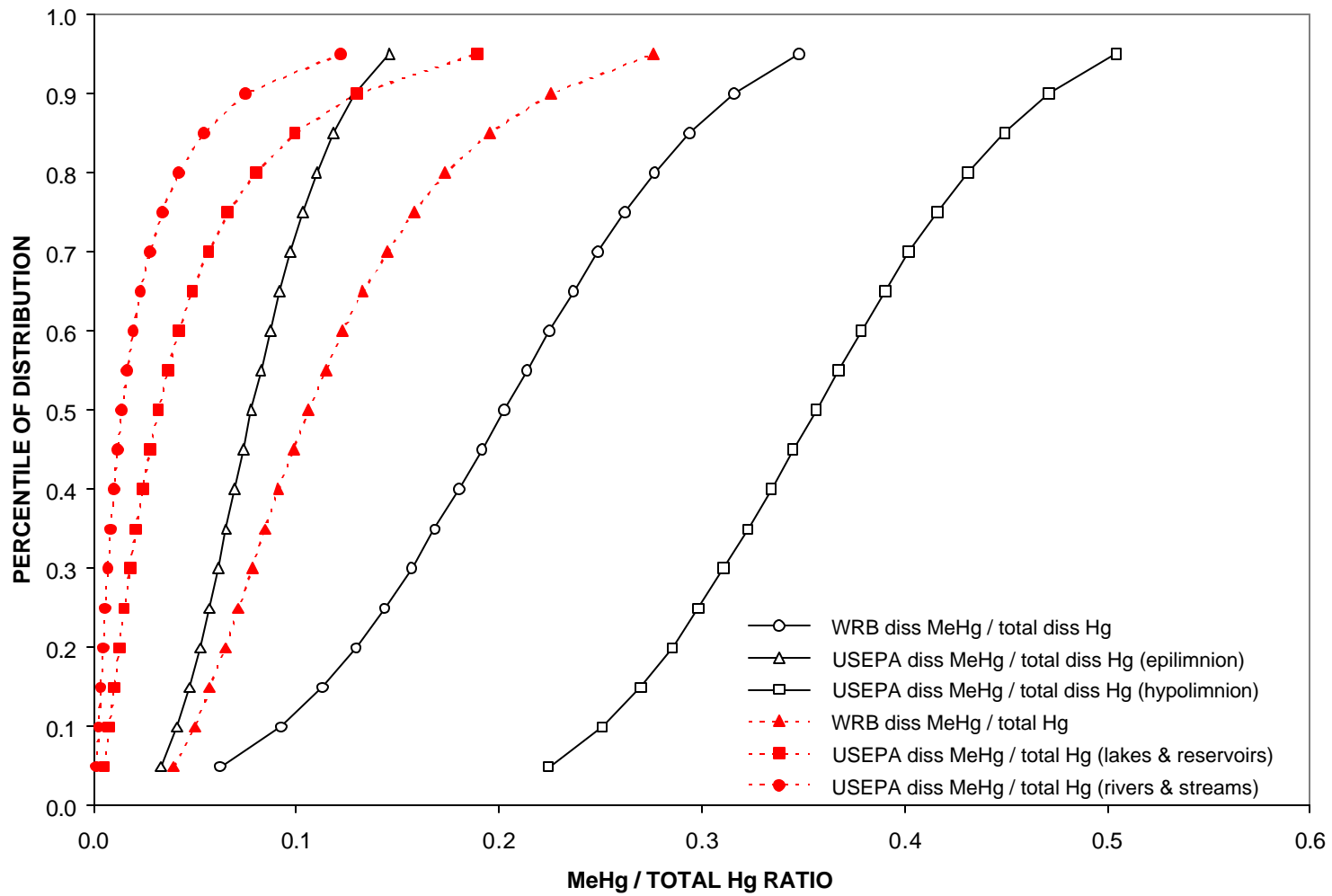


Figure 12. Distributions of calculated and measured Total mercury:MeHg ratios filtered surface water samples from the Willamette River Basin [44] in comparison to ratio distributions estimated by the U.S. EPA on a national basis for the epilimnion and hypolimnion [4].

## APPENDIX A

### Comparison of Modeled and Measured Fish Length Distributions

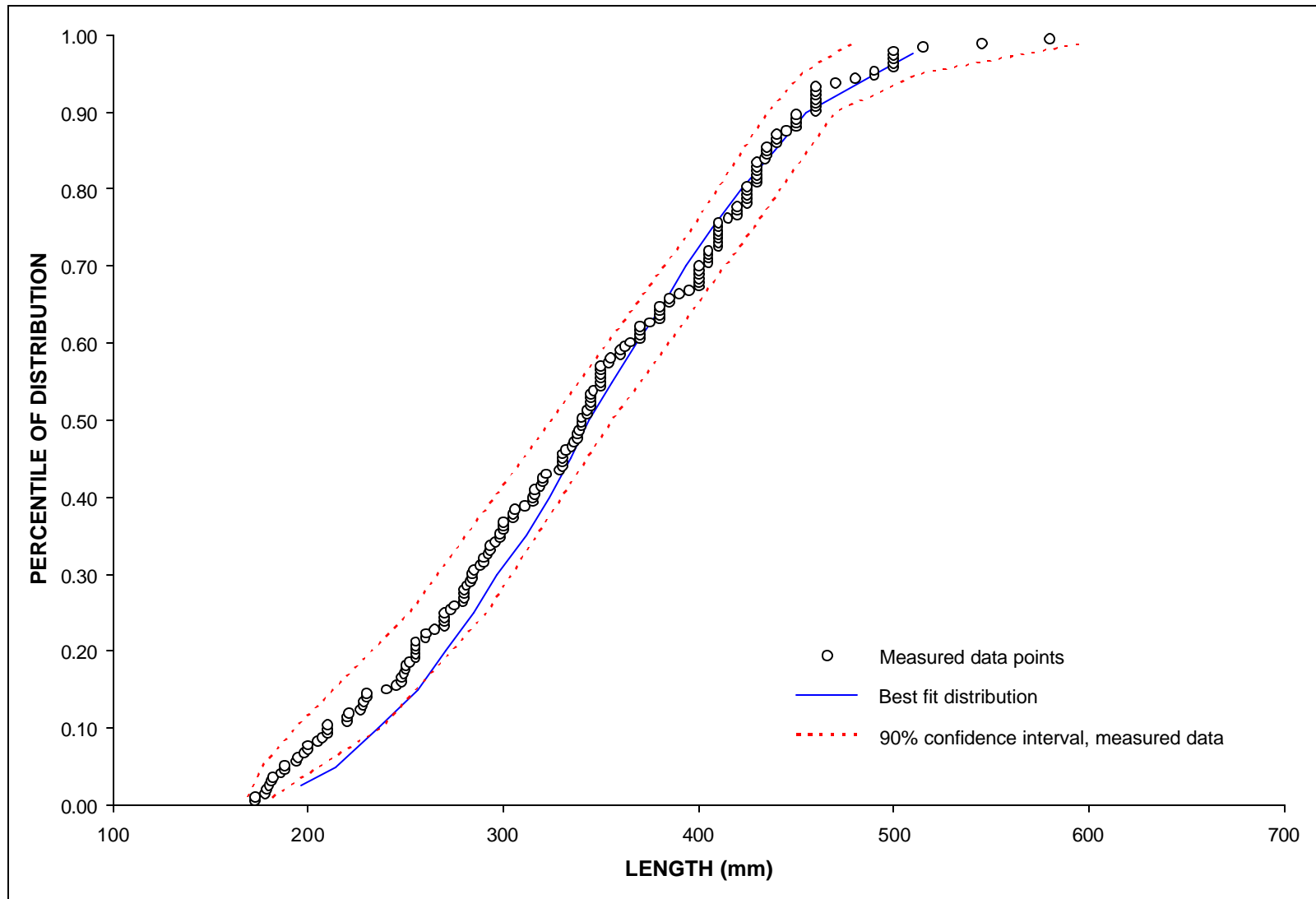


Figure A-1. Comparison of the measured adult fish body length empirical density function (○) (with 90 percent confidence interval (----)) and the best fit cumulative density function ( $\frac{3}{4}$ ) for largemouth bass (LMB).

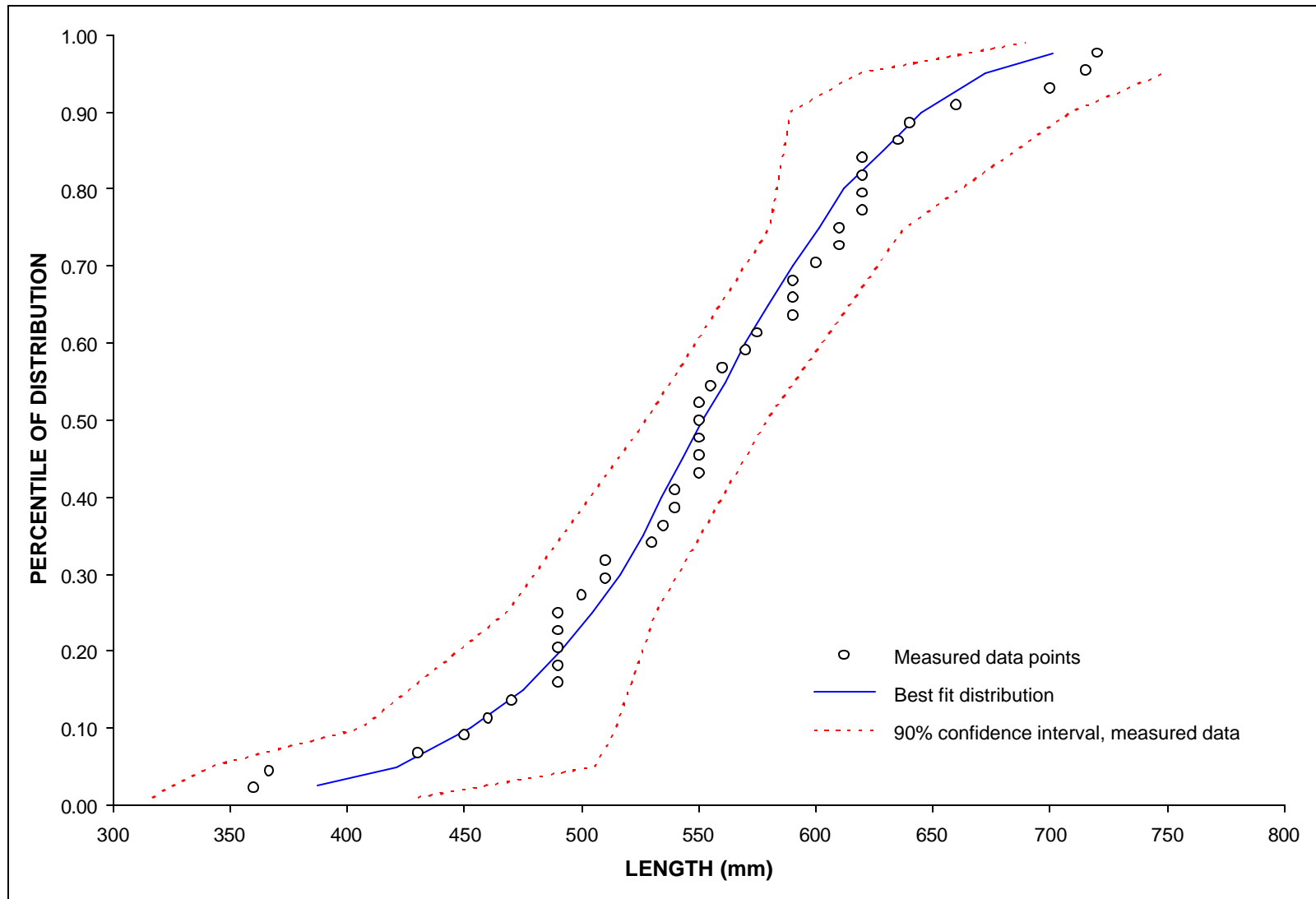


Figure A-2. Comparison of the measured adult fish body length empirical density function (O) (with 90 percent confidence interval (----)) and the best fit cumulative density function ( $\frac{3}{4}$ ) for common carp (CAR).

