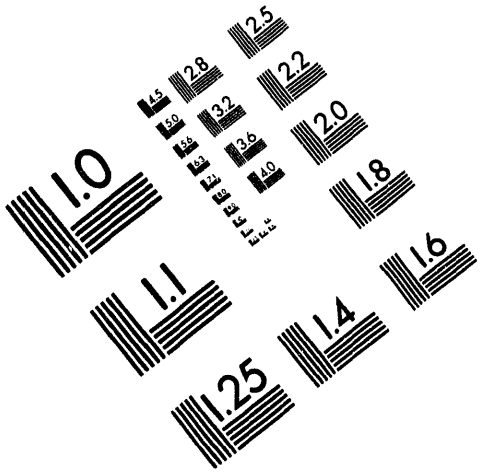

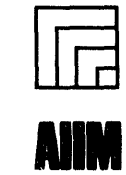

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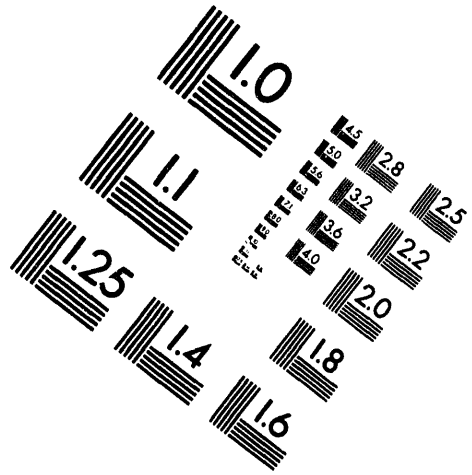

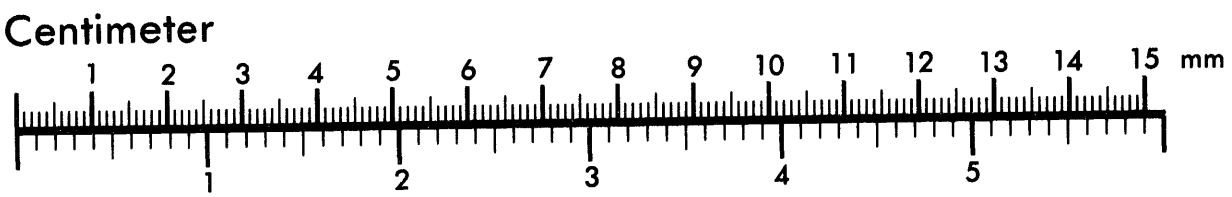

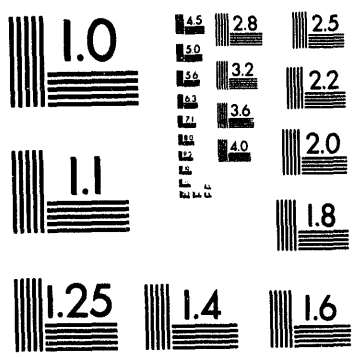

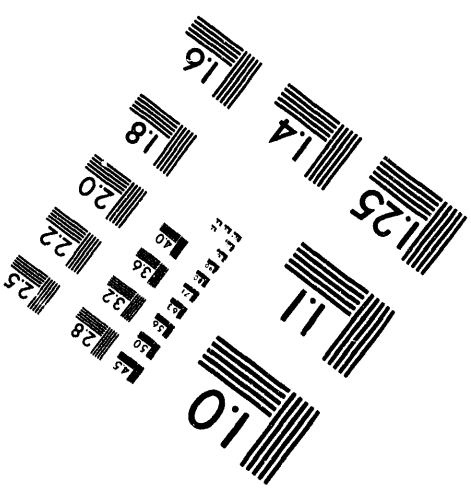

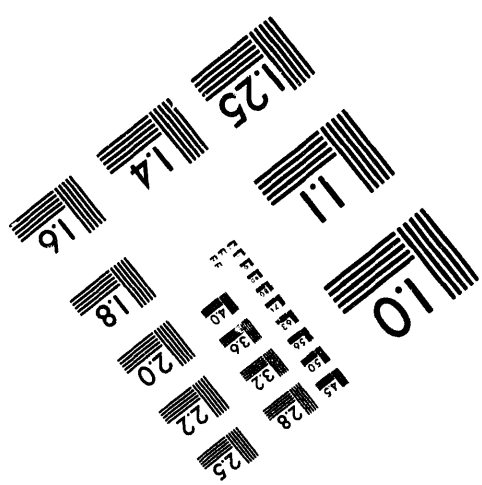

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BNL-60435

ASSESSMENT OF MERCURY HEALTH RISKS TO ADULTS FROM COAL COMBUSTION

F. Lipfert, P. Moskowitz, V. Fthenakis, M. DePhillips,

J. Viren and L. Saroff

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Biomedical and Environmental Assessment Group
Analytical Sciences Division
Department of Applied Science
Brookhaven National Laboratory
Upton, New York 11973

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ABSTRACT

The U.S. Environmental Protection Agency (EPA) is preparing, for the U.S. Congress, a report evaluating the need to regulate mercury (Hg) emissions from electric utilities. This study, to be completed in 1995, will have important health and economic implications. In support of these efforts, the U.S. Department of Energy, Office of Fossil Energy, sponsored a risk assessment project at Brookhaven National Laboratory (BNL) to evaluate methylmercury (MeHg) hazards independently. In the BNL study, health risks to adults resulting from Hg emissions from a hypothetical 1000 MW_e coal-fired power plant were estimated using probabilistic risk assessment techniques. The approach draws on the extant knowledge in each of the important steps in the calculation chain from emissions to health effects. Estimated results at key points in the chain were compared with actual measurements to help validate the modeled estimates. Two cases were considered: the baseline case (no local impacts), and the impact case (maximum local power-plant impact). The BNL study showed that the effects of emissions of a single power plant may double the background exposures to MeHg resulting from consuming fish obtained from a localized area near the power plant. Even at these more elevated exposure levels, the attributable incidence in mild neurological symptoms (paresthesia) was estimated to be quite small, especially when compared with the estimated background incidence in the population. For example, in a population of 10,000 heavy fish eaters, about one case of paresthesia due to fish consumption would be expected in the absence of a power plant, fewer than three cases with the plant, and about 220 cases due to all other causes. Many implicit and explicit sources of uncertainty exist in this analysis. Those that appear to be most in need of improvement include data on doses and responses for potentially sensitive subpopulations (e.g., fetal exposures). Rather than considering hypothetical situations, it would also be preferable to assess the risks associated with actual coal-fired power plants and the nearby sensitive water bodies and susceptible subpopulations. Finally, annual total Hg emissions from coal burning and from other anthropogenic sources are still uncertain; this makes it difficult to estimate the effects of U.S. coal burning on global Hg concentration levels, especially over the long term. MeHg is also suspected of contributing to delayed childhood development due to effects arising from the suspected sensitivity of the fetus to exposure during pregnancy. These suspected risks may play a critical role in the debate regarding the need to control Hg emissions and are now being examined by BNL and other organizations.

EXECUTIVE SUMMARY

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Emissions and Atmospheric Processes

The hypothetical power plant examined in this study was assumed to burn coal having the U.S. average content of Hg (0.08 ug/g). The emission controls (electrostatic precipitator) used at this plant only reduce the Hg emissions by about 10%. This results in an estimated total Hg emission rate of 180 kg/y from the hypothetical plant, comprising elemental, reactive, and particulate Hg. A Gaussian plume model was used to estimate the local annual average air concentrations of the three species and deposition rates were estimated for each within 50 km. In modeling deposition, Hg speciation was important because of the variations in water solubility of the different Hg species. Dry deposition was modeled by assuming a value for the dry deposition velocity, defined as the ratio of the deposited flux to the air concentration. Wet deposition was modeled in two different ways, using either the washout ratio (ratio of concentration in precipitation to air concentration) or a dynamic plume depletion algorithm. The modeling results suggest that Hg emissions from the plant could double the total local background deposition at the point of maximum impact, but that about 95% of the Hg emissions from a tall stack would travel beyond the 50 km radius. At this distance, the incremental Hg deposition from the plant would be of the order of 1% of background levels. Measurements of Hg in rain near an incinerator were used to validate the local deposition model.

Fish Consumption Rates and the Distribution of Baseline MeHg Doses

The U.S. population is exposed to MeHg principally through the consumption of fish. Fish consumption statistics from various sources were used to estimate the MeHg dose to the population. Surveys and national fishery statistics suggest that the overall average per capita fish consumption rate is about 25 g/d (about 1 meal per week), with a 95th percentile level of about 80 g/d. About 95% of the U.S. population consumes some fish over the course of a year. Mercury levels tend to be higher in freshwater sportfishing species; the BNL assessment selected the upper Midwest region for study because freshwater fish consumption tends to be higher there; the average daily (baseline) total MeHg dose was estimated to be about 4.6 μg/d. The 99th percentile was about 34 μg/d, which is only about 10% of the EPA reference dose that defines the safe limit for adults. Because of the large uncertainties remaining in the specification of aquatic methlyation processes, the estimated increase in freshwater fish MeHg due to Hg deposition from the hypothetical plant is assumed to be proportional to the change in local Hg deposition. Thus, the MeHg dose to the populations consuming locally-caught fish near the plant would increase in proportion to their freshwater fish consumption.

Metabolic Processes

In contrast to both carcinogenic and irritant air pollutants, the dose metric of concern for MeHg is neither the total accumulated dose nor the maximum acute level, but the equilibrium body burden that is attained as a balance between steady intake and excretion. Body burden not only controls health responses, but can be directly related to measurable levels of MeHg in blood and hair. Because a high body burden of MeHg can only be obtained by eating fish more often, the averaging process that takes place with respect to individual doses obtained from eating disparate meals over time is an important feature that must be included in a risk assessment. Monte Carlo simulations were used to develop an empirical model of this process. The resulting body burden estimates were then used to predict the baseline distribution of MeHg in blood and in hair, and these values were found to compare satisfactorily with the available measurements.

Dose-Response Functions

The central nervous system is the principal target for MeHg, with the potential for effects on sensory, visual, and auditory functions. Individuals may vary greatly in their responses. Paresthesia is perhaps the mildest symptom of MeHg poisoning, and response data on 122 Iraqi adults who consumed Hg contaminated bread in 1971-1972 were used to derive a continuous dose-response function. A logistic regression model that best fit the observations in the region of low dose (which is of primary interest here) was selected for use in this risk assessment. The uncertainties in the parameters of this function were seen to be the primary source of uncertainty in the overall assessment. The implications of using a threshold-based or deterministic hockey-stick function were also explored.

Assessment of Baseline and Incremental Risks from a 1000 MW_e Plant

Using a probabilistic approach, the human uptake of MeHg from fish consumption was estimated for baseline and power-plant impact cases. The effects of power plant emissions on marine species were assumed to be negligible, since these species are primarily affected by global levels of Hg, and the U.S. utility industry contributes less than 1% of the existing global pool each year. The risk analysis was performed for several different assumptions involved in the metabolic model and predicted a baseline average risk level of about 0.002-0.003% (2-3 chances in 100,000), with a 95th percentile risk range of 0.006-0.012%. When power plant increments were added, the expected average risk level increased to 0.004-0.007% with an upper 95th percentile risk of 0.013-0.027%. If a deterministic hockey-stick dose-response model had been used instead of the logistic model, the incremental frequency of paresthesia would have had about a 99% chance of being zero.

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1.0 INTRODUCTION

1.1 Background Information on the Health Effects of Mercury

Mercury has been feared and respected among the elements since antiquity. Based on their experiments with destructive heating and distillation, the Greeks believed that metals were composed of only two elements: sulfur and mercury. Sulfur was identified as the "soul" of the metal and mercury was its intelligence (Salzburg, 1991). This classification now seems ironic in terms of modern society's priorities for environmental control, in which these two elements seem to weigh so heavily.

Mercury occupies a special place in the hierarchy of environmental pollutants. Unlike many of the "air toxics," adverse health effects from Hg have been convincingly demonstrated in the past, because of several unfortunate poisoning incidents. Although mercury is distributed throughout the environment as an air pollutant, its toxic effects result only from ingestion of methylmercury (MeHg), which is defined as the CH_3Hg^+ ion in whatever form it appears. MeHg is normally only found in seafood, as a result of bioaccumulation processes; however, there have been isolated incidents of human consumption of foodstuffs contaminated by mercury fungicides. Unlike most other air pollutants, there are few permanent sinks for mercury in the environment, and emissions from a given source may eventually be spread around the planet. As a result, adverse effects must be considered on scales ranging from local to global.

For the purposes of assessment of health effects, it is important to divide the mercury compounds found in the environment into two groups: inorganic Hg, primarily elemental mercury vapor or Hg^0, and organic mercury, primarily methylmercury, MeHg. The toxicity of mercury has been known for millennia, mainly as poisoning from inorganic Hg from occupational exposures (the "mad hatter" syndrome). The toxicity of organic mercury was demonstrated more recently in humans by poisoning events in Japan due to consuming fish from Minamata Bay (WHO, 1990) and in Iraq from eating seed grain which had been coated with MeHg (Bakir et al., 1973). The effects of organic mercury poisoning are neurotoxic due to accumulation in brain tissues and range from a slight numbness in the extremities (paresthesia) to death. Organic mercury (i.e., MeHg) readily passes the placenta, leading to concerns that maternal exposure may lead to neurological problems in offspring (Clarkson, 1993).

The extreme variability typical of responses to organic mercury poisoning was shown in an unfortunate incident in New Mexico in 1969 (Pierce et al., 1972). Over a 14-week period, 7 family members ate pork from an animal that had been fed contaminated seed grain. Although they all were reported to have eaten approximately equal amounts, only 3 members of the family (ages 8, 13, and 20) became seriously ill. In addition, a child that was *in utero* during this period was born with severe neurological difficulties. The three children were stricken in inverse order of age: the youngest remained permanently blind and unable to converse; the next recovered to a state of some self-sufficiency, but with vision capable of light perception only, and the oldest recovered to the point where she only suffered from constriction of visual field. The other family members "remained asymptomatic," although the mother was reported to have had slight slurring of speech during a 2-week period. Given this extreme variability in response to even very high doses of MeHg, the need for a probabilistic approach to health risk assessment seems obvious.

1.2 Environmental Regulations for Mercury

Mercury was originally designated as a hazardous air pollutant by the U.S. Environmental Protection Agency (EPA) in 1971 (36 FR 5931; March 31, 1971). It was subsequently included as one of eight substances listed in the National Emission Standard For Hazardous Air Pollutants (NESHAP) provisions (40 CFR 61).

In 1975, standards were set for stationary sources which process mercury ore to recover mercury, use mercury chlor-alkali cells to produce chlorine gas and alkali metal hydroxide, or incinerate or dry sludge from wastewater treatment plants. Emissions to the atmosphere from mercury ore processing facilities and mercury cell chlor-alkali plants were limited to 2300 grams of mercury per 24-hour period. Emissions from sludge incineration plants, sludge drying plants or a combination of these that process wastewater treatment plant sludges were limited to 3200 grams of mercury per 24-hour period (40 FR 48302, Oct. 14, 1975). Owners and operators were required to

perform emission testing and sludge sampling. Specific testing and sampling methods and reporting requirements were finalized in 1987 (52 FR 8727 March 19, 1987).

The 1990 Amendments to the Clean Air Act may eventually supplant NESHAPS, by requiring percentage reductions for 189 listed Hazardous Air Pollutants from identified source categories. Title III of the Amendments (the Act) lists mercury compounds as one of the identified Hazardous Air Pollutants. The section "Specific Pollutants" [(c)(6)] directs the EPA to list sources that account for 90% of the aggregate mercury emissions to air which are subject to standards, by 1995. Such standards are to be promulgated by 2000. This provision, however, does not include air pollutants emitted by electric utility steam generating units.

Consequently, to date there is no regulation of mercury emissions from electric utilities, including clean coal technology plants. Section (n) of Title III, "Other Provisions," directs the Administrator to conduct three studies which will assist the EPA in determining whether toxic air pollution regulations are needed, as well as providing enough data to determine appropriate standards for utilities.

Two of the three studies deal with mercury:

> "The Administrator shall conduct...a study of mercury emissions from the electric utility steam generating industry..." This report is to be submitted to Congress by November 1994.
> "The National Institute of Environmental Health Sciences shall conduct...a study to determine the threshold level of mercury exposure below which adverse human health effects are not expected to occur." A draft of this report is currently undergoing agency review.

It is difficult to make any prediction as to the outcome of these studies because of the ambiguity of the Act, which does not define the scope of these studies nor how they are to be performed. Further, it does not identify any analytical protocols for analyzing the emissions, nor does it identify an "acceptable" level of risk. However, it can be assumed that, because of the large amounts of data being assembled for the studies, if standards are proposed, they will be promulgated relatively quickly. In contrast to the source points covered in section (c)(6), there will likely not be a five-year period between identification and promulgation of standards.

Section 129(a)4 of the Act calls for the EPA to specify numerical limitations for mercury emissions from Municipal Solid Waste incinerators. Although imminent, there is no set standard to date. There is, however, a draft standard (EPA Docket #A90 45 II-F-1) which states that incinerators must achieve either 80% reduction in mercury emissions or a discharge Hg concentration less than 100 μg/m3 at 7% O_2. To place the latter emission limit in the context of coal combustion, consider that the flue gas concentration of Hg from firing coal containing 0.08 ppm of Hg is about 8-10 $\mu g/m^3$. However, typical power plants operate at much higher firing rates than incinerators, so that the actual mass emission rates may be more comparable.

1.3 Outline of This Report

The DOE studies of toxic air emissions from coal combustion are intended to provide support for the forthcoming regulatory agenda. This report includes the following sections. First, the extant assessments of MeHg in the environment are summarized and critiqued, after which the framework of this assessment of the effects of coal combustion is laid out. Data on emissions from power plants are then reviewed and the types of clean coal technologies that should prove effective in reducing Hg emissions are discussed. Considerable effort was devoted towards developing an empirical understanding of the factors that affect (existing) mercury levels in freshwater fish, since these are the species most likely to be impacted by local coal combustion. Similarly, a variety of sources of data on rates of seafood consumption was examined, in order to derive a realistic statistical distribution of MeHg doses. Those doses are converted to body burdens using an empirical model that accounts for the averaging process, and a logistical dose-response function is developed that uses body burden as input. The risk assessment results are reviewed in the context of background neurological symptom levels, and overall conclusions from this phase of the work are presented and out standing research needs are discussed.

This report has evolved over a 1-year period as analytical techniques and the available data bases have improved. The most recent information available has been incorporated, accepting the uncertainties that may result from lack of complete validation of the new data. For example, reviewers have expressed reservations about the speciation of mercury emissions, about the definitions of "anthropogenic" and "natural" sources in the context of historical increases in the Hg content of biospheric reservoirs, about the need for mesoscale transport and deposition models, and concerning the effects of a possible relationship between fish consumption and body mass (Moskowitz et al., 1994). Readers are therefore urged to use this report as a step along the way towards a more complete understanding of mercury health risks and to monitor the emerging technical literature for further developments.

2.0 PREVIOUS MERCURY ASSESSMENTS

Previous assessments of the health risks of mercury have been of two general types: assessment of the baseline risks due to all sources of atmospheric mercury, and assessment of the incremental effects of emissions from specific sources or types of sources. As discussed below, a good understanding of the baseline is a prerequisite for considering the incremental risks. Virtually all parties agree that the *average* health risks from MeHg are trivial; what may be at issue are the estimates of *extreme* values, which must be derived from probabilistic considerations. Probabilistic methods have come into use for such purposes only relatively recently. Some of the previous assessment efforts have been rather simplistic when viewed in that light, but are included in this section for completeness and perspective.

2.1 Assessment Methods

There are perhaps two basic methods of risk assessment that have been used in these studies. The more comprehensive analyses compare methylmercury doses to those leading to actual observed neurological responses or to mathematical models of those responses. The major poisoning incidents mentioned above are often the source of such dose-response data. The other analyses rely on dose thresholds or "no observed adverse effect" levels (NOAELs) that have been established by various regulatory or advisory agencies, such as the Food and Drug Administration (FDA), the EPA, or the World Health Organization (WHO). In all cases, it is necessary to estimate the distribution of MeHg doses to the various populations being considered. In most general populations, the average doses of MeHg are well below the steady-state levels at which neurological responses might be expected, so that the adequacy of the assessment depends on the methods by which extreme cases are considered and the levels of realism incorporated in the assumptions used.

2.2 Assessments of Baseline Risks

2.2.1 The National Academy of Sciences (NAS) Study, "Seafood Safety." The Institute of Medicine of the NAS prepared a comprehensive account of many of the risks involved in eating seafood, including natural toxins, organic pollutants, and trace metals including mercury (Ahmed, 1991). Their methodology included individual variability in MeHg uptake and metabolic half-life and was based on mathematical models of the responses to the Iraqi grain poisoning incident (Marsh et al., 1987; Bakir et al., 1973). The objective of this analysis was to estimate the actual risks entailed at the regulatory thresholds most often used, with and without a safety factor of 10. Although the report presented data on seafood consumption and mercury contamination levels, it did not go on to combine the two distributions in order to estimate the actual population risks. The report concluded, nevertheless "risks that may be significant include reproductive effects from polychlorinated biphenyls (PCBs) and methylmercury. and, possibly, parkinsonism in the elderly from long-term mercury exposure."

2.2.2 The 1986 FDA Analysis. The NAS study described above criticized a previous FDA effort by Tollefson and Cordle (1986) for its reliance on acceptable daily intake levels (ADIs) rather than on dose-response functions, and on its failure to consider a wide range of fish species (the latter criticism seems unjustified since consumption and mercury data were provided for 6 species and mercury data for another 20 or so). Tollefson and Cordle concluded "The majority of fish consumers in the United States could easily double their intake and still remain below the ADI." They went on to defend the FDA action level of 1 μg/g Hg in fish as providing adequate protection for adults and children.

2.2.3 The 1993 FDA Risk Assessment. The FDA (C. Carrington, personal communication, August 1993) is developing a spreadsheet-based probabilistic model to assess the health risks from mercury in tuna fish. This model samples from discrete distributions of mercury in tuna and tuna consumption levels, develops equilibrium blood Hg levels for a number of individuals, adds background blood Hg levels, and then evaluates their risks based on the Iraqi poisoning data. Only fragmentary information about this model is available, but it appears that the fish consumption data are based on only the tuna-consuming portion of the population, with a mean consumption rate of 31 g/d. According to the data in Section 7.0, this value seems inordinately high. The endpoint in question here is the transfer of effects of maternal consumption of MeHg to fetal and child development. Details and results from this model have not yet been published.

2.2.4 National Institute of Environmental Health Sciences (NIEHS). NIEHS was requested by Congress to "....determine the threshold level of mercury exposure below which adverse human health effects are not expected to occur." A preliminary draft copy of this report (J. Fouts, personal communication, 1993) was obtained; it discusses the available dose-response information and provides some fish consumption data and exposure estimates for methylmercury. The report evaluates the available data on fish consumption and their uncertainties, including subgroups of high fish consumers, but makes no estimates of population exposures to MeHg. To satisfy the Congressional mandate, the NIEHS report cites the doses or fish concentrations that have been deemed "tolerable" by the FDA, EPA, and WHO.

2.2.5 The New Jersey Baseline Risk Assessment. The most recent general assessment of baseline risks is that of Stern (1993), who used the data on Iraqi mothers and children, as analyzed by Marsh et al. (1987) to estimate the risks entailed in the current U.S. EPA reference dose (0.3 μg/day per kg of body weight). Stern also discusses other relevant epidemiological studies of MeHg and neurological symptoms. He concluded that the reference dose should be reduced to 0.07 μg/kg/day to preclude effects on fetal development. It was not clear from this paper whether this was also an official position of the New Jersey Department of Environmental Protection and Energy.

The assumptions and input data used by Stern could be considered a point of departure for the present assessment. Data on mercury content of various marine species were taken from the 1978 survey by Hall et al. and combined with consumption data from a nationwide survey from 1982-87, in a probabilistic analysis. The average mercury concentration, weighted by percentage of the catch, was 0.11 μg/g, with a maximum of 1.0. This algorithm implicitly assumes that the variability in the MeHg concentration of the fish eaten in various amounts comes from variations by species, rather than variations within species due to age, weight, length, etc., and ignores typical consumption patterns involving a mixture of species. The maximum consumption rate considered was 227 g/d (0.5 lb/d, 7 days a week), with an average of 32 g/d (slightly more than 1 meal per week), based on an unpublished FDA document. These consumption rates pertain only to that segment of the population that consumes fish on a regular basis, which Stern estimated to be about 85% (based on surveys). On this basis, he estimated that 3% of the (fish-eating) 70 kg adults exceeded the present EPA reference dose, and that 23% of the 62 kg females exceeded the recommended lower standard for fetal effects.

2.3 Risk Assessments for Mercury Sources

2.3.1 New Jersey Municipal Solid Waste Incinerators. In 1992, the New Jersey Task Force on Mercury Emissions Standard Setting released an analysis of six specific incinerators for which mercury emission rates had been determined. Their method of analysis was to predict downwind air concentrations using standard dispersion models, estimate wet and dry deposition rates and run-off, and then to estimate rates of Hg accumulation in aquatic sediments and subsequent bioaccumulation in fish. In order to estimate the incremental MeHg dose, estimates were made of the amount of locally-caught fish consumed, which was taken as equal to the national average for all types of fish. The consumption value used (32 g/d) was obtained from an internal FDA publication. These doses were then compared to the reference dose. Such an assessment approach does not actually estimate health risks *per se*, since dose-response information underlying the reference dose levels is not considered. This report also includes the baseline risk estimates published by Stern (1993), which were discussed above, in order to provide an estimate of the MeHg background upon which the incremental risk is imposed.

The New Jersey analysis used the maximum annual average air concentrations from each incinerator as a starting point, and applied these values to hypothetical lakes, assuming that all of the mercury was emitted as soluble $HgCl_2$. These concentration increments were about the same as the existing ambient mercury background (see Section 5.8 for a discussion of actual measurements of Hg wet deposition near a New Jersey incinerator). This mercury was assumed to be deposited wet and dry into the watershed, where it accumulated in lake sediments for 20 years. The bioconcentration factor used was based on sediment Hg concentrations and was chosen to be representative of New Jersey lakes, but no consideration was given to the implied fish species involved and their actual rates of consumption by humans (the basic data were obtained from northern Minnesota lakes). The resulting fish MeHg concentrations due to the incremental effects of incinerators ranged from 0.7 to 2.68 mg/kg, while the baseline concentrations due to all other mercury sources averaged only 0.11 mg/kg, with a maximum of 1.0 mg/kg. As discussed in Section 5.8, point sources with modest stack heights may result in local hot spots of Hg deposition; however, for site-specific assessments such as this, the existence of water bodies providing edible fish must be shown, and Hg deposition should be averaged over their entire watershed areas.

Consumption rates of these locally-caught fish were assumed to be the same as the U.S. average for all types of fish, adjusted downward to reflect absence of fishing during the winter, to yield a figure of 24 g/d/person. This corresponds roughly to one fishing trip per week over the 9 remaining months of the year, based on New York State's survey of anglers (Pat Festa, personal communication, August 1993). The overall conclusions of the New Jersey assessment were that the current EPA reference dose should be reduced, and that emissions from existing incinerators may cause "significant increase in the ingestion of methylmercury."

2.3.2 Power Plant Risk Analysis. Constantinou et al. (1993) presented a power plant risk analysis for mercury with both deterministic and probabilistic elements. They considered a range of exposure pathways, including inhalation, drinking water, and ingestion of plants and fish. Ingestion of fish was found to be the most important pathway in their deterministic (worst case) analysis. The endpoint of this analysis was the fraction of the reference dose implied by the incremental effects of a single power plant, which they identified as the "hazard index." Inorganic and organic mercury hazards were combined in this analysis. The modeling inputs for the deterministic analysis featured "conservative" assumptions for the point estimates that might be suitable for a regulatory perspective. The probabilistic analysis used distributions of parameters from the literature, which may or may nor encompass the deterministic point estimates (personal communication, E. Constantinou, April 1994). The assumptions of the two analyses are compared in Table 2.1. The power plant was assumed to have emissions controls, and the modeling incorporated the actual positions of lakes within its near-field impact area.

Table 2.1 Modeling Inputs for the Analysis of Constantinou et al. (1993)

Parameter	Deterministic Value	Probabilistic Values
Hg emissions	1.4 mg/s	1.4 mg/s
Hg species	Hg^{++}(particulate)	Hg^0
Hg reactions	none	1st order rate
Hg(gas) scav.	none	2.4 E-4
Hg(part) scav.	4.3 E-4	7.2 E-4, GSD*=0.67
Hg land run-off	98%	85-95%
Hg methylated	100%	1-10%
fish bioconc.	33,000	4675, GSD=0.85
fish consumption rate	37 g/d	2.5 g/d, GSD=1.99

*GSD = geometric standard deviation; see Appendix A.

For the deterministic analysis, the total hazard index due to the plant was estimated to be about 0.09; for the probabilistic analysis, the expected (i.e., mean) value was 0.0014 with a GSD of about 4.5. The deterministic value fell at about the 99th percentile of the probabilistic dose distribution. This analysis provides a good example of the expansion of uncertainty that can occur in a chain calculation involving many individually uncertain inputs.

2.3.3 New York State Methodology for Estimating Externalities. Rowe et al. (1993) proposed a methodology for estimating health effects of coal combustion, for application to New York Power plants. The Hg emission rates cited are about twice the national average, according to the most recent EPRI data (D. Porcella, personal communication, 1993). They assume all Hg is emitted in soluble form ($HgCl_2$). All deposition is concentrated in one catchment area (watershed). Much of the analytical methodology follows the New Jersey model described above for incinerators. The mechanism for making Hg available for bioconcentration by fish is through uptake by sediments during the plant's lifetime (60 y), for release to the water column. The bioconcentration factors (BCF) used are keyed to sediment Hg concentrations. A single BCF is used for all fish species; no account is taken of chemical or geomorphic factors associated with the lake or watershed. The model considers background levels of MeHg in fish and in humans, but assumes a national average seafood consumption rate of 32 g/d (no source cited). A parametric approach was taken to the ingestion of locally-impacted freshwater fish: 1 meal/mo, 4 meals/mo, and 10 meals/mo. Data from the Iraqi grain poisoning accident were used to develop dose-response functions for adult paresthesia and retardation in children through placental transfer. In addition, the EPA "lowest observed adverse effect levels" (LOAELs) were used to set thresholds of no effect.

This model is applied to 3 specific New York lakes, and it is noted that small lakes may concentrate the Hg emissions to a greater extent, but that the population exposed to the fish in such a lake will be small, a compensating factor. All told, this is probably the most thorough investigation of power plant impacts, but it would have been greatly improved by the application of probabilistic techniques.

2.4 Summary of Previous Mercury Risk Assessments

The extant assessments of mercury risks are based on data assembled from a wide range of sources and disciplines; a common methodology has not yet evolved. With the benefit of hindsight, it appears that improved accuracy and relevance would result from incorporating the following features:

1. Use of probabilistic rather than deterministic methods.
2. Disaggregation of the total seafood diet into separate components for which distributions of mercury contents and rates of consumption can be estimated.
3. Identifying particular dietary components that might be sensitive to power plant impacts.
4. Considering equilibrium rather than instantaneous blood levels of Hg.
5. Incorporating the dose-response functions for the endpoints of interest directly into the assessment, including the associated uncertainties.
6. Using area-averaged deposition to a water body.

3.0 FRAMEWORK OF THE PRESENT ASSESSMENT AND OVERVIEW OF INPUTS AND ASSUMPTIONS

This assessment has been designed to take maximum advantage of the available experimental observations, as a means of reducing the degree to which worst-case assumptions may enter. In this phase of the assessment, paresthesia in adults is used as the health endpoint of concern. A subsequent assessment phase could then substitute maternal-fetal effects into the basic framework. The basic elements are summarized in Table 3.1.

3.1 Source Characteristics

The study is based on a hypothetical power plant, as a paradigm. Extension to the U.S. population of all U.S. power plants is thus only a matter of acquiring the necessary data on source and receptor characteristics. The fraction of mercury emitted as Hg^{++} is assumed to depend mainly on the Cl^- content of the coal. The presence of flue-gas clean-up devices is an option. The plant is assumed to have a nameplate rating of 1000 MW_e, a capacity factor of 80%, and a stack height of 200 m, and to be located in the upper midwest in reasonably level terrain in the vicinity of sport fishing lakes. Average mercury content in the coal is assumed (0.08 ppm). Statistical distributions of these deterministic parameters may be added later as appropriate data become available.

Table 3.1 Features of the LNL Mercury Assessment Model

Mercury emissions	Fraction of Hg emitted as Hg^{++} is estimated based on Cl^- content of coal. Effects of air pollution controls are estimated.
Transport, dispersion, and deposition	Gaussian plume transport and dispersion model for three Hg species for distances up to 50 km. Constant values of deposition velocity and washout ratio for each class of Hg compounds. No atmospheric chemical reactions.
Accumulation in surface waters	All Hg deposited within the watershed is assumed to enter the water body. Incremental wet+dry deposition is estimated.
Contributions to global background	Any Hg not deposited within 50 km is assumed to enter global pool. Global deposition is increased in proportion to (net) emissions.
Effects of local sources on fish Hg content	In the absence of better information, the average Hg in each fish species is assumed to increase in proportion to the Hg deposition increment (local plus global).
Background Hg dose from seafood	The dose distribution is calculated by using probabilistic methods to sum (log normal) distributions of the product of Hg concentration and consumption rate for fresh-water and marine species and for canned tuna.
Equilibrium metabolic model	The equilibrium level of the body burden of MeHg is estimated by considering the frequency of the three different types of fish meals, in addition to the total MeHg dose. The distribution of body burden is estimated using probabilistic methods, from distributions of Hg dose, body mass, and half life of Hg, as a baseline and with power plant contributions to MeHg in freshwater fin fish .
Dose-response functions and risk analysis	Distributions of the parameters of a continuous dose-response model are developed from the Iraqi paresthesia data and used to estimate levels of risk for the baseline case and for the incremental effects of a 1000 MW_e hypothetical power plant.

3.2 Dispersion and Deposition of Airborne Hg

Results from a standard EPA dispersion model are used to estimate annual air concentrations up to 50 km downwind. A pattern of rainfall frequency by wind direction is assumed and wet and dry deposition rates of mercury are estimated, based on washout ratios and dry deposition velocities. Mercury not deposited within 50 km is assumed to contribute to the global background. This calculation is based on a mass balance of annual mercury input from coal, the removal characteristics of control devices, and the depletion of the plume by wet and dry deposition. Extension to downwind distances greater than 50 km is only a matter of applicability of the dispersion model.

3.3 Receptor Characteristics and Pathways

Fish ingestion is assumed to be the only important pathway for exposure to MeHg (Gunderson, 1988). The watershed drainage area is assumed to be twice the lake area, for the purpose of estimating mercury inputs. The basis for estimating changes in fish MeHg content due to the power plant is by means of the incremental mercury deposition ratio: power plant contributions in relation to (global) background. Typical measurements are used to estimate background levels. These calculations are carried out by major categories of fish species, in order to preclude the need to estimate bioconcentration factors for each species. Thus far in the calculation chain, all data are deterministically based on single-point estimates; probabilistic methods could be applied to this part of the analysis as data become available. This approach allows identification of the separate uncertainty contributions of source and receptor characteristics in relation to those of the dose and response characteristics.

3.4 MeHg Dose Distributions

The (adult) population at risk is assumed to consume a mixture of four types of seafood: fresh and/or frozen marine species, shellfish, canned fish, and locally caught freshwater species. Only the last category is assumed to be directly affected by the power plant in question; the other three categories are assumed to be affected only by increases in global Hg background resulting from the Hg not deposited locally. Survey data on fish consumption are used to generate these probability distributions. Survey data on mercury in fish are also used, and the product of these two distributions defines the distributions of average (steady-state) daily mercury dose to the population at risk accrued over time. Both the baseline doses and the incremental doses from the power plant are considered. Their relationship with EPA and WHO reference doses is noted, but such comparisons are incidental to the framework of the risk assessment.

3.5 Health Responses

The response to ingestion of MeHg is an increase in blood MeHg, which in turn affects central nervous system function, since MeHg passes the blood-brain barrier. The rate of transfer from diet to blood is a dynamic process which reaches steady state in a matter of months, given a steady intake (ingested Hg not retained in the body is excreted and re-enters the biosphere). The parameters of this process are body mass, fraction of body weight as blood, and the half-life of mercury in blood. Experimental data on all three parameters are used to generate a distribution of metabolic transfer functions. Since the model is based on the cumulative response to methylmercury over a period of several half-lives, it is necessary to estimate the statistical properties of the dose integrated over this time, as opposed to the distribution of individual meals. Those individuals eating fish more often will tend to have less statistical variation than those eating fish only rarely, because of the averaging process.

The product of long-term dietary intake and transfer function distributions yields a distribution of MeHg concentrations in blood. This distribution in turn drives dose-response functions for adult paresthesia, which have their own uncertainties. These uncertainties include the choice of the functional form of the model (continuous risk vs. threshold response functions), background paresthesia levels, and the statistical uncertainties in each of the models. The resulting distributions of risks are then fed back to the source characteristics module to consider the need for (additional) source controls on mercury emissions.

