

# PROBABILISTIC ASSESSMENT OF REGIONAL MERCURY EXPOSURE

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**Abstract.** A methodology for the probabilistic assessment of regional exposure to mercury is presented. The methodology is based on a multimedia model of the atmospheric fate and transport of mercury over a continental scale, and the aquatic simulation of the transformation and bioaccumulation of mercury in lakes in the region of interest. A probabilistic approach allows one to take into account the spatial variability of environmental characteristics and the variability of dose-related variables (e.g., fish consumption, body weight) among individuals. The methodology is demonstrated for total mercury deposition in the Great Lakes region of the U.S.

## 1. Introduction

Previous work has focused on the health risk assessment of mercury (Hg) emissions from industrial facilities such as power plants at local spatial scales, i.e., of the order of 100 km (e.g., Constantinou *et al.*, 1995a). Since recent studies have shown that the local impacts (i.e., within 100 km) of Hg emissions from individual power plants are typically not significant and that the majority of these power plant emissions is transported beyond 100 km from the sources (Allan *et al.*, 1994; Constantinou *et al.*, 1995b), it is of interest to address the potential impacts of Hg emissions at regional scales (i.e., at scales ranging from 100 km to several 1,000 km). We address here the potential regional impacts of atmospheric Hg transported over the contiguous United States of America (U.S.). Figure 1 depicts the overall methodology used to assess the potential exposure to Hg transported over such regional scales. This methodology consists of the following steps:

- (1) Simulation of the long-range transport, transformations and deposition of atmospheric Hg over a continental scale;
- (2) Simulation of the aquatic chemistry and bioaccumulation in fish of Hg in lakes of a selected region;
- (3) Estimation of the human dose of Hg through consumption of local freshwater fish.

Clearly, it is not feasible to simulate the fate and bioaccumulation of Hg in fish for every surface water body of the contiguous U.S. Moreover, consumption of fish from local lakes varies significantly among individuals. Therefore, a probabilistic approach was selected that could account for the spatial variability of environmental characteristics relevant to the atmospheric deposition and bioaccumulation of Hg, as well as for the variability in local fish consumption among the U.S. population. In this