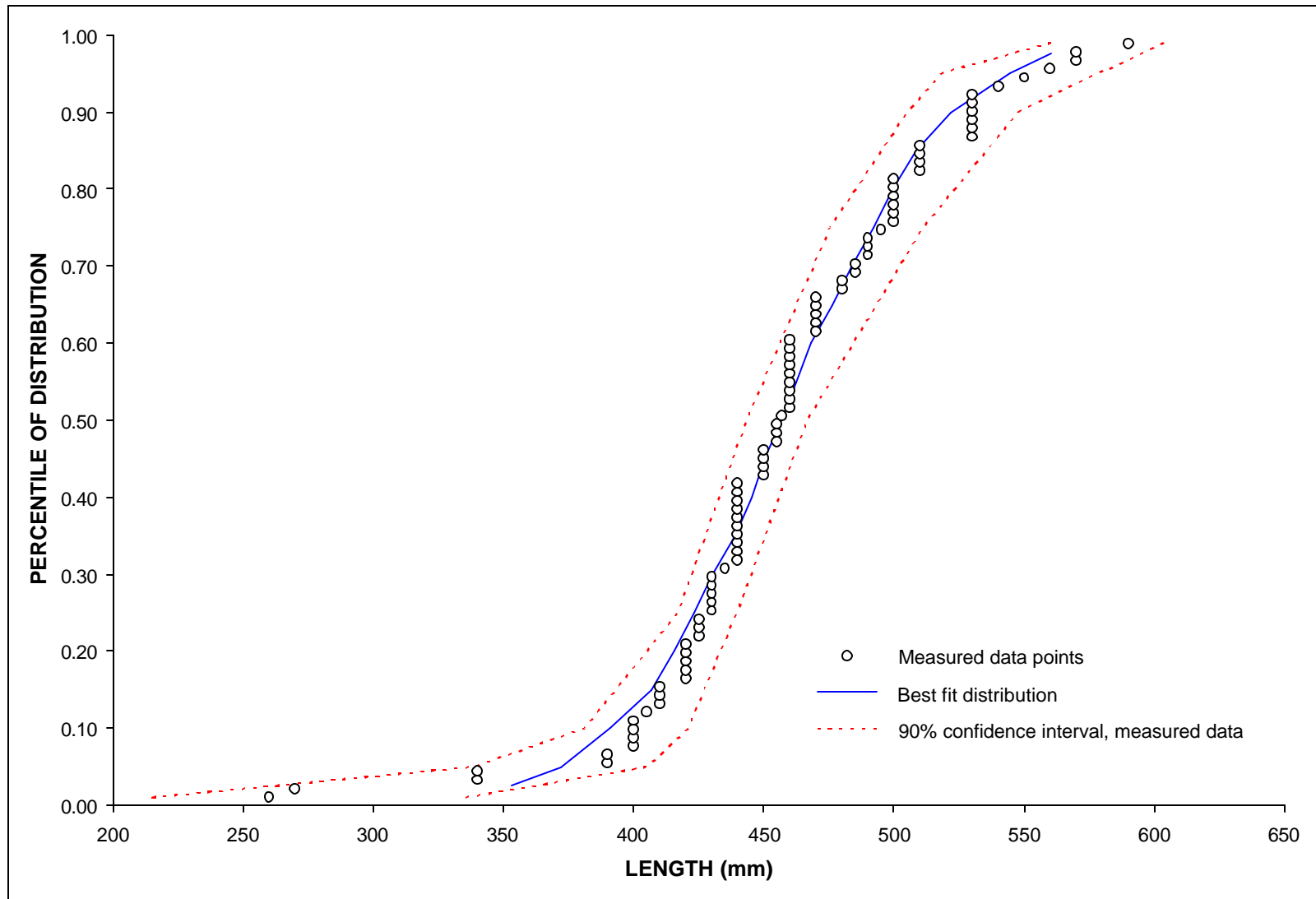


Figure A-3. Comparison of the measured adult fish body length empirical density function (○) (with 90 percent confidence interval (----)) and the best fit cumulative density function ( $\frac{3}{4}$ ) for largescale sucker (LSS).

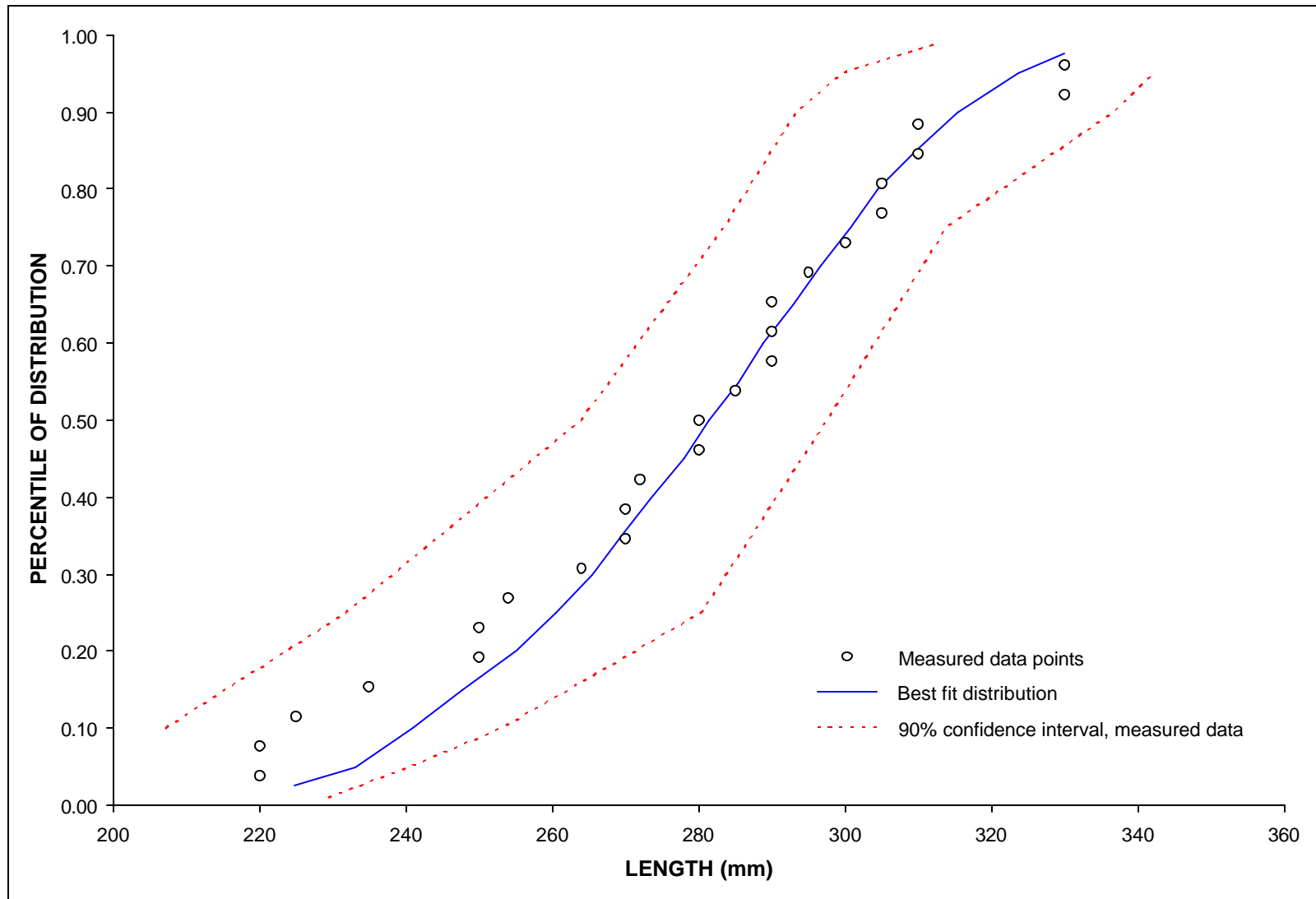


Figure A-4. Comparison of the measured adult fish body length empirical density function (○) (with 90 percent confidence interval (----)) and the best fit cumulative density function ( $\frac{3}{4}$ ) for cutthroat trout (CTT).

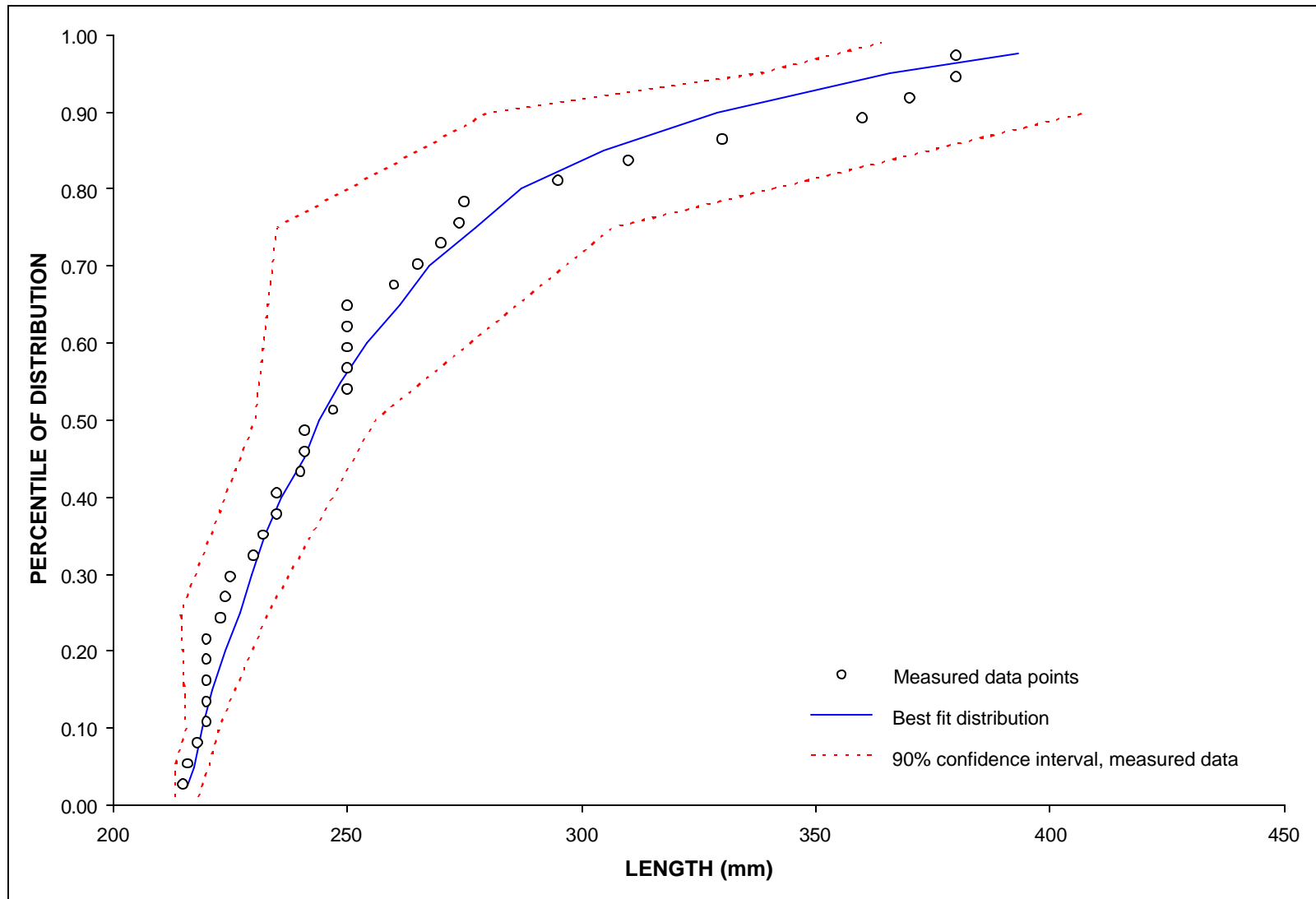


Figure A-5. Comparison of the measured adult fish body length empirical density function (○) (with 90 percent confidence interval (---)) and the best fit cumulative density function ( $\frac{3}{4}$ ) for rainbow trout (RB T).