3.6 Probabilistic Risk Assessment Methods

Traditional (i.e., deterministic) risk assessments combine a series of average, conservative, or worst case point estimates to derive a combined point estimate of the overall risk (here "conservative" is taken to mean biased in the direction of higher risk). As noted by Burmaster et al. (1990), there are several major limitations with this procedure. First, when point estimates are used, risk managers have no way of knowing the probabilities associated with the worst-case risk estimates. Second, the results can be biased by artificially assigning a high degree of conservatism to key parameters. Third, traditional sensitivity analyses may be misleading when key parameters are at or near their existing maximum values. For these reasons, the traditional risk assessments are useful only when the estimated risks fall below a *de minimus* value. When the risks exceed the *de minimus* value, the outputs from most deterministic risk assessments are of little value.

For these reasons, in this assessment, estimates of risk were prepared using probabilistic sampling techniques. In a probabilistic analysis, each of the many parameters within the equations that comprise the risk model is defined by a probability distribution function (PDF) instead of by single point estimates. The PDF is an explicit reflection of the full range of possible values combined with some measure of the probability of occurrence for each parameter value. The analysis proceeds by sampling independently from each PDF and carrying each result or "realization" through the calculation chain until a distribution of output values is derived. This distribution provides an "expected" value, defined as the average of all the realizations, and a number of other statistics, such as median (50th percentile), 95th and 99th percentiles. Note that the maximum risk developed from such probabilistic methods is an unstable statistic which can vary greatly from trial to trial; furthermore, the maximum predicted risk will increase with the number of realizations in each trial. Even if the input distributions are well-defined, such as normal (Gaussian) or log normal (see Appendix A), the output distributions are not limited to such prescribed definitions. They may be distributions best described as log normal with a very long tail. The shapes of the output distributions relate in part to the algorithms used to calculate risk and to the inclusion of multiplicative or nonlinear equations. The number of successive realizations should be adequate to describe the input parameter distributions; most of the results in this assessment were based on 5000 trials.

The computational package used in this assessment (@RISK) allows a choice of either "Latin Hypercube" or "Monte Carlo" sampling techniques, which are basically similar. They differ principally in their sampling strategy. Latin Hypercube sampling techniques stratify the input PDFs in order to emphasize the most likely values, while Monte Carlo sampling uses completely random sampling. Because of the stratified inputs, Latin Hypercube sampling techniques can be more computationally efficient than the better-known Monte Carlo approach.

The calculated PDFs give the risk manager an explicit description of the uncertainty surrounding the risk estimate. On the basis of these PDFs, it is possible for the decision maker to make explicit judgments about the degree of conservatism to be included in the final decisions. This contrasts with the alternate approach, where the degree of conservatism is implicitly built into the model results by the modeler, is not explicitly displayed, and may not be understood by the decision-maker.

For this assessment, three different types of risk estimates are considered for the endpoints in question (initially, adult paresthesia). First, the expected individual risk, averaged over the entire population being considered. For the baseline case, this is the entire U.S. population. When the effect of the hypothetical power plant is considered, this statistic is interpreted as applying to the population that consumes fish from the affected water body. Next, the "maximally exposed" individual is considered, taken as the 95th or 99th percentile of the distribution of exposed individuals. This estimate may be interpreted as applying to a subsistence fisherman. Finally, the number of additional cases of paresthesia to be expected in the exposed populations at risk is estimated. For this latter risk estimate, it is important to have reliable estimates of the actual population densities to be expected in areas with important recreational fishing activities.

4.0 EMISSIONS OF MERCURY FROM POWER PLANTS

In this section, background information on mercury emissions from coal-fired power plants is presented. Total mercury emissions depend on the coal mercury content and the types of emissions control equipment; Chu et al. (1993) indicate that recently measured Hg emissions are in the range 4-8 $lb/10^{12}$ Btu, roughly corresponding to Hg contents of 0.05-0.10 ppm. Earlier data on mercury in coal had indicated a higher mean and a much larger range of values (0.04-0.34 ppm [Obermiller et al., 1991]). The changes appear to be due to improved methods of sampling and analysis. The chemical form of the mercury emitted appears to depend on the chloride content of the coal; in addition, some mercury compounds are removed more efficiently by control devices than others.

Based on 1992 coal consumption of 895 million tons, of which 780 million tons is consumed by utilities (EIA, 1993), U.S mercury emissions from coal burning would be estimated at 72 tons, while an estimate based on previous data would be about 140 tons. In comparison, the municipal solid waste industry estimates its annual emissions of Hg at 44 tons; the mercury content of the total U.S. solid waste stream was estimated at about 360 tons for 1989; not all of this waste was combusted, however (ICF Kaiser Engineers, 1992).

4.1 Mercury Species of Interest

Almost all of the mercury in flue gas is in the gaseous state (Lindberg, 1980). However, recent studies indicate that 50% to 90% of the mercury in flue gas from coal firing may be oxidized (Bloom et al., 1993; Chu et al., 1993); most of these oxidized species are probably also gaseous. The conversion of elemental to oxidized Hg depends on the interplay of several factors, including temperature of the combustion zone, residence times and temperatures in heat exchangers, and the chloride content (Cl^-) in the coal. According to Felsvang et al. (1993), the Cl^- content appears to have a strong influence in converting Hg^0 to Hg^{++} (Figure 4.1). As shown in this figure, in four of the five plants using coal with Cl^- content above 0.02%, the fraction of oxidized mercury was 0.70 or more. The estimated range of Cl^- in U.S. coals is 0.04-0.2% (C. Schmidt, personal communication, April, 1994), so that it is expected that a large fraction of the emitted mercury will be reactive.

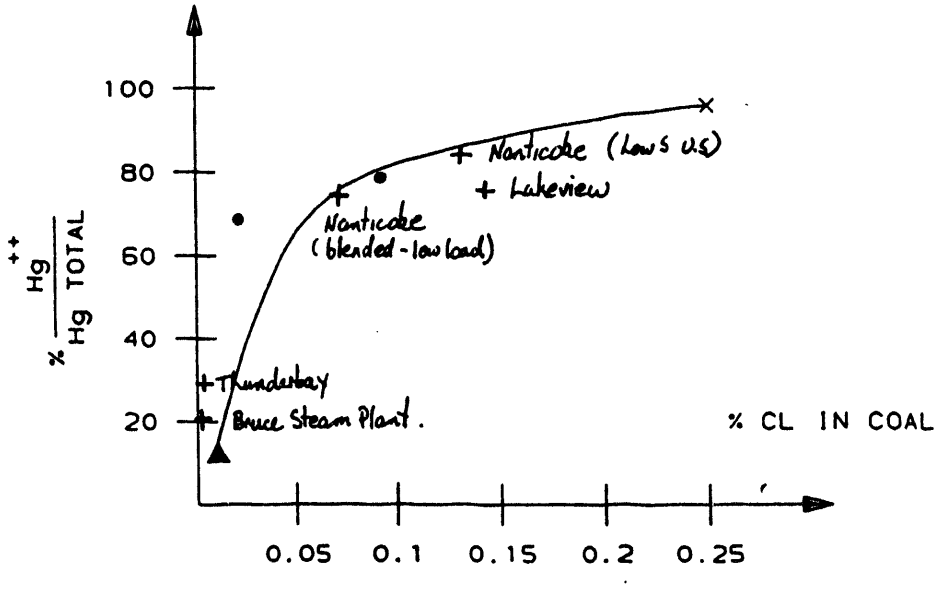


Figure 4.1. Data on the fraction of total emitted Hg measured as reactive Hg, as a function of fuel chloride content. Graph from Felsvang et al.(1993); additional data (+ symbols) from Ontario Hydro (O. Melo, personal communication).

4.2 Mercury Emissions Control

As discussed above, most of the Hg in coal is vaporized, while about 10% is associated with the fly ash. The latter can be removed by particulate control equipment, whereas removal of gaseous mercury requires additional control equipment. The gaseous species include both elemental and oxidized forms. More than 90% of the oxidized form of Hg, primarily $HgCl_2$, is likely to be removed by conventional wet and dry (e.g., activated carbon) scrubbing processes, while very little of the elemental Hg will be removed by scrubbing, because of its low solubility. Accurate estimates of Hg removal effectiveness are difficult to make, because the low Hg concentrations (0.1-10 $\mu g/m^3$) typically encountered in flue gases present a difficult challenge for sampling and detection methods. All of today's U.S. coal-burning power plants have some kind of particulate control device, usually electrostatic precipitators (ESPs).

4.2.1 Electrostatic Precipitators (ESPs). Preliminary measurements at plants burning bituminous coal indicate that conventional ESPs remove less than 20% of the Hg present in flue gases (Chu et. al, 1993). Since most of the Hg is in the vapor phase, ESPs can not be effective control devices unless Hg condenses onto droplets at or before the device (Benson et al., 1993). Meij (1991) suggests that Hg removal efficiency across an ESP improves with increasing chloride content in coal, suggesting that Cl^- may reduce the electrical resistivity of the particulates and aid the efficiency of particulate collection on the ESP plates. In this assessment, a 10% removal efficiency is assumed for the reference case. This efficiency would be increased if the presence of control systems was assumed, as discussed below.

4.2.2 Wet Scrubbing. The removal of Hg by a combination of ESP and wet scrubbing is reported by several sources. Meij (1991) (reported by Nobblett et al., 1993) indicates removal efficiencies from 10 to 70% for such systems. Nobblett et al. also report that Hg removal depends on the chloride content in the coal, simply because of the oxidation to water-soluble $HgCl_2$. For plants burning western U.S. low chloride (<0.01%) coals, the Hg removal ranged from 5% to 25%, while for plants burning eastern coals (chloride from 0.1-0.3%), the efficiency of Hg removal ranged from 45% to 96%.

4.2.3 Dry Scrubbing. There are two major types of dry scrubbing systems: spray dryer absorption (SDA) and dry injection. SDA has been applied to hundreds of coal-fired boilers, municipal solid waste, and hazardous waste plants, to remove acid gases and particulates from emission streams. In a spray dryer, the sorbent solution or slurry (usually lime slurry), is atomized into the incoming flue gas streams to increase the liquid-gas interface and to promote the mass transfer of the contaminant from the gas to the slurry. Simultaneously, the thermal energy of the gas evaporates the water in the slurry droplets to produce a dry powder that contains the contaminant and some unreacted alkali. After leaving the spray dryer, the powder-carrying gas passes through a fabric filter or ESP where the dry product is collected. Dry injection generally involves pneumatic introduction of a dry, powdery material into the flue gas stream with subsequent collection of the contaminated powder on a fabric-filter or ESP. The injection point in this process can be anywhere from the boiler-furnace area all the way to the flue gas entrance to the bag-house, depending on operating conditions and design criteria (Holmes et al., 1993).

Efficient removal of both elemental and oxidized mercury may require a combination of SDA and solvent injection. In a combined system, a powdered dry sorbent (usually activated carbon) is injected in the duct carrying the effluent gas, usually upstream of the spray dryer absorber. Elemental mercury and mercury oxides are physically and chemically adsorbed on the surface of carbon. A fabric filter (bag-house) is used downstream of the injection chamber to collect the contaminated carbon along with other particulates in the flue gas. Felsvang et al. (1993) report mercury removal efficiency of several SDA systems in coal-fired power stations operating in the US and Europe. As shown in Table 4.1, these efficiencies range from 6% to 96%. Also shown in this system is a strong influence of the coal Cl^- content on the removal efficiency. The efficiency of the SDA system in Plant D (last entry in Table 4.1) increased from 96% to more than 99% when a carbon injection system was added.

Felsvang et al. (1993) also report the results of a pilot plant study which further quantifies the effect of adding sorbent injection to the SDA. Two types of coal of different chloride content were used in this study: coal A, with 0.019% Cl^-, and coal B, with 0.094% Cl^-. With SDA alone, the mercury removal efficiency from coal A emissions

was 57%, whereas from coal B, it was 78%. It appears that the efficiency of the SDA is strongly dependent on the fraction of oxidized Hg. Combining SDA with activated carbon increased the efficiency for both types of coal to 90%. However, it was found that coal A required five times more carbon than coal B. This is probably due to the fact that coal B, with higher Cl^- content, produces a higher fraction of soluble Hg (mainly as $HgCl_2$, which is also more reactive) and less (insoluble) elemental Hg than coal A. In a SDA system without carbon injection, the inherent removal of elemental Hg is practically zero, whereas the inherent removal of water-soluble oxidized Hg is 95%. With active carbon injection, removal of elemental Hg can increase up to about 5-60% (Figure 4.2). Obviously, the higher the quantity of elemental Hg in the emission stream, the larger the quantity of carbon required. In addition, Jozewicz et al. (1993), and Felsvang et al. (1993) report that chemically activated carbons are several times more effective in removing Hg than thermally activated ones. Chemical agents used in activation include compounds of iodine, sulfur and chlorine, with iodine appearing to be the most effective impregnant (Figure 4.3).

Table 4.1 Effect of Coal Chlorine Content on Removal of Hg from Flue Gas

Plant	Fly Ash Loading	Coal Cl^- Content	% Hg Removed
A	High	Low (0.01%)	14
B			23
C			6
G			16
E	Low	High (0.1-0.3)%	55
H			44
F	Medium		89
D	High		96

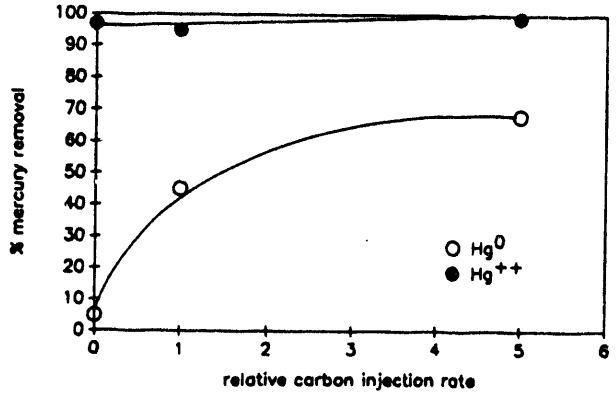


Figure 4.2. Effect of carbon injection on Hg removal by spray-dryer absorption systems.

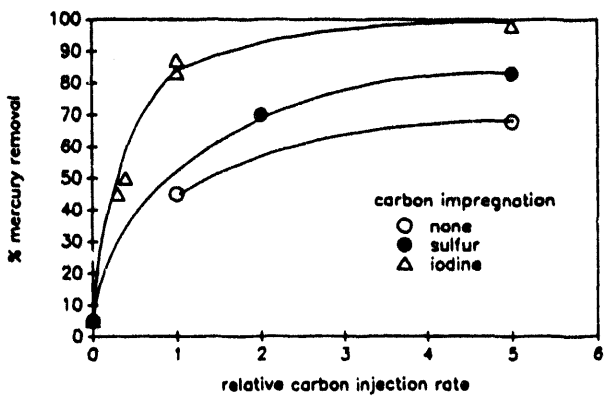


Figure 4.3. Effect of chemical carbon activation agents on Hg removal.

4.2.4 Condensing Heat Exchanger. This device is reported to be effective in removing particles along with gases and vapors. Such removal is accomplished as water condenses around particles, forming droplets that can be captured in a scrubbing device. There are a number of condensing heat exchangers operating on district heating plants in Scandinavia, and, according to McIlvaine (1993), mercury is effectively removed by these devices.

4.2.5 Activated Char Filter. In many waste incinerator installations in Europe, mercury is removed from the gas streams by a final-stage filter with activated char placed after the conventional air pollution control equipment. McIlvaine (1993), in a literature review of several control options (e.g., wet scrubber, activated char, condensing heat exchanger, carbon injection), reports that char filters are the most efficient devices for mercury removal. However, he does not refer to Hg speciation and the variation of Cl^- in the flue gas streams is not considered. Also, this option is probably the most expensive to implement in power plants, given the high flow rates of stack gases.

4.3 Summary of Data on Mercury Emissions.

Recent data on Hg emissions from coal indicate lower and less variable values than previously reported. In addition, the chemical form of the emitted Hg appears to be important, since the soluble compounds may be more efficiently removed by flue gas treatment. Rates of Hg removal range from about 10% to 80-90%. Definitive data on these trends await release of results from the EPRI PISCES program (Chu et al., 1993); in the interim, an average value of Hg in coal (0.08 ppm) is assumed, with minimal (10%) removal by control equipment.

5.0 MERCURY IN THE ENVIRONMENT AND THE FATE OF MERCURY EMISSIONS

In this section, the information necessary to consider mercury emissions in the context of environmental impacts is presented. This includes comparisons of total Hg emissions and the rates of dispersion and deposition from point sources such as power plants. Finally, a protocol for estimating deposition from a hypothetical power plant is derived.

5.1 Background Information

There are many sources of mercury in the environment. Mercury is extracted from the earth primarily as cinnabar (HgS). Annual worldwide production is about 10,000 tons, and smelting and ore roasting can produce local emissions of mercury vapor. Global annual anthropogenic emissions have been estimated at about 3000 tonnes; natural emissions may be up to twice that amount (WHO, 1991). Coal combustion releases mercury to the atmosphere primarily as gases, while municipal incineration releases both gaseous and particulate forms. Certain industrial processes, chiefly chlor-alkali plants, were known to release large amounts of mercury to air and aqueous waste streams (Wheatley, 1979), but these have now been largely brought under control in the developed world. Natural emissions emanate from volcanoes, from soil and biota, and from sea spray and forest fires. While there are considerable uncertainties, natural and anthropogenic emissions of mercury are thought to be roughly comparable. Suffice it to say, there are no major (individual) sources of mercury emissions on the planet, and the problem should be regarded as global in scope.

The atmospheric mercury cycle is quite complicated, since the residence time of Hg vapor in the atmosphere is long (weeks to ca. 1 y) and there are few permanent sinks. Aquatic sediments provide relatively long-term storage, but organic Hg released to the water column can be reduced to mercury vapor and reemitted to the atmosphere. Even mercury in dental amalgams can be recycled through cremation.

The pathways for human exposure to mercury include inhalation of airborne mercury, which is mainly inorganic, and through ingestion of foods containing mercury, mainly as MeHg. Many studies have shown that the most important pathway is through consumption of predatory fish, since they can bioconcentrate methylmercury by many orders of magnitude as it works up the food chain. Mercury in drinking water and in other foods does not constitute an important pathway (except perhaps for the low doses characteristic of non-fisheaters).

This assessment deals with two of the most important environmental cycles of mercury, the atmospheric and aquatic-biological cycles. The atmospheric cycle involves anthropogenic emission of mercury, conversion in air to the oxidized state, deposition by aerodynamic and precipitation processes, conversion in soil and water of deposited oxidized (Hg^{++}) species to gaseous elemental mercury (Hg^0) and dimethyl mercury $(CH_3)_2Hg$, and subsequent re-oxidation of elemental mercury to water-soluble forms. This cycle ends with the deposition of non-volatile forms to soils and aquatic sediments, where the mercury may remain for long periods.

The aquatic-biological cycle of mercury involves the formation of methylmercury (CH_3Hg^+), its enrichment in organisms and nutritional chains, and finally, destruction (i.e., demethylation) of methylmercury. Methylmercury is the dominant form of mercury in higher organisms, although it represents only a small amount of the total mercury in the atmosphere and in precipitation. The rates of formation and destruction of methylmercury are therefore very important for determining the enrichment of mercury in aquatic organisms.

Once mercury is in the atmosphere, it may react with other atmospheric constituents and be transformed from one oxidation state to another. The primary transformations taking place in the atmosphere involve oxidation of gaseous Hg^0, by O_3, HCl, Cl_2 or O_2, to inorganic compounds of Hg^{++} (only small quantities of insoluble methylmercury and soluble methylmercury chloride may be formed in the atmospheric aqueous phase). Because of these reactions and the fact that most mercury emitted at the stack is oxidized already, it is reasonable to assume that most mercury in the plume will be in water-soluble form. However, it is possible that some reducing reactions may take place in the plume, which could reduce the fraction of soluble mercury.

In the aqueous phase (e.g., rain, cloud, or fog aerosols), reactions involve the oxidation of Hg^0 to Hg^{++}, the reduction of Hg^{++} to Hg^0 and the dissociation/complexing of the oxidized species.

5.2 Mercury in Natural Fresh Waters

Several studies have been done in Europe and the U.S. on deposition of atmospheric mercury in lakes. A study of Hg levels in 220 lakes in southern and central Sweden showed an implied relationship with atmospheric deposition. The reported mean values of mercury concentration in fish (pike) were 0.68 and 0.86 mg/kg for the

two regions respectively, while the natural background level of fish is estimated to be in the range of 0.05-0.2 mg/kg (Bjorklund et al., 1984). A study of 80 lakes in northeastern Minnesota (Sorensen et. al., 1990) shows similar elevated Hg concentrations in pike (i.e., 0.44 mg/kg) The estimated atmospheric annual Hg inputs to these lakes together with Hg concentrations in the water are shown in Table 5.1. These data give an idea of the variability of mercury inputs in a location relatively remote from strong anthropogenic sources and show the range of bioconcentration in sediments and plankton.

Table 5.1 Summary of Mercury Concentrations in Northeastern Minnesota Lakes

Atmospheric inputs	mean	minimum	maximum
average precipitation concentration (ng/L)	18.7	16.8	24.2
annual wet deposition* ($\mu g/m^2/y$)	12.6	10.4	15.4
Concentration measurements			
surface waters (ng/L)	2.47	0.9	7.0
surface sediments (ng/g)	174	34	753
zooplankton (ng/g)	87.9	9.5	209

* wet deposition falling directly on lake surface.
Source: Sorensen et al. (1990), based on 1988 Hg concentrations and long-term precipitation data.

Data compiled by Lindqvist (1985) indicate that the predominant mercury compounds in fresh waters are inorganic and monomethyl mercury compounds (Table 5.2). The chlorides $HgCl_2$ and CH_3HgCl are the main mercury compounds in neutral to acidic water environments, whereas in alkaline environments the hydroxide compounds $Hg(OH)_2$ and CH_3HgOH prevail. Thus, lake pH is expected to play an important role in the bioaccumulation of mercury in aquatic organisms.

5.3 Mercury in the Atmosphere

Mercury is present in the atmosphere in three oxidation states: elemental, Hg^0; monovalent, Hg^+; and divalent mercury, Hg^{++}. Mercury is emitted by anthropogenic sources in elemental and various oxidized forms. Elemental Hg and $(CH_3)_2Hg$ also enter the atmosphere via biological processes. Oxidized inorganic forms (e.g., $HgCl_2$) are extremely water-soluble, methyl chlorides (e.g.,$HgCH_3Cl$) are also quite soluble, whereas elemental Hg and dimethyl mercury $(CH_3)_2Hg$ are insoluble forms. Until recently it was generally assumed that the main form of volatile mercury in air is elemental Hg; this is still the case for Hg generated from natural sources, but there is evidence that plumes emitted from coal-burning power plants may contain a large fraction of water-soluble $HgCl_2$, at least initially (Bloom, 1993), as shown in Figure 4.1. Background ambient air concentrations in remote areas are in the range of 1-3 ng/m^3, and as high as 10 ng/m^3 in urban areas. Lindberg (1980) measured concentrations of total gaseous mercury in excess of 1000 ng/m^3 in the plume of a coal-fired power plant within a few km from the source. Since concentrations of 5-10 $\mu g/m^3$ are expected at stack exit, these data imply somewhat less dispersion than might have been expected for that downwind distance.

The degree that these compounds are removed from the atmosphere by rain depends on their solubility in water. A list of these compounds and their Henry's Law constants is shown in Table 5.3. These Henry's Law constants are expressed as the ratio of the volumetric concentration of a compound in a gas-phase which is in equilibrium with its aqueous phase, over its concentration in the aqueous phase. A high value implies a high degree of volatility; note the increases with temperature.

Table 5.2 Distribution of Mercury Compounds in Natural Fresh Waters
(values are percentages of either total inorganic (a) or total monomethyl mercury (b)

Compound	pH of water body				
	4	5	6	7	8
a. Inorganic Compounds					
$HgCl_3^-$	0.2	0.2	0.1	0.1	-
$HgCl_2$	99.6	98.8	91.1	34.1	0.1
HgOHCl	0.1	0.9	8.5	49.4	6.2
$HgCl^+$	0.2	0.2	0.2	0.1	-
$Hg(OH)_2$	-	0.002	0.2	16.4	93.7
Total	100	100	100	100	100
b. Monomethyl Compounds					
CH_3HgCl	97.8	97.1	89.8	33.5	1.9
CH_3HgOH	0.1	0.8	8.4	65.8	98.1
CH_3Hg^+	2.1	2.1	1.9	0.7	0.04
Total	100	100	100	100	100

[1] Adapted from Lindqvist (1985).

Table 5.3 Henry's Law Constants of Atmospheric Mercury Compounds*

Compound	Formula	Henry Law Constant	Temp(^{0}C)
Elemental mercury	Hg^0	0.29	20
Methyl mercury	$(CH_3)_2Hg$	0.15 0.31	0 25
Methyl mercury chloride	CH_3HgCl	0.9e-5 1.6e-5 1.9e-5	10 15 25
Mercury hydroxide	$Hg(OH)_2$	1.6e-6 3.2e-6	10 25
Methyl mercury hydroxide	CH_3HgOH	~10e-7	
Methyl chloride	$HgCl_2$	1.2e-8 2.9e-8	10 25

**adopted from Lindqvist (1985).*

Mercury can be retained in the atmosphere for long periods and consequently is transported over long distances (Lindqvist, 1985). Elemental mercury has a residence time between 0.5 and 1.5 years, whereas most oxidized species have lifetimes of hours or days in the atmosphere (Iverfeldt and Lindqvist, 1986). Short lifetimes imply rapid deposition and thus local impacts. Assuming a lifetime of one day, these compounds would be transported several hundred kilometers downwind before they convert to other forms; this provides an estimate of the scale of interest for local impacts. In this assessment, the fraction of soluble to total mercury is assumed to remain roughly the same within the geographical scale considered (i.e., 50-100 km radius).

5.4 Mercury in Terrestrial Systems

Nater and Grigal (1992) sampled organic litter and surface and subsurface soils along a broad transect in Minnesota, Wisconsin, and Michigan. They noted a slight increase in surface Hg from west to east, which is consistent with increasing anthropogenic deposition. The subsurface Hg appeared to be invariant with direction, and comparison of the difference between the range of values found at the surface with subsurface Hg implied from 35 to 165 years of deposition, at the estimated present deposition rate of 15 $\mu g/m^2/y$. Hg concentrations in organic litter were much higher than in soil, and also higher than in most U.S. coals. The amount of Hg currently stored in

the terrestrial reservoir appears to be much higher than the annual rates of deposition, even around point sources (see below).

5.5 Plume Dispersion

The Hg concentrations used in this study are annual average ambient concentrations at ground-level, estimated from a Gaussian dispersion model, assuming point-source releases (Lipfert et al., 1986). Hourly average estimates from models of this type are considered to be accurate within a factor of 2 at least 50% of the time (Turner et al., 1985), when applied to flat terrain and downwind distances from a few hundred meters to about twenty kilometers. The statistical properties of the distributions of plume impacts are not well known, but since values cannot be negative, a log normal distribution may be a reasonable assumption for this purpose. This assumption translates into a geometric standard deviation of about 2.8, such that the 95% confidence limits for an individual impact with a median value of 1.0 would be about 0.13 and 7.8 (plus or minus a factor of 7.8). However, based on a peak-to-mean concentration ratio of 30, there would be about 260 such impact events annually, such that the corresponding uncertainty in the annual average would be plus/minus 13%. If the model were run for a longer period, the confidence limits would become even tighter. These calculations pertain to a single point source; the impacts from multiple sources are additive on an annual basis, but rarely on an individual impact event basis (which would require the multiple plumes to coincide in direction). The uncertainty of these predictions when applied over complex terrain and longer distances is significantly greater.

The atmospheric Hg concentrations used in this study are annual averages at ground level. Although rates of total deposition may depend on the vertical distribution of mercury within the plume, ground level concentrations are used because the dry deposition velocity (V_d) and the washout ratio (WR) are defined in reference to ground level concentrations:

$$V_d = \frac{\text{Deposition flux of compound to the ground}}{\text{Concentration of compound in surface-level air}}$$

$$WR = \frac{\text{Concentration of compound in precipitation}}{\text{Concentration of compound in surface-level air}}$$

The assumption embedded in these definitions is that the vertical distribution of Hg concentrations downwind of the release is uniform over a certain height H. This is true at distances greater than about 2 km under most atmospheric conditions, as discussed below; it may not be true at the point of maximum ground-level impact. However, it is not clear whether the values of V_d and WR available from the literature are applicable to plume impact conditions, in any event.

The lower part of the troposphere, the atmospheric boundary layer (ABL), is closely coupled with the earth's surface by turbulent exchange of mass, momentum and energy. The depth of the ABL varies from tens of meters when the air near the surface is stably stratified, to several kilometers when the atmospheric conditions are highly unstable. In the lowest part of the ABL, the surface layer (SL), concentration of pollutants may vary significantly with height. However, the thickness of the SL is small, about 10% of the thickness of the ABL. Above the SL, in the bulk of the ABL, in most locations and for most of the time, turbulent eddies predominantly affect vertical transport, mixing is very efficient, and vertical concentration gradients are very small (Dabberdt et al., 1993).

Goldberg (1972) has suggested that all atmospheric Hg is included in a column not higher than 5 km, whereas Williston (1968) showed that the highest concentrations of Hg are close to the ground. Based on these studies, Airey (1982) concluded that it is more realistic to use average concentrations within heights of 0.5 km or less for scavenging calculations. Wet deposition estimates in this assessment are based only on Hg concentrations calculated at ground level.

5.6 Atmospheric Removal Processes for Hg

Mercury is removed from the atmosphere by two mechanisms: 1) "dry" deposition, (i.e., deposition in the absence of hydrometeors); 2) wet deposition, (i.e., absorption into droplets followed by droplet removal and precipitation). Both mechanisms are driven by the concentration of Hg in the air, that is, higher ambient Hg concentrations result in proportionately higher deposition rates.

Mercury fluxes from soils and natural waters have been studied in the laboratory and in the field. Dry deposition has been found to be quite variable in terms of both magnitude and direction - under certain conditions, both soils and water bodies can emit mercury vapor, especially as temperature increases. In remote areas away from anthropogenic mercury sources, only wet deposition has a known (constant) direction of mass transfer.

5.6.1 Dry Deposition. Dry deposition can be parameterized by a deposition velocity, V_d, which was defined above. The dry deposition velocity of particles depends on their physical state and size distribution, whereas the deposition velocity of gases is mainly dependent on the characteristics of the surface on which the particles are deposited. Only a small fraction of Hg exists in particulate form in U.S. power plant emissions, since these plants are equipped with particulate controls. In this study, it was assumed that the fraction of particulates in the post-control emission stream is 1% of the total mercury emitted, based on the finding of a particulate content of 0.7% in a Tennessee plant by Lindberg et al. (1991).

Although recent data indicate that a high proportion of mercury in coal-fired power plant stacks may be Hg^{++}, no information is available on its composition as it travels downwind in the plume. It has been suggested that the Hg^{++} may be reduced back to Hg^0 by SO_2 (S.E. Lindberg, personal communication, October 1993). However, in the presence of HCl, Hg^{++} will be complexed as $HgCl_2$, which would allow little reduction; Hg^{++} may be distributed between gas and droplet phases, which can affect its dry deposition velocity (C. Seigneur, personal communication, April, 1994). As a reactive gas (such as HNO_3), it may be expected to deposit rapidly. The dry deposition rates for particles depend on their sizes; fine particles dry deposit much more slowly than reactive gases, for example.

Values of deposition velocity of mercury vapors reported in the literature range from 0.006 to 0.5 cm/s, the lowest deposition rates occurring in a deciduous forest during the dormant period. Clark (1993) reports values for forests which vary from 0.001 cm/s in winter to 0.03 cm/s in summer. For an initial estimate of dry deposition of gaseous Hg^0, an estimated value of 0.02 cm/s is used. It is important to note that the more reactive forms of Hg such as Hg^{++} or $HgCl_2$ may be deposited at higher velocities than elemental Hg^0. Deposition of reactive compounds is not limited by leaf pathways (e.g., stomata and mesophyll) but is primarily controlled by aerodynamic resistance. Deposition velocities of reactive gases can range from about 10^{-3} cm/s up to 10 cm/s; their values depending primarily of the chemical characteristics of the gas and the roughness of the surface (see Table 5.4). From these values it appears that a good approximation for $HgCl_2$ dry deposition is 1.9 cm/s.

The rate of dry deposition can be expressed as

$$DD=V_d\, C(r,0)\, I_d \qquad [5\text{-}1]$$

where V_d is expressed in m/s, C(r,0) is the ambient ground level concentration at distance r from the source [g/m^3], and I_d is the ratio of the time that is either dry or rains only lightly, i.e., not more than 0.1 inch per hour. The R_d value used in the current study is 0.92, which corresponds to typical Northeastern U.S. climate.

Table 5.4 Typical Dry Deposition Velocity (V_d) Values of Some Reactive Gases

Compound	Surface	V_d range (cm/s)
Cl_2	alfalfa	1.8-2.1
fluorides	forage	1.4-2.4
iodine	grass	0.5-4.0
NO_2	alfalfa	1.9
SO_2	forest	< 2
SO_2	medium grass	0.8-1.9
SO_2	water	0.46

source: Sehmel (1980)

Table 5.5 Measured Values of Wet Deposition of Hg

location	wet dep rate ($\mu g/m^2y$)	precipitation (m)	reference
NE Minnesota	12.6	0.67	Sorensen et al., 1990
southern Sweden	20	~0.8	Johansson et al., 1991
New England coast	10	1.0	Fogg&Fitzgerald,1979
New Jersey backgr.	10-12	1.0	Greenberg et al., 1992
Italy (background)	6-7	~1.0	Ferrara et al. (1986)
Italy (urban)	10	~1.0	Ferrara et al. (1986)
Italy (near Hg sources)	12-22	~1.0	Ferrara et al. (1986)

5.6.2 Wet Deposition. Wet deposition rates depend on the solubility of the mercury compounds present and the rate of precipitation. Table 5.5 presents data from the literature.

The annual flux of mercury deposition on ground-level surfaces can be estimated by

$$WW = WR\ C(x,y,0)\ P \qquad [5\text{-}2]$$

where WR is the washout ratio [dimensionless], C(x,y,0) is the ambient ground level concentration [g/m^3], and P is the annual average precipitation [m/yr].

Washout ratios in the range of 10^3 to 10^6 are reported in the literature; these are listed in Table 5.6. The low ratios listed in the first column of this table correspond to total mercury, whereas the highest value, 10^6 corresponds to particulates emitted from forest sources. In this assessment, a mid-point value of 10^5 is used for the deposition of water-soluble mercury in the plume. This number is based on soluble mercury and is one to two orders of magnitude higher than the ratios based on total mercury.

Table 5.6 Estimates of Approximate Hg Washout Ratios*

*ratios of total Hg concentrations in rain to various Hg concentrations in air, dimensionless

Mercury compounds in air		location	reference
total Hg	soluble Hg		
10^4	10^5	rural Sweden	(a)
10^4		rural Italy	(a)
10^4		rural UK	(a)
3×10^4		urban Japan	(a)
$0.8\text{-}1.4 \times 10^3$		central Italy	(b)
10^6		tropical Pacific	(a)

*ratios of total Hg concentrations in rain to various Hg concentrations in air, dimensionless
References: (a) Lindqvist (1985) (b) Ferrara et al. (1986)

5.7 A Model of Anthropogenic Hg Accumulation in Lakes

Mercury contamination of remote lakes in the North-central United States has been attributed to increasing deposition of atmospheric mercury. Swain et al. (1992) measured Hg concentrations in sediments of seven lakes in Minnesota and Wisconsin and calculated that the annual atmospheric Hg deposition (wet + dry) increased from 3.7 to 12.5 $\mu g/m^2$ since 1850. The lakes in this study were not in the vicinity of any power plants or other anthropogenic source of mercury and the deposition increase was similar among lakes, implying regional or global sources for the mercury entering these lakes. These investigators also found that atmospheric Hg deposition to an area of terrestrial catchment several times the size of a lake is transported to the lake. According to their study, the current deposition flux can be described from the equation

$$F_{td}[\mu g/m^2 yr] = 12.5 + 3.27\ A_d/A_L \qquad [5\text{-}3]$$

and the preindustrial deposition flux from

$$F_{td}[\mu g/m^2 yr] = 3.7 + 0.83\ A_d/A_L \qquad [5\text{-}4]$$

where A_d is the area of the terrestrial catchment around the lake and A_L is the area of the lake.

The deposition flux due to anthropogenic sources is estimated by subtracting Equation [5-4] from [5-3]:

$$F_{td}[\mu g/m^2 yr] = 8.8 + 2.44\ A_d/A_L \qquad [5\text{-}5]$$

For direct to deposition to a lake, this equation results in a flux of about 11 $\mu g/m^2 yr$. This estimate includes both direct deposition of Hg from current anthropogenic sources and deposition of atmospheric Hg which may have come from outgassing from previously deposited Hg. For $A_d/A_L = 0$, the estimate of current deposition from Eq. [5-3] is similar to other values measured in the United States for wet deposition alone (see above), implying a modest contribution of dry deposition of elemental Hg. This also implies that these wet deposition measurements must also reflect the presence of anthropogenic Hg contributions, even in remote locations.