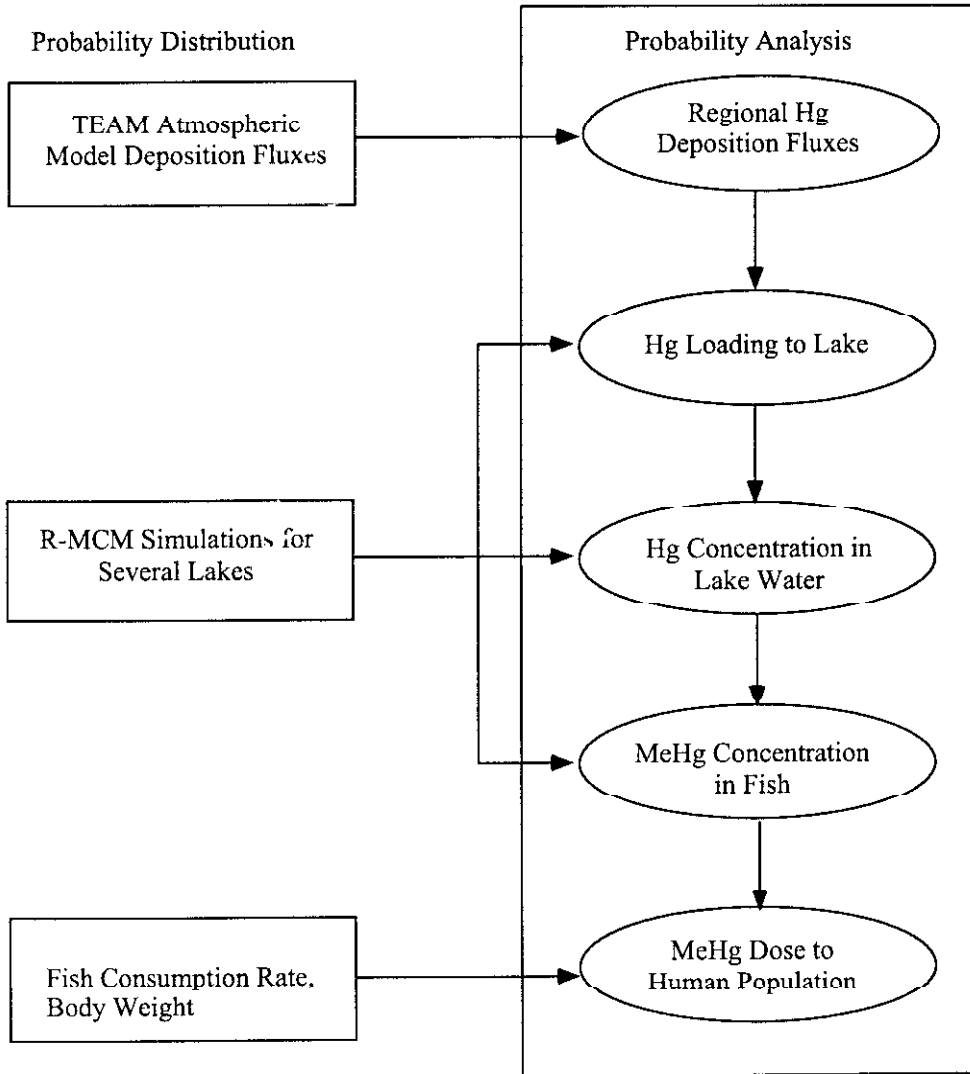


Fig. 1. Schematic Description of the Methodology for the Probabilistic Assessment of Regional Hg Exposure.

approach, we use probability distributions of the major relevant variables to construct probability distributions of the human exposure to Hg through fish consumption. It must be emphasized that there are large uncertainties that remain in various components of the analysis (i.e., atmospheric modeling, aquatic modeling, dose modeling). Our analysis addresses only the variability of key input variables and does not include a treatment of input data uncertainties or model formulation uncertainties. Therefore, the results presented here should be seen as a demonstration of the methodology rather than definitive exposure assessment values. It must be noted also that our regional assessment addresses all sources of Hg, not only power plant emissions. Global background Hg is included through the boundary conditions of the atmospheric model. However, since our

analysis addresses the regional impacts of Hg emissions, the localized impacts of specific sources (air emissions, water discharges or land contamination) are not considered. Also, it should be noted that at this point, uncertainties still remain in emission speciation, boundary conditions, meteorological inputs and model formulation that prevent us from using the models for the purpose of estimating individual contributions from specific sources.

We present first the overall modeling approach. Next, the probability distributions selected for the relevant variables are discussed. Then, the methodology is demonstrated using the Great Lakes region of the U.S. as an example.

## 2. Modeling Approach

As depicted in Figure 1, the overall model of the fate and transport of Hg and its dose to humans consists of several components. We briefly discuss each of these components.

**Atmospheric transport, transformation and deposition of Hg:** The TEAM regional atmospheric model (Pai *et al.*, 1997) was used to simulate the long-range transport, chemical transformations, and dry and wet deposition of Hg over the contiguous U.S. Comparison of the simulation results of the TEAM model with observations for total gaseous Hg (TGM) ambient concentrations and wet deposition fluxes showed reasonable agreement (Pai *et al.*, 1997). Therefore, the dry and wet deposition fluxes of Hg calculated by the TEAM atmospheric model were used as input to the watersheds.

**Hg loading to the lake:** The amount of Hg that enters a lake depends on the total Hg loading to the watershed from atmospheric deposition (see above), the ratio of the watershed area to the lake area and the fraction of Hg deposited in the watershed that is transported to the lake. The Regional Mercury Cycling Model (R-MCM) (Harris *et al.*, 1996) was used to simulate Hg loading to lakes from dry and wet deposition fluxes to their watersheds.

**Hg methylation in lake water and bioaccumulation in fish:** The aquatic concentration of methylHg and its bioaccumulation through the food chain depend on the Hg loading to the lake, the characteristics of the lake (water quality, acidity, watershed type, biological characteristics) and the bioenergetics of the various trophic levels from benthos to predator fishes. R-MCM was used to simulate the cycling of Hg in lakes and its bioaccumulation in fish.

**Ingestion dose:** The methylHg dose to the human body depends on the methylHg concentration in fish, the consumption rate of fish caught in local lakes and the human body weight. Note that we consider only methylHg as the Hg species of interest. Also, we only address here the consumption of local freshwater fish. Consumption of commercial fish from freshwater and the ocean/sea will contribute to the total body burden, but their methylHg concentrations are not addressed in our modeling approach, since some of those fishes reside out of our modeling domain. Consumption of commercial fish can be considered to be a "background" fish intake and we address here the incremental fish intake due to consumption of fish caught by local anglers.