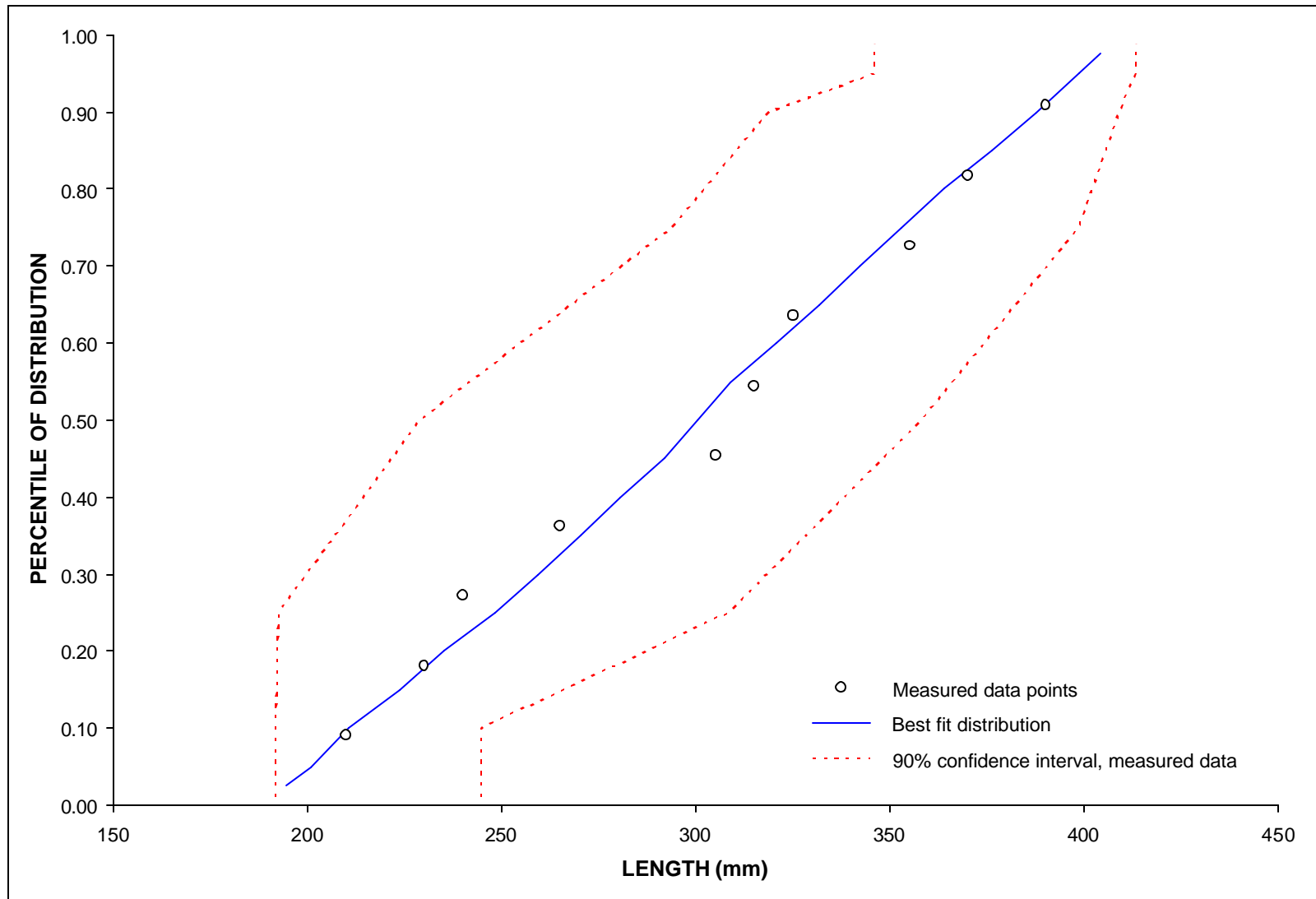


Figure A-6. Comparison of the measured adult fish body length empirical density function (○) (with 90 percent confidence interval (---)) and the best fit cumulative density function ( $\frac{3}{4}$ ) for smallmouth bass (SMB).

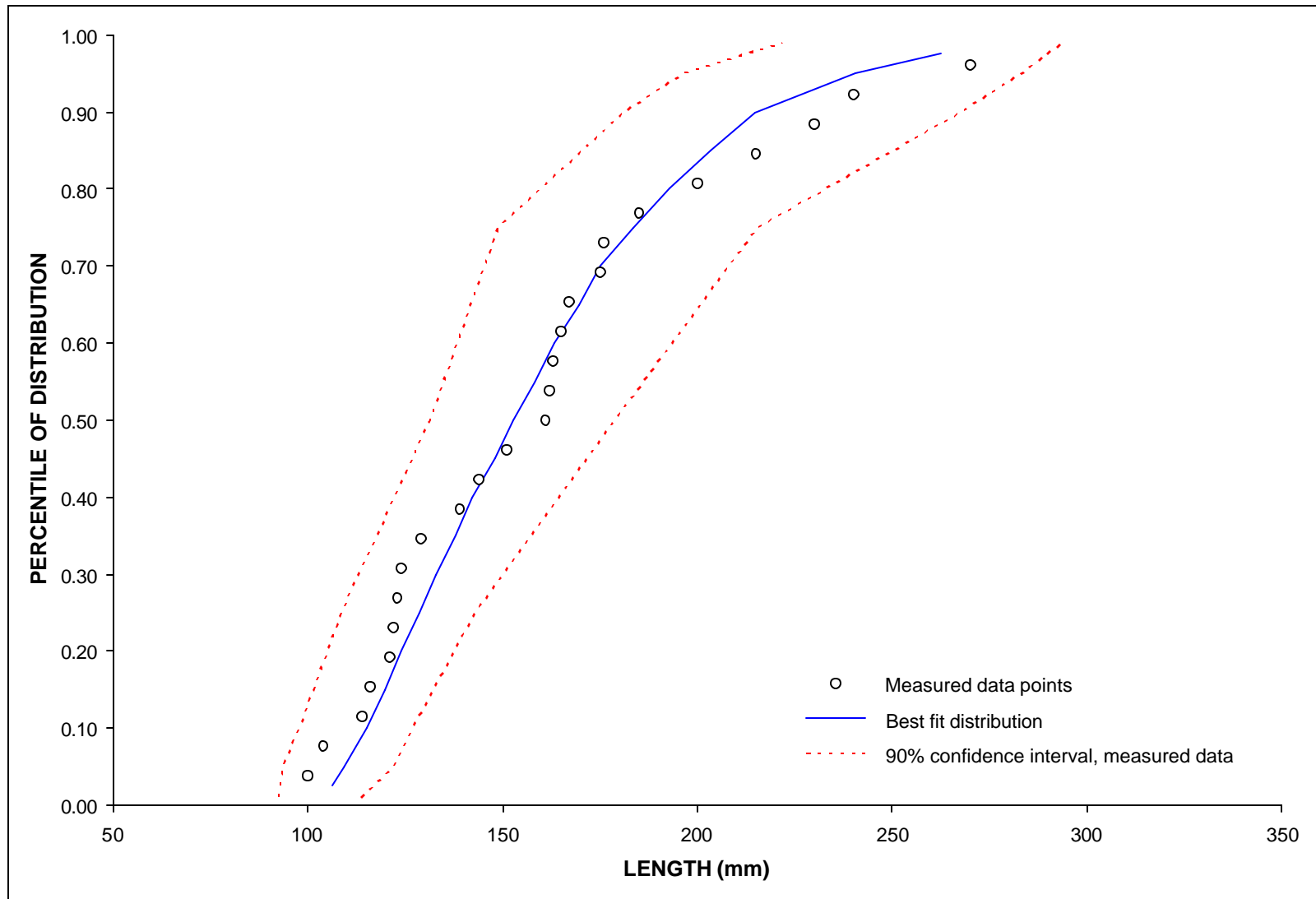


Figure A-7. Comparison of the measured adult fish body length empirical density function (○) (with 90 percent confidence interval (----)) and the best fit cumulative density function ( $\frac{3}{4}$ ) for bluegill (BLU).

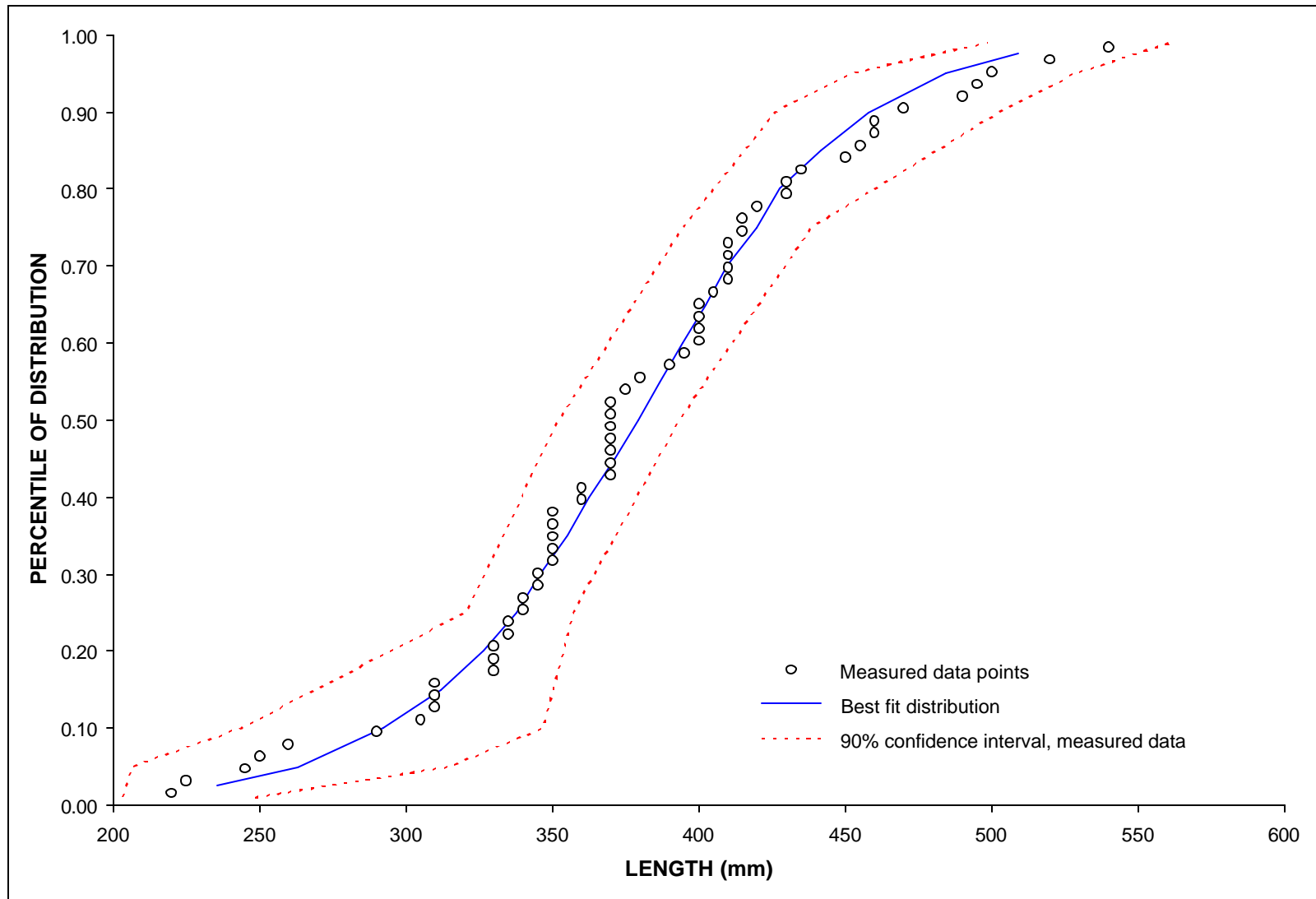


Figure A-8. Comparison of the measured adult fish body length empirical density function (○) (with 90 percent confidence interval (----)) and the best fit cumulative density function ( $\frac{3}{4}$ ) for northern pikeminnow (NPM).

## APPENDIX B

# Comparison of Modeled and Measured Fish Length-Weight Relationships

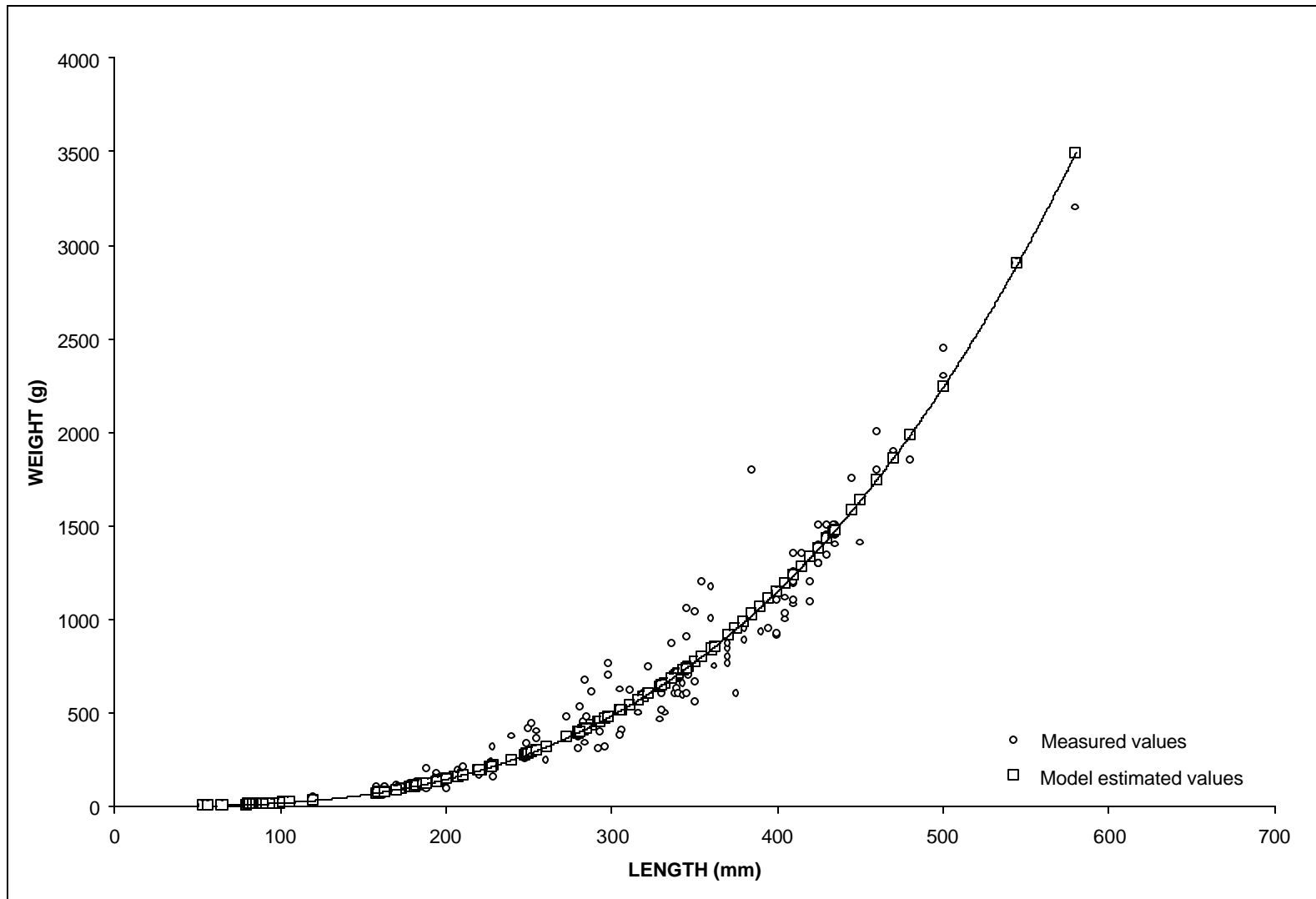


Figure B-1. Comparison of measured (○) and modeled (◻) length-weight relationships for largemouth bass (LMB) in the Willamette River Basin. Spearman rank correlation ( $r_s$ ) = 0.979,  $p < 0.001$ .