An alternative scavenging model derived from field measurements (J. Shannon, personal communication, 1993) is based on the assumption that any rain event will remove approximately half of the mercury in the atmosphere. Mass balance calculations showed agreement with this hypothesis, under the assumptions of: a) washout occurs through an atmospheric boundary layer of 0.5 km thickness; b) the concentration of soluble Hg reduces exponentially to 50% of its initial value during a single precipitation event; c) one hundred precipitation events

occur in a course of a year and the time between such events is sufficient for atmospheric mercury to resume its previous concentration.

5.8 Measured Deposition Patterns Around Point Sources

Greenberg et al. (1992) measured Hg concentrations in rain near a large resource recovery plant in New Jersey. The maximum allowable emission rate is 45 g/h, and the stack height is about 60 m (Greenberg, 1994). Using the methods of Lipfert et al. (1986), a maximum annual average ground-level concentration of total Hg due to this plant of about 1.3 ng/m^3 is estimated, located about 0.5 km downwind. The data on Hg in precipitation over the six rain events sampled were then averaged by approximate radius from the plant (Figure 5.1). The data clearly show the maximum rain concentration and thus wet deposition occurs close to the source (within about 1 km). This is reasonably consistent with the present model, and the washout ration implied by this comparison is about 1.1 x 10^5, which is consistent with the data of Table 5.6 if a high fraction of soluble Hg in the stack plume is assumed. Given the likely presence of Cl^- from burning plastics, the assumption seems reasonable. Although Greenberg et al. estimated the background Hg concentration at about 18 ppt, the plot shows that 10-12 ppt is a more likely range. With 1 m of annual precipitation, the background wet Hg deposition would be 10-12 μg/m^{2}y. Figure 5.1 also shows that the envelope of maximum Hg deposition decreases with distance from the plant at a rate somewhere between r^{-1} and r^{-2}, depending on how much credence is given to the data point based on only 1 sample. Hogstrom et al. (1979 estimated that about 30% of the mercury emitted from a chlor-alkali plant was deposited within the first 4 km., although wet deposition appeared to be still elevated at 10 times this distance.

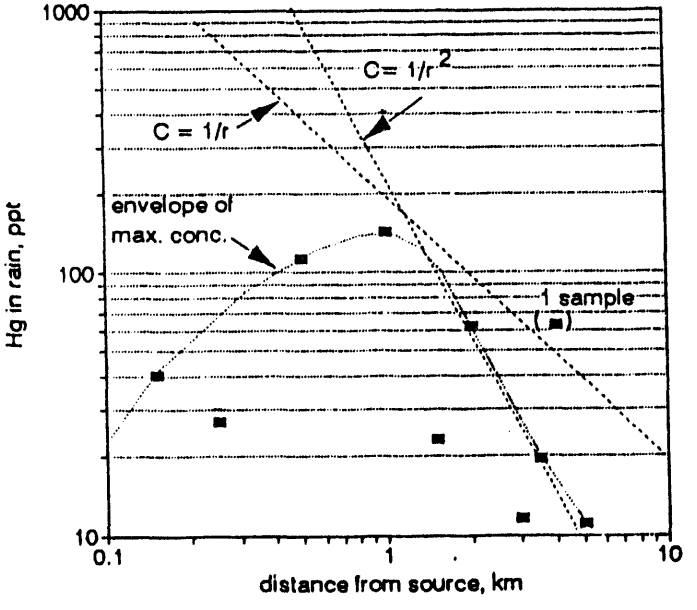


Figure 5.1. Average concentrations of Hg in precipitation, measured for 6 rain events, near a New Jersey incinerator. Data from Greenberg et al. (1992).

5.9 Plausibility Checks Based on Global Mass Balance Calculations

One way of verifying the reasonableness of these parameter estimates involves examining global averages. Natural sources of mercury were estimated to release from 2700-6000 tonnes annually (WHO, 1991); anthropogenic

sources add about another 3000 tonnes. When referenced to the surface area of the planet (510×10^6 km^2), these amounts would correspond to average flux rates of 11-18 μg/m^2/yr, assuming a uniform distribution with no point sources. Laboratory experiments have found flux rates of a similar magnitude (Xiao et al., 1991), with a negative sign (emission) in summer and a positive (deposition) sign in winter. This is consistent with the finding by Hogstrom et al. (1979) of atmospheric mercury gradients radiating away from a point source even upwind of the source, which was taken as evidence of the re-emission of previously deposited mercury. Now, if a global average Hg^0 concentration of 1.5 ng/m^3 (Lindqvist, 1985) and an average dry deposition velocity of 0.02 cm/s are assumed, the resulting average deposition flux is about 10 μg/m^{2}y. The global average wet deposition flux appears to be of the order of at least 5 μg/m^{2}y, so that the total annual Hg deposited appears to be about 15 μg/m^2, which agrees with the figures quoted by Nater and Grigal (1992) and with the estimates of Swain et al. (1992) from lake sediments, mentioned above.

An additional mass balance consideration includes the ratio of deposition to the apparent total Hg content of the earth's atmosphere. Based on an estimated global average ambient concentration of 1.5 ng/m^3, the atmospheric reservoir would contain about 4500 tonnes of Hg. Using the estimated dry deposition velocity of 0.02 cm/s, the annual rate of deposition would be about 3200 tonnes. When wet deposition is added, these figures are roughly in balance. The ratio of the atmospheric pool of Hg to the emission rate gives an estimate of the average Hg residence time, in this case about 1 year. If the atmospheric mercury is all trapped in a lower portion of the atmosphere, the residence time would be shorter.

5.10 The BNL Model for Incremental Hg Deposition from Point Sources.

The total deposition of mercury on ground level surfaces can be estimated once the distribution of ground level ambient concentration is known. In these atmospheric dispersion calculations, Hg emissions from a hypothetical 1000 MW_e power plant working 75% of the time were assumed, together with an Hg content in coal of 0.08 ppm (the U.S. average), and that 90% of it is emitted from the stack; the average Hg emission rate from the plant is thus 5.7 mg/s (0.05 lb/hr). The assumption of 75% reactive Hg is consistent with a coal chloride content of about 0.1% or more, if the data of Figure 4.1 hold.

The model calculations for a hypothetical 1000 MW_e power plant are based on the following parameters:

stack height:	200 m.		
total Hg emission rate	5.7 mg/s (180 kg/y)		
maximum annual average ground level concentration ratio (X/Q)	6×10^{-9} sec/m^3		
radius at which this occurs	2.0 km		
distribution of wind and precipitation direction	uniform		
annual precipitation	1 m.		
fraction of particulate Hg	0.007		
fraction of soluble (Hg^{++}) Hg	0.75		
(Hg species)	Hg^0	Hg^{++}	$Hg_{part.}$
dry deposition velocities (cm/s)	0.02	1.9	~0
washout ratios	~0	10^5	10^6

In addition to these fixed parameters, the model requires information on the rate at which annual average air concentrations decrease with radius downwind of the plant. Data obtained by Lipfert et al. (1986) from model runs using the Cincinnati (OH) wind rose were used. The rate of decrease of Hg deposition downwind of the maximum point is seen to be more gradual than most of the data observed near an incinerator in New Jersey (Figure 5.1).

Should data on ambient concentrations specific to wind directions become available, then the relative frequency of precipitation for winds in the appropriate direction should be included in the model.

In this model, mercury not deposited within 50 km of the source is assumed to remain airborne and to eventually become part of the global background. Thus the rate of airborne mercury flux beyond the 50 km radius, is

$$R_{airborne} = Q - R_{td} \qquad [5\text{-}6]$$

where Q is the emission flow rate and R_{td} is the rate of total deposition within the specified radius, calculated by integrating the product of deposition flux (F_{td}) and incremental deposition area from the plant out to the desired radius. Note that circular symmetry is assumed in this case, for simplicity.

$$R_{td} = 2\pi \int_0^{50} F_{td}(r)\, r\, dr \qquad [5\text{-}7]$$

The deposition flux is then estimated as the sum of the contributions from wet and dry deposition for the 3 groups of mercury species indexed by i: soluble, non-soluble and particulate forms. Thus,

$$F_{td}(r) = \sum_i DD_i(r) + \sum_i WW_i(r), \quad i=\text{soluble, non-soluble, particulate} \qquad [5\text{-}8]$$

where DDi(r) and WWi(r) are the fluxes of dry and wet deposition introduced earlier in section 5.6. These fluxes are calculated by

$$DD_i(r) = V_d\, a_i C(r,0)\, I_d \qquad [5\text{-}9]$$

$$WW_i(r) = WR\, a_i C(r,0)\, P \qquad [5\text{-}10]$$

where C(r,0) is the total ambient ground level concentration of total mercury at distance r from the plant, a_i is the fractions of the concentrations of soluble, non-soluble and particulate forms of mercury, I_d, is the ratio of time it does not rain heavily in a particular location, and P is the annual average precipitation in same location.

The results of these calculations are shown in Figure 5.2; note that the dry deposition contribution is the difference between the two curves shown. The extension to a radius of 100 km is shown as a dotted line since Gaussian models of this type are traditionally limited to 50 km. The maximum predicted total incremental deposition rate is 17 $\mu g/m^2y$, which drops slowly with radius for the first few km. This value of local incremental deposition thus only applies to small watersheds in the immediate vicinity of the plant. Since larger watersheds would see a lower average incremental rate of deposition but would augment the direct deposition to the lake with run-off from the watershed, this maximum value is used in the impact estimates. A factor of 2 is taken as the nominal estimate of the local change in total Hg deposition due to the hypothetical power plant.

The total fraction of Hg deposited out to 50 km is about 5.2%; to 100 km, about 6.7%. These fractions would be increased considerably if a shorter stack height had been assumed, since maximum concentrations increase approximately with the square of stack height. This probably accounts for Hogstrom's (1979) finding of a higher fraction of deposition near a chlor-alkali plant. The wet deposition rates are based on ground-level Hg concentrations, which are assumed to be uniform with height as a long-term average. Deposition in the first 2 km from the stack is thus very uncertain, since some Hg could wash out of the plume while it is still aloft. However, the deposition measurements shown in Figure 5.1 support the BNL model, and the surface area affected by this uncertainty is relatively small. Thus, this initial region has been neglected. The characteristic curves for wet, dry and total deposition are parallel because all deposition is based on the same downwind profile of annual average air concentration and because uniform terrain and rates of precipitation are assumed.

The total deposition rate within an area of 50 km around a 1000 MW_e power plant is about 10 kg/yr; the residual amount of about 170 kg/yr is assumed to contribute to the background mercury. Since global Hg emissions from natural and anthropogenic sources are about 4000 and 3000-6000 tonnes per year, respectively (WHO, 1991), it may be assumed that the global Hg impacts of a single power plant may be neglected. This may also be the case for the entire U.S. utility industry, based on an estimated total emission rate of 72 tonnes/y. However this analysis

is essentially stationary in time - it does not consider the possible long-term effects of increases in the global pools of Hg.

Although this is intended to be a probabilistic risk assessment, we have not studied the effects of uncertainties in the parts of the overall risk model that deal with transport and deposition from a source. We have tried to allow for some portion of this variability by varying the incremental deposition relative to background from 50% to 200%, with the intent of including uncertainties about the model formulation as well as in the model parameters in this range. Sensitivity studies would be useful to identify those elements of the local impact calculations that have the largest effect.

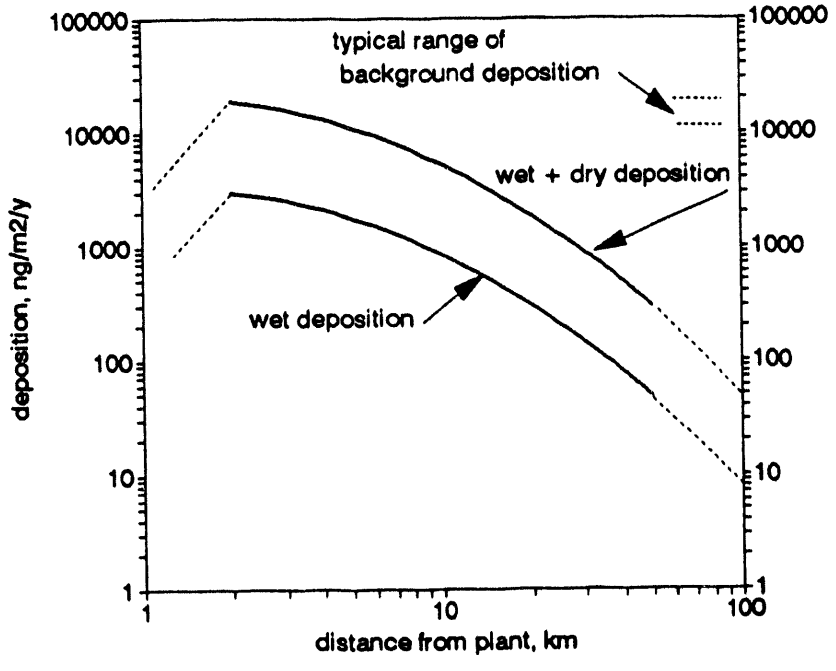


Figure 5.2 Estimate rates of total and wet deposition of mercury from a hypothetical 1000 MW_e power plant burning coal with average Hg content.

6.0 MERCURY CONCENTRATION LEVELS IN SEAFOOD

Studies have shown that the only significant pathway for methylmercury to humans is through consumption of contaminated seafood (Constantinou et al., 1993; Gunderson, 1988). This results from the extremely low concentrations of mercury in the atmosphere and in deposits and from the ability of predatory fish to bioconcentrate MeHg by many orders of magnitude.

As discussed below, the important fish contributions to the U.S. diet include shellfish, canned fish products, and fresh and frozen fin fish. Marine species tend to dominate the last category, but freshwater species are far more likely to be affected by local and regional sources of mercury emissions. Mercury levels in each category are discussed below, from the perspective of estimating the statistical properties of their distributions.

6.1 Mercury Levels in Freshwater Fish

Much of the concern about mercury as an environmental pollutant stems from the high concentrations of MeHg found in sport fish. Twenty-six states now have advisories with regard to consumption of certain fish species (Figure 6.1); concentration thresholds range from 0.5 to 1 μg/g, and concentrations in excess of these advisory levels occur with some regularity (Gloss et al., 1990). Moreover, lakes and streams in the interior of the United States are the locations most likely to be impacted by local sources of airborne mercury emissions, including those from coal burning. The success of any assessment of these effects thus depends critically on a detailed understanding of the baseline levels of mercury in fresh-water species.

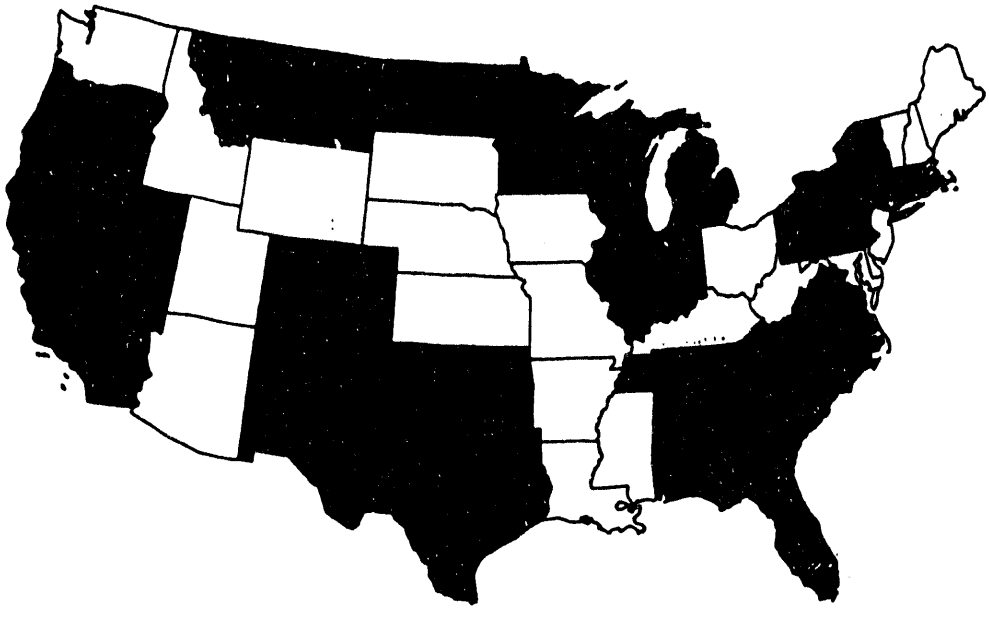


Figure 6.1. States having advisories for fish consumption based on mercury content.

6.1.1 EPA Data Base. There is only one known national database on contaminants in fresh-water species (U.S. EPA, 1992), but these data were deliberately obtained from polluted locations (not necessarily polluted with mercury, however) and thus may not comprise a representative baseline. Figure 6.2 shows the locations of sampling and Figure 6.3 presents a frequency distribution of the mercury levels found, together with consumption Hg advisory levels for several states. Most of the freshwater species are included in the EPA data base, with the notable exception of yellow perch (a small and generally less desirable species for sport fishing). The median of this distribution is 0.12 μg/g with a GSD of about 2.5 (based on the upper tail). One can see from Figure 6.3 that the upper and lower tails of the distribution are quite different; this probably results from the inability to measure very low Hg concentrations.

6.1.2 Data from Upper Michigan. A more useful data set for the present purpose is that of Gloss et al. (1990) and Cusimano et al. (1989, who sampled 49 lakes in the Upper Peninsula of Michigan as part of the National Surface Water Survey. These lakes were thought to be remote from local sources of mercury pollution. A variety of species were caught, and detailed data were presented for 864 fish taken from 37 different lakes. These data were made available to BNL for this analysis. The species for which mercury analyses were done included brook trout, large and small-mouth bass, northern pike, walleye, white sucker, and yellow perch. In terms of numbers of fish, yellow perch was the dominant species. A detailed analysis of this data set by individual fish and by lake characteristics is given in Appendix B.

The data on each fish include age, length, weight, and mercury content. The mercury data reported are total mercury from muscle specimens; the MeHg content was reported to be about 99%. The lake data available in Cusimano et al. (1989) included pH, DOC, certain chemical parameters, lake and watershed areas, sampling depth, and hydrological type. Deducing the importance of lake properties on mercury levels can be important for a national assessment of the effects of coal burning because lake properties (especially acidity) vary regionally and also affect fish abundance.

This analysis was intended to explore the dependence of Hg levels on fish characteristics, vis-a-vis lake characteristics. This analysis was conducted at two levels. First, data on the individual fish caught are used, in order to test for the dependence of mercury levels on various physical and chemical parameters. Then averages for each lake and major species (large mouth bass, northern pike, white sucker, and yellow perch) are used to examine the influence of lake characteristics in more detail; data on mercury levels in the lake waters were not available. Finally, the fish catch statistics were used to examine the probabilities of sport fishermen catching high-mercury content fish by angling.

Figure 6.4 plots the frequency distributions of Hg, for individual species (Figure 6.4a) and averaged over all fish species in each lake (Figures 6.4b to 6.4c). Not only are the distributions of Hg within each lake positively skewed, suggesting a log normal distribution, the distributions of the lake averages and medians are similarly skewed. The average of these medians is 0.21 μg/g, with a GSD of 2.8, and the median lake had a median Hg level of 0.17 μg/g. Weighting by the number of fish caught (species analyzed for Hg), the median Hg level was 0.28 μg/g. There was no relationship between median mercury level and total mass of fish harvested (Figure 6.5). However, the most productive lakes had mercury levels slightly below the median for all lakes in this data set.

The distribution of median mercury levels in individual lakes is somewhat more variable than the distributions within lakes. Thus, if it is assumed that a subsistence fishing population may be utilizing any randomly selected lake, a GSD of about 3.0 seems appropriate, with a median Hg concentration of about 0.17 μg/g. This implies 5% and 95% lake medians of about 0.03 and 1.05 μg/g, respectively. However, if more than one lake is utilized, as would seem likely for lakes of this size, the variability in mercury levels averaged over the population's total fish harvest would likely decrease. The larger the population at risk, the closer their mercury intakes will approach the average for the region. This may explain why epidemiological surveys of indigenous populations find so few cases of adverse health effects: sampling variability dominates studies of small populations, and the tendency toward more nearly average consumption levels affects large populations.

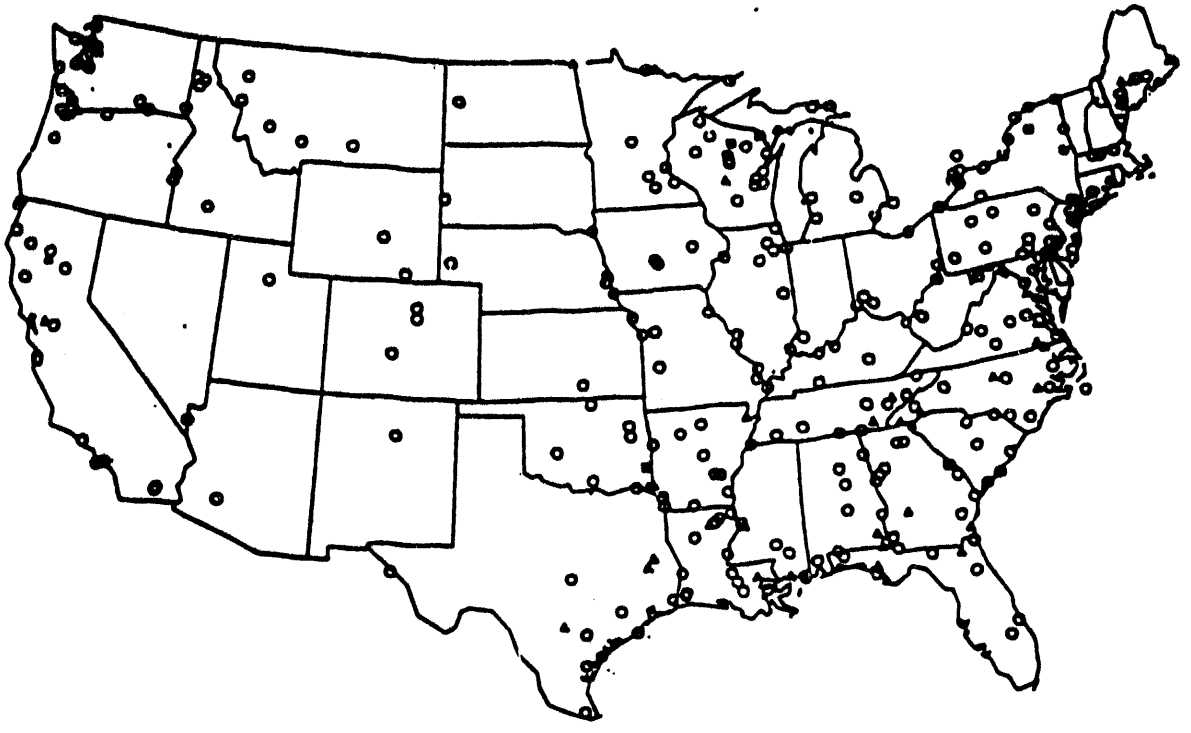


Figure 6.2. Locations of sampling by EPA for contaminants in fish. Source: U.S. EPA (1992).

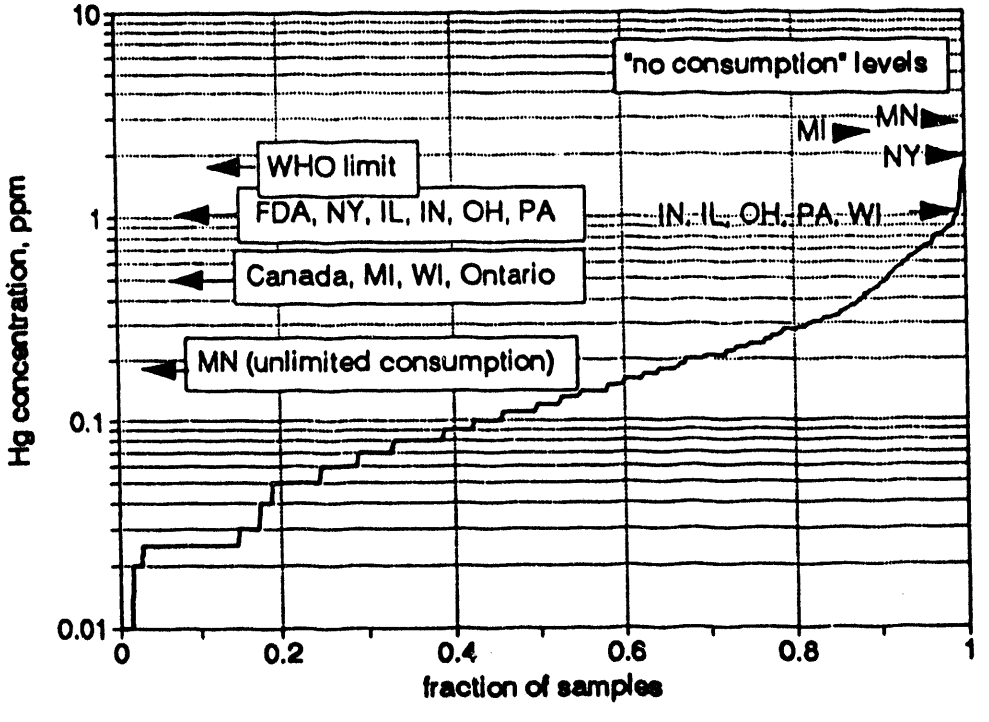


Figure 6.3. Overall frequency distribution of mercury concentrations in individual fish samples taken by EPA, with advisory levels used by selected states.

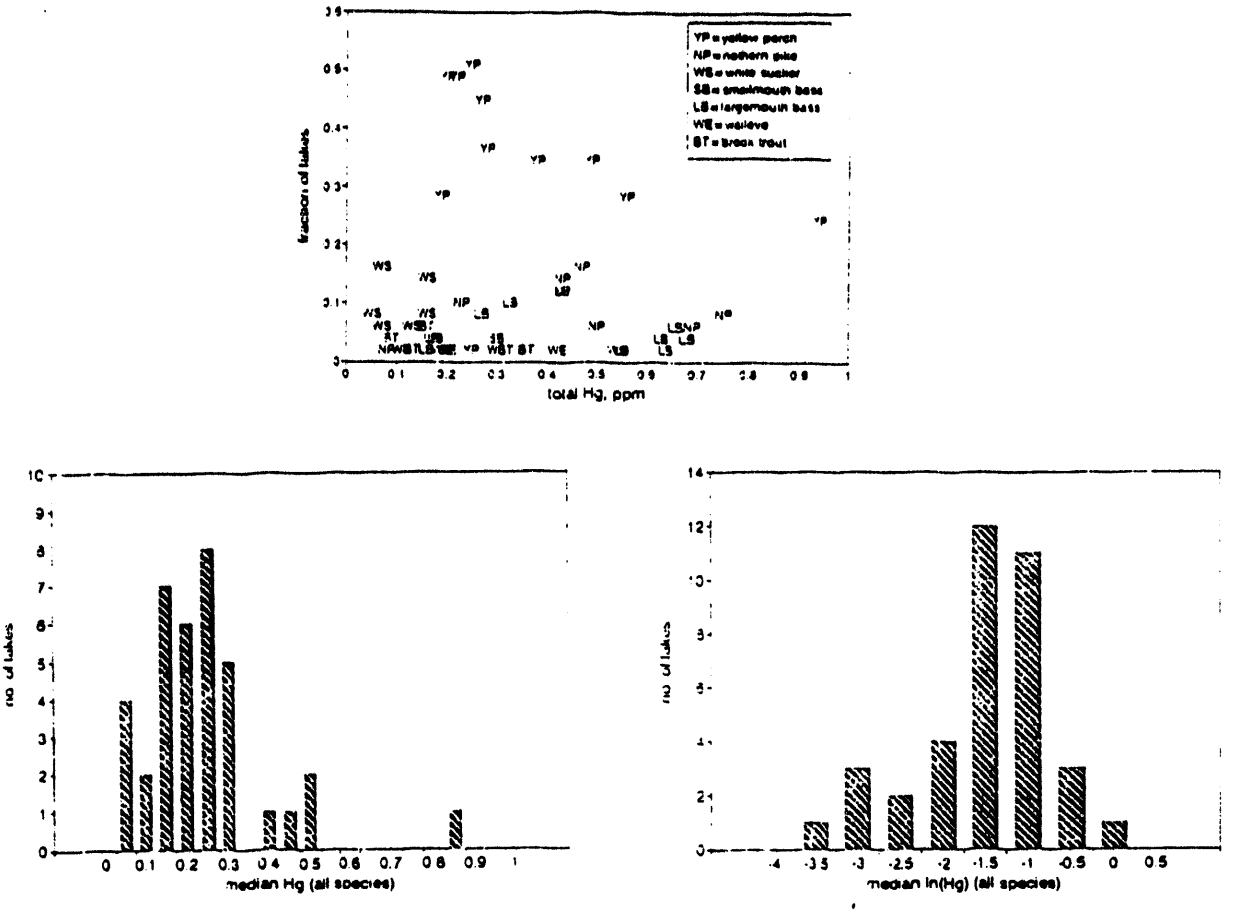


Figure 6.4. Data on the distribution of Hg in 49 Upper Michigan lakes, from Gloss et al. (1990). (a) Probability of fishing a lake with specified average fish Hg levels, by species. Plotting symbols are: BT = brook trout, LMB = large mouth bass, NP = northern pike, SMB = small mouth bass, WE = walleye, WS = white sucker, YP = yellow perch. (b) Frequency distribution of median Hg levels for all fish species. (c) as in (b), for the median log of Hg concentration.

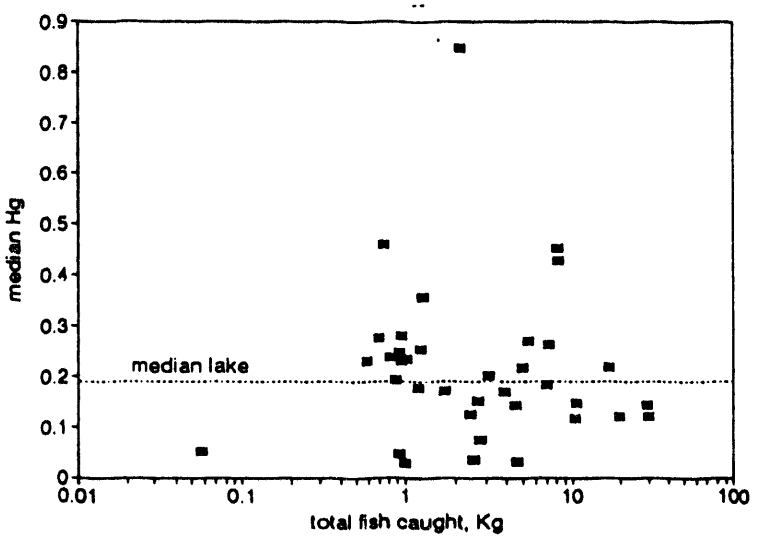


Figure 6.5. Median Hg concentration by weight of fish caught, for each of 49 Upper Michigan lakes. Data from Gloss et al. (1990) and Cusimano et al. (1989).

The detailed analysis of mercury in Upper Michigan fish (Appendix B) was limited by the relatively few observations, but suggests the following conclusions:

1. The effects of lake characteristics on fish mercury levels vary by species; maximum levels for different species (after accounting for weight) will not occur in the same lake, in general. This in turn suggests that assessments should be specific to defined fish species, and that generic bioaccumulation factors should not be used.

2. When statistically significant, the effects of DOC and pH on Hg have the expected signs, which reinforces the conventional wisdom about the relevant mechanisms for bioaccumulation of Hg.

3. The relative size of the catchment area for atmospheric mercury was never a statistically significant parameter, in spite of its large range, and was negative in 3 of the 4 cases. This finding suggests the influence of other factors with regard to terrestrial transport of deposited Hg, which could include the nature of the watershed terrain and ground cover and lake volume. It would appear from these data to be problematic to assume that a substantial fraction of the Hg deposited in the watershed will end up in fish, although doing so constitutes a conservative assumption.

6.1.3 Data on Mercury in Freshwater Fish from Other Sources. There are several additional sources of data on mercury in freshwater fish; a summary of 3 of them is provided in Table 6.1. The average and median mercury concentrations for fresh-water species sampled by EPA are 0.19 and 0.14 μg/g, respectively. The standard deviation (SD) and geometric standard deviation (GSD) are 0.15 and 2.46, respectively. These values provide an estimate of the maximum likely mercury concentration (based on 1.65 standard deviations, or approximately the 95th percentile) of about 0.67 μg/g. Since the mercury concentration can never be negative and the maximum species average was 0.68, it is concluded that the distribution among species in Table 6.1 is approximately log normal and may be represented by these statistics.

Figure 6.6 compares the results from the 3 surveys represented in Table 6.1. In most instances, the NOAA data have higher mercury levels, suggesting a downward trend over time that might be due to a tendency toward catching smaller fish. It is also possible that some of the discrepancy may be explained by changes in laboratory techniques over time.

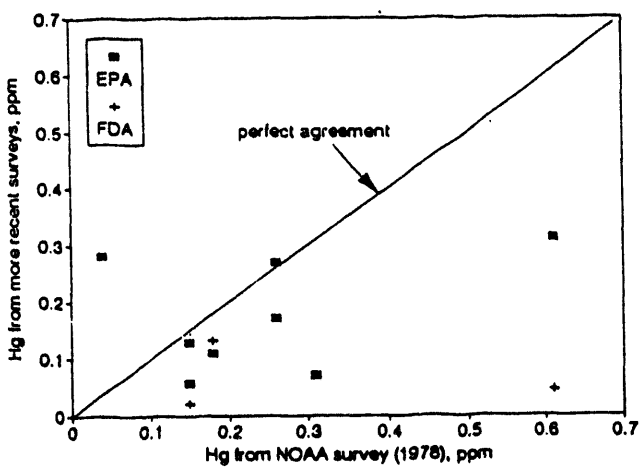


Figure 6.6. Comparison of Hg levels in freshwater fin fish as determined by EPA (1992) and NOAA (1978).

Table 6.1 Mercury Levels in Freshwater Fish According to Three Surveys

	EPA data (1992)			FDA data (1992)			NOAA data (1978)		
species	av'g Hg	std dev	# sampled	av'g Hg	range	# sampled	av'g Hg	max Hg	# sampled
American Eel	0.190		1						
Bass	0.267	0.025	3						
Bigmouth Buffalo	0.065	0.035	4						
Black Buffalo	0.120		1						
Black Bullhead	0.073	0.048	2						
Black Crappie	0.150	0.105	4						
Black Drum	0.057	0.017	3				0.150	0.60	137
Black Redhorse	0.160		1						
Blacktail Redhorse	0.580		1						
Blue Catfish	0.209	0.141	4						
Bluegill	0.268	0.296	5				0.260	1.01	49
Bocaccio	0.020		1						
Bowfin	0.300	0.280	2						
Bridgelip Sucker	0.240	0.172	4						
Brook Trout	0.105	0.025	2	0.025	0-0.13	5			
Brown Bullhead	0.093	0.047	3						
Brown Trout	0.128	0.083	9						
Bullhead	0.025		1						
Carp	0.109	0.136	153	0.130		1	0.180	0.54	52
Catfish	0.128	0.091	13	0.020	0-0.06	11	0.150	0.38	35
Chain Pickerel	0.677	0.280	3						
Channel Catfish	0.121	0.082	28						
Chiselmouth	0.025		1						
Chub	0.140		1						
Coast Sculpin	0.025		1						
Composite Bottom	0.160		1						
Crappie	0.170	0.000	4				0.260	1.39	212
Crayfish (whole)	0.025		1						
Creek Chubsucker	0.030		1						
Croaker	0.050	0.000	2						
Cutthroat Trout	0.070		1						
Dolly Varden	0.050		2						
Flathead Catfish	0.223	0.258	8						
Freshwater Drum	0.157	0.054	3						
Gizzard Shad	0.025		1						
Golden Redhorse	0.240		1						
Goldfish	0.210		1						
Gray Redhorse	0.150		1						
Green Sunfish	0.420		1						
Greenfish	0.070		1						
Greyfin Sucker	0.070		1						
Hardhead Catfish	0.380		1						
Lake Chubsucker	0.130		1						
Lake Trout	0.220		1						
Lake Whitefish	0.080		1						
Largescale Sucker	0.110	0.030	2						
Large-mouth Bass	0.448	0.331	69	0.310		1			

Table 6.1 (cont'd) Mercury Levels in Freshwater Fish According to Three Surveys

	EPA data (1992)			FDA data (1992)			NOAA data (1978)		
species	av'g Hg	std dev	# sampled	av'g Hg	range	# sampled	av'g Hg	max Hg	# sampled
Longear Sunfish	0.145	0.015	2						
Longnose Sucker	0.053	0.026	2						
Mountain Whitefish	0.100		1						
N. Redhorse	0.220		1						
North Hogsucker	0.230		1						
Perch				0.130	0-0.31	3			
Northern Pike	0.311	0.166	9	0.044	0-0.16	5	0.610	1.71	87
Pickerel	0.640		1						
Quillback	0.040		1						
Quillback Carpsucker	0.366		1						
Rainbow Trout	0.047	0.031	8						
Red Drum	0.337		3						
Redbreast Sunfish	0.070		1				0.310	1.20	174
Redear Sunfish	0.110		1						
Redeye Bass	0.550		1						
Redhorse Sucker	0.256	0.220	16						
River Carpsucker	0.063	0.038	2						
Rock Bass	0.160	0.036	3						
Rotten (catfish)	0.010		1						
Sacramento Sucker	0.122	0.089	3						
Sauger	0.367	0.062	3						
Sculpin	0.253	0.055	2						
Shorthead Redhorse	0.125		1						
Silver Redhorse	0.140		1						
Small-mouth Bass	0.323	0.259	22						
Small-mouth Buffalo	0.158	0.088	4						
Spot	0.260	0.360	2				0.040	0.18	60
Spotted Bass	0.410	0.170	2						
Spotted Drum	0.020		1						
Spotted Sucker	0.123	0.057	10						
Squawfish	0.420	0.306	9						
Striped Bass	0.370	0.020	2				0.750	2.00	231
Sucker	0.120	0.083	37	0.110	0.05-0.19	3			
Tilapia	0.048	0.032	2	0.010	0-0.02	8			
Trout	0.113	0.066	2	0.025	0-0.13	5			
Walleye	0.494	0.387	21						
Warmouth	0.260		1						
White Bass	0.352	0.209	6						
White Catfish	0.060	0.022	3						
White Crappie	0.227	0.239	7						
White Perch	0.108	0.101	3						
White Sucker	0.116	0.077	36						
White Surfperch	0.130		1						
Yellow Bullhead	0.460		1						

Data sources: U.S. EPA (1992). FDA: Cramer (1992). NOAA: Hall et al. (1978).