The mathematical relationships for each one of these model components are presented below. Note that in each case, a relatively sophisticated model (e.g., TEAM, R-MCM) is used to develop a range of parameter values (e.g., atmospheric deposition flux, bioaccumulation factor) for input into the probabilistic model.

The atmospheric deposition flux (ADF) ( $\mu\text{g}/(\text{m}^2 \cdot \text{y})$ ) is calculated as the sum of the dry deposition flux, DDF, and wet deposition flux, WDF, obtained from the TEAM atmospheric model.

$$\text{ADF} = \text{DDF} + \text{WDF} \quad (1)$$

The Hg loading to lakes, MLL ( $\mu\text{g}/(\text{m}^2 \cdot \text{y})$ ) is calculated by R-MCM for each given lake.

$$\text{MLL} = \text{ADF} * W \quad (2)$$

where  $W$  is a dimensionless parameter that is lake-specific. This parameter accounts for the ratio of the watershed area to the lake area, as well as the fraction of Hg retained in the terrestrial area surrounding the lake.

The total Hg concentration in the lake water column,  $C_w$  (mg/l) depends on the Hg loading to the lake as well as some lake-specific characteristics such as the sedimentation rate.

$$C_w = \text{MLL} * d * 10^{-6} \quad (3)$$

where  $d$  is a lake-specific parameter (y/m) that is calculated by R-MCM.

The methylation of Hg in lake water and its bioaccumulation in fish were found to be slightly negatively correlated in the application of R-MCM to 29 lakes throughout the U.S. That is, there were no lakes with both high methylation rate and high bioaccumulation factors. Therefore, the processes of Hg methylation and subsequent bioaccumulation were grouped into a single equation that represents the ratio of methylHg concentration in fish,  $C_f$  (mg/kg) to total Hg concentration in lake water,  $C_w$ .

$$C_f = C_w * \text{BAF}_f \quad (4)$$

where  $\text{BAF}_f$  represents a bioaccumulation factor referring to total Hg in lake water (l/kg).  $\text{BAF}_f$  is a lake-specific parameter that is calculated by R-MCM.

The ingestion dose rate,  $D$  ( $\text{mg}/(\text{kg} \cdot \text{day})$ ), is calculated from the methylHg fresh fish concentration,  $C_f$ , the fish consumption rate, FIR ( $\text{g}/\text{day}$ ) and the human body weight, BW (kg).

$$D = C_f * \text{FIR} * 10^{-3} / \text{BW} \quad (5)$$

### 3. Probability Distributions of Input Variables and Parameters

For the purpose of demonstrating the methodology, we apply it to the Great Lakes region. We present here the probability distributions selected for the relevant variables for this specific application. As mentioned above, the TEAM atmospheric model was used to estimate the deposition fluxes of Equation (1), and R-MCM was used to calculate the parameters  $W$ ,  $d$ , and  $\text{BAF}_f$  in Equations (2) through (4).

The atmospheric model, TEAM, was applied to an area that covers the contiguous U.S., southern Canada and northern Mexico. Hg deposition fluxes due to the global background and anthropogenic Hg emissions from within the TEAM domain were simulated over the entire domain. Results are presented here for the Great Lakes region. A comparison of TEAM model simulation results for atmospheric TGM concentrations and Hg wet deposition fluxes with observations collected in Minnesota, Wisconsin and Michigan showed the model calculations to be consistent with the observations (Pai *et al.*, 1997). For the purpose of this probabilistic assessment, we selected the full range of modeled deposition values for three states of the Great Lakes area (Minnesota, Wisconsin and Michigan) to represent the variable DDF and WDF of Equation (1). These three states correspond to states where lakes simulated with R-MCM were located. Values of the deposition fluxes were calculated by the atmospheric model for each surface grid cell of 100 km x 100 km area. Lognormal distributions represented most closely the deposition flux histograms and, therefore, were selected for both dry and wet deposition fluxes. The characteristics of these distributions are presented in Table I.

Table I.  
Input Variables used in the Probabilistic Exposure Assessment.