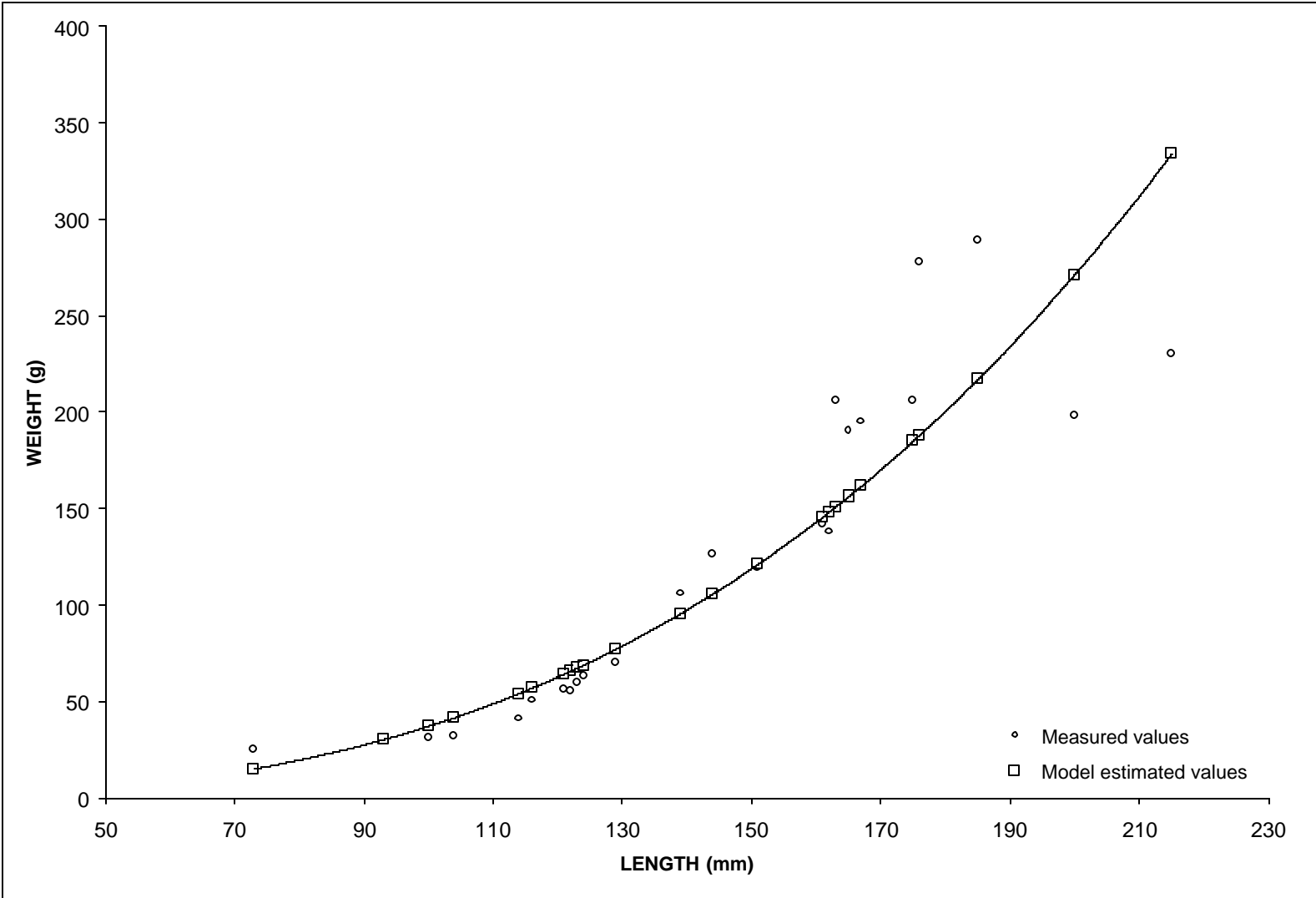


Figure B-2. Comparison of measured (○) and modeled (◻) length-weight relationships for bluegill (BLU) in the Willamette River Basin. Spearman rank correlation ( $r_s$ ) = 0.736,  $p < 0.001$

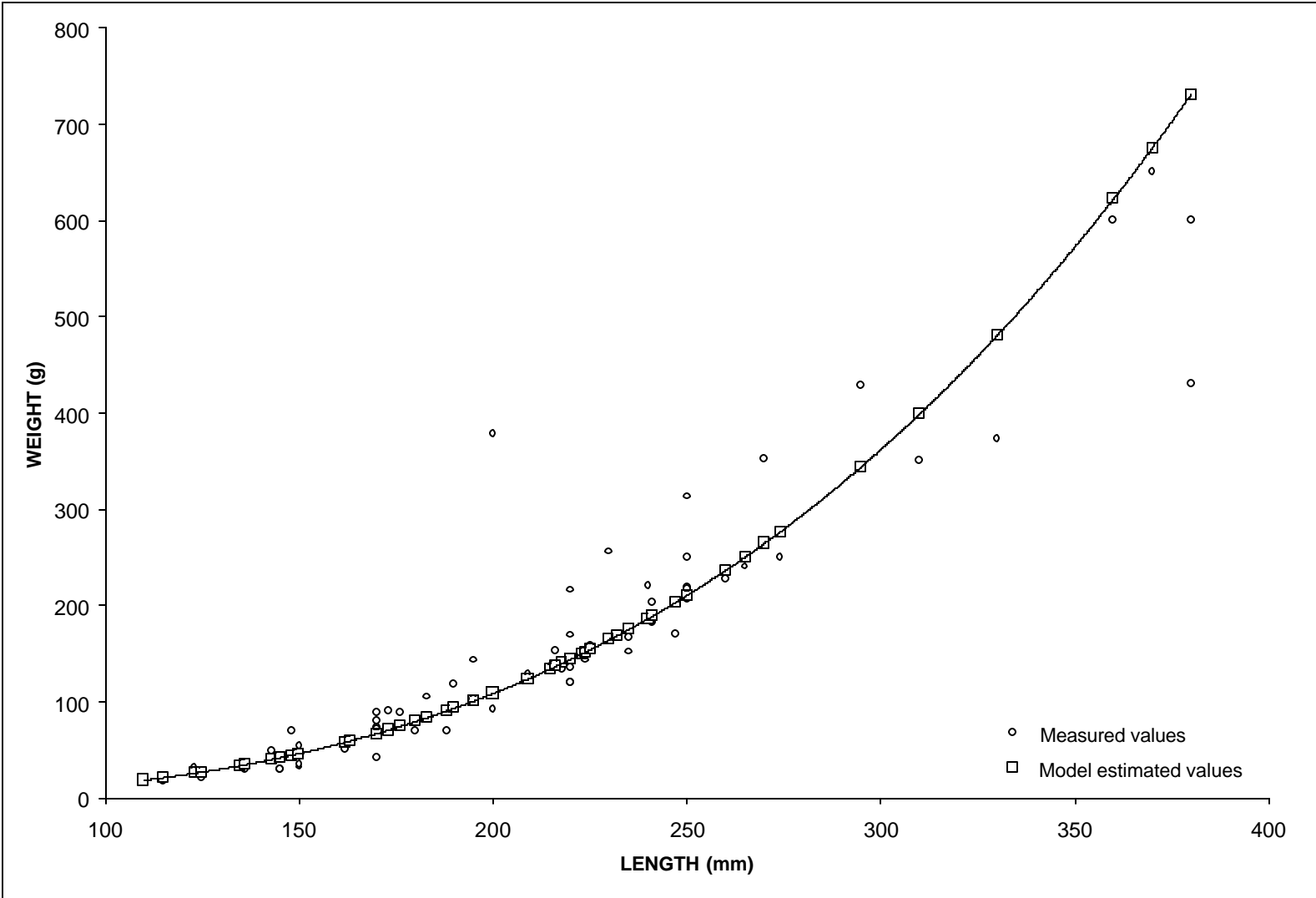


Figure B-3. Comparison of measured (O) and modeled (E) length-weight relationships for rainbow trout (RBT) in the Willamette River Basin. Spearman rank correlation ( $r_s$ ) = 0.964,  $p < 0.001$ .

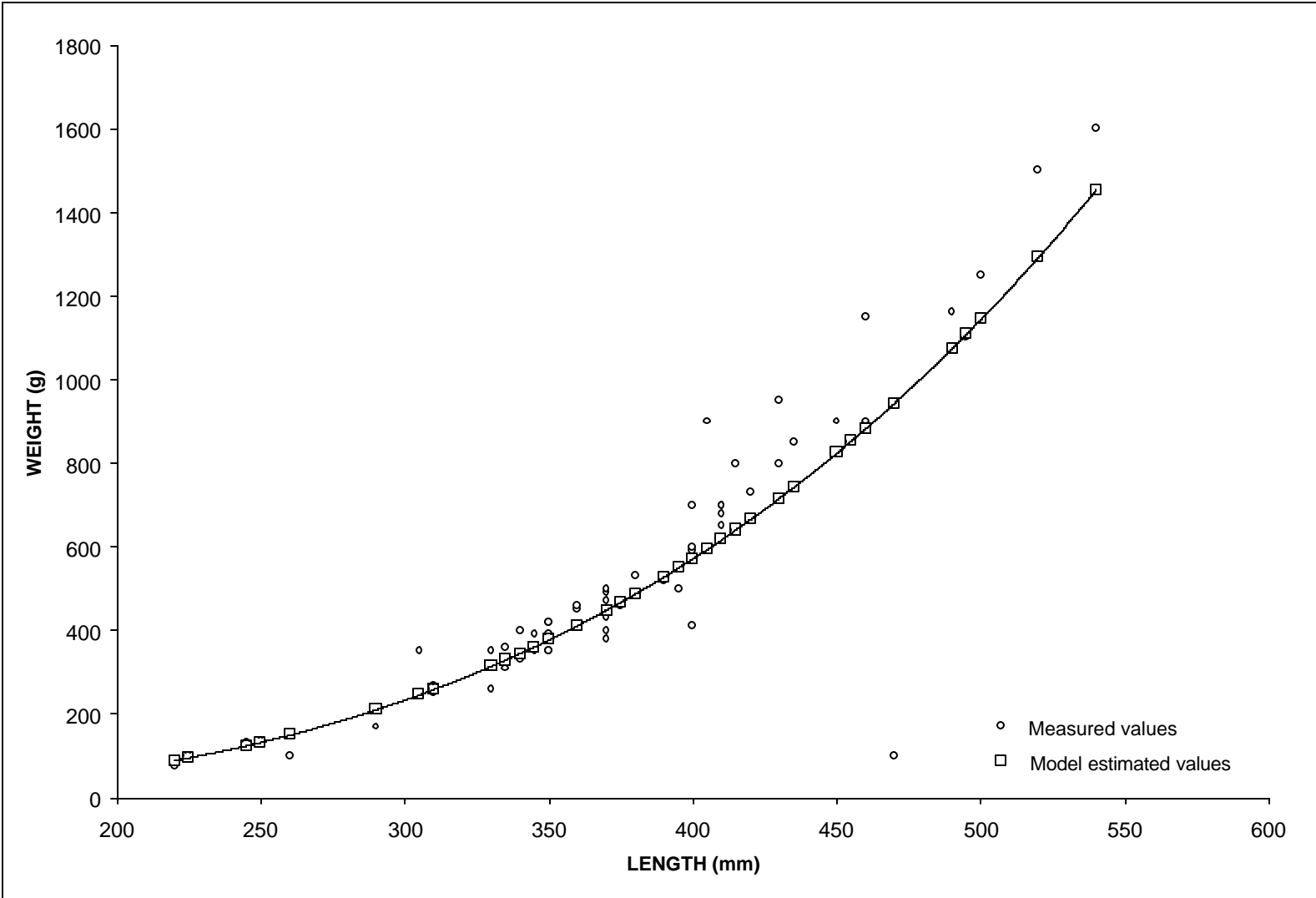


Figure B-4. Comparison of measured (○) and modeled (◻) length-weight relationships for northern pikeminnow (NPM) in the Willamette River Basin. Spearman rank correlation ( $r_s$ ) = 0.962,  $p < 0.001$ .