The variation among these data and with respect to the Michigan data discussed above is considerable, which is further suggestive evidence of the role of environmental factors. For example, Sloan and Schofield (1983) investigated the influence of Adirondack lake pH and of the effects lake liming as a means to reduce pH on mercury levels in brook trout, and found that acid drainage lakes had the highest mercury levels regardless of liming status.

Sorensen et al. (1990) sampled northern pike in 65 Minnesota lakes and found Hg levels ranging from 0.14 to 1.52 μg/g, after standardizing to a fish length of 55 cm. Thus, the variation just due to location (i.e., lake), implied a GSD of about 2.0, while the Michigan data imply a GSD of about 3.0 when both fish species and length are also varying. The lakes sampled by Sorensen et al. were much larger on average than the Michigan lakes (328 ha vs. 9 ha, and they found a negative relationship between lake size and lake mercury content. The mercury levels in pike in the two data sets are thus seen to be consistent. Sorensen et al. (1990) also found that the strongest correlates with mercury in northern pike (after adjusting to a standard length) were mercury levels in lake water and in zooplankton, which in turn were correlated with a number of lake and watershed physical and chemical factors. Their regression equation for northern pike implied a small effect of lake pH, about 7% increase for a pH change from 7 to 5.

Walleye Hg concentrations were examined in 219 Wisconsin lakes by Lathrop et al. (1991). They found that concentrations increased with fish size and decreased with lake alkalinity. The least acid lakes had Hg levels from 0.2 to 0.5 μg/g; the most acid lakes had Hg concentrations from 0.3 to 1.3 μg/g. However, only 42% of the individual fish variation in Hg was explained by these parameters.

Richardson and Currie (1993) cite the following data on mercury in fish found in Ontario lakes:

			Hg level (μg/g)	
species	no. sampled	no. lakes	median	range
lake trout	2968	182	0.20	0.01-6.13
walleye	5620	334	0.50	0.01-5.42
northern pike	4657	360	0.45	0.02-5.00
all combined	13245	607	0.39	0.01-6.13

These data are reasonably consistent with those from Upper Michigan but are weighted towards the high-Hg species. The ranges of Hg imply GSD values of about 3.0 based on Figure A.4 (see Appendix), which is also reasonably consistent with the U.S. survey data.

Hg data for 4 species from Lake St. Clair, MI (R. Hesselberg, personal communication, 1993) show a strong downward trend in Hg from 1970 to 1976, especially after the data are adjusted to a common weight for each year. This trend is due to the phase-out of a chlor-alkali plant; the gradual decline in the levels of Hg in fish reflects the lag in the biological processes after the Hg input was reduced. Presumably, a similar lag would be seen if a new source were introduced.

It would appear from these results that a comprehensive assessment of the effects of lake morphology and chemistry will require data on lake type (and probably watershed/lake area ratio and the types of terrain and ground cover in the watershed), pH and DOC. The relative frequency of drainage vs. seepage lakes varies regionally; seepage lakes are common in Florida and in the upper Midwest, but less common in the Northeast (J. Baker, personal communication, September 13, 1993).

6.2 Mercury in Marine Species

The most comprehensive source of data on mercury in marine species is the NOAA survey reported by Hall et al. (1978), which is characterized by relatively large numbers of samples for each species (20-1000), almost 19,000 fish samples in all. A more recent source of marine mercury data was provided by the U.S. Food and Drug Administration, based on very limited sampling (Cramer, 1992). These data are compared to the EPA data set in Table 6.2, and crossplots are provided for comparison in Figures 6.7a and b. The average and median mercury

contents of shellfish sampled by EPA are 0.051 and 0.026 μg/g, respectively. For marine fin fish, the figures were 0.21 μg/g and 0.11 μg/g, respectively. Note that the highest mercury levels are found in swordfish and shark, which are species whose consumption by humans tends to be limited. Mercury levels in the more commonly consumed marine species, including shrimp, flounder, pollock, salmon, and whitefish, are much lower. Tuna is discussed below, as are weighted average mercury levels.

The effect of the local marine environment in Southern California was assessed for dover sole by Fowler et al. (1975). They found no difference in average mercury levels between 11 fish taken from a coastal basin contaminated with toxic metals in the sediments and 12 fish from an uncontaminated coastal basin off Santa Barbara. This raises questions as to the role played by local dry deposition and urban run-off (although Southern California was probably not the best place to test this hypothesis for mercury because of its typically modest precipitation volumes).

Figures 6.6 and 6.7 show consistently higher mercury levels in the 1978 NOAA survey. The question arises as to whether the differences may be due to a decreasing temporal trend or to differences in the sampling characteristics. Note from Table 6.2 that the older data set has far more samples per species, but the mean and medians of log normal distributions are unaffected by the number of samples. The conclusion thus follows that the data support a decreasing trend in average mercury in fish, but it is not possible to determine whether this is a real trend or an artifact of sampling and analytical procedures. Figure 6.8 tries to estimate GSD's from the ratios of maximum to average concentrations, recognizing that these will be underestimates because the medians are always lower than the averages in such distributions. Most of the data fall between GSDs of 1.5 and 3 and there is no consistent difference between freshwater and marine species in this regard.

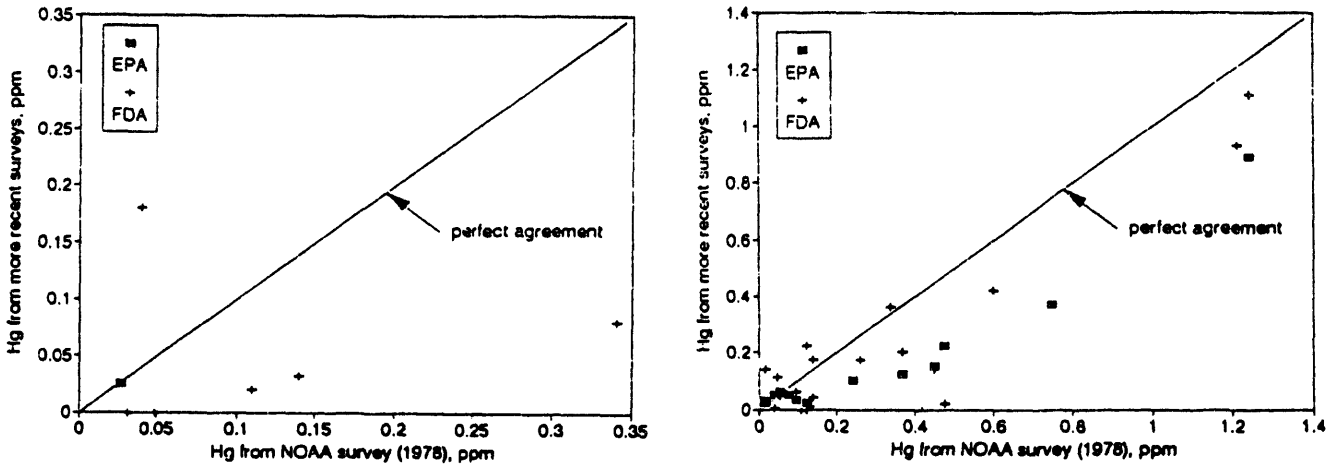


Figure 6.7. Comparison of Hg levels in marine species as determined by EPA (1992) and NOAA (1978). (a) shellfish. (b) marine fin fish.

Table 6.2 Mercury Levels in Marine Species According to Three Surveys

	EPA data (1992)			FDA data (1992)			NOAA data (1978)		
species	av'g Hg	std dev	# sampled	av'g Hg	range	# sampled	av'g Hg	max Hg	# sampled
Abalone							0.018	0.12	30
Clam				ND		5	0.049	0.26	584
Crab				0.032	0-0.09	34	0.140	0.61	314
King Crab							0.070	0.24	93
Lobster				0.060	0-0.14	3	0.340	1.60	770
Spiny Lobster				0.020		1	0.110	0.37	65
Shrimp				0.180	0-2.0	11	0.040	0.33	353
Mussel	0.000		1						
Shellfish	0.000		1						
Soft Shell Clams	0.017	0.009	3						
Squid, Octopus				ND			0.031	0.40	339
Scallops							0.050	0.22	138
Oysters	0.025		1				0.027	0.43	280
Pacific Oysters	0.025	0.000	2						
Flounder	0.033	0.020	14	0.060	0-0.08	4	0.096	0.88	1179
Ocean Perch				0.008	0-0.03	4	0.130	0.59	268
True Cod	0.025		1	trace		2	0.125	0.59	134
Haddock				trace		1	0.110	0.36	88
Atlantic Croaker	0.025		1	0.220	0.13-0.32	2	0.124	0.81	217
Surf Smelt	0.030		1				0.016	0.06	53
Flathead Sole	0.033	0.008	1						
Pollock				0.040	0-0.10	7	0.140	0.93	227
Atlantic Salmon	0.050	0.000	2	0.005	0-0.11	22	0.040	0.21	806
Giant Kingfish	0.050		1				0.078	0.33	19
Whitefish	0.060		1	0.044	0-0.31	8	0.054	0.23	86
Blackfish	0.060		1						
Stingray	0.080		1						
White Sturgeon	0.095	0.005	2						
Spotted Seatrout	0.100	0.036	3				0.242	1.19	201
Weakfish	0.110		1	0.110		1			
Mackerel				0.110	0-0.23	3	0.048	0.19	111
Pink Salmon	0.110		1						
Diamond Turbot	0.110		1						
Bluefish	0.120	0.061	9	0.200		1	0.370	1.26	94
Brown Trout	0.128	0.083	9						
Mullet	0.025		1	0.140	0-0.27	2	0.016	0.28	191
Red Snapper	0.150		1	0.140	0.07-0.26	5	0.450	2.17	759
Tuna				0.170	0-0.75	245	0.260	0.90	261
Dolphin				0.170	0.12-0.21	3	0.140	0.53	73
Saltwater Catfish	0.220	0.150	7	0.020	0-0.06	11	0.475	1.20	81
Bonito				0.360		1	0.340	0.74	435
Grouper				0.420	0.35-0.48	2	0.600	2.45	928
Chinook Salmon	0.320		1						
Sheepshead	0.323	0.221	3						
Striped Bass	0.370	0.020	2				0.750	2.00	231
Swordfish				0.930	0.26-3.22	99	1.210	2.72	115
Longnose Gar	0.660		1						
Leopard Shark	0.890		1	1.110	0.23-2.95	71	1.240	4.53	588

Data sources: U.S. EPA (1992). FDA: Cramer (1992). NOAA: Hall et al. (1978).

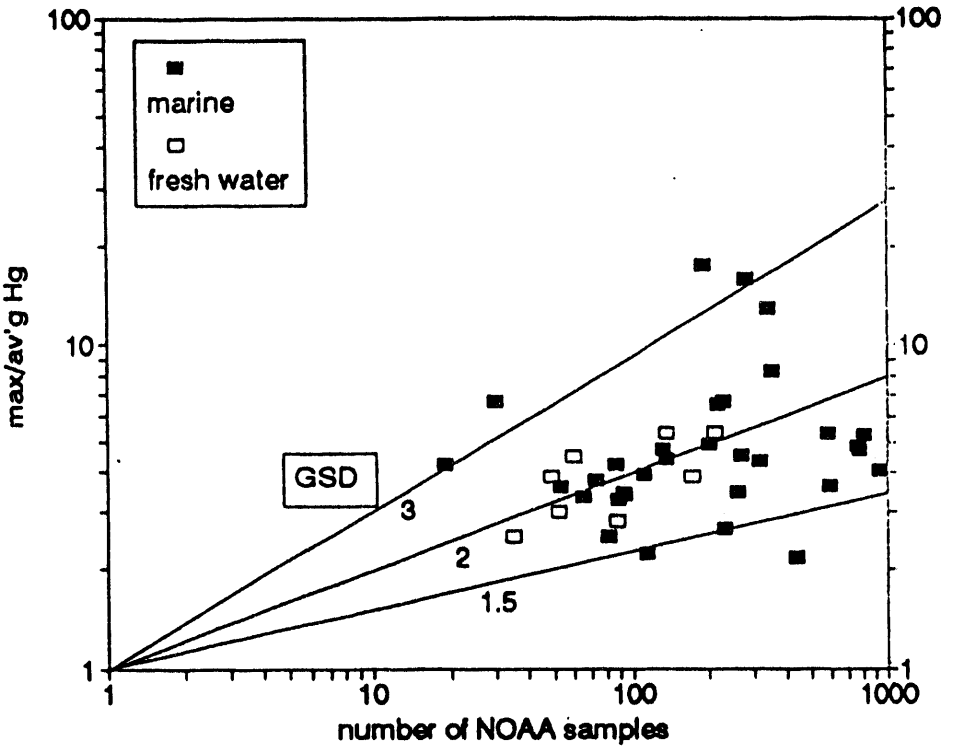


Figure 6.8. Ratios of maximum to average Hg concentrations in NOAA fish samples, by species. Lines of constant geometric standard deviations (GSD) are shown.

6.3 Mercury in Canned Fish Products

Canned tuna fish represents an important portion of average American fish consumption, and special attention is warranted to obtain an accurate measure of its mercury content. The other important canned fish species are salmon and sardines, both of which have average MeHg around 0.02 μg/g (Hall, 1974). Hall also analyzed 20 samples of a variety of canned seafood for methyl and total mercury. The average methylmercury for tuna was 0.32 μg/g, which was about 89% of the total mercury. The GSD of this distribution was about 1.5.
The 1978 NOAA survey contained total mercury data for three tuna species:

species	# of samples	av'g Hg	max Hg
light skipjack	70	0.144	0.385
light yellowfin	115	0.271	0.870
white	76	0.350	0.904

The weighted average of these samples is 0.26 μg/g and the GSD is about 1.8. If the data of Hall (1974) are used to correct the data to a methylmercury basis, the average would be about 0.23 μg/g.

Carrington (1993) derived a distribution for mercury in canned tuna which had mean, median, and maximum values of 0.195, 0.15, and 0.87 μg/g, respectively, and SD and GSD values of 0.144 and 2.16. This GSD value, representing mercury differences due to fish age and weight and environment, is about the same as the GSD's found among species. It is assumed that these data are for total Hg, of which MeHg represents about 90% (Hall, 1974). If so, the corresponding MeHg level would be 0.175 μg/g.

Cramer (1992) reports a mean concentration of methylmercury in tuna for 1991-2 (variety not identified) of 0.17 μg/g, with a maximum of 0.75 μg/g based on 245 samples; these data are seen to be consistent with the earlier samples. Data from the Seychelles Islands (Mathews, 1983) show somewhat higher tuna mercury levels (based on 5 samples each): skipjack, 0.29 μg/g; yellowfin, 0.23 μg/g; and dogtooth, 1.22 μg/g. These data showed that fish weight was an important parameter (Figure 6.9); dogtooth and skipjack tuna had about the same mercury levels after accounting for weight, but yellowfin tuna was an order of magnitude lower. In 1992, skipjack and yellowfin accounted for the bulk of commercial landings of tuna by U.S. vessels; 66% and 29%, respectively.) Figure 6.9 also supports the existence of important interspecies gradients in mercury for marine fish taken from the same waters, after accounting for differences in weight. This figure reflects the results of a multiple regression of log(Hg) against log(weight), with species types as dummy variables; all variables were statistically significant and the effect of weight was very nearly linear. Note that the mercury content of fish could also be related to fish length or age; bioaccumulation results in part from the relatively long half-life of MeHg in fish. For these purposes, fish weight is preferred rather than age, since weight has more relevance to the numbers of fish meals and to actual MeHg doses.

Further data on average methylmercury levels in canned tuna were reported by Yess (1993), who found an average level of 0.17 μg/g in 220 samples analyzed by various FDA laboratories. White tuna had higher Hg levels and tuna packed in oil had substantially lower levels. The median mercury level was 0.14 μg/g. However, the minimum detectable level (MDL) was 0.10 μg/g, and these values were treated as 0.0; treating them as half of the MDL would increase the mean to 0.18 μg/g. The GSD was about 2.3 after this correction was made. These data on mercury levels in tuna support a decreasing trend over time during the 1970s; it is not clear whether this trend might be continuing. Anecdotal evidence suggests that such a trend could result from the tendency to harvest smaller fish, in part because of the increasing scarcity of the resource; smaller fish will have lower mercury levels, on average.

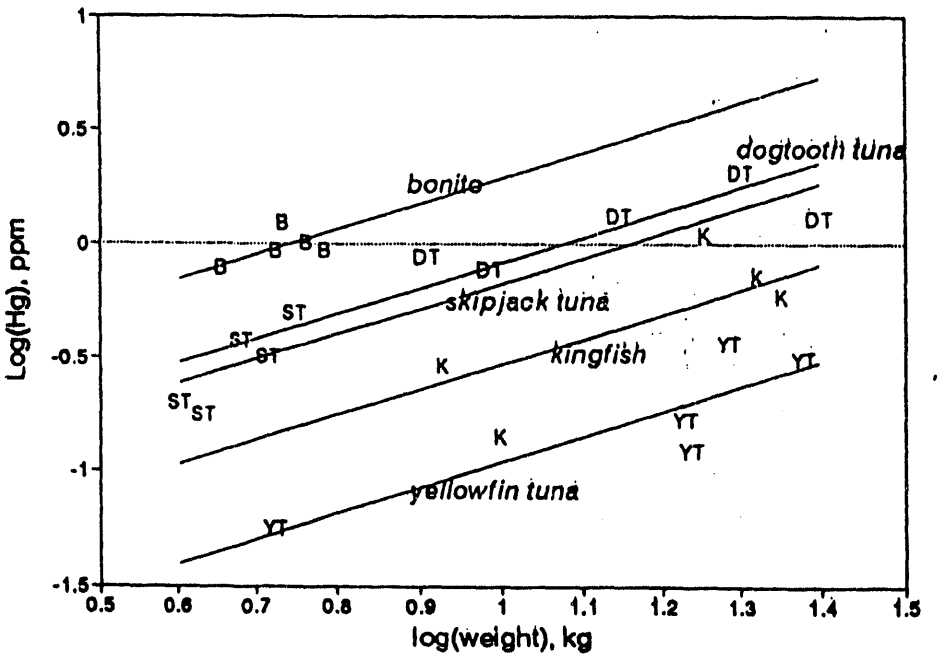


Figure 6.9. Mercury concentrations in selected species of fish caught in the Seychelles Islands. Lines are best-fit slopes from a multiple regression against weight and dummy variables for each species. Plotting symbols: B=bonito, DT=dogtooth tuna, K=kingfish, ST=skipjack tuna, YT=yellowfin tuna. Data from Mathews (1983).

6.4 Summary of Data on Mercury Levels In Fish

Table 6.3 summarizes the parameters of the distributions of mercury in fish for the major data sets that were examined, based on the assumption of log normal distributions. In summary, substantial variations in the mercury concentrations reported for a given fish species are found. Some of this may be due to differences in laboratory techniques over time and the use of total Hg vs. MeHg, but the main sources of variability within a given species are fish size and age, and for freshwater species, lake DOC and pH. The effect of watershed/lake surface area ratio on Hg content could not be identified from these data (Upper Michigan data; see Appendix B). The variations among species averages were of about the same order as those within species, and were probably related to the trophic level of the fish. If tunafish are considered separately, there was no difference between mercury levels in marine shellfish and marine fin fish, when weighted by the quantities caught. Mercury levels for freshwater game fish were higher and more variable, in general. There was the suggestion of a downward trend in Hg levels over time, especially for canned tuna, but this could not be confirmed statistically. Better data on current mercury levels in seafood are badly needed.

Table 6.3 Summary Data on Mercury Concentrations in Fish

Reference	Year (data)	Species	Location	Median Hg	GSD	Notes
				(μg/g)		
EPA,	1992	all	U.S.	0.12	2.5	
		freshwater	U.S.	0.14	2.5	
Gloss et al.		freshwater	Michigan	0.21	2.8	(within lakes)
(1990)		"	"	0.17	3.0	(between lakes)
		"	"	0.22		(indiv. fish)
Sorensen et al.		freshwater	Minnesota	0.39	2.0	(standardized (1990) to 55 cm)
Richardson & Currie		freshwater	Ontario	0.39	3.0	
Hall (1974)		tuna		0.32	1.5	
Hall (1978) NOAA	1978	tuna		0.23	1.8	
Carrington		tuna		0.15	2.16	
Cramer		tuna		0.17		
Yess		tuna		0.14	2.3	

7.0 FISH CONSUMPTION RATES AND ESTIMATES OF POPULATION Hg DOSES

As mentioned previously, for the purposes of this assessment it is assumed that the major pathway for MeHg is ingestion of seafood. This is less likely to be true for low doses, but the assessment is aimed at high doses of MeHg. The population-average rates of consumption of fish may be estimated from fisheries production data, but their distributions must be obtained from surveys. One of the important statistics needed from surveys is the fraction of non-consumers in the population. The mercury concentration data referred to in this section were taken from Tables 6.1 and 6.2; dose estimates based on numbers of meals consumed assume fish servings of 200 g per meal.

7.1 Production Statistics

The National Marine Fisheries Service (NMFS) publishes an annual report that gives U.S. production statistics and trends, with emphasis on marine species. Great Lakes fish production statistics are available from the U.S. Fish and Wildlife Service's Great Lakes Laboratory; this seems to be the only reliable source of production data on freshwater species.

7.1.1 Marine Species. The National Marine Fisheries Service (NMFS) publishes an annual report on the U.S. commercial fish catch and its disposition, including human consumption. Figure 7.1a presents consumption trends since 1910; overall consumption has risen since the late 1960s, peaking in 1986. The subsequent decline in consumption has been attributed to a world-wide decline in the resource (Brown et al., 1993), and, presumably as a result, prices have risen. This situation may add to the demand for recreational fish catches. However, the New York Times (August 9, 1993) reported that regional (NY, NJ, CT) commercial marine catches have increased 59% over the past 5 years and recreational fishing has declined, in part because of a limit imposed on the number of bluefish that may be taken per day.

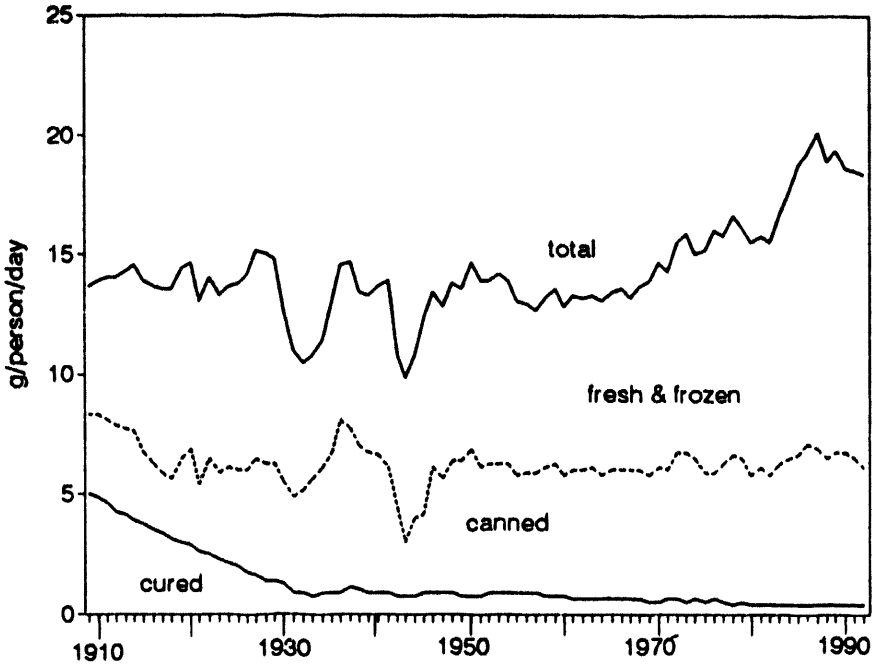
Ahmed (1991) cited the following 1990 NMFS per capita production figures:

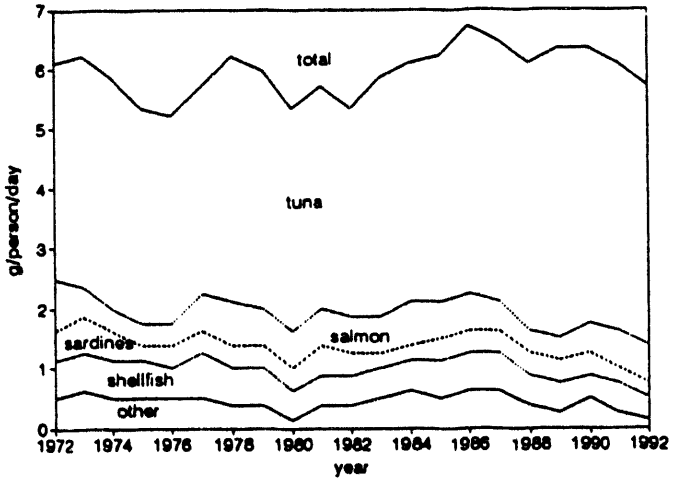
total commercial fish and shellfish:	19.8 g/d
fin fish	8.8 g/d
shellfish	4.2 g/d
canned seafood products	6.3 g/d

In addition to the 4 billion pound commercial catch, NMFS reports that 600 million pounds of fin fish were caught by 17 million anglers, and 200-300 million pounds of shellfish were harvested (an estimate not based on statistics). Assuming that 50% of the catch was eaten and was distributed over the anglers' households, additional 8 g/d is estimated for that portion of the population. Assuming that the figure for the recreational shellfish harvest represents only edible meat, these figures would be increased by another 30-50%, but there is no way to identify the population involved in recreational shellfishing. Ahmed (1991) also reports that consumption of commercially caught seafood increased 60% from 1979 to 1989; the NMFS figures published in 1993 show an increase of only 20%.

The 1992 U.S. seafood consumption rate from commercial sources is about 18 g/d per person, of which about 6 g/d is from canned product and about 0.3 g/d represents smoked or salted fish (NMFS, 1993). The largest source of the increase in fish consumption was for fresh and frozen fish. All of these production statistics are based on the entire U.S. population, rather than the population of fish-eaters.

Figure 7.1b shows recent trends in the consumption of canned fish products. Tuna fish accounts for the bulk of this category, currently about 4.5 g/d, on average. Salmon and shellfish are the next most important canned fish categories. Figure 7.1c displays recent trends for the fresh and frozen fish categories (this is not a cumulative plot), which totaled about 3 g/d for shrimp, 3.5 g/d for fillets and steaks, and 1.2 g/d for fish sticks and portions.





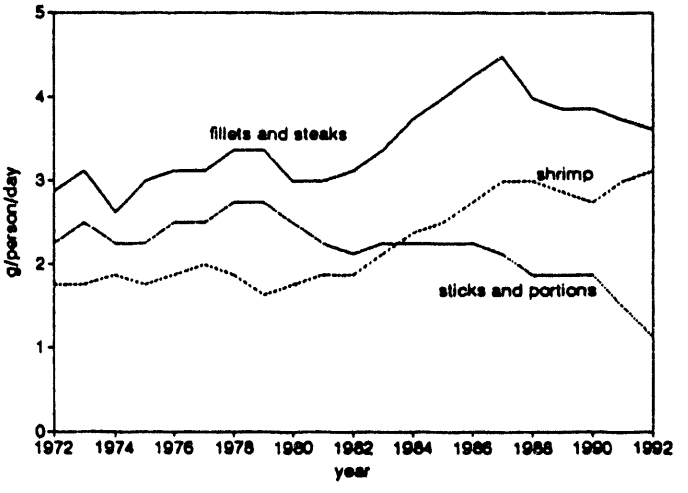


Figure 7.1. Trend in U.S. per capita fish consumption. (a) Major categories (cumulative plots). (b) selected canned fish products (cumulative plots). (c) Trends in consumption of selected fresh and frozen seafood (individual trend lines). Data from National Marine Fisheries Service (1993).

It is assumed that this last category represents mainly whiting and allied species, which tend to be low in mercury. These 3 categories account for about 8 g/d of the approximately 12 g/d "fresh and frozen" fish in Figure 7.1a.

Figure 7.2 displays the distribution of commercial fish landings by location of the ports. The most important region is the Pacific Coast and Alaska, followed by the Gulf Coast. Great Lakes landings (freshwater fish) accounted for only a tiny fraction (0.3%) of the total in 1992; these fisheries have been declining in commercial importance for decades.

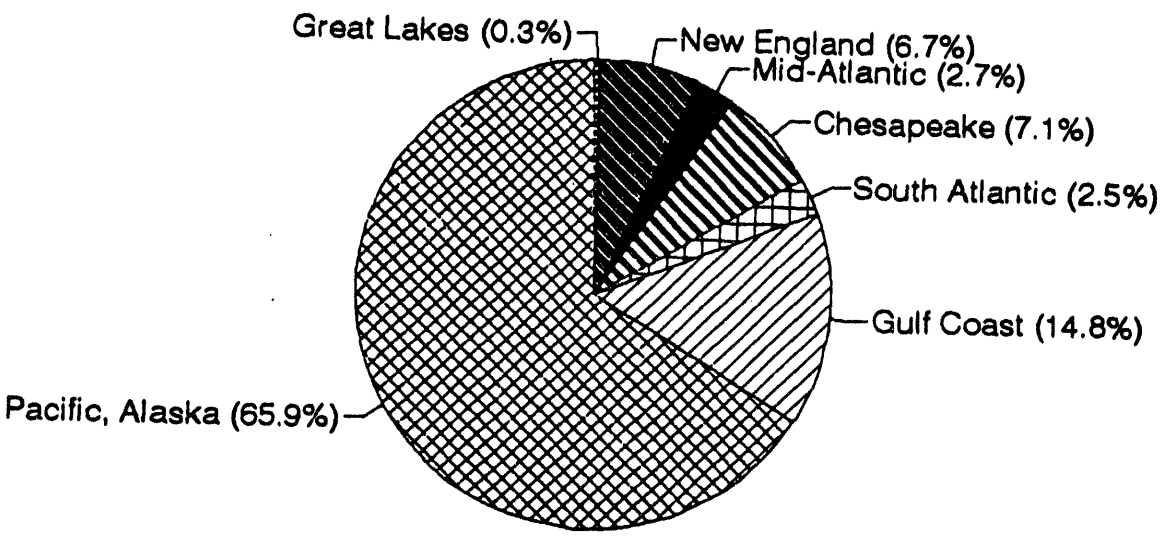


Figure 7.2. Live weight shares of the 1992 U.S. commercial domestic fish catch, by geographic region of landing port. Data from National Marine Fisheries Service (1993).

7.1.2 Freshwater Species. The Great Lakes Research Center of the U.S. Fish and Wildlife Service supplied data on 1991 commercial fish production, by state and individual Great Lake. The total catch for human food was given as about 30 million pounds, which checks reasonably well with the 39 million pound figure (live weight) reported by NMFS for Great Lakes ports. This amounts to about 0.6 g per person per day for those 7 states (excluding Pennsylvania because of its limited lake shoreline), so that it is apparent that Great Lakes fish must not constitute a major source of fish food, even for the local region. In addition, whitefish was the most common species, which is very low in mercury. The distribution of mercury intake by species for this fish source is given in Figure 7.3; yellow perch slightly exceeds whitefish, because of its much higher Hg content.

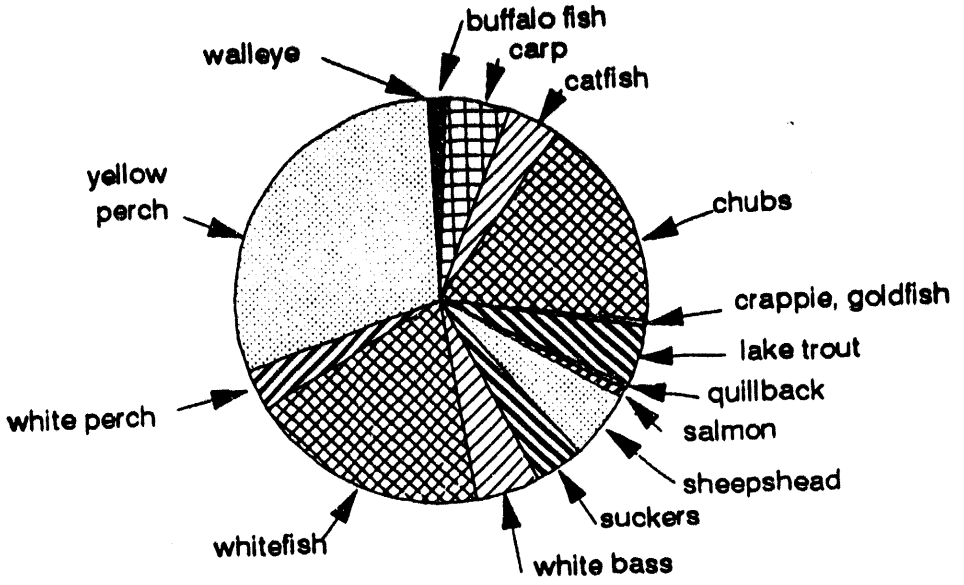


Figure 7.3. Shares of the total mercury burden in 1991 Great Lakes commercial fish landings, by species. The weighted average concentration was 0.11 ppm. Data on fish landings from U.S. Fish and Wildlife Service; data on Hg concentrations from various sources.

7.2 Surveys

Data from production statistics provide overall consumption rates, but not their distribution within the populations at risk. This type of information can only come from surveys, several of which are reviewed in this section. Although the extant fish consumption surveys vary considerably in design, some useful information is available from almost all of them. All of these figures are given in units of g/person/day (unless otherwise stated).

7.2.1 NHANES. The U.S. National Center for Health Statistics surveyed about 28,000 people of all ages from April 1971 to June 1974, as part of their National Health and Nutrition Examination Survey (NHANES) (Carroll and Abraham, 1979). This survey showed that about 40% of adult females and about 45% of adult males "seldom or never" ate fish/shellfish and that most of the remaining population ate fish 1-6 times a week. For persons below the poverty level, 0.6% of the sample ate fish every day, 48.4% ate 1-6 times per week, and 51%, seldom or never. For those above the poverty line, 0.1% ate fish twice a day, 1% ate every day, 54% 1-6 times per week, and 44.9%, seldom or never. Because most of the population fell into the very broad categories of "seldom or never" or 1-6 meals per week, this survey was not very useful for estimating detailed consumption rates within that range. "Seldom" could be as often as once per month, for example. However, these data do confirm that seafood was not a staple element of the U.S. diet in the early 1970s.

7.2.2 The NPD Survey - Great Lakes States. Survey data from the National Marine Fisheries Service were made available by the FDA (G. Cramer, personal communication, 1993), consisting of portions of a 1-year national sample of about 7000 families. About 26,000 panelists completed this survey, reporting the species of fish that they had consumed within a month; the panel was subdivided evenly to cover an entire year. This portion of the NPD survey pertained only to fish consumers in the 8 Great Lakes states, and give average daily consumption of 9 selected freshwater species and of total fish consumption. The actual survey sample size for the 8 states was 2201 persons and was conducted by NPD Research during 1973-74 (funded by the Tuna Research Institute). The consumption of total and selected freshwater fish are given in Table 7.1; the marine fish consumption rates were obtained by difference.