Input Variable	Distribution Function	Characteristics	Source
DDF	Lognormal truncated <sup>1,2</sup>	Mean = 5.1 $\mu\text{g}/(\text{m}^2 \cdot \text{y})$ $\sigma = 2.8 \mu\text{g}/(\text{m}^2 \cdot \text{y})$	TEAM
WDF	Lognormal truncated <sup>1,3</sup>	Mean = 8.4 $\mu\text{g}/(\text{m}^2 \cdot \text{y})$ $\sigma = 4.6 \mu\text{g}/(\text{m}^2 \cdot \text{y})$	TEAM
W	Lognormal truncated <sup>2,3</sup>	Mean = 2.42 $\sigma = 0.38$	R-MCM
d	Lognormal truncated <sup>2,3</sup>	Mean = 45.4 y/m $\sigma = 26.3 \text{ y/m}$	R-MCM
BAF <sub>t</sub>	Lognormal truncated <sup>2,3</sup>	Mean = 390,000 l/kg $\sigma = 218,000 \text{ l/kg}$	R-MCM
FIR	0 up to 65th percentile, Exponential truncated above 65th percentile	Mean = 3.7 g/day Maximum value = 200 g/day	This work
BW	Lognormal <sup>3</sup>	Mean = 66 kg $\sigma = 13 \text{ kg}$	AIHC, 1994

<sup>(1)</sup>Distributions were truncated at the minimum and maximum values obtained in the TEAM simulation.

<sup>(2)</sup>Distributions were truncated at two standard deviations above the mean or at the maximum value obtained in the 8 R MCM simulations, whichever was the greater.

<sup>(3)</sup>Arithmetic mean and standard deviation.

R-MCM was applied to a data base of 29 lakes distributed throughout the U.S. (Mitchell *et al.*, 1995) with 8 of those lakes located in the Great Lakes region, in Minnesota, Wisconsin and Michigan. These eight lakes include drainage lakes as well as seepage lakes. Table II summarizes the major characteristics of these eight lakes.

Table II  
Summary of Lake Characteristics

Lake number & name	Lake Type	pH	DOC (mg/l)	Predatory Fish Species
1. McGrath L.	Seepage	5.4	3.0	Largemouth bass
2. L. Mendota	Drainage	8.5	10.0	Walleye
3. Big Portage L.	Drainage	8.6	3.0	Northern pike
4. L. Mitchell	Drainage	7.7	3.0	Walleye
5. Park L.	Seep./Drain.	8.3	12.6	Largemouth bass
6. Platte L.	Drainage	8.3	2.0	Northern pike
7. Grove L.	Drainage	8.1	10.0	Northern pike
8. Mountain L.	Drainage	7.4	10.0	Northern pike

The median, mean and maximum values for methylHg concentration in predator fish for the eight lakes simulated are 0.46, 0.67 and 2.1 ppm, respectively. These results are commensurate with observed methylHg concentrations in fish observed in the Great Lakes region. For example, a survey of 56 lakes in Michigan gave an arithmetic mean concentration of methylHg in largemouth bass (all ages) of 0.44 ppm and a maximum concentration of 1.8 ppm (Day, 1996). Figure 2 presents the modeled methylated fraction of Hg for these lakes; Figure 3 presents the modeled bioaccumulation factor for methylHg (the product of these two parameters correspond to BAF, in Equation (4)). The methylHg concentrations in fish were found to be sensitive primarily to the dissolved organic carbon concentration in lake water, lake temperature, pH and sediment burial velocity. The results of the R-MCM simulations showed significant variation in methylHg concentrations and methylHg bioaccumulation in fish, within a given region (e.g., in the Great Lakes region as shown in Figures 2 and 3). These variations result from the differences in water quality that can be observed among surface water bodies within a given region. Therefore, it would not be feasible to represent a region by a single "typical" lake, and a probabilistic approach that integrates the variations in environmental characteristics observed within that region is the preferred approach. The R-MCM simulation results for these eight lakes were used to develop probability distributions of the parameters  $W$ ,  $d$ , and BAF. The application of R-MCM to the data base of 29 lakes suggests that the variability of these parameters can be approximated by lognormal distributions. The specific distributions derived for the Great Lakes region are presented in Table I.