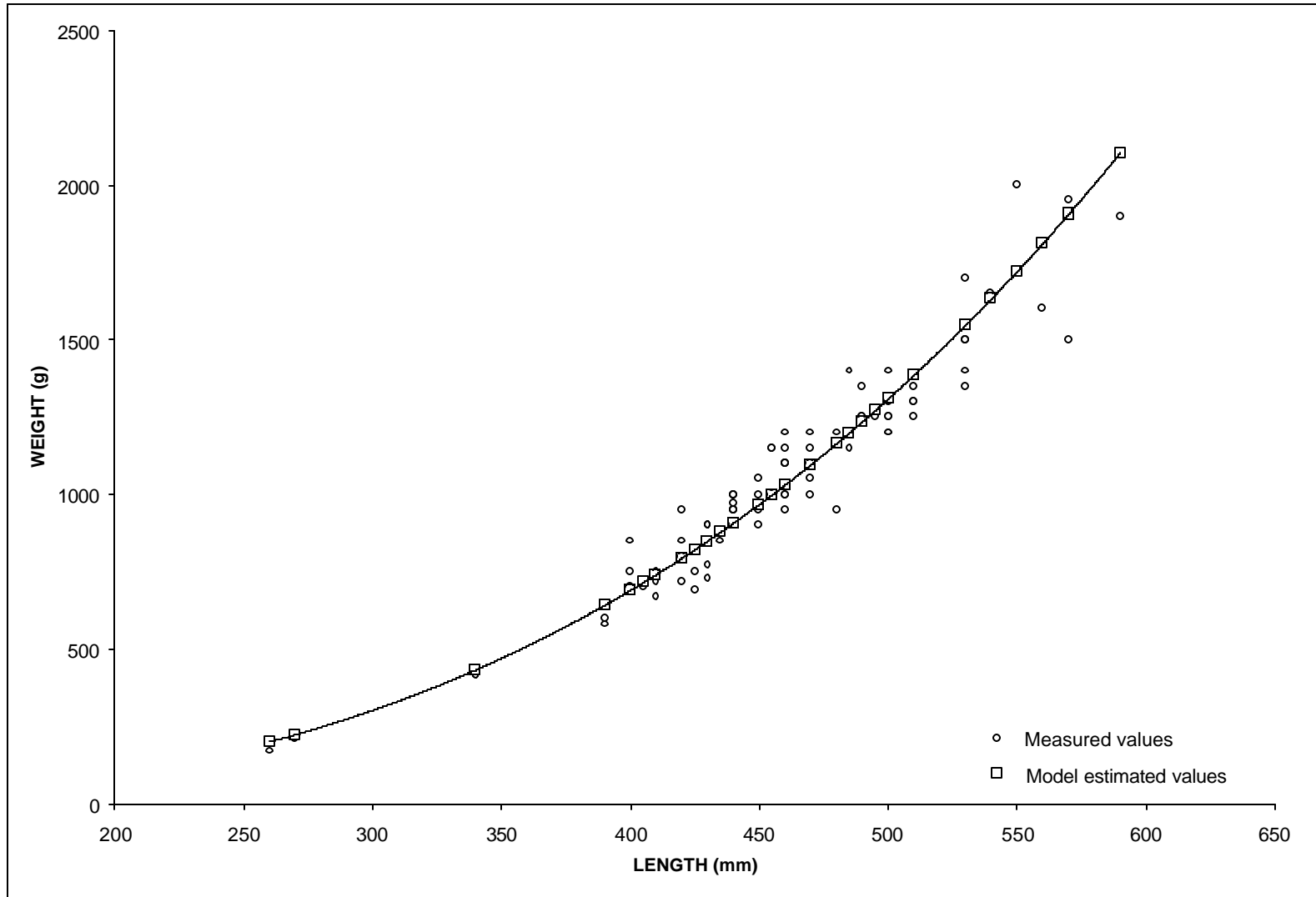


Figure B-5. Comparison of measured (○) and modeled (◻) length-weight relationships for largescale sucker (LSS) in the Willamette River Basin. Spearman rank correlation ( $r_s$ ) = 0.964,  $p < 0.001$ .

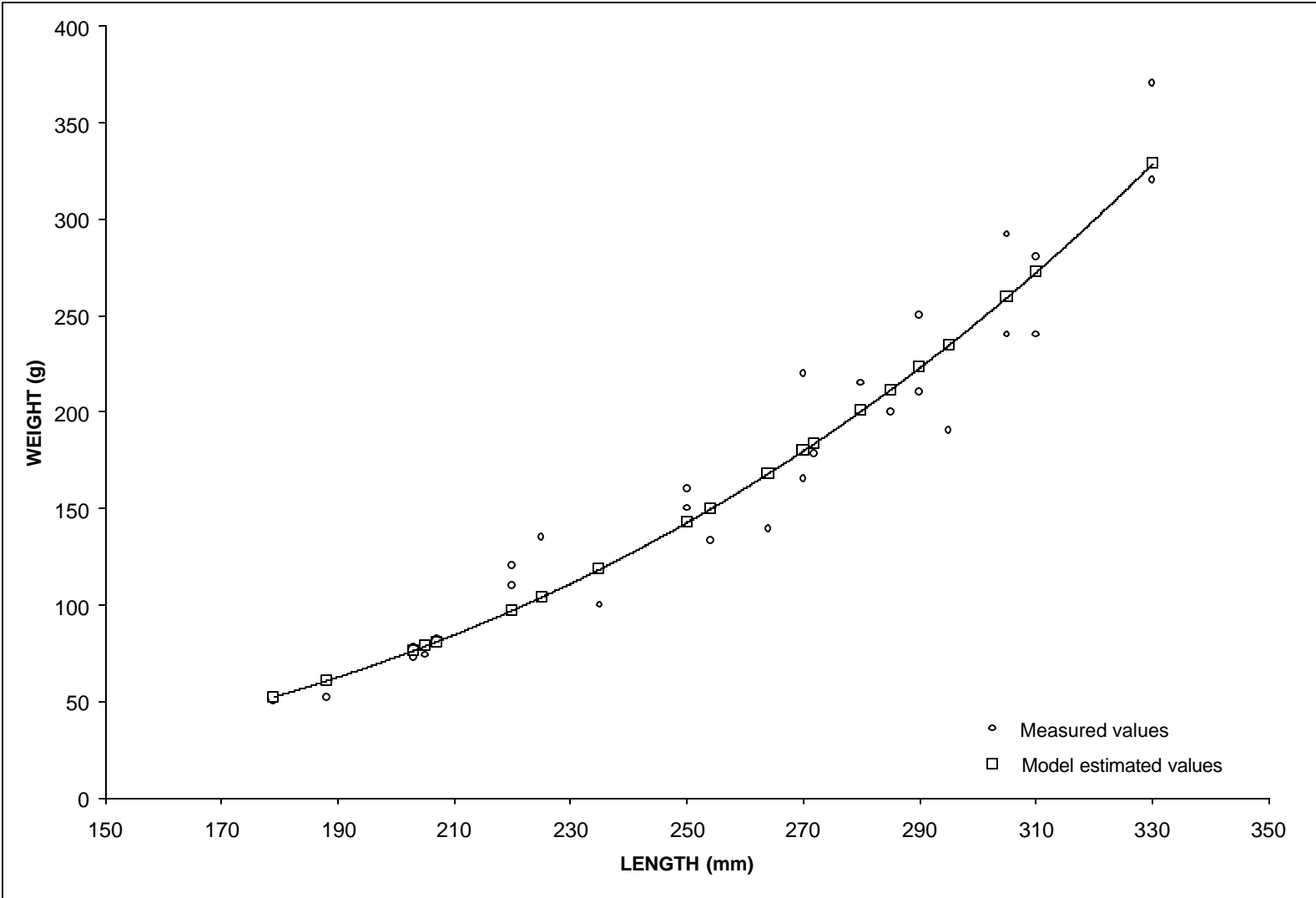


Figure B-6. Comparison of measured (○) and modeled (◻) length-weight relationships for cutthroat trout (CTT) in the Willamette River Basin. Spearman rank correlation ( $r_s$ ) = 0.848,  $p < 0.001$ .

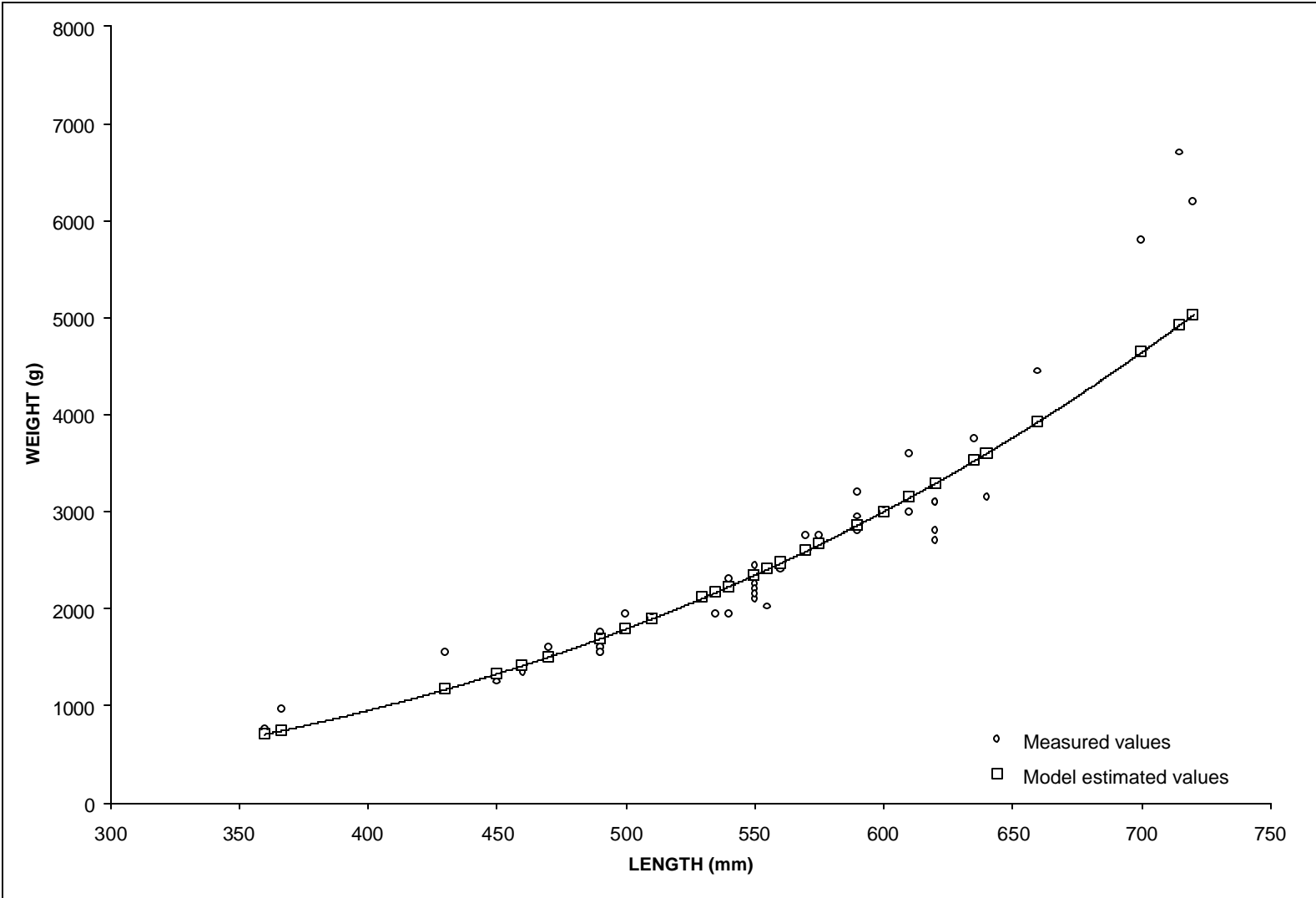


Figure B-7. Comparison of measured (○) and modeled (◻) length-weight relationships for common carp (CAR) in the Willamette River Basin. Spearman rank correlation ( $r_s$ ) = 0.933,  $p < 0.001$ .