Table 7.1 Results of a Survey of Fish Consumption (g/d) in 8 Great Lakes States, 1973-74

State	Selected fresh-water	Marine fish	Total
IL	8.9	13.0	21.9
IN	6.8	9.6	16.4
MI	7.3	9.9	17.2
MN	9.8	8.2	18.0
NY	6.2	18.3	24.4
OH	8.2	11.4	19.6
PA	6.6	12.4	18.9
WI	10.2	9.5	19.7

Source: NPD Research, Inc.

For all 8 states. the average per capita consumption of selected freshwater species was 7.7 g/d; marine species averaged 12.6 g/d, for a total of 20.2 g/d, which is substantially higher than the national average for those years (15.5 g/d). To place these figures in the context of the 1992 production values cited above, the non-canned portion (as estimated from national production figures) should be multiplied by 1.34 to account for the temporal trend; this gives an estimate of 24.7 g/d for this segment of the population on a 1992 basis. Incidentally, the EPA Exposure Factors Handbook cites a figure of 6.5 g/d for the average non-marine fish consumption rate, averaged over the entire population, including nonconsumers. Comparing the national total fish consumption figure from this survey, as reported by by Goyer et al. (1985), with the NMFS commercial production totals for those years shows that about 13% of fish consumption is from noncommercial sources. The NPD survey (as reported by EPA) also showed that people in Census Regions with coastal states consumed about 20% more fish on average than those in the interior regions. The GSD for the data reported by EPA for the NPD survey was about 2.15.

The value for marine species in Table 7.1 is slightly lower than the value for commercial landings from Figure 7.1a, whereas it would be expected to be higher since non-fisheaters were excluded from this survey. The difference could be due to wastage or non-food use of some portion of the catch, poor recall from the survey, or errors in the weights of the average meals assumed (which increased with age and were higher for adult males). The minimum, 90th percentile, 99th percentile, and maximum values for all 8 states were 0.1, 15.7, 36.8, and 160.2 for selected freshwater species, and 0.3, 41.7, 92.3, and 201.4 for total fish consumption. This maximum value corresponds to about 1.2 meals per day for an adult male and 1.5 meals per day for an adult female. The implied GSDs of these distributions are about 2.5 for freshwater species and 2.0 for all types of fish. Figure 7.4 gives the distribution of mercury dose by freshwater species; pike is the main source. This survey did not provide data on the fraction of non-fisheaters in those states, but about 95% of the national sample reported eating some seafood.

7.2.3 USDA Surveys. The U.S. Department of Agriculture (USDA) surveyed food consumption by 38,000 individuals stratified into 16 age-sex groups (Pao et al., 1982). These data were collected for 3-day periods in the 48 conterminous states; only 24.5% of the sample reported eating any fish or shellfish within a 3-day period. 0.4% reported eating fish on all 3 days; the median number of fish meals for fish consumers was 1.2 in 3 days, or 146 meals per year. The median amount consumed (by fisheaters) in 3 days was 37 g/d, which is substantially higher than any of the other sources of data; the GSD (as reported by EPA, 1990) was about 2.3, which is also slightly higher, probably because of the short period of record. Assuming that the 75.5% of the population not eating fish during the 3-day survey were in fact non-fisheaters, the national average consumption rate would be about 9 g/d, which seems too low by about a factor of 2.

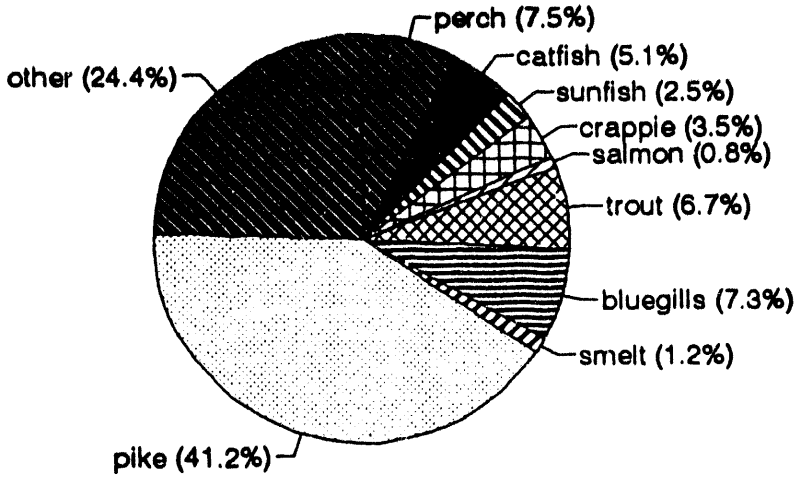


Figure 7.4. Shares of dietary Hg based on freshwater fish consumption data from 8 Great Lakes states. Consumption data from the NPD survey (1973-74).

For adults from age 35 to 75, these consumption rates are slightly higher. Assuming a weekly fish consumption cycle for everyone, i.e., that some portion of the 75.5% apparent nonconsumers ate fish on the other 4 days, and that the other 24.5% did not eat fish on the nonsurvey days, the average long-term consumption rate of all users in this survey would be diluted by 3/7, yielding a median daily rate of about 16 g/d on a long-term rather than a 3-day basis. This value agrees fairly well with the other surveys and the production data. Another approach is to multiply the mean serving size by 52 to get an annual rate; this results in a mean daily rate (for the whole population) of 16.6 g/d, which also checks fairly well with other estimates.

Even though the USDA survey may not be very useful for absolute values, these data may be useful for estimating relative distributions by age and sex, by geographic region, and by other demographic characteristics. The survey also gives breakdowns for shrimp, canned tuna, and fin fish other than canned, dried, and raw. Figure 7.5 plots consumption rates by household income for the Northcentral states, for central cities, suburbs, and nonmetropolitan areas, on the basis of "users" (during the 3-day period) (Figure 7.5a) and the whole population (Figure 7.5b). Suburban respondents tended to consume the least fish at most income levels, and central city residents the most; however, high income nonmetropolitan respondents showed the highest consumption rates, abou' a meal every other day (during the 3-day survey period).

Although the USDA survey does not present actual figures on annual mean consumption, it does give data in the same formats for all fish, canned tuna, shrimp, and "fin fish other than canned, dried and raw." By comparing similar statistics for these various categories, estimates of the fractional consumption rates may be obtained, assuming that the survey respondents had similar eating patterns (i.e., 3-day vs. long-term) for each category. For example, 0.4% of the sample are some fish on all 3 days, 0.1 % ate fin fish, 0.1% ate tuna, and less than 0.1% ate shrimp. Similar relationships are seen among species for the other frequencies of consumption, but for these purposes, serving sizes must be considered. To do this, the product of the quantities eaten per occasion and the fraction of the population using the product during the 3-day period are used. This gives the following breakdown of total fish consumed:

canned tuna:	18.3%
fin fish:	73.3%
shrimp:	5.8%

The remaining 2.6% is assumed to be other forms of seafood. The tuna share of 18.3% translates into 3.3 g/d by this methodology, based on a value of 18 g/d (from commercial production statistics) for all types of seafood. Since the USDA survey includes fish from noncommercial sources, this figure is expected to be on the low side (the tuna production figure is 4.5 g/d). However, this methodology does not work for shrimp consumption; the 5.8% share is not compatible with the previous estimate of 4.5 g/d. The difference between the survey figures and the commercial production figures could also be due to differences in dietary habits between tuna and fin fish consumers; if people are more likely to eat tuna every day than they are fin fish (which, incidentally is not supported by the survey), then this methodology will underestimate the tuna contribution. The problem with the shrimp data could also be that infrequent consumption cannot be accurately measured with a 3-day survey.

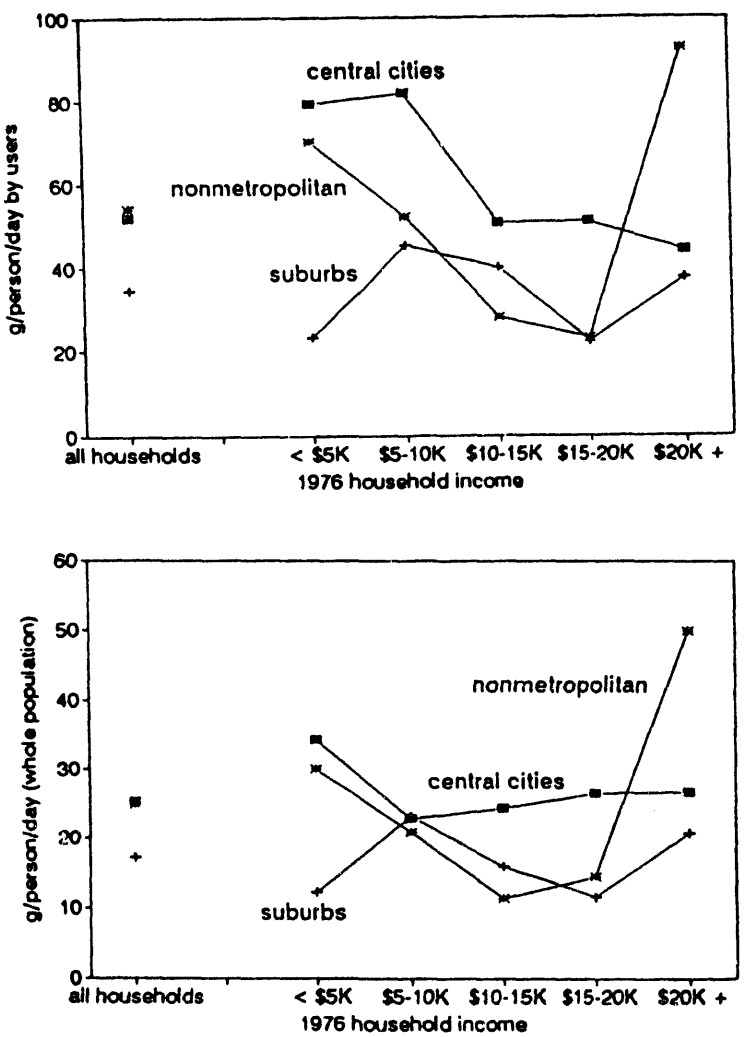


Figure 7.5. Survey results on consumption of fish and shellfish in the Northcentral U.S., spring 1977, by residential location and household income (data from USDA). (a) consumption rates for users during a 3-day period. (b) consumption rates for the whole population.

The USDA 3-day survey data were used by the Environmental Protection Agency (EPA) to provide national estimates of fish/shellfish consumption on an acute basis, i.e., the distribution of consumption on any given day (U.S. EPA, 1992, Appendix F). These estimates are not particularly useful for estimating the long-term consumption patterns needed to estimate equilibrium levels of mercury in human blood. However, the GSDs of these distributions were in the range 1.6 to 2.5.

7.2.4 Surveys of Fishermen. New York State surveyed 17,000 freshwater fishermen in 1988 and collected data on fish consumption and time spent angling for selected species (Connelly et al., 1990). The most popular species were bass (type not specified), trout, walleye, and yellow perch, all of which tend to have elevated mercury levels. 4530 respondents reported eating an average of 45 fish meals per year (about 22 g/d), and includes all kinds of fish, whether obtained from sport fishing or not. This value checks well with the earlier survey (NPD) reported above. The number of meals eaten increased with age, education, and income, but was reasonably uniform throughout the state. Those anglers who fished Lake Ontario reported consuming about 7 meals per year from this source (3.5 g/d); 15% reported eating more than 12 meals per year (in contravention of the state health advisory). The NPD survey of freshwater fish consumers in New York reported above found about 12 meals per year from all waters. Although about 70% of those surveyed reported that food was an important motivation for fishing in New York, about half of the fish caught were released or disposed of.

In 1993, the U.S. EPA sponsored a survey of anglers using the Ohio River for the 12 months following April 1, 1991. The main emphasis of the survey was on knowledge of and compliance with health advisories on contaminants in fish. Five thousand questionnaires were distributed to holders of fishing licenses in counties near the Ohio River in 6 states; responses were received on the importance of health advisories, fish consumption by species, and fish preparation methods (Knuth et al., 1993). Responses indicating the numbers of meals of different species were received from 1084 people; another 109 indicated consumption without giving the numbers of meals. Figure 7.6 displays the frequency distribution of meals, which is approximately log normal, with the low end truncated at 1 meal per year. The highest consumption rate for an individual species was 350 meals per year of large mouth bass; the highest combined species consumption rate was 364 meals per year. 57.4% indicated they ate no fish from the Ohio River, and the average number of meals per year (including zeroes) was about 7, which checks well with the figure for Lake Ontario (Connelly et al., 1988). On the basis of fisheaters only, the average was about 19 fish meals per year. Average mercury concentrations for each species were obtained from the EPA survey and weighted by the total numbers of meals consumed; this yielded an average mercury concentration of about 0.28 μg/g, which checks well with the average mercury levels from Michigan and Minnesota. Using this figure, the average daily dose was 1.07 μg/d including non fisheaters and 2.91 μg/d for fisheaters only. The highest individual daily dose by species was for large mouth bass (86 μg/d), followed by walleye (29 μg/d) and sauger (20 μg/d). The distribution of doses by species within the 7566 fish meals reported is given in Figure 7.7; large mouth bass and sauger are the largest contributors. The frequency distribution of mercury doses to fish eaters, based on the overall average concentration of 0.28 μg/g, could either be described by Figure 7.6 or by a log normal distribution with a median dose of 1.5 μg/d and a GSD of 3.5. The minimum dose was assumed to be given by a single meal of the species with the lowest average mercury level.

Data on coastal recreational fishing are included in the EPA Exposure Factors Handbook (1990). A 1-year survey of 1059 recreational fishermen in the Los Angeles area found a median fish consumption rate of about 37 g/d, with a GSD of 4.5. This very wide distribution had a 95th percentile of 339 g/d, which represents about 2 fish meals per day, every day of the year, which seems high for an urban area. By species, the distribution was dominated by "California halibut" at 143 g/person/d. Another survey was reported for Commencement Bay, WA (near Tacoma), which showed that about 9% of those surveyed fished daily and consumed 381 g/d, of which the 2 largest contributors were reported to be Pacific hake (77 g/d) and "walleye poilock" (sic) at 180 g/d. These values are reported as averages over two seasons and over the fishermen's families. If the daily figure for the Washington survey is interpreted as the approximate 90th percentile of the entire sample, the two surveys are in reasonable agreement. These values were used to develop the guideline recommendations for freshwater fish consumption and for recreational fishing in the EPA Exposure Factors Handbook (1990):

freshwater fish (general public) 6.5 g/d average
recreational fishermen, 30 g/d average with a 90th percentile value of 140 g/d
subsistence fishermen (Alaska), 304 g/d.

These values should be compared with the summary statistics developed for this assessment, which are given in Table 7.2, below. Note that the factors for recreational fishing were developed exclusively from coastal waters.

7.2.5 Surveys of Subsistence Populations. Richardson and Currie (1993) report surveys of 4327 Amerindians residing in Ontario reservations, using mercury in hair and in the fish consumed to deduce apparent consumption rates. Data are presented by age, sex, and reservation location. Males had an apparent median consumption rate of 19 g fish per day; females, 14 g/d. These rates were higher than the reported Canadian national average of 11 g/d. Apparent consumption rates also increased with latitude, which the authors took as an index of community isolation. The highest values were about 120 g/d as a reservation median.

Wolfe and Walker (1987) report on the dietary components of 98 communities in Alaska, obtained from 1981 to 1987. Yearly per capita subsistence harvest figures ("dressed" weights) were given for fish, marine mammals, land mammals, and other foods, and an economic analysis was performed. Data on the species consumed were not reported. Urban communities in Alaska consumed an average of about 22 g/d of fish, which is remarkably similar to the figures for the "lower 48" states. The remaining (non-urban) communities displayed a wide range of fish consumption rates, from 31 to 1541 g/d with a median of about 220 g/d (about 1 meal daily). The range of consumption rates for marine mammals was similar, but showed little overlap with the fish consumers. The highest consumption rates were in the interior of the state, in Yukon-Koyukuk County. This county had a population density of 1 person in 20 mi^2 in 1980. The species most likely to be consumed in the interior regions of Alaska were reported to be the medium-to-small sized salmon such as chum or silver salmon (R. Mikkelson, personal communication, 1993), which would have only moderate levels of Hg.

Wolfe and Walker also noted that communities without road access had higher subsistence fish consumption by a factor of 4, and that road access brings in urban dwellers to share in the harvest, thus reducing the catch per capita. Thus, it seems unlikely that the upper range of the consumption figures they reported would be applicable to a lake or stream in the immediate vicinity of a power plant.

7.3 The Distribution of MeHg Doses to Adults

The previous sections presented data on mercury concentrations in selected species and on their rates of consumption for food. This section combines the two datasets to provide estimates of the distributions of daily mercury doses through seafood.

7.3.1 Dose by Major Groups of Species. The total dose of MeHg is assumed to be comprised of the independent contributions of canned tuna, fresh and frozen marine fin fish, shellfish, and fresh and frozen freshwater fin fish, each of which has a log normal distribution. Also, equivalence between total Hg in fish and MeHg is assumed, as has been shown in several studies. In order to assess the contribution of marine fin fish, the weighted average Hg concentration is computed, using the distribution of landings by species for 1992 (NMFS, 1993). These are wet

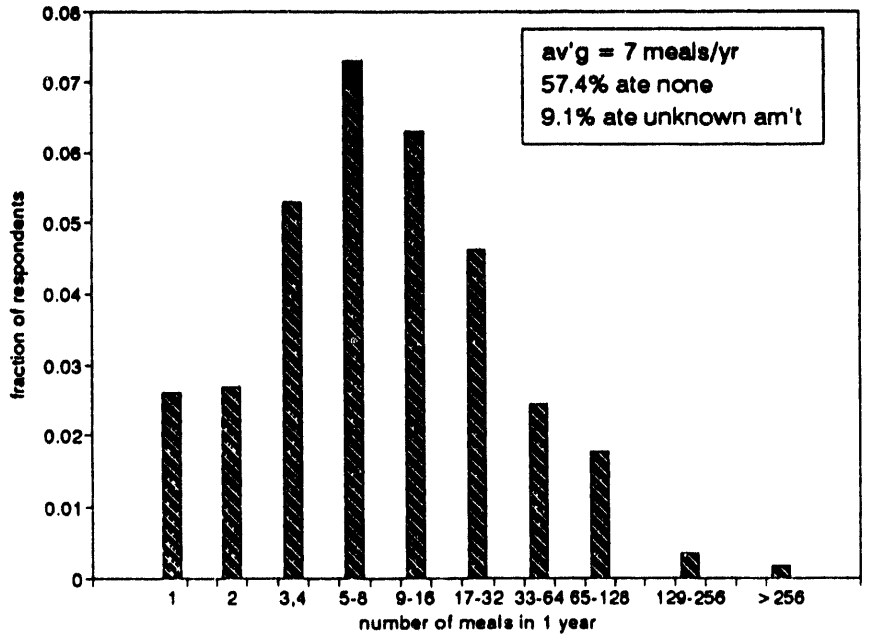


Figure 7.6. Frequency distribution of meals eaten of recreationally caught Ohio River fish. Data source: Knuth et al. (1993).

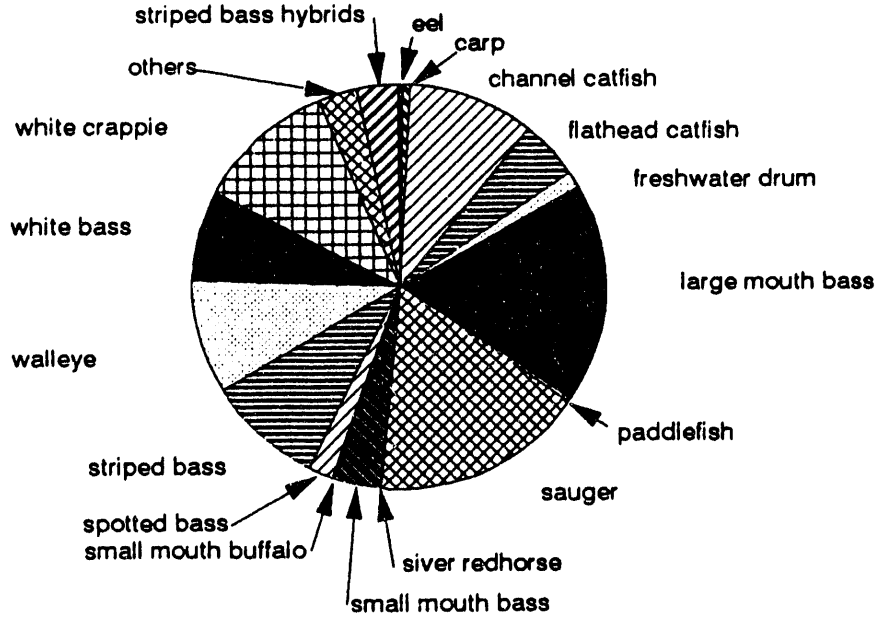


Figure 7.7. Shares of Hg in the total recreational catch of Ohio River fish, by species. Data source: Knuth et al. (1993).

weights, as opposed to edible weights, but this statistic would still be acceptable if the edible fraction of weight were the same in all species. The mercury concentration data used for marine species were taken from Table 6.2, based on averages of the available Hg data for each species. Shellfish and fin fish were examined separately and a weighted average Hg level for fin fish was computed, excluding tuna (because most tuna is consumed as canned product, for which separate consumption statistics are available). Since only edible species have mercury concentration data, this procedure automatically excludes non-food species such as menhaden.

Figure 7.8 presents these distributions. Figure 7.8a shows that shrimp and crab are the largest contributors to the average Hg dose from shellfish; however, Table 6.2 shows relatively large discrepancies (factors of 4-5) between the FDA and NOAA determinations of average mercury levels, the FDA data being high for shrimp and the NOAA data being high for crab. Obviously, better data are needed to resolve these uncertainties.

The distributions of mercury doses from fin fish are shown in Figure 7.8b. Here the largest contributors are pollock and tuna, pollock because of the very large catch and tuna because of the relatively high mercury levels. Table 6.2 also shows a wide discrepancy in the mercury levels for pollock, but less so for tuna. A high priority should be given to determining reliable mercury levels in pollock, given its high apparent consumption rates.

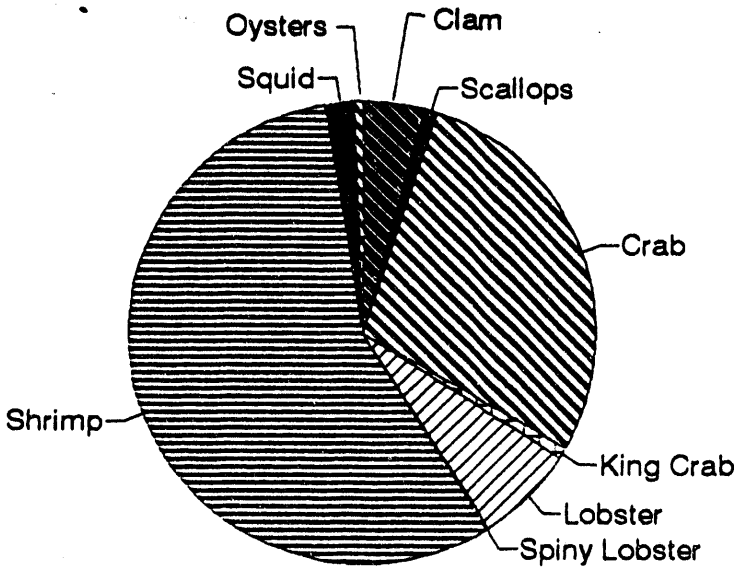
The weighted average mercury level for shellfish was 0.09 μg/g; for fin fish without tuna, 0.08 μg/g. These figures compare reasonably well with the unweighted data from Table 6.2 for shellfish, but the weighted average fin fish mercury level is much lower because of the high contribution of pollock. Because of the close correspondence of weighted average Hg levels for shellfish and marine fin fish (less tuna), these two categories were combined for the purpose of estimating the distribution of doses.

The 1973-74 NPD survey mentioned above (Goyer et al., 1985; EPA, 1990; NOAA, 1978) provides an alternative source of data on weighted average mercury levels and doses. About 65% of the panelists reported eating tuna; the next most common categories were shrimp and "not reported." These relative consumption reports were used to weight the mercury concentration data of Tables 7.3 and 6.2, in order to generate weighted averages (the "not reported" and "other" categories were assumed to be processed fish products such as fish sticks, composed mainly of pollock). The weighted average Hg concentrations were as follows:

All species:	0.129 μg/g
fin fish:	0.141 μg/g
shellfish:	0.084 μg/g
fin, no tuna:	0.071 μg/g

Tuna was the largest contributor to the weighted average mercury in all species, at 43%. The "other" category was next at 9.9%, followed by shrimp (8.6%). Fresh-water game fish contributions were appreciable: large mouth bass, 4.2%; northern pike, 1.7%. The combined contribution of pollock and "other" was 11.7%. The contributions of large predatory fish such as shark and swordfish were quite small, since only 41 panelists reported eating swordfish and only 3 reported eating shark. However, both the availability of these species and dietary preferences may have changed considerably in the 20 years since this survey was taken.

7.3.2 Hg Doses from Individual Species. Additional data, which apparently came from the 1973-74 NPD survey, were provided by Goyer et al. (1985), including the rates of consumption of individual fish species. These data were used to examine the relative contributions to the national average Hg dose in more detail (Figure 7.9). These data are similar to, but not entirely consistent with, the data given in the EPA Exposure Factors Handbook. Figure 7.9a shows that tuna contributed almost half of the "national dose" in 1973-74, primarily because of the large fraction of consumers. The next largest contribution was from (freshwater) bass, because of its relatively high average Hg content. No other single species played an important role in the weighted national average Hg dose.



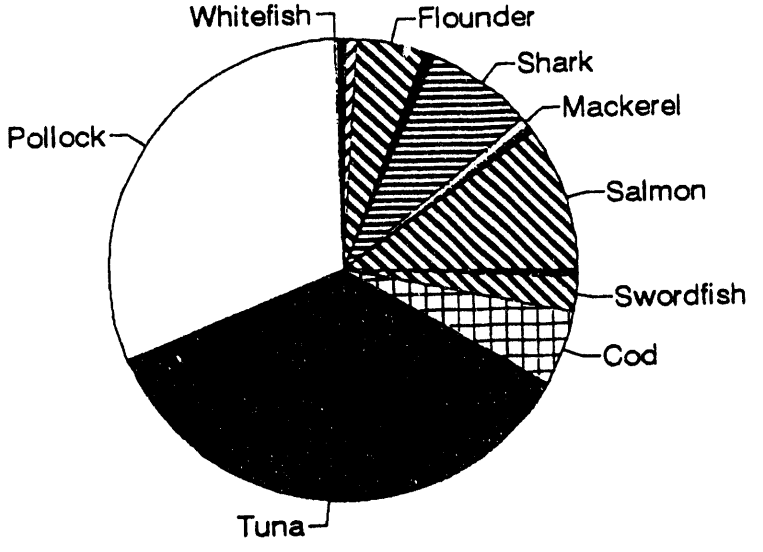


Figure 7.8. Shares of total Hg doses in 1992. Consumption data based on NMFS (1993); Hg concentrations from Table 6.4. (a) shellfish. (b) marine fin fish.

However, when the data are ranked by the doses to consumers of selected species (rather than being averaged over the entire U.S. adult population), a different picture emerges (Figure 7.9b). Consumers of pike or bass (both freshwater species) receive almost twice the national average Hg dose from these species alone, because both the Hg levels and the consumption rates for users were high; consumption of other species would add to their average daily dose levels. Tuna ranks far down in this ranking because of the relatively low average consumption rates by users (6.1 g/d according to Goyer et al.; 3.5 g/d according to the EPA Exposure Factors Handbook).

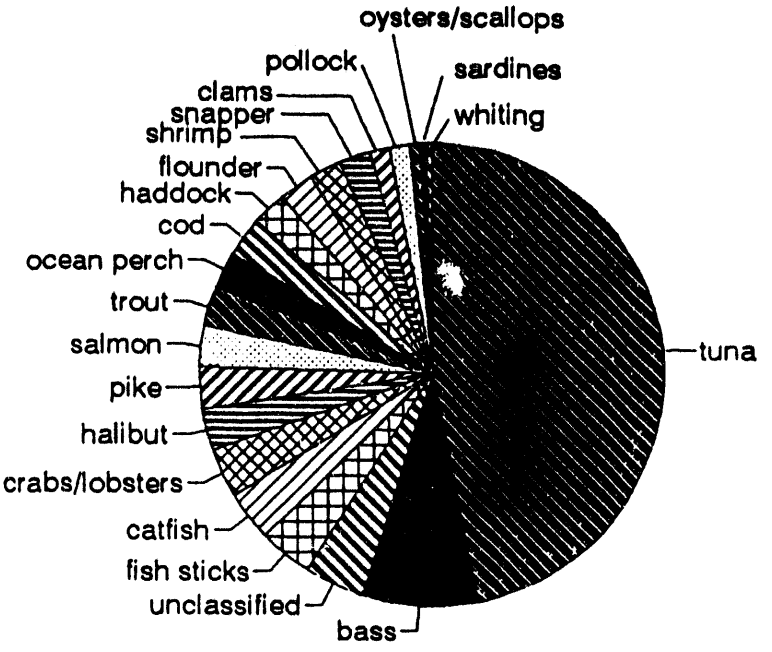
The NOAA report citing the NPD survey results (NOAA, 1978) also provided some information on the upper part of the distribution of doses. They used the "acceptable daily intake" (ADI) of MeHg of 30 μg for a 70 kg person as a reference. 99.81% of the panelists were within this dosage. Based on an average consumption of 20 g/d and a weighted average Hg level of 0.13 μg/g, the average dose is 2.6 μg/d. Assuming a GSD of 3.0 would result in an estimated 99% of the sample within the ADI (neglecting the effects of body weight on ADI and dose). This indicates that exposure may have increased since 1973-74, which is consistent with the trend towards higher rates of consumption shown in Figure 7.1a.

The FDA surveys contaminants in food periodically by means of a "market basket" survey, in which retail foods are purchased at different locations in the United States and analyzed for the presence of a number of contaminants, including mercury. The market basket comprises about 120 different foods, 20-30 samples of are collected each year. The latest such survey (Gunderson, 1988) was conducted from 1982 to 1984 and found an average total daily intake of mercury of 3.38 μg/d. The values were higher for males and increased with age; these trends are also seen in seafood consumption, and Gunderson reported that 77% of the total Hg intake came from seafood. This provides an excellent check on the national average dose from seafood estimated above, since 0.77 x 3.38 = 2.6 μg/d (the figure given above). The FDA market basket survey was based on 16 samples each of haddock or cod fillet (0.12 μg/g average Hg), canned tuna (0.277 μg/g), fresh or frozen shrimp (0.028 μg/g), and commercial fish sticks (0.025 μg/g). The total fish consumption level implied by these figures is 22 g/d, assuming equal quantities of all 4 items. The FDA report did not state whether the mercury found in foods other than seafood was inorganic or MeHg; the only non-seafood item with noticeable mercury levels was canned mushrooms, with 0.03 μg/g Hg.

7.3.3 Data for Use in Probabilistic Simulations of the Distribution of Fish Consumption and Hg Doses. This assessment is concerned with the impact of coal combustion on mercury deposition and subsequent health effects. Accordingly, the Upper Midwest was selected as the geographic area of interest. This region is reasonably densely populated, has many lakes with game fish, and generates electricity from coal. The specific case considered is the adult population in the Great Lakes states that consume freshwater fish. The mean and median concentrations of MeHg and their GSDs, and the mean and median consumption rates and their GSDs are given in Table 7.2.

Table 7.2 Representative Statistics for Components of the Seafood Diet in Northcentral States

Component	Means		Medians		GSDs		Apparent Dose
	diet g/d	Hg μg/g	diet g/d	Hg μg/g	diet g/d	Hg μg/g	mean μg/d
canned tuna	4.5	0.195	2.7	0.15	2.7	2.16	0.88
freshwater fin fish	10.3	0.28	6.8	0.15	2.5	3.0	2.88
other marine species	9.9	0.077	7.2	0.061	2.0	2.2	0.76
total seafood	24.7						4.52



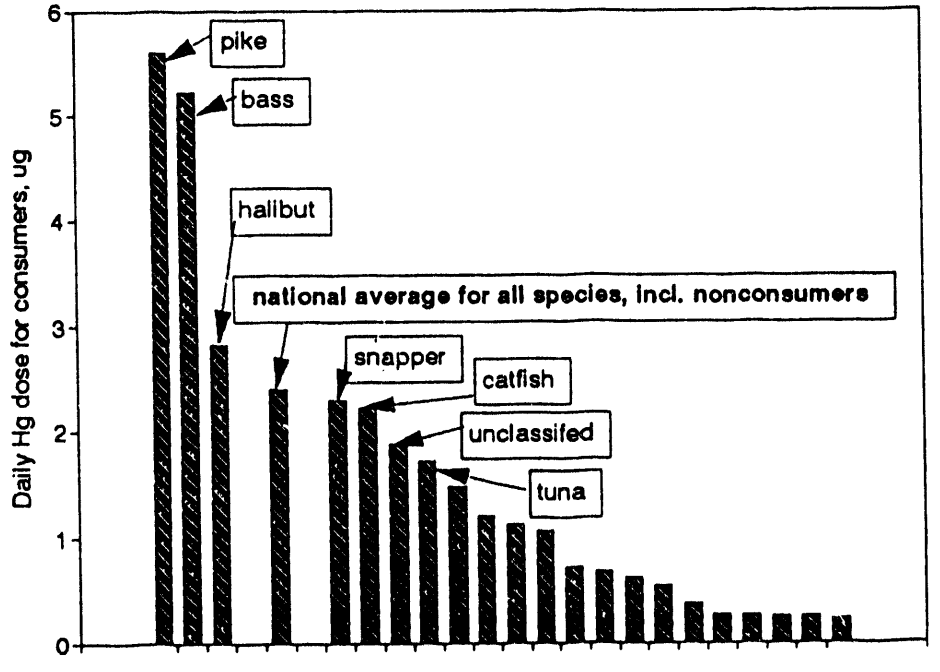


Figure 7.9. Distribution of seafood Hg by species. Consumption data from Goyer et al. (1985). (a) shares of total U.S. consumption. (b) daily dose rates for consumers of selected species.

The GSD for tuna consumption was developed by assuming that the maximum consumption of canned tuna is one 3-oz can every day. The total and freshwater consumption figures were taken from the NPD survey, adjusted to 1992, and the marine species figure was obtained by difference. In this case, marine seafood includes fin fish, shellfish, and canned product other than tuna.

A probabilistic simulation of these distributions with 10,000 iterations produced the statistics for the components and the total dose shown in Table 7.3. Note that each line in Table 7.3 represents a separate distribution; the individual percentiles do not sum to the total.

Table 7.3 Probabilistic Simulation of Hg Dose in Northcentral States (μg/d)

Component	Mean	Median	5%	95%	99%
canned tuna	0.92	0.42	0.054	3.3	7.5
freshwater fin fish	2.90	1.03	0.10	10.5	29
marine species	0.75	0.44	0.076	2.4	4.7
total seafood	4.59	2.70	0.72	13.4	34.2

Freshwater fish are seen to contribute the largest amount of Hg, by far, resulting in a mean dose for the Northcentral region that is somewhat higher than the FDA market basket figure for the whole U.S. However, it is possible that all consumers tend to eat the same total amount of fish and thus that the freshwater contribution would be replaced by another source for a different population without access to fresh water. If the substituted seafood had an average mercury concentration of 0.09 μg/g (corresponding, for example, to a mixture of other marine species), the above analysis would conform quite well to the FDA figure for seafood of 2.6 μg/d. The mean and medians of this distribution correspond to about one adult meal per week, depending on the mercury concentration. Figure 7.10 presents histograms of the total dose and its logarithm; note that even the distribution of logs is positively skewed, because of the addition of several log normal distributions with different GSDs comprising the total.

The extremes of this distribution may be evaluated as follows. The 99th percentile of the total dose obtained in 10000 samples was 34 μg/d and the maximum was 174 μg/d, dominated entirely by freshwater fish. This maximum value (which is not a robust statistic) corresponds closely to 2 meals per day at 0.5 μg/g (or 3 meals/day at 0.33 μg/g), which would be appropriate for subsistence on a single species of game fish such as large mouth bass or walleye. If this distribution were extended to include the maximum subsistence population in Alaska (where species such as pink salmon tend to be the mainstay of the diet), the maximum daily dose would be about 170 μg/d. These values are consistent with a dosage distribution developed from a survey in the late 1960s (reported by Clarkson, 1990), which found a mean daily intake of 36 ng/d per kg of body weight (about 2.5 μg/d for an adult) with a 99th percentile value of 243 ng/kg/d (17 μg/d for an adult). If the approximate 40% increase in average per capita fish consumption that has occurred since then is taken into account, these rates would be 4.2 and 28 μg/d, respectively. Note that the appropriate Hg concentration to be used for such an extreme case is not the maximum individual fish mercury level, but the species average, since the only way an individual can accumulate such a large daily average consumption rate is by eating fish frequently. This process will drive the yearly average mercury level closer to the average for that species, because of the central limit theorem.

However, the minimum levels of fish consumption may have been overstated. The NPD survey found that 95% of the population ate some seafood during a 1 month period; assuming that this month was not atypical, then 5% of the population would be estimated to eat no seafood. The 5% point of the total seafood distribution of Table 7.3 corresponds to about 7 fish meals per year, which is significantly higher than zero in this context.