The probability distribution for the fish consumption rate was developed as follows. The mean consumption in the U.S. population, including those who eat freshwater fish and those who do not, is estimated to be 2.2 g/day based on the 1989-1991 Continuous Survey of Food Intake by Individuals (CSFII) conducted by the U.S. Department of the Agriculture (EPA, 1996). This freshwater fish consumption includes both locally caught fish and farm raised fish. The latter amounts to a production of over 500 million pounds per year, i.e., about 2.4 g/day per capita. However, only about half of this amount is likely to be eaten due to wastage and the fact that not all of the production weight is edible. This analysis suggests that the average consumption of locally caught

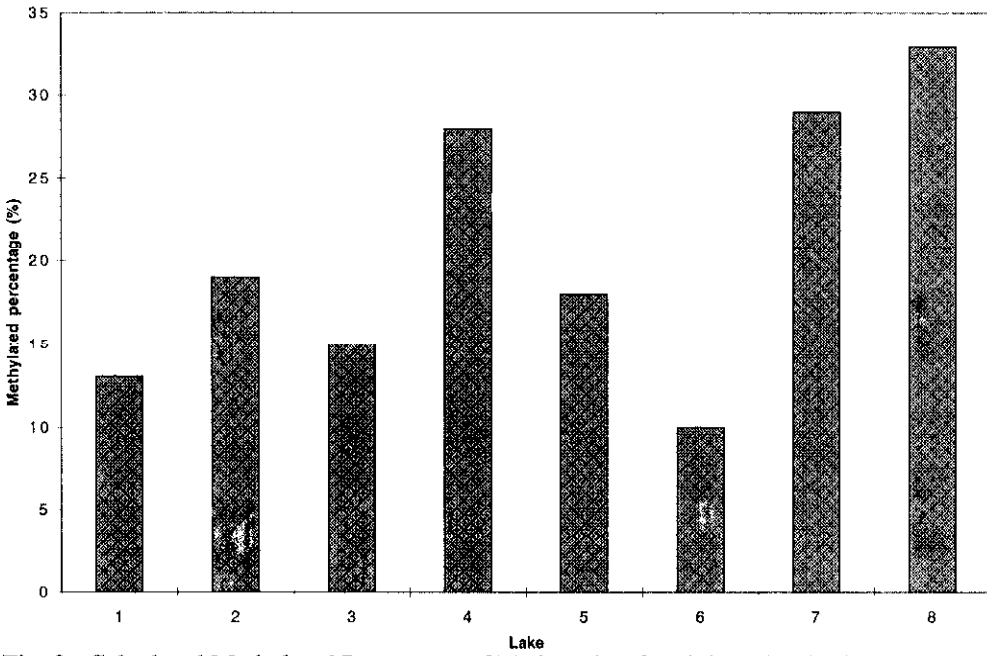


Fig. 2. Calculated Methylated Percentage of Hg in Lakes for eight Lakes in the Great Lakes Region.