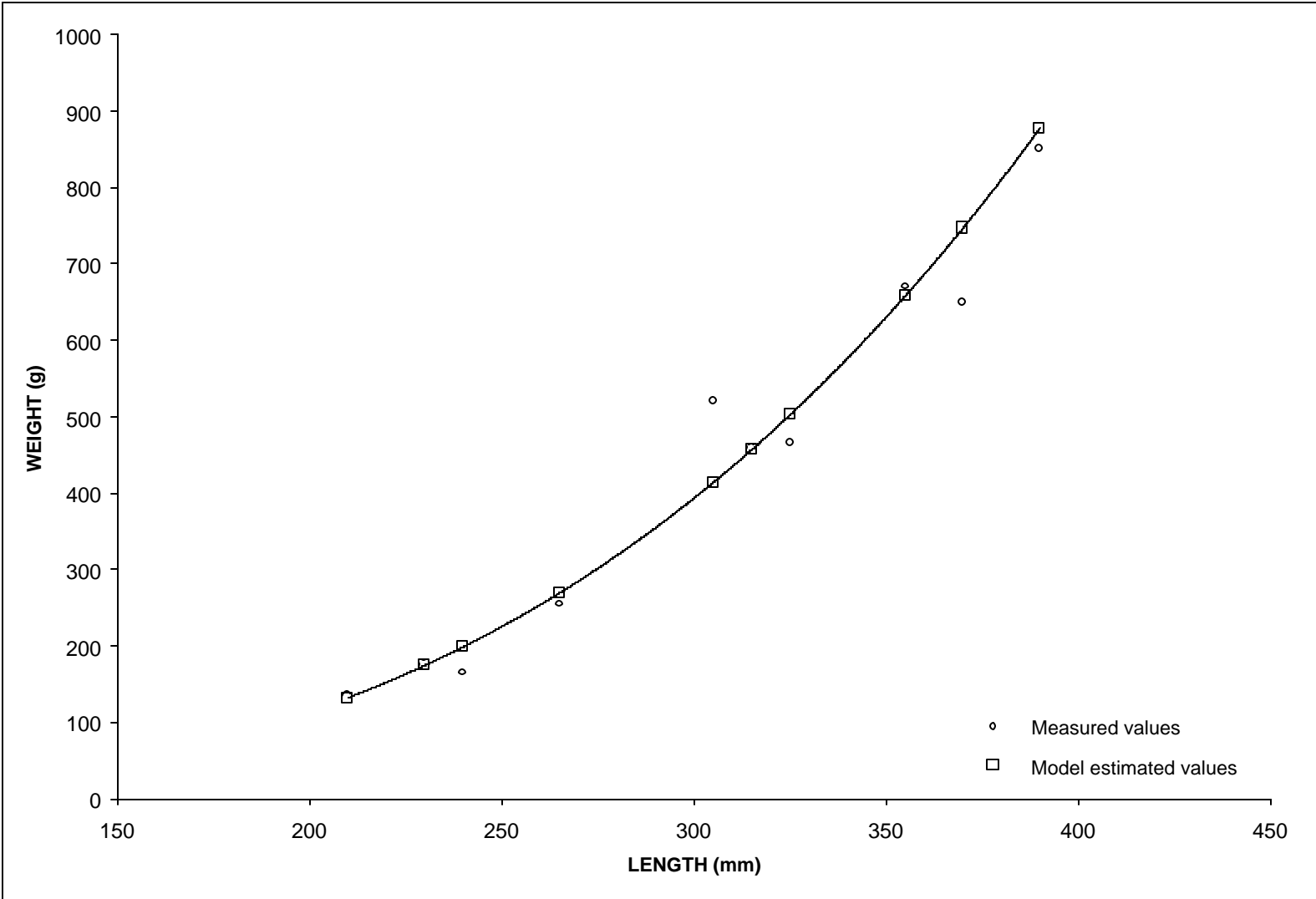


Figure B-8. Comparison of measured (○) and modeled (◻) length-weight relationships for smallmouth bass (SMB) in the Willamette River Basin. Spearman rank correlation ( $r_s$ ) = 0.729,  $p < 0.001$ .

## APPENDIX C

### Comparison of Modeled and Measured Fish Tissue Concentration - Body Length Relationships

**Table C-1. Summary of regression statistics for measured and model estimated tissue concentration - body length relationships.**

	slope	intercept	Pearson ( $r^2$ )	Pearson p
<b>MEASURED</b>				
BLU	0.0002	-0.73	0.02	0.93
NPM	0.0020	-1.00	0.57	<0.001
LMB	0.0008	-0.64	0.26	<0.001
LSS	0.0039	-2.54	0.66	<0.001
CAR	0.0018	-1.60	0.65	<0.001
RBT	0.0034	-1.78	0.34	0.05
CTT	0.0042	-2.28	0.29	0.15
SMB	0.0041	-1.86	0.84	0.005
<b>PRE-CALIBRATION</b>				
BLU	0.0019	-1.59	0.18	<0.001
NPM	0.0011	-1.26	0.15	<0.001
LMB	0.0016	-1.36	0.28	<0.001
LSS	0.0002	-1.44	0.02	0.68
CAR	-0.0004	-1.11	-0.08	0.07
RBT	0.0019	-1.60	0.20	<0.001
CTT	0.0014	-1.44	0.09	0.04
SMB	0.0015	-1.40	0.20	<0.001
<b>POST - CALIBRATION</b>				
BLU	0.0033	-1.39	0.31	<0.001
NPM	0.0015	-0.84	0.23	<0.001
LMB	0.0017	-0.98	0.34	<0.001
LSS	0.0019	-1.66	0.23	<0.001
CAR	0.0012	-1.37	0.20	<0.001
RBT	0.0021	-1.41	0.21	<0.001
CTT	0.0028	-1.90	0.17	<0.001
SMB	0.0023	-1.42	0.27	<0.001

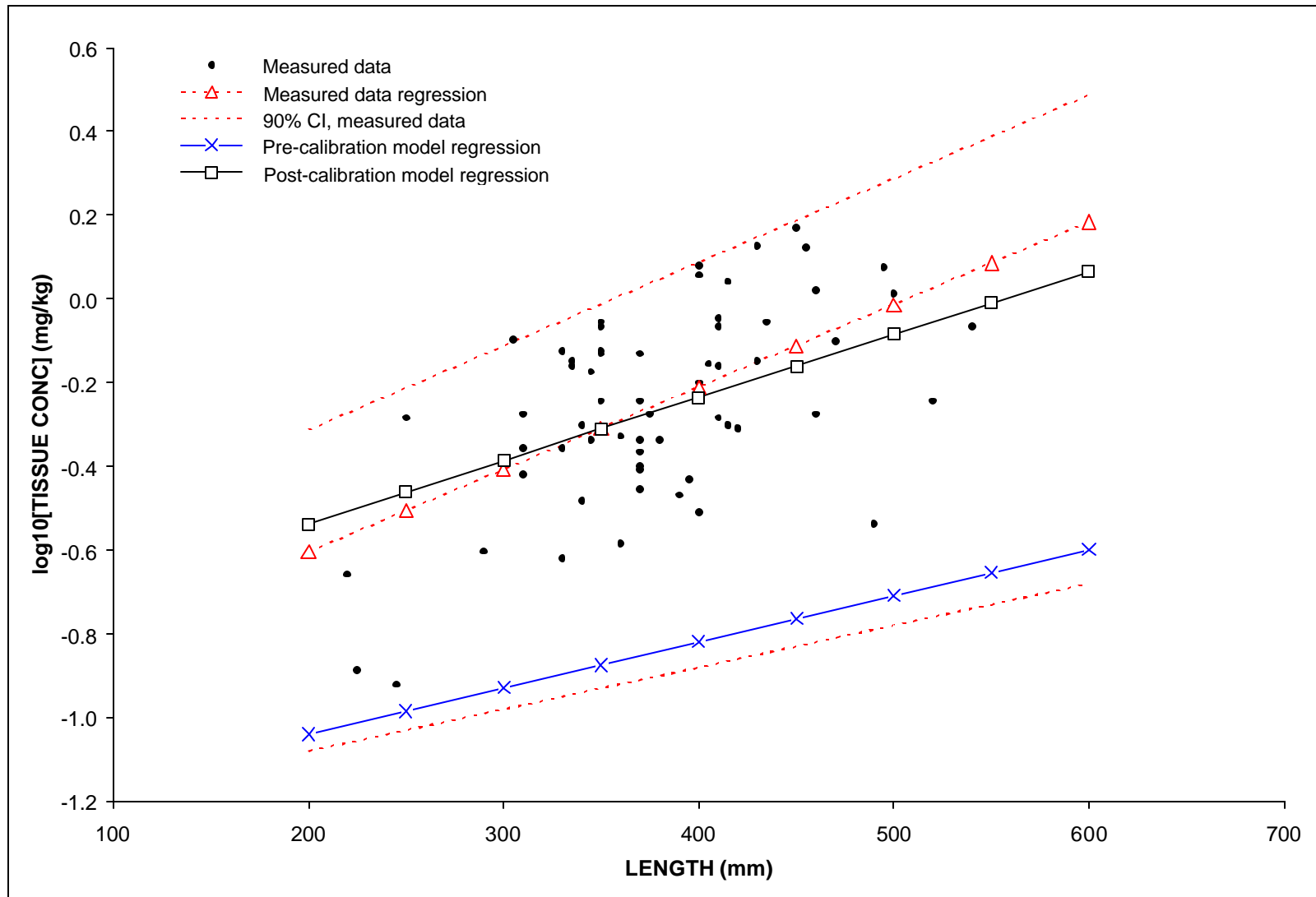


Figure G1. Comparison of (a) measured and modeled estimated tissue mercury concentration-body length relationships, (b) pre- and post calibration modeled relationship estimates, and (c) the 90 percent confidence interval on (----) the measured data for northern pikeminnow (NPM).

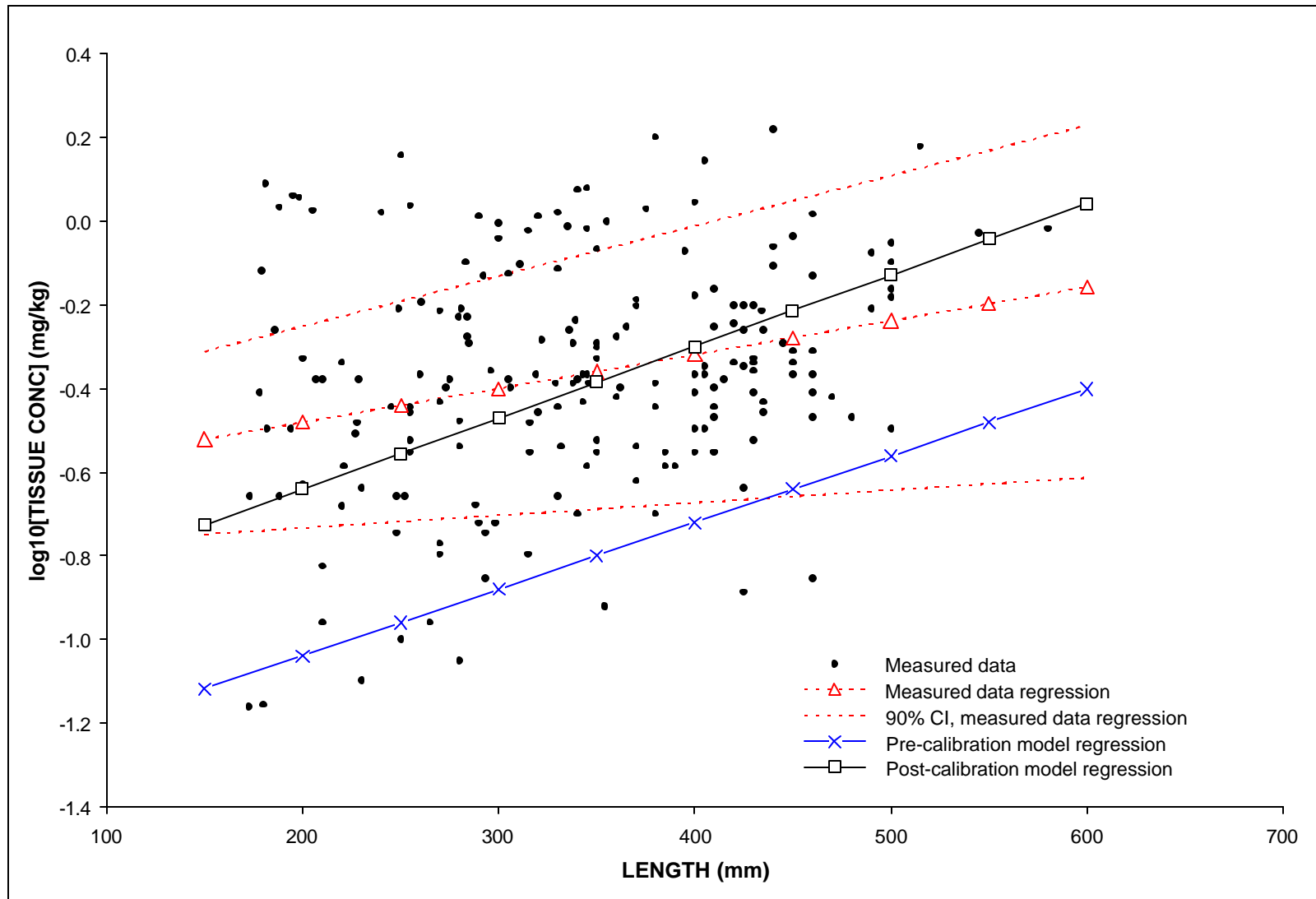


Figure G2. Comparison of (a) measured and modeled estimated tissue mercury concentration-body length relationships, (b) pre- and post calibration modeled relationship estimates, and (c) the 90 percent confidence interval on (----) the measured data for largemouth bass (LMB).