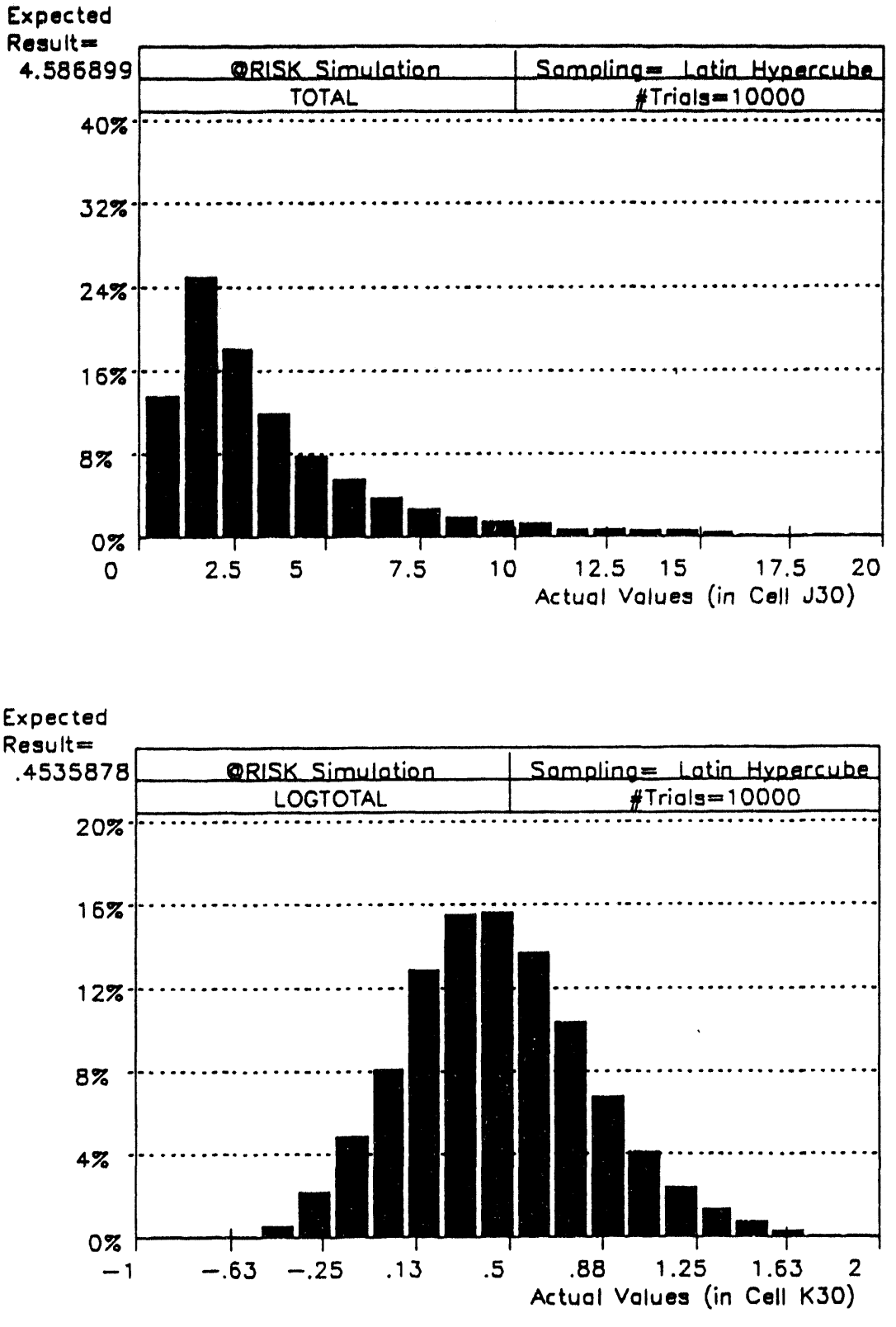


Figure 7.10. Histograms of U.S. average daily Hg dose from seafood. (a) daily dose rates (μg/d). (b) log of daily dose rates.

Tables 7.2 and 7.3 should be considered to apply to fisheaters only and the numbers should be deflated by 5% to make them applicable to the entire population. Also, as discussed below, very infrequent fish-eaters may never reach an equilibrium level of Hg in their blood, so that the dose-response functions are probably not applicable to them in any event. However, the data on tuna consumption included above appears to reflect the whole population, rather than the sub-population of tunafish eaters. Since mercury tuna is not greatly affected by coal burning, use of these data is consistent with the overall goal of the assessment.

7.4 Summary of Data on Fish Consumption and MeHg Dose Distributions

A review of the data available from various sources has shown that it is necessary to combine some of them in order to derive statistics appropriate for this assessment. For example, many of the data on distributions of consumption date from 1973-74, and overall seafood consumption has increased substantially since then. Thus, the trend data from national production statistics are used to adjust the older distributional statistics upward to more nearly reflect current consumption levels. However, if public preferences for certain species have changed over the years, this procedure may entail errors. Good support is found from both surveys and production statistics for an overall average seafood consumption rate in the Northcentral states of about 25 g/d, with a 95th percentile level of about 80 g/d. About 95% of the U.S. population consumes some seafood over the course of a year. The average MeHg daily (baseline) dose for consumers of freshwater fish in the Northcentral U.S. was estimated to be about 4.6 μg/d, and the 99th percentile was only about 11% of the lowest adult dose estimated to exhibit adverse health effects, as reported by Clarkson (1990).

8.0 UPTAKE OF MeHg: THE EQUILIBRIUM DIET-BLOOD-BODY BURDEN RELATIONSHIP

Estimation of MeHg doses takes the assessment process to the point of entry to the human body. For many air pollutants, the subsequent processes of uptake and transport to target tissues or organs are not well defined. For MeHg, however, both theory and experimental data are available.

For example, Nordberg and Strangert (1978) make distinctions between carcinogenic and noncarcinogenic responses. Carcinogenic responses are considered to be stochastic processes, because the initial growth of cancerous cells in an individual cannot be predicted from first principles. The population dose-response curve is based on an aggregation of individual responses and may often be depicted as linear, with no threshold dose. Dose may be expressed as the product of concentration and exposure time, say, over a lifetime.

For the noncarcinogenic effects of interest here, Nordberg and Strangert consider that a critical dose gives rise to defined effects in specific organs. For MeHg, the target organ is the brain, where the onset of neurological symptoms has been estimated to begin at a concentration level of about 1 ppm, which corresponds to about 200 ng/g in blood (Berlin, 1976). The concentration of MeHg in blood is a useful assessment parameter, because it can readily be measured and thus reference data are available, and because blood is the medium of circulation of poisons within the body. Variations in blood and brain concentrations may occur from individual differences in metabolic processes.

The task of estimating the distributions of blood MeHg levels and the resulting body burdens resulting from specific levels of dietary intake is considered in this section. Note that concentrations of Hg in human hair are also used as an indicator of body burden or dose, especially because gradients along a length of hair provide a dynamic record of exposure. However, concentrations in hair are also affected by inorganic Hg and may be compromised by external artifacts. The baseline risk assessment then provides an opportunity to compare the distributions of body burdens, blood and hair levels of MeHg with actual measurements as a reality check on the methods used.

8.1 A Theoretical Model

8.1.1 Basic Processes. The absorption of MeHg in the intestinal tract is essentially complete (Nordberg and Strangert, 1985). MeHg is then excreted from the body as a first-order decay process (Clarkson, 1990), such that

the fraction remaining from an individual dose ingested at time t may be represented as e^{-kt}, where k is the elimination constant. If repeated doses of magnitude m (μg/d) are ingested on a steady basis, an equilibrium body burden is reached that may be estimated by

$$BB_e = mt_b/\ln(2) \qquad [8\text{-}1]$$

where t_b is the biological half-life. Eq. [8-1] results in an equilibrium body burden of about 100 times daily intake for a half-life of 70 days.

Figure 8.1 depicts the cyclical increase in body burden of a substance with a half-life about 70 days (k=0.01) and a cycle of fresh intake of unit magnitude every 4 weeks (m=1/28). The figure shows that equilibrium body burden level (BB_e = 100/28 = 3.57) is never reached in a strict sense because of the cyclical intake; a blood sample taken immediately after the intake cycle would read about 30% higher than one taken just before. However, Eq. [8-1] holds if body burden levels are averaged over an intake cycle, and a quasi-steady state is reached after 3-4 half-lives have elapsed (97% of equilibrium is reached after 5 half-lives). When individual variations in observed half-lives are considered (discussed below), quasi-equilibrium times would range from about 4 months to almost 2 years. As the frequency of fish consumption (m) increases, the total body burden increases proportionately according to Eq. [8-1], and the relative cyclical effect of each meal decreases. For consumption of fish on a daily basis, for example, this variation would amount to a swing of only 1%. However, the rate of progress toward equilibrium does not depend on the rate of intake of MeHg.

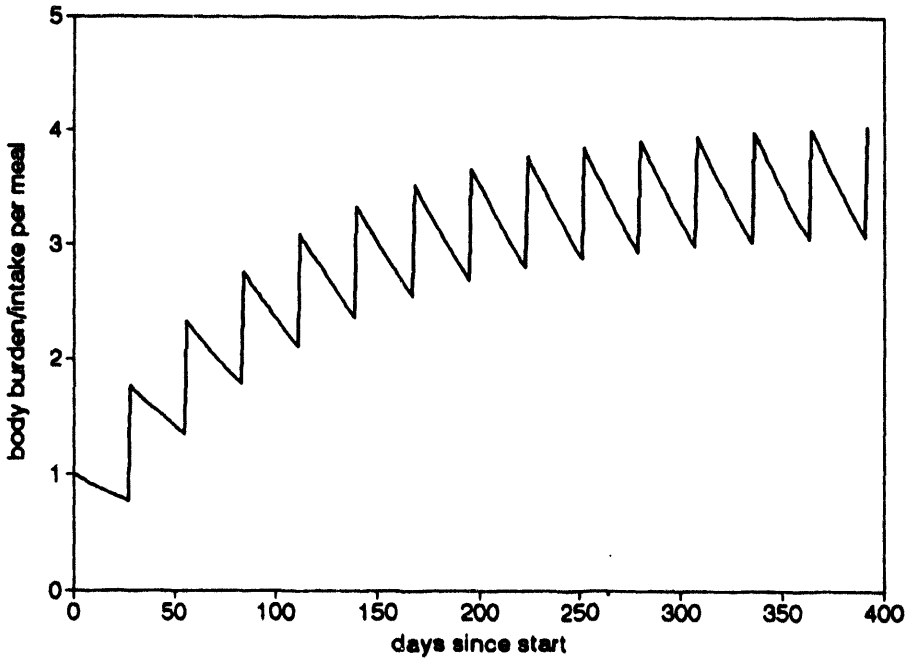


Figure 8.1. Simulated metabolic build-up of a substance that is ingested every 28 days, with a half-life of 70 days.

MeHg concentrations in blood may be predicted from Eq. [8-1] by referring the body burden to the volume of blood. Kershaw et al. (1980) studied the dynamics of mercury retention based on consumption of single fish meals by 5 volunteers and developed a theoretical model of the process. The output from this model is the factor (A) linearly relating equilibrium blood MeHg level (EB, μg/L) with dietary intake (m, μg/d):

$$A = EB/m = (ft)/(0.07W \ln[2]) \qquad [8\text{-}2]$$

where f is the fraction of the dose deposited in blood, t is the biological half-life of MeHg, and W is body mass (kg). The factor 0.07 was assumed by Kershaw et al. to represent the ratio of blood volume (L) to body weight (kg), based on research by Miettinen (1972), who compared blood concentrations with whole-body counts of radiolabeled MeHg. Kershaw et al. used the data from five volunteers to evaluate Eq. [8-2] and derived a value of 0.9 for (A) for a 70 kg person. Note that the factor (A) represents the fraction of the body burden of MeHg in blood, which would be expected to be less than unity, due to uptake of some of the MeHg by tissue and organs.

Miettinen (1972) reported that the half-life of MeHg is blood was "considerably" shorter (50 days) than in the whole body (76 days), but his decay plots show that much of the excess decay occurs in the first 2 weeks and that the long-term decay rates in blood are closer to those in the whole body. The difference between the two group means was only marginally significant.

8.1.2. Individual Variability. Miettinen's data for whole-body half lives were normally distributed with a mean of 76 days and a standard deviation (for individuals) of about 11 days. This would yield a 5%-95% range from 46 days to 96 days for the population whole-body half-lives and a GSD of 1.25.

The experiments of Kershaw et al. (1980) also determined half-lives of Hg in blood for 5 subjects, in hair for 1 subject, and hair/blood ratios at near-peak Hg levels. The range in blood half-lives was from 47 to 67 days, with a mean of 52 days. The data on hair were shown only as a plot, and the rate of decay of Hg concentrations appeared to be about the same as in blood for that subject (t=~70 days).

The half-lives of Hg in hair varied from 35 to 189 days in 48 Iraqi poisoning victims (Al-Shahristani et al., 1974). This distribution was found to be approximately log normal by Ahmed (1991), with a median value of 68.2 days and a geometric standard deviation of 1.395. Others have represented it as normal but bimodal, with one peak at about 65 days applying to about 87% of the group, and another smaller peak at about 130 days, applicable to the remainder. Sherlock et al. (1984) also found a range in biological half-lives among 20 volunteers, with an average of about 52 days (measured in whole blood). According to IPCS (1990), half-lives in blood tend to be shorter than in the whole body, but hair and blood values are about the same, with a wider range observed in hair. There is suspicion that a part of the wider range in hair might be artifactual.

From first principles, it follows that a given amount of MeHg from fish consumed by a light-weight person would have more effect than if consumed by a heavier person, *ceteris paribus*. Thus, a probabilistic risk analysis must include the expected variability in body mass in the population at risk. Since there are no published data on actual fish consumption patterns by body mass, it is necessary to make assumptions about the relationship between fish consumption and body mass.

The range in body mass of the U.S. population is appreciable; the National Center for Health Statistics gives mean weights of about 157 lb (71.4 kg) for both sexes, ages 18-74, with a standard deviation of 34.8 lb (15.8 kg) (Robinson, 1981). However, the distribution appears to be more nearly log normal, since 3 standard deviations below the mean corresponds to about 53 lb., which is very light for an adult, and 3 standard deviations above the mean corresponds to 261 lb., which is also somewhat on the low side of extreme weights. Converting the standard deviation to a geometric basis gave more reasonable results in a log normal model: mean = 157 lb (71.4 kg), 0.64% less than 88 lb (40 kg), and 0.68% greater than 264 lb. (120 kg).

8.2 Epidemiological Determinations of the Metabolic MeHg Relationships

Several studies have provided concurrent data on MeHg in human diet and in blood (or hair) that could be used for experimental verification of the model of Kershaw et al. (1980), over a longer term. Of course, such studies may be limited by the numbers of subjects studied and by the accuracy with which dietary intake can be determined, but it is important to determine if the results are compatible with theory after allowances are made for these limitations. Kershaw et al. provided results from several of these older studies (converted to a whole-blood basis),

and noted that the results for the dietary constant (A) tended to be higher from tracer studies than when (A) was determined from dietary studies. This apparent discrepancy could result from errors in dietary recall.

8.2.1 Dietary Hg Studies. Early studies in Britain were based on the "duplicate diet" studies (Haxton et al., 1979; Sherlock et al., 1982), in which volunteers supplied exact duplicates of their diets for a 7-day period, which were then analyzed for mercury content. Blood and hair samples were also taken during this period. The resulting dose-response functions were shown to be highly variable, with experimental values of (A) in the range 0.04-0.2. Figure 8.2 (from Sherlock et al., 1982), displays some of the results from other studies. Sherlock and Quinn (1988) presented a statistical analysis of the sources of variation in experimental diet-blood MeHg studies.

As the independent regression variable, uncertainties in dietary Hg will bias the slope (A) downward. Assuming that these uncertainties are responsible for a portion of the "lack of fit" of the blood versus dietary Hg data (as exemplified by the typically low correlation coefficients), these values for (A) would be increased into the range of other studies.

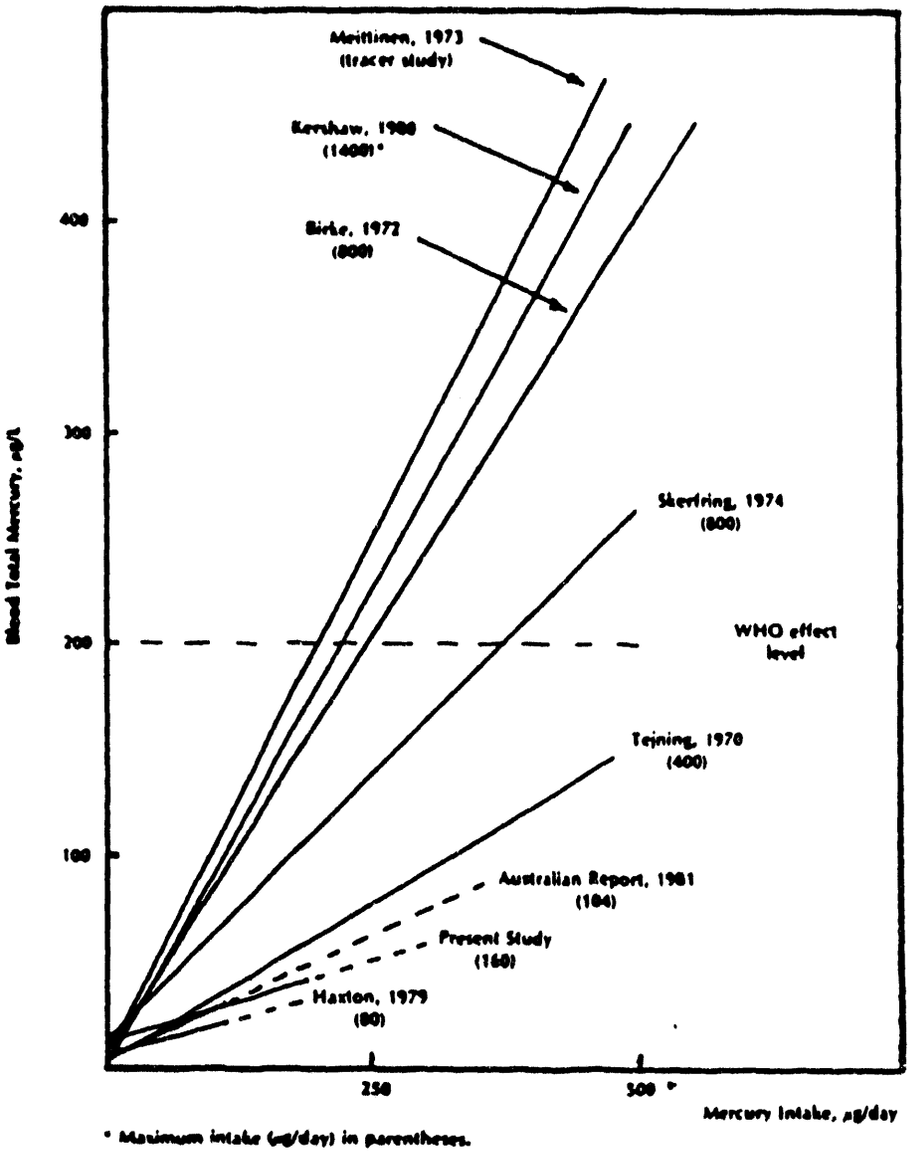


Figure 8.2. Relationships between blood mercury concentrations and mercury intake from various studies. Source: Sherlock et al., 1982.

Clarkson (1990) reports Swedish data in which blood mercury was determined and plotted against self-estimated dietary intake of MeHg for 6 long-term fisheaters. This relationship appeared to be described by the log-log model

$$\text{Log (blood)} = -1.75 + 1.12 \log (\text{diet}) \qquad [8\text{-}3]$$
$$(0.16)^* \quad (0.11)^*$$

Since the relationship should theoretically be linear (regression coefficient = 1.0), and such a relationship falls within the confidence limits of Eq. 8-2, a "background" value to be added to the data set was found that would yield a coefficient of unity. This value was a blood Hg of 0.55 at an intake of 10 ng/kg/day, or about 1 fish meal per month for a 70 kg person. This model is

$$\log (\text{blood}) = -1.365 + 1.0 \log (\text{diet}) \qquad [8\text{-}4]$$
$$(0.16)^* \quad (0.07)^*$$

or, in terms of a linear model, blood MeHg = 0.73 dietary MeHg (A=0.73).

Sherlock et al. (1984) studied the dynamics of blood MeHg in 20 subjects who consumed controlled diets of halibut (from a common source) for about 100 days. They were subdivided into 4 dose levels, of 42, 77, 101, and 226 μg MeHg per day. At the end of the experiment, their average blood MeHg levels formed a nearly perfect (R=0.997) linear relationship with their average dietary intake levels. The average value of (A) from this study was about 0.82, with a standard deviation of 0.10, after removal of an outlier. They also found a relationship between half-life and body mass that gave a smaller dependence of (A) on body mass than shown by Kershaw et al. (1980). This relationship was judged to be the most credible among the meager data available; it justifies the use of the equilibrium model in relating daily intake to body burdens on a long-term basis.

8.2.2 Baseline Data on Mercury in Blood and Hair A number of population-based surveys have been conducted on the mercury content of human blood and hair. In some cases, the absolute concentration levels have been shown to depend on analytical techniques, especially as to whether MeHg of total Hg is being determined, and to vary with the rate of fish consumption. These data are useful to this assessment for two reasons: as a reality check on the fish dose-body burden relationship and to confirm the relative distributions of concentrations.

As discussed below, the distribution of MeHg body burden for an individual tends toward the central (median) value as additional meals are consumed from the same supply of seafood. Thus, we would expect that the distributions of MeHg found in blood samples drawn from a population stem mainly from individual differences in physiology, including body mass and the half-life of mercury, although there will be some contribution to MeHg variability from variations in diet as well. With data obtained on a random schedule, some samples may be obtained soon after a fish meal, thus deviating from equilibrium. We examined extant data on blood and hair Hg, as follows.

Wheatley et al. (1979) obtained over 35,000 hair and blood samples from native populations in Canada, including some from communities impacted by discharges from chlor-alkali plants. The distribution data are shown in Figure 8.3. For the entire sample (Figure 8.3a), the median blood level is about 12 ppb, and the GSD was estimated to be about 2.84. However, the distributions varied considerably by province (Figure 8.3b). The Quebec and Ontario data had higher and more variable levels than those from Manitoba, Saskatchewan, and Alberta (GSD=2.4). We might hypothesize that the presence of high-dose individuals within a population will increase both the mean and the standard deviation.

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* standard deviations

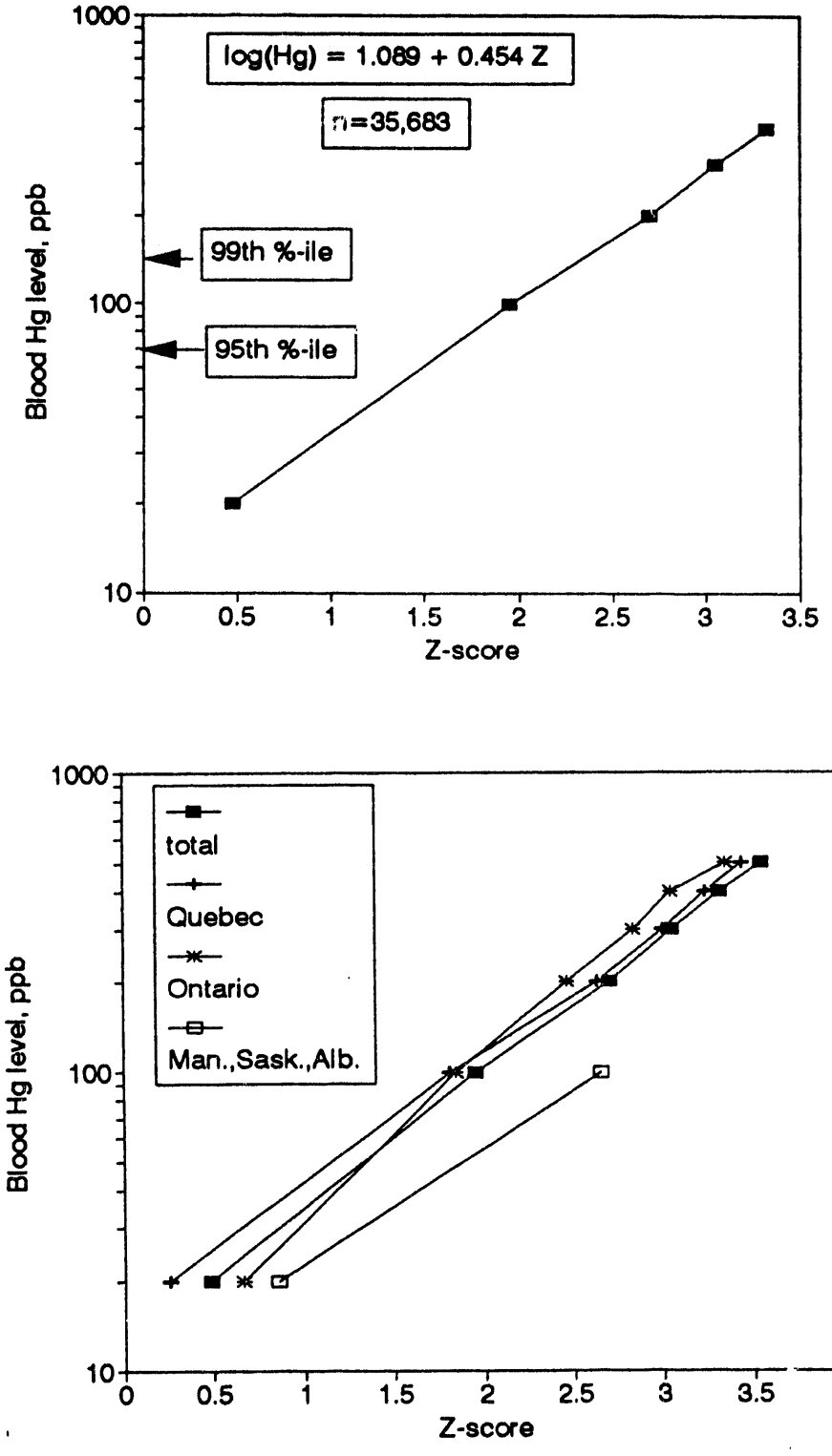

Figure 8.3 Distribution of mercury in blood for Canadian Indians and Inuits. (a) total sample (b) by Province. Data from Wheatley, 1979.

The data of Grandjean et al. (1992) from the Faroe Islands provide an opportunity to examine the effect of differences in fish consumption rate on the distributions of blood and hair Hg. The overall median blood level was 121 ppb, with a GSD of 2.32. However, the GSD of those reporting no fish consumption was much higher (4.51), suggesting that some portion of this cohort may have in fact consumed MeHg, perhaps from whale meat. The GSDs of the higher consumption cohorts ranged from 1.8 to 2.5, showing that there is still considerable variability in body burden even after consumption rate has been controlled for.

Stratification by fish consumption was also done by Den Tonkelaar et al. (1974?), for a sample from the Dutch population. Here median blood levels were much lower, from 1.5 ppb for non-fisheaters to 4.4 ppb for once-per-week eaters. The GSDs did not vary systematically by consumption level, however, and were in the range 1.7 to 2.1.

Mercury in blood and hair were also examined in various other populations, with the results given in Table 8.1. We see a wide range in mean Hg levels, but surprising uniformity in the GSD's. The mean GSD is about 2.1; the highest value is also for the largest sample (Wheatley, 1979), which comprised a variety of different populations. Those subpopulations including subsistence fisheaters tended to have larger GSDs, as might be expected.

Table 8.1 Baseline Data on Hg in Blood and Hair

Reference	population	# subjects	determination	mean*	GSD
Grandjean et at. (1992)	Faroe Isl. mothers	997	cord blood maternal hair	24.2 4.5	2.3 2.3
Wheatley (1979)	Canadian natives	35683	blood (some values based on hair)	12.2	2.8
Ahmed (1991)	South Haven, MI Algonac, MI	14 30	blood blood	1.6 2.3	2.3 2.0
Turner et al. (1980)	Peruvians fisheaters controls	 128 190	 blood blood	 82 9.9	 ~1.9 ~2.1
Hecker et al. (1974)	Ann Arbor, MI S.Amer. Indians	100 90	blood blood	0.95 1.4	1.4 1.7
Dennis & Fehr (1975)	Saskatchewan (native & non-native)	679	blood	6.7	2.2
Petersen et al. (1994)	Wisconsin Chippewas	326	blood	3.9	~2.2
Shimomura et al. (1980)	rural Japanese	1324	hair	2.53	2.1
Smith et al. (1985)	sample of U.S. women of childbearing age	1431	hair	0.48	2.35

* blood values in μg/L; hair values in μg/g

8.2.3 Use of Mercury in Hair as an Indicator of MeHg Dose. Use of mercury in hair as an indicator of exposure has the advantage of providing retroactive information. By using the rate of hair growth, typically 1-1.5 cm/month, and Hg concentrations at different distances from the scalp, an exposure profile may be constructed. Airey (1983) reviews sampling and analysis procedures, and notes that interference can result from dyes, shampoos, and bleaches, as well as from dust and dirt. Head hair typically contains more mercury than body hair. Airey cites the work of others showing that while inorganic Hg is about 4-5% of MeHg in red blood cells, it is 16-20% in hair. She notes that hair Hg can be much higher in people who have been occupationally exposed to Hg. Airey also reviews the literature on the MeHg concentration levels at which neurologic effects have been noted, and provided data on average hair Hg levels from different countries. For the United States, the weighted average was 2.9 ppm. She also developed several regression equations relating hair Hg to fish consumption, for example

$$\text{Hair Hg (ppm)} = 1.05 + 0.095 \text{ fish intake (kg/pers/y)} \qquad [8\text{-}5]$$

Based on the U.S. average consumption rate of 25 g/d, the U.S. average hair concentration should be about 1.9 ppm, according to this equation. Figure 8.4 is a plot of international data on Hg in hair vs. estimated daily MeHg dose; the regression line is approximately (without the Kenya outlier)

$$\text{hair Hg (ppm)} = 1.6 + 0.14 \text{ dose } (\mu g/\text{day}) \qquad [8\text{-}6]$$

This yields an average U.S. hair Hg level of about 2.3 ppm. Although Airey's paper was not specific on this point, it appears that her values are for total Hg in hair.

The country-average hair Hg data suggested higher values in midlatitude northern hemisphere countries which could be due to fossil fuel burning, but it was also suggested that lower soil temperatures in the north could retard Hg volatilization and thus re-emission of Hg back into the atmospheric cycle.

Other data on background levels of Hg include those of Chattopadhyay and Jervis (1974), who found median Hg levels of 1.2 ppm in rural Ontario (100 km from industrial areas) and 2.0-2.3 ppm in urban Toronto. Gowdy et al. (1977) report a mean concentration of about 14 ppb of total Hg in whole blood from 210 individuals in the Washington, DC area, which was reduced to about 8 ppb after deleting 9 "abnormal" values over 50 ppb. These figures would convert to approximately 3.5 and 2.0 ppm in hair, but Gowdy et al. also reported that "more than half" of the blood Hg was organic. Their ratio between whole blood and red blood cells was estimated to be about 0.76. No "stigmata" of Hg poisoning were noted in the 9 patients with more than 500 ppb in blood.

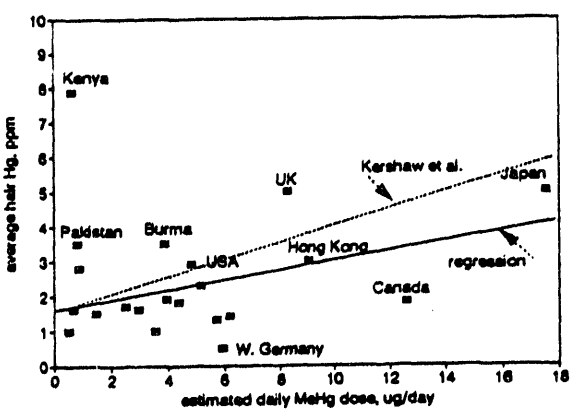

Figure 8.4 Average total Hg in hair for selected countries, as a function of estimated MeHg intake. Data from Airey (1983).

Kyle and Ghani (1982) presented data on fish consumption and hair Hg for Papua New Guineans which showed a relationship between the two parameters. This population may be of some interest since the probability of contamination by hair preparations may be lower than in more developed countries. The following relationship between inorganic and methylmercury in hair was derived from these data:

$$\text{Inorganic Hg} = 0.35 + 0.18\ \text{MeHg} \qquad [8\text{-}7]$$

the highest MeHg level used in developing this equation was 40 ppm. If we use Eq. 8-7 to adjust Airey's data to a MeHg basis, we get about 1.1 ppm as the baseline (no fish consumption) and 1.65 ppm with average fish consumption.

The highest hair Hg level reported in Ontario Amerindians by Richardson and Currie (1993) was 128 μg/g. Mercury levels (and apparent fish consumption) were also higher in summer and increased with age. Values for teenagers were about 1/3 of those for persons over 50.

8.3 An Algorithm for the Steady-State Distribution of MeHg Body Burdens in a Heterogeneous Population

Methylmercury does not accumulate indefinitely in humans, but follows a first-order decay process with elimination mainly in feces. This decay process is characterized by the biological half-life (t_b), which varies from about 50 to 150 days or more. The effects of MeHg, and its concentrations in organs, blood, and hair, will be a function of body burden per unit of body mass (BM), which is a kind of gross concentration metric. This unit body burden (UBB) will depend on the rate of intake m (which we assume to be steady) and the body mass and biological half-life for the individual. Variations within a population of these three parameters will give rise to variations in blood and hair concentrations and in neurological effects, if any.

The distributions of these parameters within a population will reflect two separate sources of variation: for each individual, variations in dietary intake; between individuals, variations in body mass and biological half-life. Data are available on the distributions of body mass and biological half-life but we must first estimate the distribution of individual intake rates and the corresponding equilibrium levels. To do this, we examine the dynamics of accumulation and excretion, according to the first-order decay law which has been found to be appropriate for MeHg:

$$BB_t = m(1-e^{-kt})/k \qquad [8\text{-}8]$$

where m is the daily intake rate and k is the elimination constant [ln(2)/ t_b]. At equilibrium, Eq. [8-8] integrates to

$$BB_{eq} = m/k \qquad [8\text{-}9]$$

We are interested in the distribution of BB_{eq} for an individual who consumes seafood by means of a random selection process from a supply that varies in mercury content. We wish to estimate the long-term probability that an individual could consistently obtain a sample that is biased either low or high with respect to the mean MeHg level, either within a given fish species or for all species. The parameters here are thus the mean and standard deviation of the mercury content of seafood, the rate of consumption, and the elimination constant.

Rate of consumption in turn is given by the average meal size and the frequency of eating, which specifically accounts for the fact that the only practical way an individual can increase his mercury dose from a random seafood supply is by eating fish more often. We take 200 g for the average (non-tuna) fish meal and 70 g for the average tuna meal. The frequency (f) of eating is thus given by m/200 or m/70, depending on the meal. For daily consumption, f=1.

The elimination constant k depends on t_b, which varies among individuals:

$$k = \ln(2)/ t_b \qquad [8\text{-}10]$$

Individuals with high t_bs will retain MeHg longer, which increases the body burden of MeHg for a given intake rate, without a concomitant increase in variability due to variations in MeHg in the food supply.

8.3.1 Simulation of Regression Toward the Mean for an Individual

We used Monte Carlo simulations to study the regression toward the mean of a distribution of sequential inputs with a first-order decay process for an individual seafood consumer. Without this decay process, the variance of the mean would be reduced directly with the number of independent meals. However, with decay, the most recently consumed meal counts more, and a closed-form solution is not convenient. We used @RISK (Palisades Corp., 1988) to simulate this process, for a two-year period (about 10 half-lives), for daily, weekly, and monthly consumption rates. In this example, we assumed a 200 g meal, drawn from a seafood supply characterized by a median Hg concentration of 0.2 μg/g and geometric standard deviations of 1.4, 2.2, and 3. t_bs of 35, 70, and 140 days were assumed. A lognormal distribution was assumed to represent the underlying distribution of mercury concentrations in individual meals. The distributions simulated by @RISK were expressed as base_{10} logarithms and then converted to actual values for the purpose of plotting.

The results are given in Figures 8.5 to 8.9. We see that both the mean values of BB_{eq} and their standard deviations are affected. Figure 8.5 shows that the values obtained from the simulation are consistently higher (both medians and means of the distributions) than the values obtained from Eq. [8-9]. This results from the non-linear nature of the process and the variability of the underlying mercury distribution, since the "excess" increases strongly with GSD (Figure 8.6). Changes in the elimination constant k had only minor effects on the equilibrium mean body burden. However, in terms of the extreme values of the equilibrium distributions, this gain in average body burden is offset by the reduction in variability that results from averaging over many meals. Figures 8.7a-c show that the GSD at equilibrium is sharply reduced, but levels off in exactly the same way that the body burden does. The product of GSD_{eq} and the square root of the number of meals is approximately constant for a given underlying GSD. Figure 8.8 shows the relationship between initial and equilibrium GSDs; we see that the elimination constant has a large effect for infrequent consumption and a much smaller effect for daily consumption. The percentage changes in GSD due to changes in k were about the same for both frequencies.