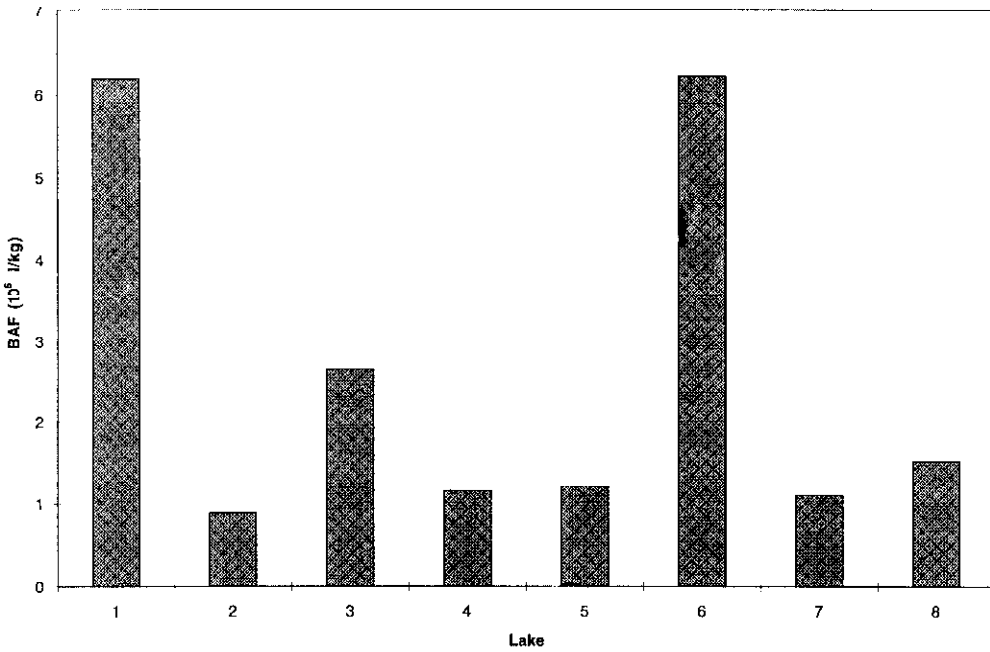


Fig. 3. MethylHg Bioaccumulation Factor calculated for the Predator Fish for eight Lakes in the Great Lakes Region.

fish is of the order of 1 g/day. The U. S. Environmental Protection Agency (EPA) Mercury Study Report to Congress cites a U.S. Fish and Wildlife Service report indicating that 26% of the U.S. population fished in 1985 (EPA, 1996). The EPA report also provides a review of fish consumption among groups of anglers. Ten studies are reported in the EPA report. There is a large variation in the amount of locally caught fish consumed by the anglers. The mean (or, if not available, the median) value reported in these studies ranges from 1.8 g/day (Soldat, 1970) to 60 g/day (Sekerke *et al.*, 1994). Ebert *et al.* (1993) reported in a study of Maine anglers a mean value of 5.0 g/day for fish consumption by all surveyed anglers and 3.7 g/day for fish consumption by the anglers and their family. Combining the results reported by Ebert *et al.* and the EPA estimate that 26% of the population fishes recreationally suggests that 35% of the population eats locally caught freshwater fish. These values result in a mean consumption rate of 1.3 g/day for the entire U.S. population, which is consistent with the estimate based on the CSFII data base. Based on this review of available data, we assumed that 65% of the population does not eat locally caught freshwater fish. For the 35% of the population that does consume locally caught freshwater fish, an exponential distribution with a mean value of 3.7 g/day and truncated at a maximum value of 200 g/day was used. An exponential type distribution is consistent with the review of freshwater fish consumption assessments.

The probability distribution for the weight of an adult woman is based on the work of Brainard and Burmaster (1992) and is recommended in the American Industrial Health Council's Exposure Factors Sourcebook (AIHC, 1994). The distribution is lognormal with an arithmetic mean value of 66 kg. A correlation between the fish consumption rate and the body weight was introduced to account for the fact that heavier individuals tend to eat more. A value of 0.33 was used for the correlation coefficient, following Hoover *et al.* (1997), who derived their correlation based on data reported in Liptert *et al.* (1996).

#### 4. Probabilistic Exposure Assessment

The model presented in Section 2 was applied in a probabilistic mode to the Great Lakes region using the probability distributions presented in Section 3. The Crystal Ball software (Decisioneering, 1992) was used to conduct a Monte Carlo simulation with 15,000 iterations. (The sensitivity of the probabilistic assessment was tested by comparing the results obtained with 15,000 and 20,000 iterations; no significant changes were observed between the two simulations.)

The results for methylHg concentrations in predator fish (i.e., largemouth bass, walleye or northern pike) are presented in Figure 4. The mean value is 0.46 ppm and the maximum value is 4.4 ppm. The 50th (median), 60th, 70th, 80th and 90th percentiles of the distribution are 0.36, 0.44, 0.54, 0.68 and 0.92 ppm, respectively. These results are commensurate with the observed methylHg concentrations in fish in Michigan lakes that were mentioned above. The maximum value modeled in the probabilistic analysis is greater than the maximum value obtained in lake-specific simulations because the former combines atmospheric deposition, hydrology and water quality variables conducive to high Hg fish concentrations. The results for the human dose simulation are presented in Figure 5. The distribution shows a median value of zero since it is assumed that 65% of the population does not eat fish caught in local water bodies. Therefore, according to our analysis, their incremental dose is zero. The mean value of the modeled dose is  $8.9 \times 10^{-6}$

mg/(kg•day). The 70th, 80th and 90th percentiles of the distribution are  $2.4 \times 10^{-6}$ ,  $1.0 \times 10^{-5}$  and  $2.6 \times 10^{-5}$  mg/(kg•day), respectively; the maximum value is  $5.7 \times 10^{-4}$  mg/(kg•day). It must be noted that uncertainties remain in the various steps of this exposure assessment model and that the results presented here must be seen as exploratory and primarily as a demonstration of a methodology of the regional assessment of Hg potential exposure.