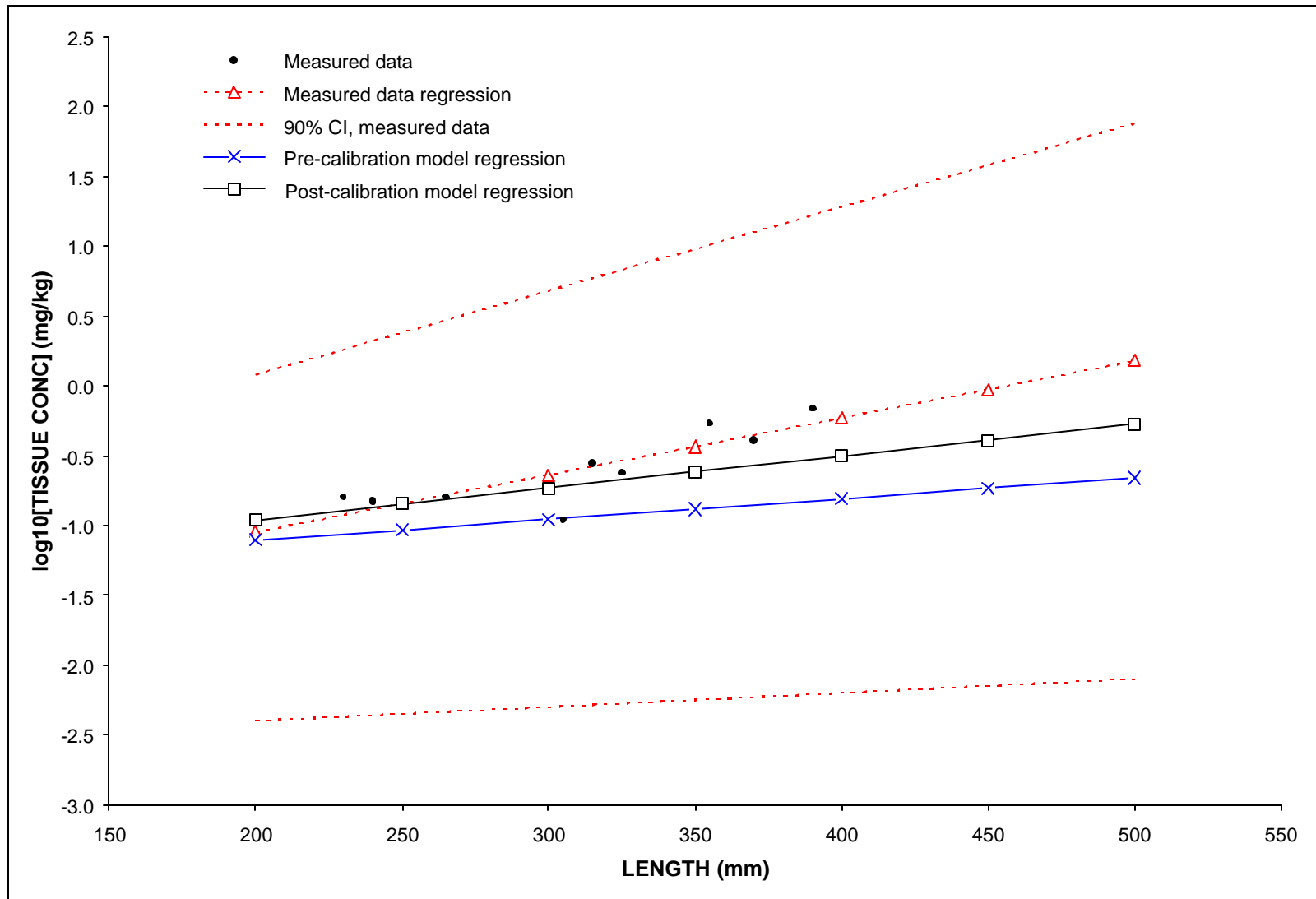


Figure G3. Comparison of (a) measured and modeled estimated tissue mercury concentration-body length relationships, (b) pre- and post calibration modeled relationship estimates, and (c) the 90 percent confidence interval on (----) the measured data for smallmouth bass (SMB).

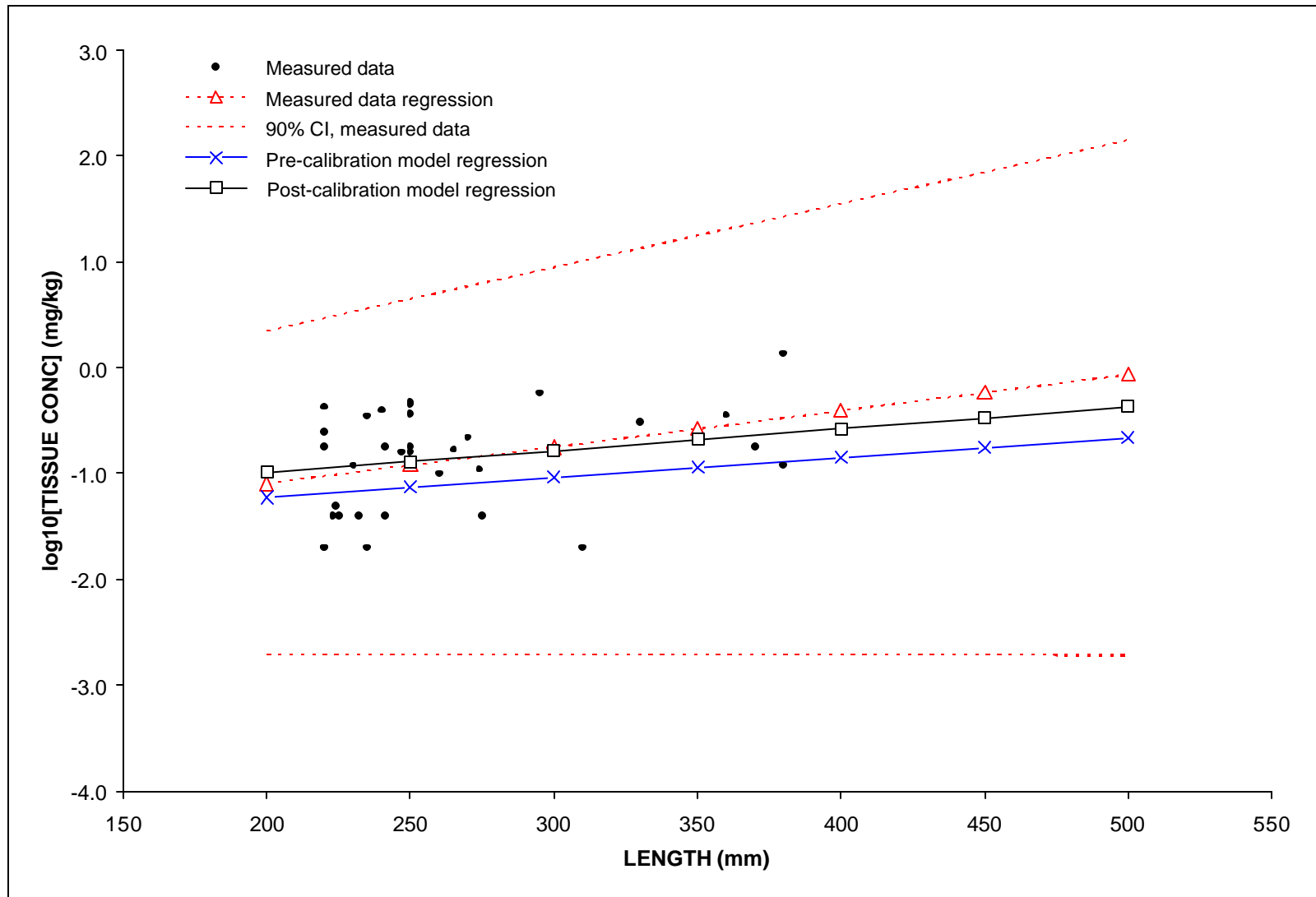


Figure G4. Comparison of (a) measured and modeled estimated tissue mercury concentration-body length relationships, (b) pre- and post calibration modeled relationship estimates, and (c) the 90 percent confidence interval on (---) the measured data for rainbow trout (RBT).

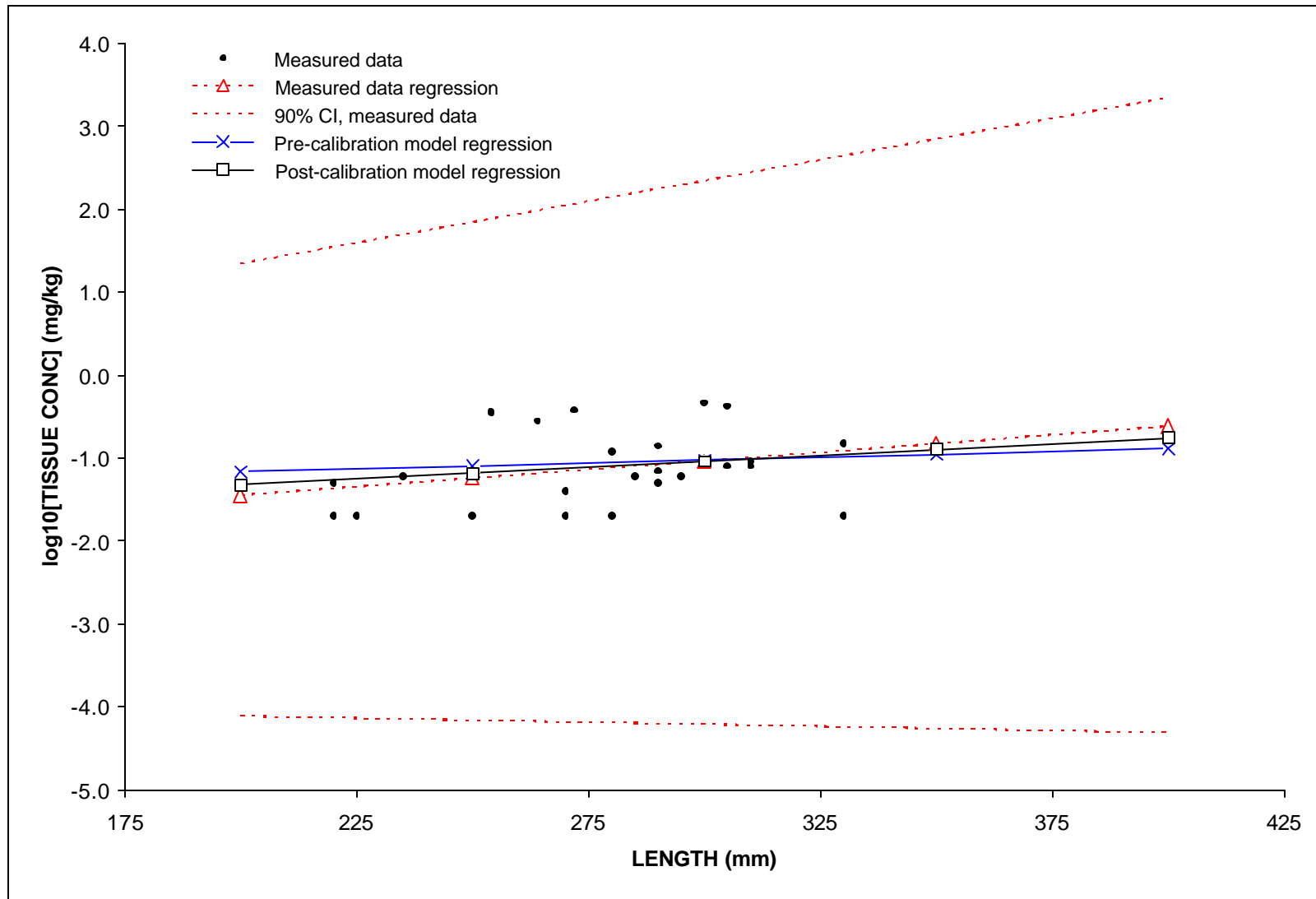


Figure G5. Comparison of (a) measured and modeled estimated tissue mercury concentration-body length relationships, (b) pre- and post calibration modeled relationship estimates, and (c) the 90 percent confidence interval on (----) the measured data for cutthroat trout (CTT).