We used multiple regression analysis of the simulation results to develop an empirical expression for the data shown in Figure 8.8 that would be suitable for use in the risk analysis. Basically, we need to predict the equilibrium standard deviation of the body burden of MeHg as a function of the underlying standard deviation of the food supply, the frequency of consumption, and the individual t_b. The best fit in terms of R^2 was obtained by fitting the logs of the standard deviations derived from the simulation:

$$\log(\text{sigma}) = -1.334 + 0.438 \log(k) + 0.485 \log(1/f) + 1.795 \log (GSD_0) \qquad [8\text{-}11]$$

This expression comes very close to the 1/2 power relationships for k and f that would be expected from theory, and we found that the error entailed by substituting 0.5 for these coefficients was negligible. This yielded the following empirical relationship:

$$\log(\text{sigma}) = -1.211 + 1.766 \log(GSD_0) + 0.5 \log (k/f) \qquad [8\text{-}12]$$

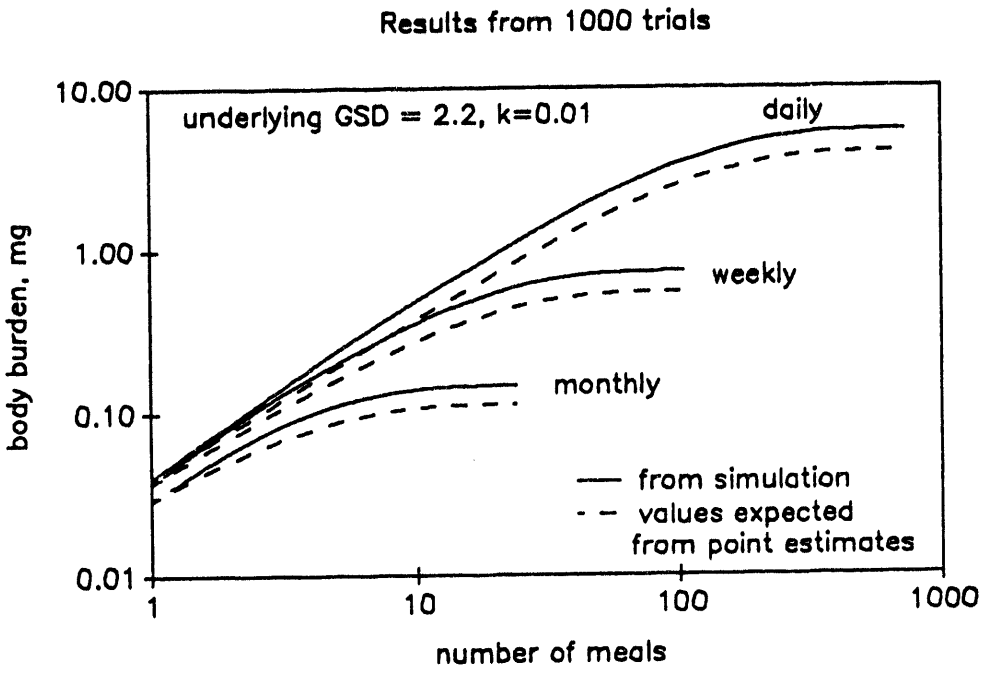

Figure 8.5. Results from simulations of the approach to equilibrium body burden of MeHg for various rates of fish consumption.

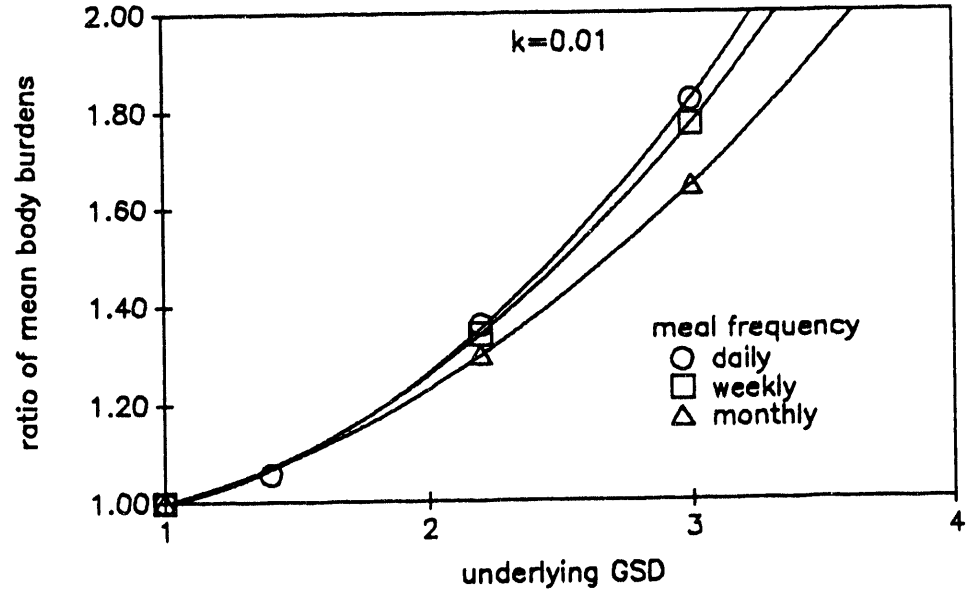

Figure 8.6. Effect of underlying GSD and meal frequency on the ratio mean equilibrium body burden values.

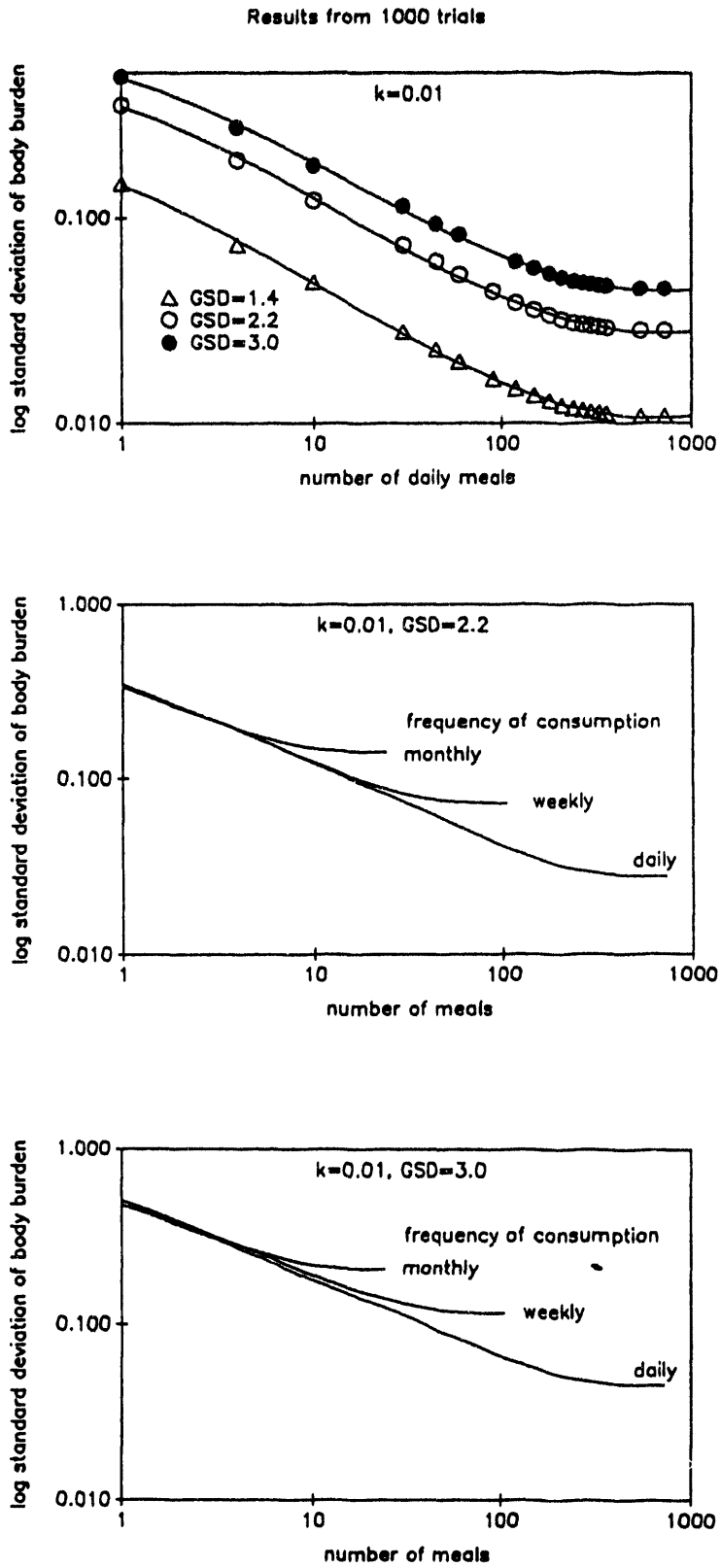

Figure 8.7. Reduction in the standard deviation of body burden as a function of number of fish meals consumed in the time to reach equilibrium. (a) for different underlying GSDs. (b) as a function of meal frequency for GSD_0 = 2.2 (c) as a function of meal frequency for GSD_0 = 3.0.

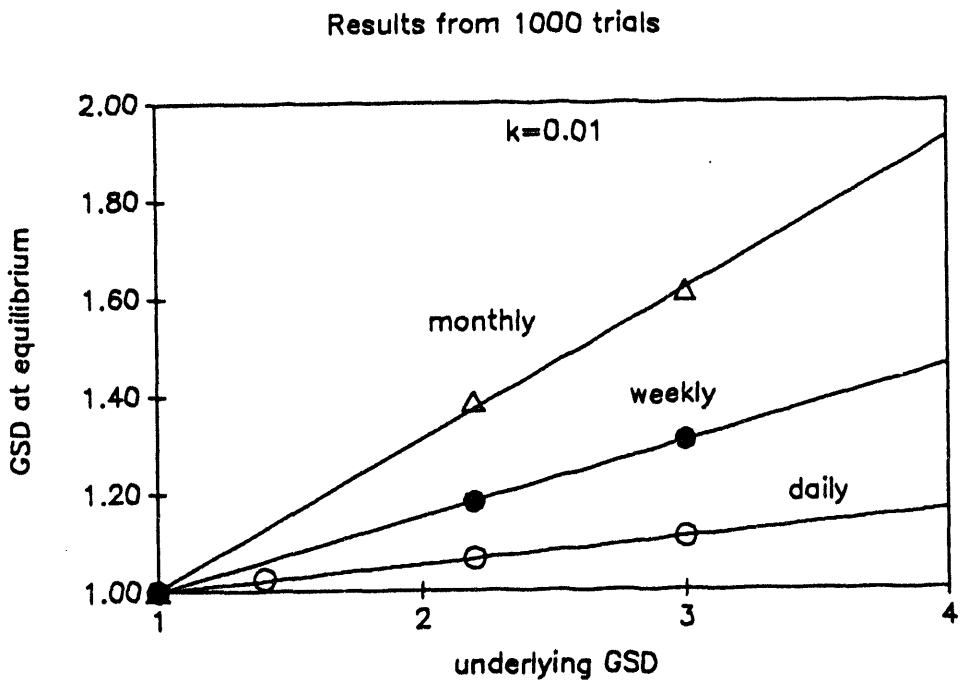

Figure 8.8 Effect of underlying GSD and meal frequency on equilibrium body burden GSD.

Figure 8.9 compares the values of the standard deviations at equilibrium as derived from the simulations with those predicted by Eq. 8-12. We see two areas of disagreement, near the origin and at large values of sigma, both constituting overprediction of variability. The former results from the fact that we expect sigma to equal zero when the underlying GSD is unity; however, the forms of Eq. 8-11 and 8-12 do not admit zeros for sigma. The overprediction at high values of sigma result in part from plotting Figure 8.9 in terms of actual values rather than the logarithms which were used in developing the empirical fit. In the region of primary interest, small but non-zero values of sigma, the two sets of data agree very well. Since overprediction of variability will lead to increased risks and is thus "conservative," we deemed Eq. 8-12 acceptable for our purposes.

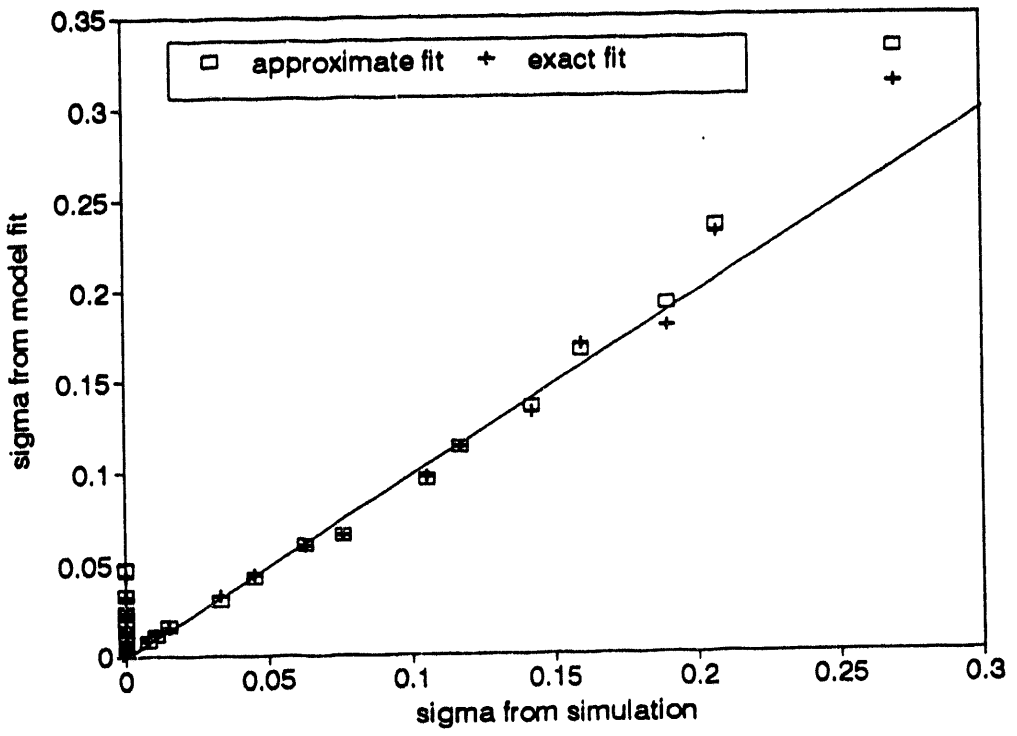

Figure 8.9 Comparison of approximate estimates of equilibrium GSD to the values obtained from the simulation, for Eq. 8.11 (exact fit) and 8.12 (approximate fit).

8.3.2 Application to Simulation of Equilibrium Body Burdens

As an example, these data show that increasing the frequency of fish consumption from monthly to daily increases the mean body burden of MeHg by about a factor of 30, but that the upper points of the distribution will only increase by about a factor of 20.

The empirical relationships from Figures 8.6 and 8.8 are then used in a Monte Carlo simulation of a population of fish consumers, in which distributions of m, t_b, and BM are used to generate a distribution of unit body burdens. The GSDs of these distributions may then be compared with the observed GSDs of mercury in blood and in hair, as a reality check on the overall simulation. These results are provided in Section 10, in conjunction with the actual risk assessment calculations.

9.0 HEALTH EFFECTS OF MERCURY AND DOSE-RESPONSE FUNCTIONS

The central nervous system is the principal target for MeHg, with the potential for effects on sensory, visual, and auditory functions. Low doses may create non-specific symptoms such as paresthesia (tingling of the extremities), malaise, or blurred vision. Higher doses may bring deafness, loss of coordination when walking, and speech disorders, and, in extreme cases, coma and death (WHO, 1990). Effects of MeHG on the developing fetus are thought to be more critical than on adults. According to WHO (1990), "it affects normal neuronal development, leading to altered brain architecture, heterotopic cells, and decreased brain size."

In this section, we use data from the Iraqi grain poisoning incident to develop a dose-repsonse function for use in the risk assessment, and we also review various epidemiological studies on chronic intake of MeHg. Adult paresthesia was selected as the endpoint for the dose-response function. Paresthesia is perhaps the mildest readily observable symptom of MeHg poisoning in adults.

One of the requirements for assessing the health risks of mercury as a pollutant is a valid dose-response function. The supporting data for such functions have largely been obtained from past incidents of acute methylmercury (MeHg) poisoning. Application of such acute response data to chronic exposures is still an open question, but, at present, these are the only data available to work with.

9.1 The Iraqi Grain Poisoning Data

The incident described below is that of a mass poisoning incident in Iraq in 1971-72 resulting from use of seed grain treated with mercury fungicides to bake home-made bread (Bakir et al., 1973). However, the data presented by Bakir et al. are incomplete and requires some manipulation and cross-referencing with other publications. The basic data set includes 122 Iraqi adults and older children, for whom blood mercury levels were determined sometime after the incident. This determination was based in part on consumption of the poisoned bread and calculations of the body burdens of MeHg; the independent (dose) variable in this study may therefore be uncertain. Various neurological symptoms were noted, up to and including death. Of these symptoms, paresthesia (numbness and tingling of the extremities) may be considered the least severe and has been selected for this exploration. There were 59 observed cases of paresthesia, distributed according to blood level grouping as shown in Table 9.1.

Table 9.1 Frequency of Paresthesia in Iraqi Adults*

group	#	estimated blood Hg range (ng/ml)	body burden (mg)	no. of subjects	no. of cases	frequency
1		0-100	4	21**	2	0.095
2		101-500	25	19**	1	0.05
3		501-1000	55	19	8	0.42
4		1001-2000	105	17	10	0.59
5		2001-3000	168	25	20	0.80
6		3001-4000	202	17	14	0.82
7		4001-5000	243	4	4	1.00

* actually, patients over age 9 ** regarded as controls

It should be noted that paresthesia is not uncommon in unexposed populations and thus that some residual or background prevalence rate should be expected, perhaps of the order of a few percent. Note also that MeHg blood levels of around 10 ng/ml are expected in normal unexposed populations and thus that it is highly likely that all of these subjects were actually exposed to contaminated bread.

Bakir et al. (1973) provided a description of the incident. Patients were seen in hospital and at home. The dose was estimated from the MeHg in flour, the weight of each loaf, and estimates by patients of the number of loaves consumed over specified periods. Symptoms were judged, and data for 120 patients were grouped into 7 exposure categories. The lowest 2 groups were considered controls (unexposed): the frequency of paresthesia in these 2 groups combined was 7.5%.

Blood MeHg data were obtained after the fact, from 20 to 115 days after exposure (average of about 65 days later). This means that the blood concentrations are low by about a factor of 2. This was mentioned by Bakir et al., but was recognized specifically by Marsh (1987).

These delayed blood MeHg determinations were used in a dynamic equation which accounted for the loss of MeHg only during the accumulation (exposure) phase to estimate body burdens, at the time of symptoms and at peak exposures. These body-burden data were not tabulated by Bakir et al., but may be read from the graphs. They correspond well with the values tabulated subsequently by Nordberg and Strangert (1978). If a blood half-life of from 62 to 75 days is used, the blood MeHg levels tabulated by Bakir et al. can be reproduced, starting with the body burdens given by Nordberg and Strangert, and indeed, Bakir et al. state that this is how their body burdens were derived (the average body weight for the patients is needed, and may be obtained from Al-Shahristani et al. (1974): 46.6 kg). This means that the symptom frequencies plotted by Bakir et al. have been associated with both blood MeHg and body burdens that are too low, by about a factor of 2.

An alternate source of information is the study based on hair MeHg by Al-Shahristani et al. (1974). This paper gives data on patient age and sex, daily input of MeHg, body weight (indirectly), and the rate of daily increase of MeHg in hair during the accumulation phase, for 30 selected patients. Near the start of exposure, it may be assumed that the rate of increase in hair is proportional to the daily intake level (since there hasn't been enough time for substantial excretion). Figure 9.1 plots the daily increase in hair MeHg against the estimated daily intake per unit of body mass.* One outlier is seen, and the regression line was calculated without this observation.

Table 9.2 Data on MeHg in Hair from Iraqi Poisoning Victims
source: Al-Shahristani et al. (1974)

Symptoms	Peak hair MeH(μg/g)	Body burden(mg)	Blood MeHg(μg/L)#	Blood/body burden
None	1-300	0.5-100	0-1200	54.5
Mild	120-600	37-200	480-2400	60-54.5
Moderate	200-800	70-280	800-3200	53.3
Severe	400-1600	140-560	1600-2400	53.3

#based on a ratio of 250.

* Curiously, the body weight data obtained from the data of Al-Shahristani et al. (1974) were largely values divisble by 5 and monotonically related to age (Figure 9.2). This strongly suggests that weights were not measured but were imputed from age instead. If so, this may be one of the sources of scatter in the dose-response plot (Figure 9.2).

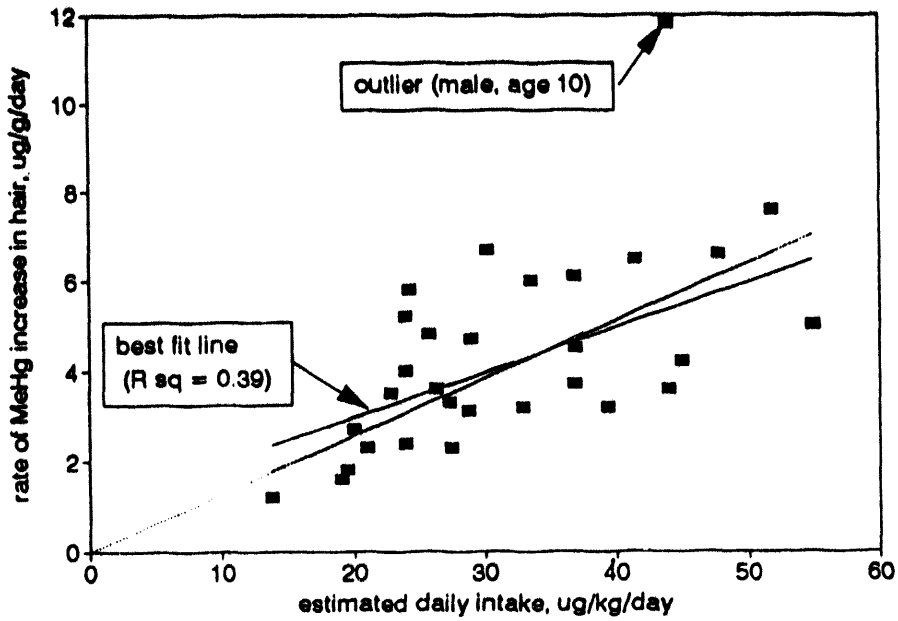

Figure 9.1. Relationship between MeHg in hair and daily intake in selected Iraqi poisoning victims.

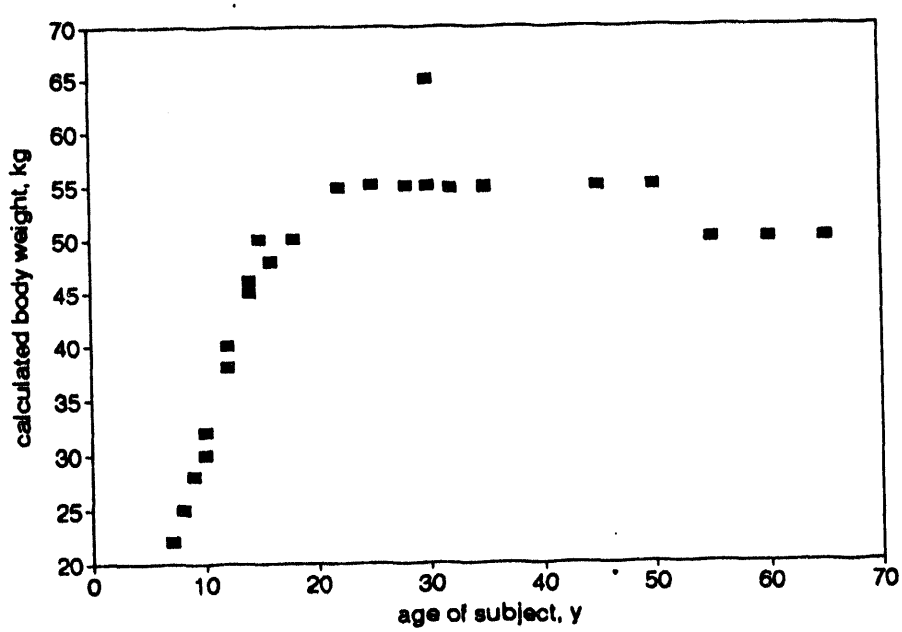

Figure 9.2. Relationship between body weight and age in the data of Al-Shahristani et al. (1974)

This paper also gives the ratio of body burden to peak hair concentration, and data on symptom severity classified by both body burdens and hair levels. Corresponding levels of blood Hg were estimated, as well as a conversion factor for body burden to blood, using a conversion factor of hair MeHg to blood Me Hg of 250, as shown in Table 9.2.

From these data, the onset of "mild" symptoms occurs at a blood level of 480 μg/L, not at 240 μg/L as implied by Bakir et al. and subsequent reports based on these data, including the (1990) WHO report (the 1976 WHO report on mercury estimates the threshold to lie "somewhere between" 240 and 480 μg/L). The corresponding threshold total body burden is also higher. It is probably safe to assume that these patients did not include cases in which deafness or death occurred, which corresponds to body burdens above about 150 mg, according to the plots of Bakir et al. (1973), which is taken as further indication that the Bakir et al. body burdens are too low.

Additional support for this hypothesis is obtained from the paper of Al-Mufti et al. (1974). They reported detailed Hg poisoning symptoms in two specific villages. Contaminated bread was consumed in one of the villages; in the other, it was not. The incidence of paresthesia was reported as 38% among consumers and 2.2% in nonconsumers. The average intake was 150 mg of MeHg over about 40 days, for a daily input of about 4 mg/d. Using the relationship between accumulated body burden and daily consumption,

$$BB = m(1-e^{-kt})/k$$

the body burden after about 40 days is estimated to have been 132 mg. If a longer consumption period, say 50 days, is selected, a body burden of 118 mg is derived. The data of Nordberg and Strangert (1978) show a value of 55 mg for about the same frequency of paresthesia (42%), which is clearly well below the figures derived from bread consumption.

Further evidence for this hypothesis is derived from the dose-response data of Al-Mufti et al. They divided the residents of the consuming village into four groups, by number of loaves consumed. These figures were converted into body burdens by using the group average numbers of loaves consumed, the average MeHg content per loaf (1.27 mg) and the numbers of days of consumption from the data of Bakir et al. (1973), either 40 or 50 days. This dose-response function is plotted in Figure 9.3 (for both time assumptions) along with the data of Bakir et al. (1973, frequencies) and Nordberg and Strangert (1978, body burdens). There is a substantial mismatch between the bread-based data and the original Bakir-Nordberg data. However, if the Nordberg and Strangert body burdens are arbitrarily doubled to account for the 65-day delay in determining the blood chemistry, as discusssd above, the two dose-response functions agree quite well. There is even an implication that the consumption time may have exceeded 50 days at the higher dose levels, which is also a reasonable inference. The 1976 WHO report discussed this problem of uncertainties, blood halflives, total ingested doses, and imputation of body burdens at some length. Based on their analysis, these body burden values may be somewhat on the high side, but the report of Marsh (1987) and the comparison of Figure 9.3 seems to be sufficient evidence to proceed.

9.2 Development of a Dose-Response Function

A quantitative health risk assessment requires a mathematical dose-response function that predicts adverse health effects in terms of the imputed dose. It must be able accommodate uncertainties in the (dose) input terms, and the uncertainties of the dose-response model itself must be defined, including those associated with the choice of model parameters or functional form. Data were used on prevalence of paresthesia in 122 patients reported by Bakir et al. (1973), in conjunction with the 1972 poisoning incident described above. These patients were grouped according to blood mercury levels at the time of examination. The upper 5 groups were known to have eaten contaminated bread; the lower two groups were regarded as "controls." Relevant statistics were given in Table 9.1. As discussed above, their unit body burdens of MeHg at the time of symptom reporting were estimated by doubling the data

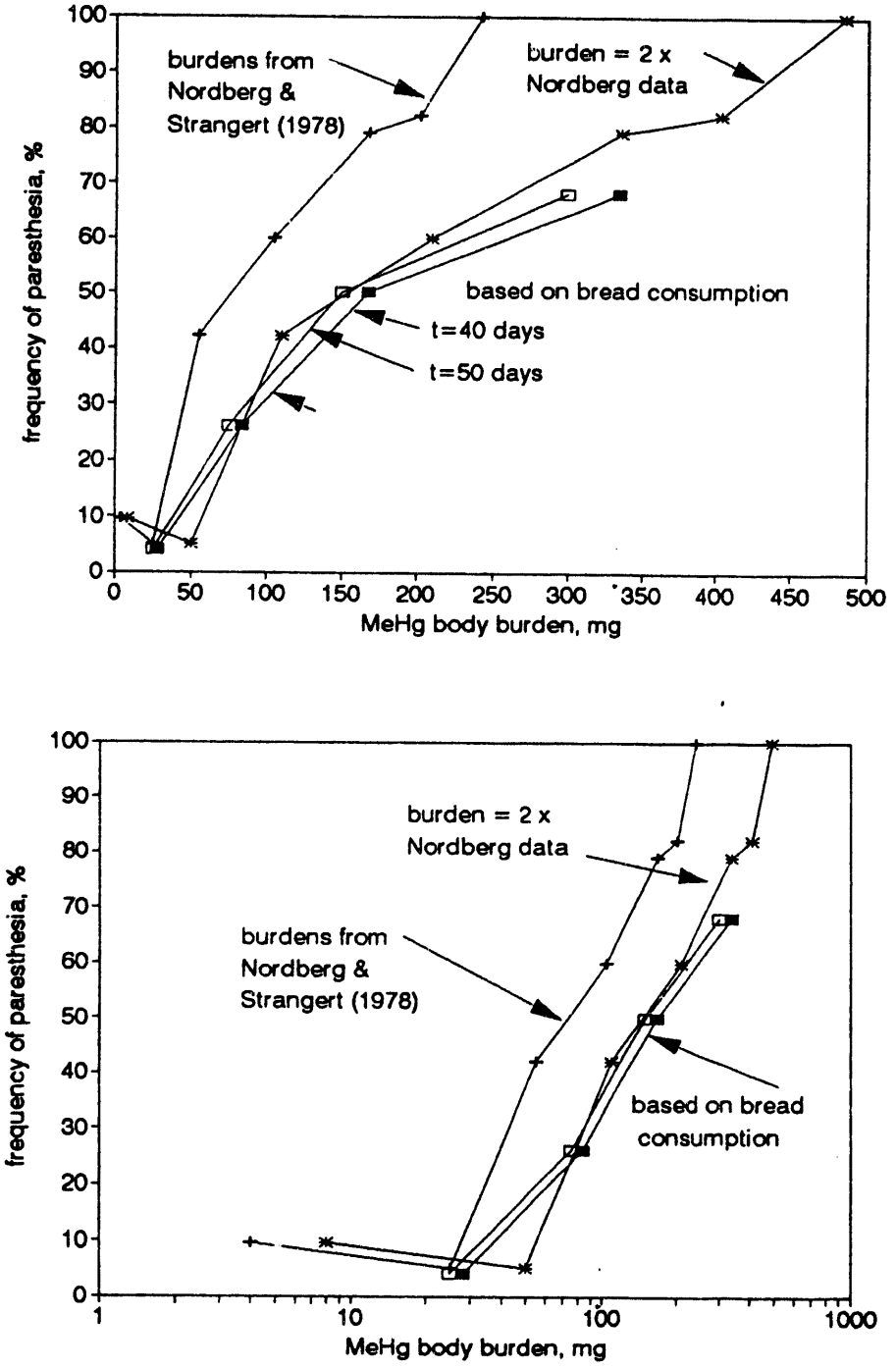

Figure 9.3. Dose-response data for paresthesia in the Iraqi poisoning incident, according to various formulations. (a) Linear dose. (b) Logarithmic dose.

given by Nordberg and Strangert (1978), and by dividing by the average body weight (46.6 kg) deduced from the data of Al-Shahristani et al. (1974). This formulation allows the frequency of paresthesia to be estimated in response to the joint distributions of dietary MeHg intake and body weight.

The logistic function was used to represent these data. This is an "S" shaped curve with asymptotes at both extremes. The "logit" is defined as ln(p/(1-p)), where p is the probability of an event or frequency of a condition or symptom. The logit function increases rapidly as p approaches 1, but never reaches 1. The use of the logarithm of dose spreads out the low end of the curve, which never reaches zero. When this function is used to represent grouped prevalence data, the statistic used to judge goodness of fit is the chi-squared, in which observed and expected numbers of cases are compared across the entire range. However, in this application, predicting paresthesia at low doses is of much more interest than at high values, so that some discretion was used in evaluating candidate models.

An alternative model that might be applied here is the "hockey stick," consisting of two intersecting straight lines. The point of intersection is the threshold of effect, which is strongly linked to the background prevalence level of the condition in question. The IPCS (1990) report cited a background level of 6.3% for paresthesia in Iraq. However, a detailed analysis by Al-Mufti et al. (1974) found only 22 cases in a presumably unexposed population of 1012, for a background level of 2.2%. If children under age 10 were eliminated from this population, the background paresthesia prevalence rate would be about 3.7%. The background rate implied by the "controls" among the 122 patients reported by Bakir et al. (Table 9.1) is 7.5 %. A range of background rates was investigated in the development of a dose-response function for paresthesia.

The "logit" regression model procedure of SPSS (Norusis, 1990) was used for this purpose, which fits a model of the form logit = $B_0 + B_1 \log(x)$. Standard errors are estimated for both regression coefficients, and these are used in a probabilistic error analysis. Figure 9.4 presents the results of some of these computations; none of the logistic curves fits all of the Iraqi data.

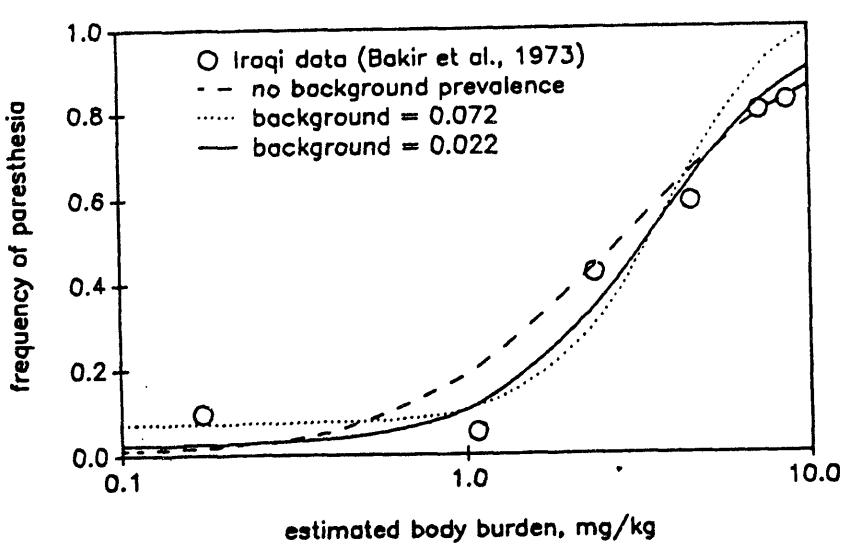

Figure 9.4. Logistic regression fits to the Iraqi paresthesia data.

With low background paresthesia assumed, the response is greatly overestimated at a body burden of 1.0. With higher assumed background levels, it is underestimated in the region of body burdens around 2.0 and tends to overestimate the response at higher burdens. The sum of the squared deviations from the lowest three data points on Figure 9.4 was used as a relative figure of merit, and found that a background paresthesia prevalence rate of 2.2% was close to the optimum. Since this is also the background level determined by Al-Mufti et al. (1974), it seemed a reasonable choice. This model is given by

$$\ln(p/(1-p)) = 2(3.80 + 2.20\log(BB) - 5) \qquad [9-1]$$

with standard errors of 0.30 and 0.43, respectively. The distribution of responses at body burdens of 0.1 and 1.0 mg/kg are shown in Figure 9.5. The median values of paresthesia risk are 0.11% and 0.083%, to which the background level of 2.2% must be added in order to compare with Figure 9.4. The 95th percentiles are 0.62% and 19.5%, respectively. It is seen from Figures 9.4 and 9.5 that the full range of dose-response models is included in the confidence limits at a body burden value of 1.0 mg/kg. Also, the GSDs of these two distributions are 2.88 and 1.72, respectively. The fact that the GSD is larger and thus that the response is less certain at the lower body burden levels is also consistent with the present understanding of the data.

It has been suggested (J. Fouts, personal communication, 1993) that the existence of an actual dose-response threshold for paresthesia may be problematic because of the small numbers of observations at low doses. After all, the probabilities of finding 0, 1, 2, 3 or 4 cases in a sample of 20 are not that different. For this reason, the lack of fit of the logistic model at low doses, where only 1 or 2 cases were observed, should not be taken too seriously.