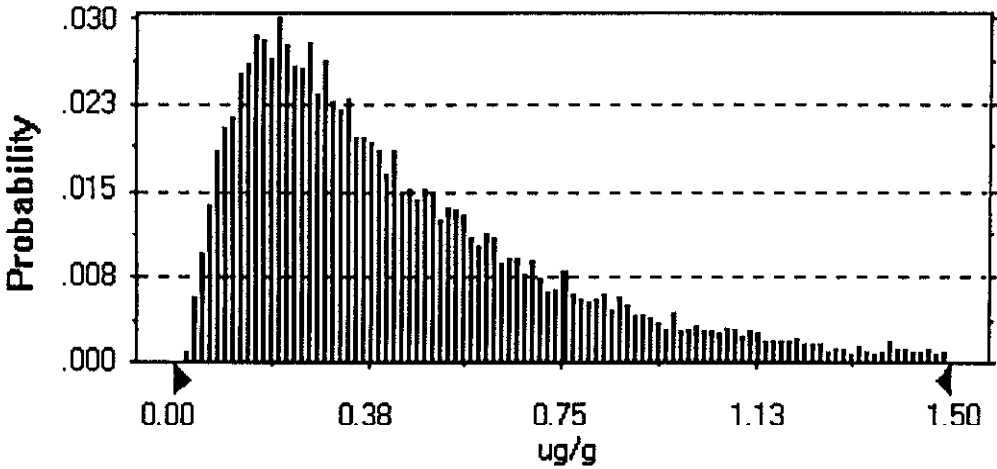


Fig. 4. Probability Distribution of the Modeled MethylHg Concentration in Fish.

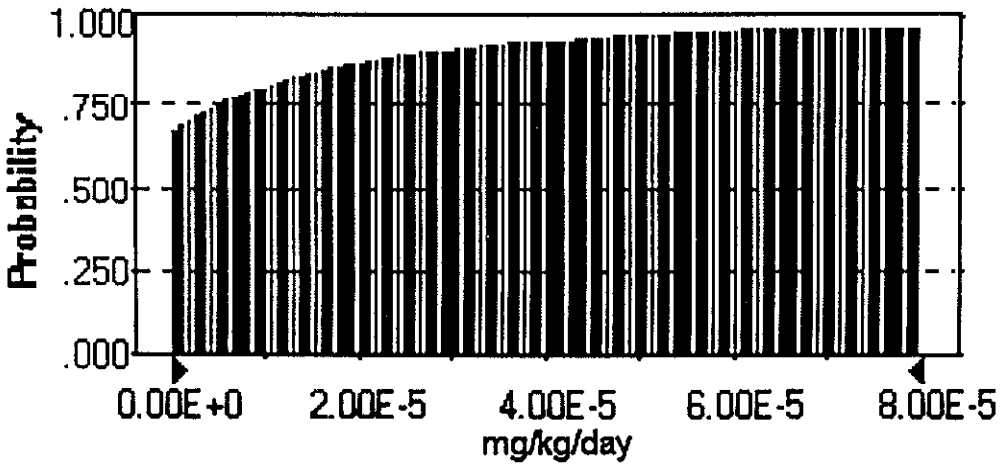


Fig. 5. Cumulative Distribution of the Modeled Human Dose due to Local Fish Consumption

### 5. Summary

A methodology for the health risk assessment of Hg at regional scales has been presented. The methodology combines atmospheric modeling at a continental scale, modeling of Hg cycling and bioaccumulation in lakes of a region within the continental domain, and modeling of Hg intake by the local population through fish consumption. Because of the spatial variability of many environmental characteristics and variability of exposure parameters among individuals, a probabilistic approach was selected. The methodology was demonstrated through its application to the Great Lakes region. Further work will focus on the following items: (1) The methodology will be extended to include uncertainties in the model input data in addition to the variability in the parameters, (2) the regional exposure assessment will be extended to other regions (e.g., the northeastern U.S. and Florida) where lake contamination by atmospheric Hg is of concern, and, (3) to the extent possible, R-MCM simulations of Hg concentrations in fish will be compared to available data for specific lakes in various regions of the U.S.

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