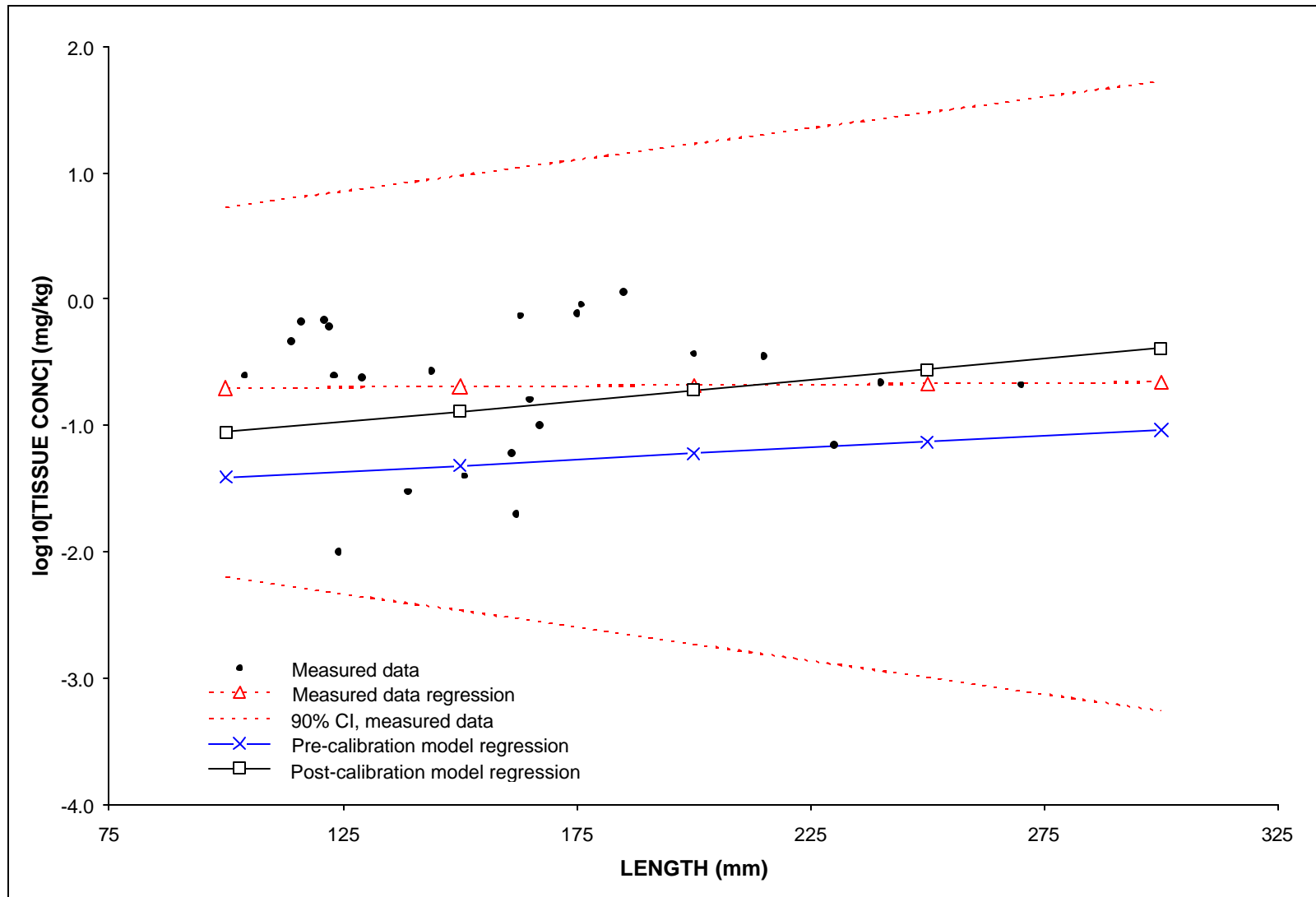


Figure G6. Comparison of (a) measured and modeled estimated tissue mercury concentration-body length relationships, (b) pre- and post calibration modeled relationship estimates, and (c) the 90 percent confidence interval on (----) the measured data for bluegill (BLU).

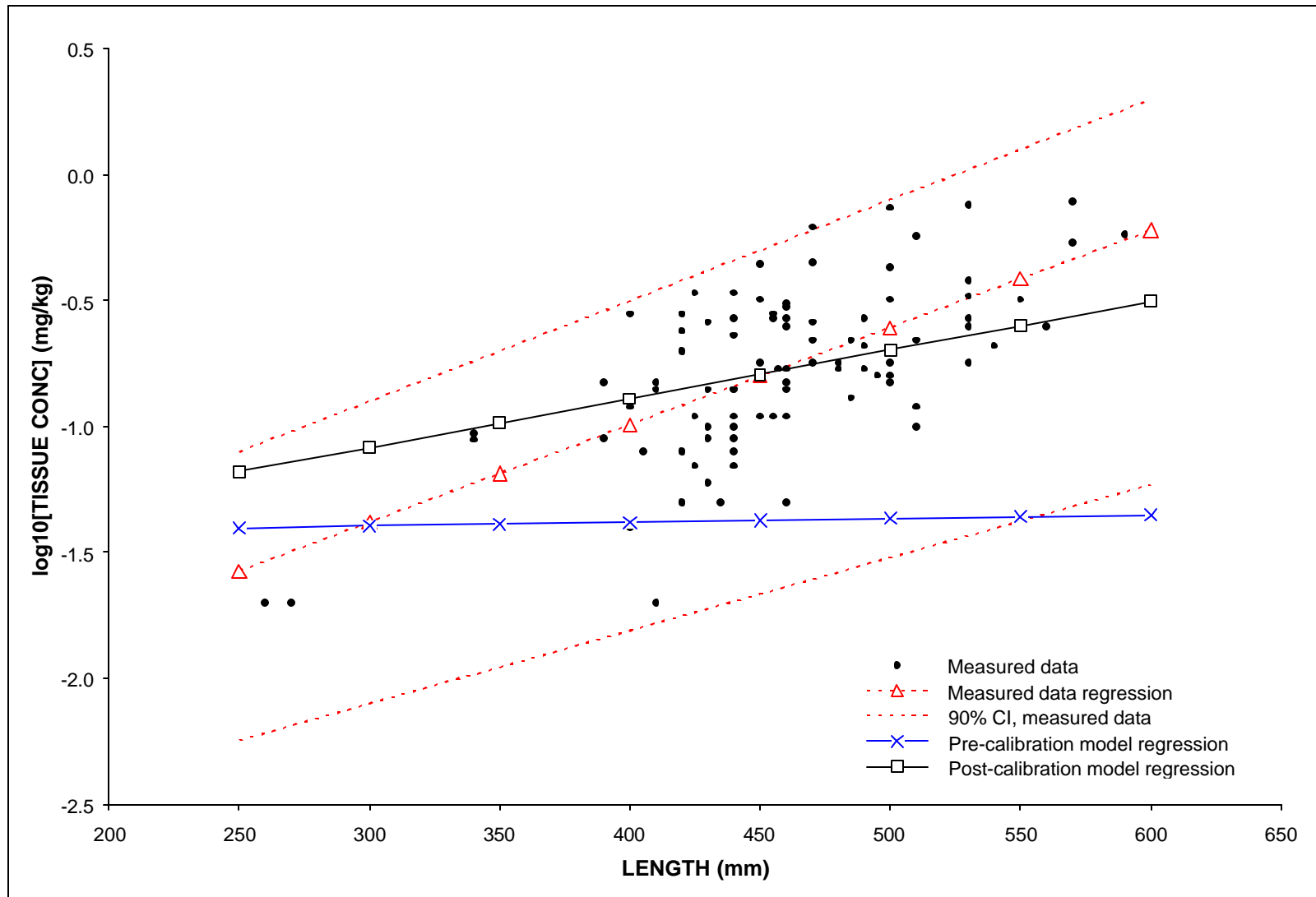


Figure G7. Comparison of (a) measured and modeled estimated tissue mercury concentration-body length relationships, (b) pre- and post calibration modeled relationship estimates, and (c) the 90 percent confidence interval on (----) the measured data for largescale sucker (LSS).

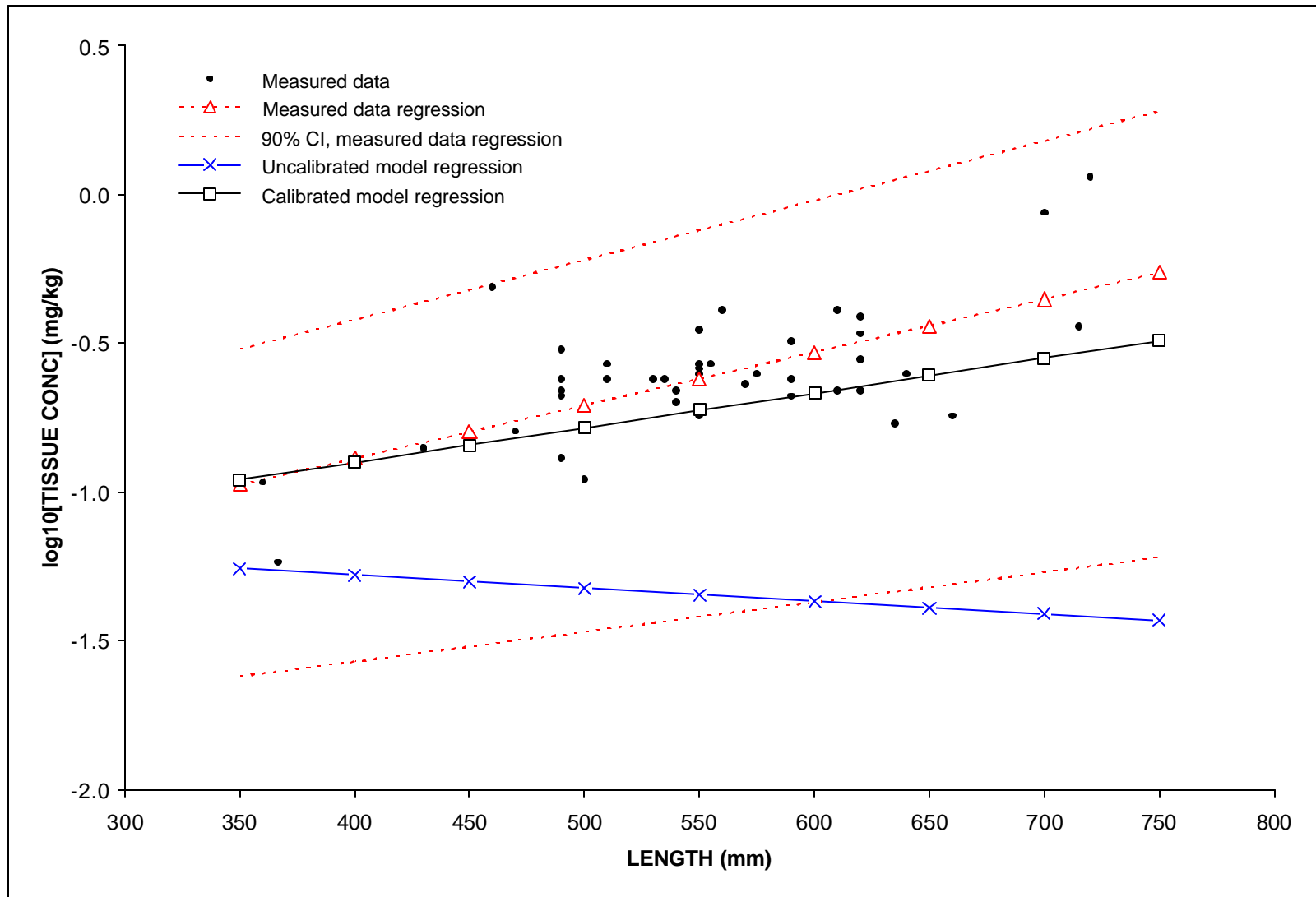


Figure G8. Comparison of (a) measured and modeled estimated tissue mercury concentration-body length relationships, (b) pre- and post calibration modeled relationship estimates, and (c) the 90 percent confidence interval on (----) the measured data for common carp (CAR).