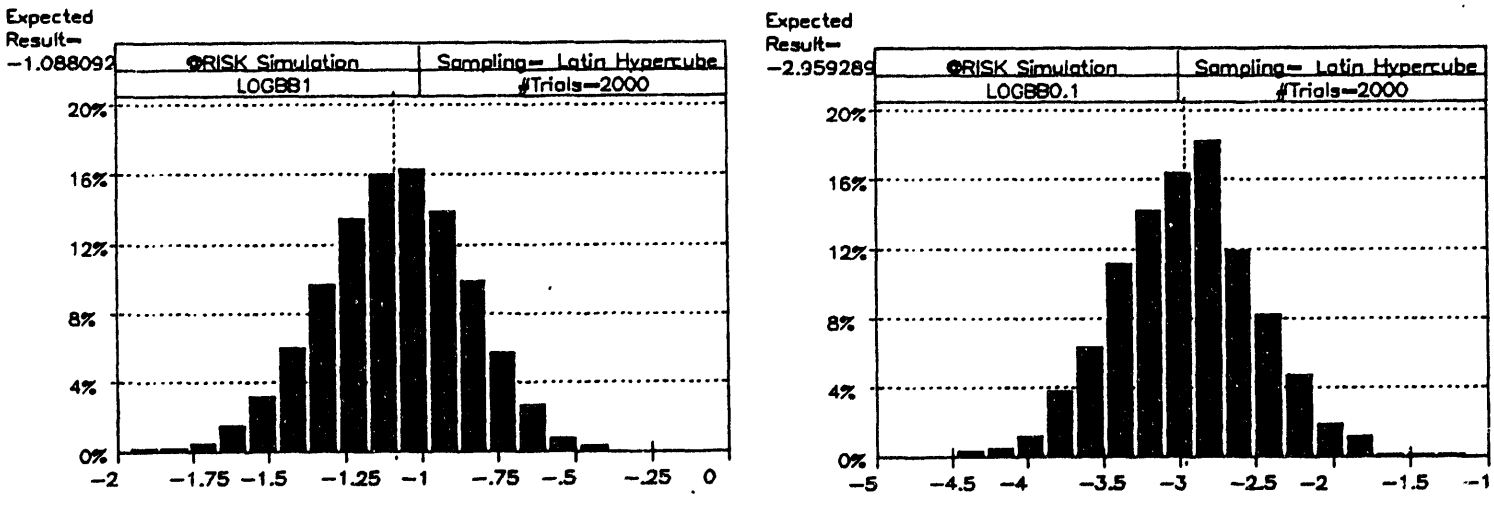

Figure 9.5. Histograms of paresthesia risk based simulations and the confidence limits of the regression equation. (a) body burden = 0.1. (b) body burden = 1.0

9.3 Other Epidemiological Observations of MeHg in Fish-eating Populations

Since the Minamata disaster in Japan, there have been many attempts to observe neurological symptoms in various fisheating subpopulations. Some of these studies use estimated dietary levels as an indicator of exposure; others use the Hg content of samples of blood or hair.

9.3.1 Results from Population Studies. In response to concerns about high levels of Hg discharges from chlor-alkali plants associated with the pulp and paper industry, an extensive population sampling and study program was launched in Canada in 1970, with emphasis on native populations that typically consume large quantities of fish (Wheatley, 1979). Over 35,000 hair and blood samples were taken in 350 communities, from 1971-78. 84 "at risk" patients were identified from these samples and given neurological examinations. 11 of these had "neurological findings possibly attributable to methylmercury." However, no definitive diagnosis of MeHg poisoning was made, and no relationship between peak Hg levels and neurological findings could be discerned.

Birke et al. (1972) studied 14 "normal" and 12 "exposed" subjects who had eaten contaminated fish. They reported total and methylmercury levels in whole blood and in blood cells and in hair. These data provide a rudimentary means of converting among commonly used mercury metrics. For example, MeHg was about 48% of total Hg in blood cells, and total Hg in whole blood was about 55% of total Hg in blood cells. MeHg in whole blood was not reported. Total Hg in hair was about 300 times that in whole blood, but not all of the hair Hg was MeHg. The relationship between total Hg in whole blood and MeHg dietary intake for 20 subjects was given by

$$\text{Hg (whole blood) (ng/g)} = 1.2 + 0.80\ \mu\text{g/d} \qquad [9\text{-}2]$$

Hg decay curves were constructed for up to 5 subjects who stopped consuming contaminated fish. After accounting for new background levels, the biologic half lives were 99 and 120 days in red blood cells and from 33 to 120 days in hair (mean=80 days). Clinical examinations showed findings of interest for two subjects. A 73 yr old male who consumed about 150 μg/d MeHg, had whole blood Hg of 125 μg/L, hair Hg of 40 ppm, and was found to have "coarse tremor of suggested intentional type and dysgraphia." The most heavily exposed individual, a 54 yr old male who consumed about 800 μg/d had a whole blood level of 650 μg/L, hair Hg of 185 ppm, and showed a "very slight coarse finger tremor in 2 examinations. However, the authors concluded that "none of the eight moderate or heavy consumers of contaminated fish showed any clear-cut clinical picture of methylmercury poisoning."

Skerfving (1974) reported blood, hair, and consumption levels of Hg for 162 Swedish fisheaters, and prevalence of 30 neurological symptoms and findings for two subgroups of "high" and "low" exposure, as judged from levels of total Hg in red blood cells. The relationship between consumption and blood for 22 subjects with relatively steady long-term intake was found to be

$$\text{Hg (blood cells) (ng/g)} = 0.02 + 0.07 \text{ daily intake } (\mu\text{g/kg/d}) \qquad [9\text{-}3]$$

When compared in the same units of measure, this slope is lower than those found by Miettinen or by Birke et al. (Figure 8.2), for example. The ratio between Hg in head hair and in blood cells was 230. The half-lives of Hg in blood cells ranged from 59 to 87 days for four subjects; a fifth had a value of 164 days. There were no statistically significant differences in neurological symptoms or findings between the high (median blood Hg = 160 ng/g) and low (median = 40 ng/g) groups. Continuous paresthesia was reported by 1/42 of the low Hg group and 2/41 of the high Hg group. This is a combined prevalence rate of 3.6%. "Impairment of superficial sensibility" affected 8 of the low Hg subjects and 6 of the high Hg subjects, for a combined prevalence rate of about 17%. Skerfving concluded that none of the 86 subjects showed a "clear-cut case of poisoning."

Bernstein (1974) reported data from 3 groups of Cree Indians in Northern Quebec, obtained in 1971. Blood Hg data were obtained from 401 persons; means of the 3 groups ranged from 22 to 41 ppb and the highest value was 306 ppb. Hair was tested from 67 people; the correlation was 0.82 and the slope was about 250. Detailed data on fish consumption were not taken, but the those individuals with the highest blood Hg appeared to be heavier consumers of fish. There was some evidence that fish Hg content was higher in the area with higher blood Hg.

Five of the individuals with high Hg levels were selected for hospital examination in Montreal. Psychological evaluation was impossible because of language difficulties. No specific mention was made of evaluation of tremors or of constricted fields; Bernstein concluded that "no significant clinical findings suggestive of organic mercury excess" were found.

Some of these same bands of Cree Indians were studied later by McKeown-Eyssen and Ruedy (1983), using the case-control methodology. 41 cases were identified through neurological examination that was blind to MeHg levels or fish consumption. Requirements for classification as a "case" included either symmetric bilateral reduction in visual fields, or the presence of neurologic disease as judged by the bilateral presence of any of a number of symptoms, including tremor. "Controls" (179 in number) were required to have none of the possible markers for cases. Since there were 460 subjects in toto, it was conclude that the remaining 240 met neither set of criteria and that the "case" prevalence rate in this population was about 9%. Although the mean blood Hg level of cases was significantly higher than that of controls, there were several potentially confounding variables to consider, including age and alcohol use. After attempts were made to control for these factors, the odds ratio for those with hair Hg between 10 and 19.9 μg/g was about twice that for those with hair Hg less than 10 μg/g. It was not possible to determine a threshold hair Hg level, and the authors suggested that the Hg levels used might be lower than previous exposure levels. They also concluded that "it remains possible that the effects are not entirely attributable to methylmercury." For example, note that the mechanism for increased exposure to MeHg within a limited geographic area is by eating more recreationally caught fish, as opposed to eating fish with higher mercury contents. There could be other differences associated with high local fish consumption, such as poverty, residence in the "bush," adherence to local customs which might involve inbreeding, etc. A more convincing demonstration might involve comparing native groups whose MeHg exposure differences came from differences in the MeHg content of the fish in their diets. No mention was made in the paper of possible differences in MeHg content of fish between the two communities studied. (Check Wheatley).

The companion to this study, McKeown et al. (1983), examined 234 Cree infants, ages 12-30 months, for neurologic, physical, mental, and psychosocial development. Only one neurologic measure was found to be significant (boys' abnormality of muscle tone), at the 5% significance level. Considering both boys and girls, this is 1 out of 16 independent tests, which is very close to expectations just due to chance. There is no evidence of delayed development at these exposure levels (maternal prenatal hair Hg up to about 24 μg/g. Since others (Stern, 1993) have suggested that prenatal exposures may be up to 4 times more sensitive than adult exposures, the findings on Cree infants do not support the findings on adults.

Harada et al. (1977) examined surveyed a group of 89 Ontario Indians who had consumed fish contaminated with Hg by a chemical plant. Fish were mainly eaten there in summer, and hair Hg was noticeably higher (up to 80 ppm) among fisheaters. The neurological examinations did not find the symptoms typical of MeHg poisoning, such as concentric visual constriction or sensory disturbances, However, the results "strongly suggested that neurological symptoms had been caused by methylmercury. The following approximate median Hg hair levels were noted, by symptom group:

1. no symptoms	5 ppm
2. only subjective symptoms	8 ppm
3. other symptoms w/o sensory disturbance	22 ppm
4. sensory disturbance with other symptoms	21 ppm
5. sensory disturbance with visual constriction	17 ppm

However, it should also be noted that all levels of hair mercury were seen within group 5, i.e., there was no evidence of a dose response relationship within this group. Also, symptom data were given for only 58 of the 89 subjects, and no mention was made as to whether the neurological evaluation was "blind." Finally, the reservation expressed above about possible confounding by lifestyle differences pertinent to heavy fisheaters applies here as well.

Valciukas et al. (1986) report a study of 200 male and 200 female Mohawk Indians suspected of exposure to MeHg through fish taken from a river contaminated by a chlor-alkali plant. Blood and hair Hg levels were determined

and a battery of mental performance tests was administered. Females were reported to consume less fish and their median blood Hg levels were about 55% of the males, although their median hair levels were the same. The distributions of Hg measures were reported to be closer to log-normal than to normal. Median total Hg levels in blood (assumed to be whole blood) were 2.6 ppb for males and 1.4 for females, of which about 70% was reported to be "organic" Hg (assumed to be MeHg). The maximum total Hg for males was 19.5 ppb (93% organic) for males and 11.3 ppb (89% organic) for females. The authors reported that "no significant associations between age-adjusted individual performance test scores and exposure were found." However, a significance level of 0.04 was noted for females on one test and of 0.13 for all three combined, with respect to total blood Hg. Since the females' test scores were less significant with respect to organic blood Hg, the authors' conclusions would probably be sustained. However, this may be a close call and it should be noted that neurological symptoms such as paresthesia were not evaluated.

9.3.2 Conclusions from Other Epidemiological Studies. Relationships between fish consumption and human Hg levels have been shown in a number of studies. A number of different bases are in use for reporting biological concentrations of Hg; care must be taken in making comparisons. There are also a number of different criteria in use for judging neurological effects and for concluding that an association might exist at mercury levels resulting from fish consumption. One possible point of contention regarding sensory disturbances is that of definitions; in the chronic studies of fisheaters, permanent symptoms are sought, while this is clearly not possible with acute poisoning incidents (except perhaps after extended follow-up). It is concluded that the evidence for neurologic effects in adults at hair levels around 20 ppm (80 ppb in whole blood) is only suggestive. This level of MeHg body burden is about 10 times the average levels expected in a "normal" population; based on a GSD of 2.3, only 0.1% of such population would exceed the 20 ppm hair level.

10.0 ESTIMATES OF RISKS OF PARESTHESIA

10.1 Bases for the Estimates

Estimates of health risks were derived by combining all the probabilistic elements derived above, as follows, using the @RISK code (Palisade Corp., 1988). The three different seafood categories defined in Section 7 (canned tuna, freshwater fin fish, and marine fin fish) were treated separately in terms of their distributions of MeHg content and rates of consumption. Their sum was used to estimate the distributions of MeHg dose and equilibrium body burden.

Simulations were performed for the baseline case and for the "impact" case, in which the Hg content of freshwater fish was increased by a factor intended to capture the incremental local effects of Hg emitted from a 1000 MW_e hypothetical power plant. This factor was assumed to be uniformly distributed over the range 1.5 to 3.0. The effects of power plant Hg emissions on marine species were assumed to be negligible, since these species are primarily affected by global levels of Hg and the U.S. utility industry emissions contribute only a small fraction of the existing global Hg pool.

The human population at risk was assumed to be adults, characterized as follows:

body weight (kg):	median = 71.4, GSD = 1.26
MeHg half-life (days):	median = 68.2, GSD = 1.395

Fish consumption rates and Hg contents were as given in Table 7.2. The dose-response function for adult paresthesia was as given in Eq. 9-1.

Two additional features were incorporated into the risk assessment as alternative simulations. First, fish consumption (individual meal weight) was linked with body weight, using a variable correlation coefficient with a uniform distribution from 0.5 to 1.0. The correlation was taken as a variable because of the lack of reliable data on this relationship. The distribution of overall consumption rate (g/d) was left unchanged by this linkage. Second,

calculations were made with and without the algorithm that mimics the metabolic MeHg dose variance reduction due to the averaging process that takes place as the equilibrium body burden is reached (see Section 8.3).

The empirical equation for variance reduction is given by

$$\log(GSD_{eq}) = 10^{(-1.211 + 1.766\log(GSD_0) + 0.5\log(k/f))} \qquad [10\text{-}1]$$

where GSD_0 is the geometric standard deviation of the mercury content in seafood, from surveys, f is the frequency of meals, k is the elimination constant, and GSD_{eq} is the effective variance of mercury content used to compute the distributions of body burden.

The elimination constant depends on half-life and the frequency of eating fish is determined from the overall time period of the simulations (5 half-lives), the average daily consumption rate (from surveys), and the meal size (with optional linkage to body weight). The nominal meal sizes were 78 g for tuna and 200 g for all other fish. The equilibrium body burden is thus estimated in four different ways, in each instance for baseline and impact cases, and the logistic dose-response function is used to estimate the risk of paresthesia for each distribution of body burdens. Figure 10.1 is a flow chart of these logical relationships.

Factors that were not linked were:

1. Body weight and half-life were assumed to be independent.
2. Fish consumption rate was assumed to be independent of fish Hg content.

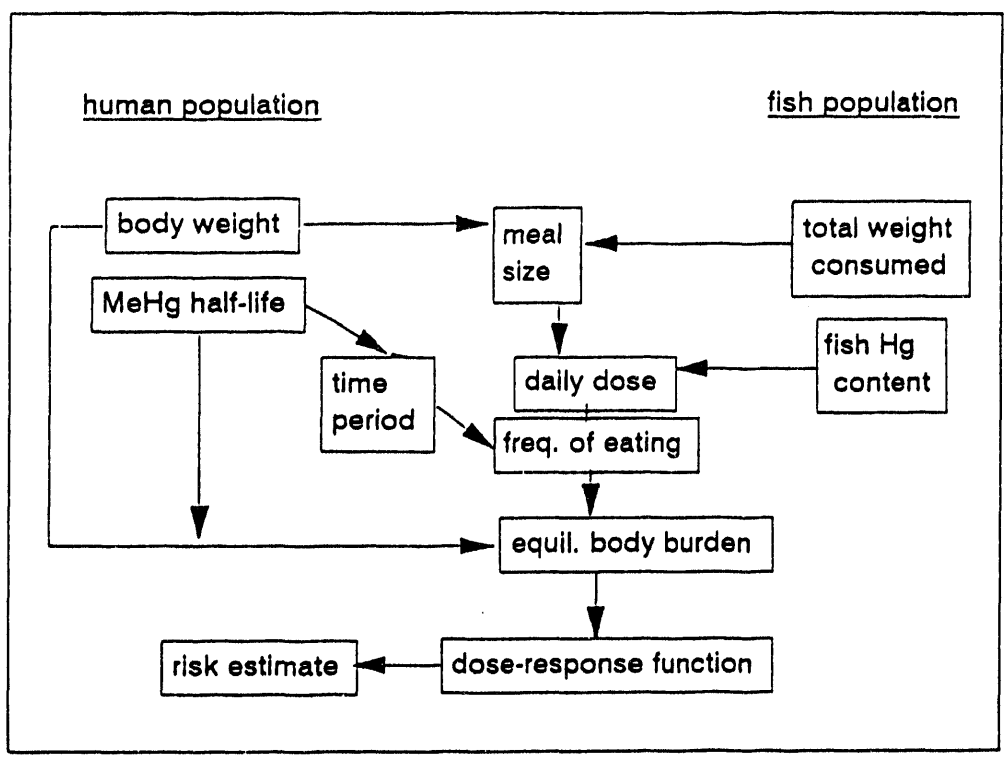

Figure 10.1 Flow chart for risk assessment logic

The simulations produced distributions of: numbers of fish meals consumed, average daily MeHg dose, equilibrium body burden of MeHg, and risk of paresthesia. Table 10.1 presents these results for the four simulation cases, in terms of medians, means, and 95th percentiles of each parameter.

10.2 Baseline Results

Among the four simulation cases reported in Table 10.1, the median number of meals consumed was reduced by about 16% by linking meal size to body weight (cases 3 and 4). This also reduced the MeHg dose correspondingly but increasing the results for cases 3 and 4 by 16% as compensation would not affect the overall findings of the risk assessment. Note that the distribution of doses is well below the EPA reference dose (0.3 μg/kg/day or 21 μg/d for a 70 kg person). The mean dose values were also quite consistent with that of Stern (1993), who estimated 3.8 μg/d at the mean with a 95th percentile of 15.3 μg/d (for fish consumers). However, Stern's apparent GSD for his distribution of doses was about 3.8, which is much broader than the present results. The WHO estimate for mean MeHg intake for the whole population is 2.4 μg/d (WHO, 1991); this somewhat lower value may reflect the absence of appreciable rates of freshwater fish consumption in the general population.

Table 10.1 Probabilistic Risk Assessment Results (5000 trials)

Case		1	2	3	4
meal size linked to body weight		N	N	Y	Y
fish Hg averaged over time		N	Y	N	Y
# of meals consumed in 5 half-lives	median	44	44	38	38
	mean	55	56	43	43
	95%	131	132	87	86
(a) Baseline Simulations					
MeHg dose (μg/d)	median	2.67	2.67	2.29	2.29
	mean	4.44	4.44	3.70	3.70
	95%	13.4	13.4	11.4	11.4
Body burden (mg/kg)	median	0.0037	0.0034	0.0032	0.0028
	mean	0.0066	0.0044	0.0048	0.0033
	95%	0.0220	0.0111	0.0142	0.0070
Paresthesia prevalence (cases per million adults above background)	median	2.4	2.0	1.7	1.3
	mean	53	29	31	19
	95%	200	110	130	62
(b) Source Impact Simulations					
MeHg dose (μg/d)	median	4.05	4.05	3.64	3.64
	mean	7.70	7.70	6.77	6.77
	95%	26.0	26.0	21.5	21.5
Body burden (mg/kg)	median	0.0057	0.0052	0.0049	0.0046
	mean	0.0115	0.0085	0.0088	0.0056
	95%	0.040	0.026	0.027	0.0132
Paresthesia prevalence (cases per million adults above background*)	median	5.7	4.5	4.5	3.3
	mean	130	73	81	38
	95%	520	270	290	133

* logistic dose-response function based on a background prevalence of 22,000 per million

However, the distribution of equilibrium body burdens reached after consuming this dose also depends on the averaging process and the numbers of independent meals consumed. The mean body burdens are approximately 100 times the dose, as expected (see Section 8.1), but the distribution is narrowed by about 45% when dose averaging is used (Cases 2 and 4). The distributions of the logarithms of MeHg body burdens are compared in Figure 10.2, and this reduction in dispersion is quite evident. The GSD for body burden in Case 1 is about 2.7; in case 4, about 1.75.

It may be useful to compare these baseline estimates with various reference values. The methods discussed in Section 8 may be used to convert from equilibrium body burdens to blood and hair concentrations. Thus the baseline case would be about 4 μg/L of MeHg in blood and 1 ppm in hair. WHO lists reference values about double these estimates (8 μg/L and 2 ppm). Other baseline hair and blood values were discussed in Section 8, and we see that these predicted values are generally consistent with them.

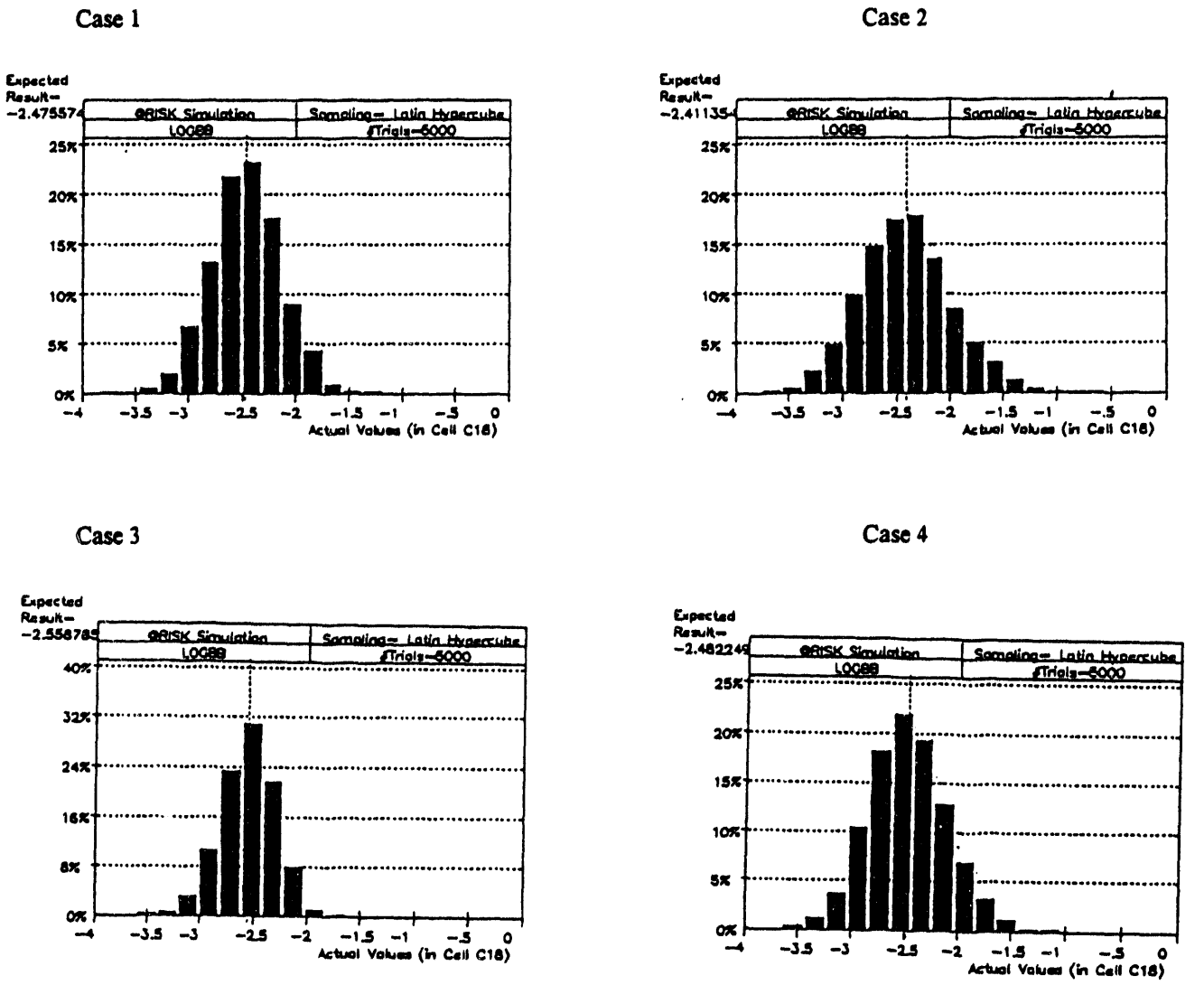

Figure 10.2. Distribution of the logarithms of the baseline body burden of MeHg.

Since the estimated baseline for this study lies among these reference values, it appears to be reasonable. Further, the GSDs agree quite well, given the fact that converting from body burden to blood and/or hair MeHg level will entail some additional dispersion due to the population variability of the conversion factors and thus we should expect that the distributions of hair or blood Hg might be wider than the distribution of body burdens (but not *vice versa*). This comparison thus lends support to the need to average the fish Hg concentrations over time.

The final entries in Table 10.1(a) are the estimated frequencies of paresthesia, which are all quite low and about 2 orders of magnitude below the estimated background prevalence of paresthesia in the general population. Note that the dispersion of the risk estimates is considerably larger than those of the body-burdens (the inputs to the DRF), because of the fundamental uncertainty of the DRF *per se* at such low doses.

10.3 Local Impacts

Section (b) of Table 10.1 presents the corresponding estimates for the power plant impact simulations. Median doses and body burdens are about 50% higher than the baseline, as expected, but the 95th percentiles are about a factor of 2 higher because of the uncertainty in the Hg concentration increment due to the power plant. The estimated increase in prevalence of paresthesia is spread even further, but remains well below background. In these simulations, only the maximum risks computed for 5000 trials begin to approximate the level of background paresthesia prevalence. As an example, the threshold body burden appears to be in the range 0.5-1 mg/kg in Figure 9.4; the maximum level reached in these four simulations of 5000 trials each was 0.47 mg/kg. This suggests that a hockey-stick dose-response function would have returned essentially zero incremental risk in all cases.

10.4 Global Effects

The ratio of transported power plant Hg emissions to total global emissions was used to estimate short-term effects on global environmental Hg and thus on MeHg in marine fish. For this hypothetical 1000 MW_e plant, this ratio is 0.17/8000 or about 2×10^{-5}. The effects of emissions of a single power plant on global mercury levels are thus seen to be quite small in the context of existing background. The incremental effect of the entire U.S. utility industry could be estimated in the same way; the incremental effect would be about 0.8%, which is also negligible. However, this analysis does not address any future risks stemming from the continuing build-up of Hg in various biospheric reservoirs, resulting from the transfer of Hg from relatively stable forms as found in ore to the more bioavailable forms of combustion products.

11.0 MAJOR UNCERTAINTIES AND RESEARCH NEEDS

This assessment has afforded an opportunity to examine the relative importance of the gaps in the data required. Table 11.1 lists the main elements of the analysis of power plant contributions to mercury in the environment and the ensuing risks to public health. Note that this listing assumes a non-linear dose-response function, and therefore that knowledge of the baseline risks is also important.

There are important interactions among these research needs. For example, if the dose-response function truly has a threshold, then the need for power plant impact assessment depends critically on the magnitude of the baseline dose, i.e., the probability that any additional health effects will be experienced due to coal burning. If the probability of exceeding the threshold is sufficiently low (which requires definition of acceptable risk levels), then research needs for the source and transport terms become moot. Worst-case assessments of such effects are necessarily local, in which a Hg source is located in the immediate vicinity of sensitive waters fished by subsistence populations (such situations must be evaluated on a case-by-case basis). Such an evaluation must include the extent to which background levels of the health end points in question are already present in the specific populations at risk.

Table 11.1 Elements of the Analysis of the Health Risks of Hg in Coal

Element	Current State of Knowledge	Research Needs
Emissions		
total Hg in coal	good	characterization by region
Hg speciation	fair	mechanistic model for Cl^-
control effectiveness	poor	effects on speciation
regional inventories	poor	need to update early estimates
Atmospheric Processes		
dispersion & transport	good	long-range, long-term models
chemical reactions	poor	validated reaction rates and models
precipitation scavenging	fair	field verification of models for plumes
long-range mass balance	poor	do surface waters act as Hg sources or sinks or both?
regional mass balance	poor	methods for dealing with "natural" flows
Terrestrial Processes		
dry deposition	poor	V_d's, mass balance
terrestrial transport	poor	how do watershed deposits affect surface waters?
Aquatic Processes		
bioaccumulation	poor	why do fish species differ? do sediments or water concentrations control? roles of pH and DOC by species factors controlling marine bioaccumulation
sedimentation rates	poor	controlling parameters
mass balance	poor	effect of Hg outgassing
Baseline Human Dose		
Hg in fish	poor	is there a time trend? levels in high-consumption species
consumption rates	fair	% of non-fish eaters no. of subsistence fishers locations, diets of subsistence populations regional data
Metabolic Processes		
model for equil. Hg	fair	is body mass independent of half life?
Dose-Response Functions		
form of model	poor	symptom data at low doses acute vs. chronic responses background prevalence rates
Assessment Data & Criteria		
source-receptor data	poor	data on sensitive waters & populations near Hg sources
acceptable risk levels	poor	depends on end point and background prevalence
relative global effects	fair	global Hg emissions and environmental levels

Unfortunately, there are major uncertainties with regard to the baseline mercury dose. There are two estimates of mercury levels in high-consumption fish species, i.e., pollock and shrimp, one from an extensive data base from the late 1970's and one comprising a few more recent samples. These two baselines differ by factors of 4.5, in opposite directions for the two species. Clearly, if environmental mercury is indeed judged to be a public health concern, the massive sampling effort performed by NOAA in the 1970s must be replicated, with emphasis on high-consumption species.

There are similar problems with outdated and sparse data on fish consumption. A large survey was conducted in the early 1970s and indicated that 95% of the sample ate some seafood. Two other surveys suggest that only about half or less of the population does so. Since there are also data on the total catch and its implied consumption rate, it is very important to know what fraction of the population is actually eating this seafood. Data on consumption of freshwater game fish (the most likely to be affected by mercury in coal) is also sparse. This analysis found major uncertainties with respect to the parameters used to estimate local deposition levels near a power plant. In addition, there are also substantial uncertainties as to the baseline deposition levels, which makes the incremental effect even less certain. Settling these questions will probably require field research, which should only be undertaken if the risk analysis confirms a need for the information.

The severity of global impacts from U.S. coal burning in general must be assessed in terms of the mercury emissions that contribute to mercury in marine fish, summed over the entire U.S. industry. As a first cut, one might ignore local deposition, realizing that this would provide a gross overestimate of the amount of U.S. mercury entering the oceans. Thus knowledge of the ultimate fate of mercury deposited within the United States, i.e., a mass balance, is required. In addition, the severity of global impacts must also be judged in terms of the incremental risks relative to existing background levels.

It is clear that substantial research is required before informed judgments can be made about the need to regulate mercury emissions from U.S. electric utilities. A modest investment in research and data could have a large return in terms of more efficient regulation.

12.0 RECOMMENDATIONS

It is clear that the risks of adult paresthesia from fish consumption are low. This analytical framework should be extended to the case of maternal fish consumption and fetal effects on retarded child development, which are thought to be considerably more sensitive. It is also clear that the basic data used in this analysis are in need of improvement:

1. Mercury levels in fish are based on conflicting and outdated data.

2. Fish consumption patterns of sensitive subpopulations (such as pregnant women) have not been established.

3. Appropriate methods for estimating Hg deposition from power plants have not been validated in the field.

4. The intersections of the sets of locations of U.S. coal-fired power plants, sensitive water bodies, and susceptible subpopulations have not been established.

5. Annual total Hg emissions from coal burning and from other sources, including natural sources, are still uncertain, which makes it difficult to estimate the combined effects of U.S. coal burning on global Hg concentration levels.

6. Since the uncertainty in the dose-response data contributed over half of the variability in the estimates of paresthesia frequency (on a log basis), improved data on health effects should result in substantially more precise assessments.

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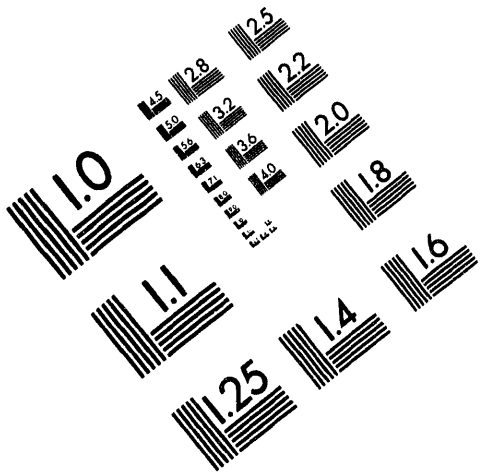

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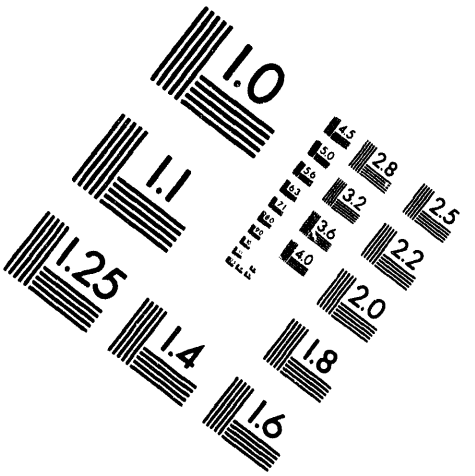

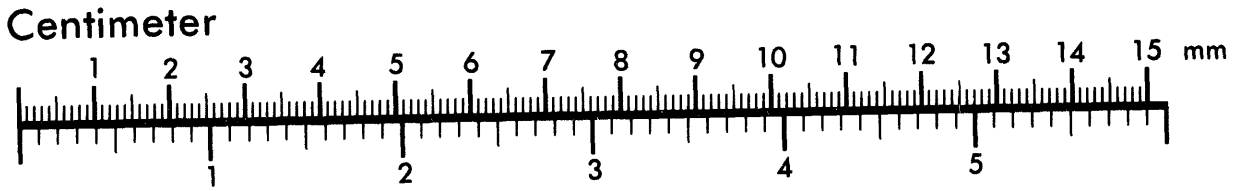

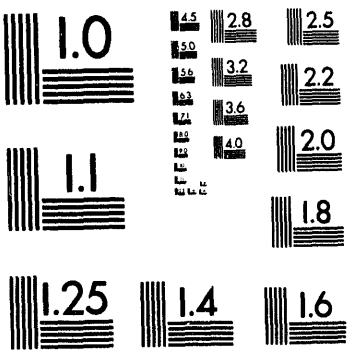

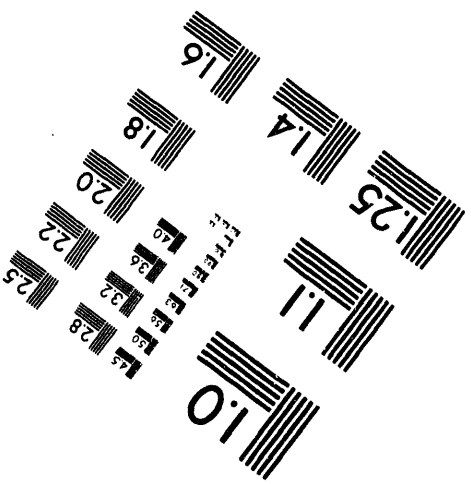

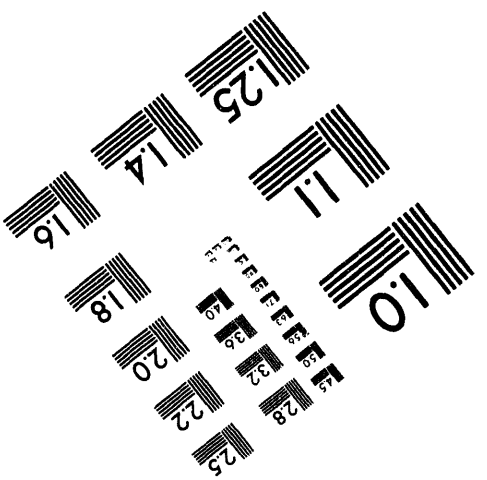

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Appendix A - Properties of Log Normal Distributions

Many of the parameters used in this assessment were perceived to have approximately log normal distributions. This means that the logarithms of the values tend to be normally distributed around the median, rather the values per se being symmetric with respect to the mean. Such distributions are symmetric in log coordinates but skewed to the right in terms of the actual values. Such a situation can arise when the values are constrained to be positive and there is no upper constraining limit.

In many cases, it was necessary to work with fragmentary data in order to deduce the properties of the distributions needed. The most common statistic reported from sampling is the mean; sometimes the range (max, min) are also reported, but the medians or other percentiles are rarely given and are needed to perform a proper simulation. It was attempted to solve this problem by constructing parametric curves describing useful properties of arbitrary log normal distributions, using the simulations of @RISK. The geometric standard deviation (GSD) and the number of points sampled were the parameters used. Most of the sampling runs used latin hypercube sampling, but no systematic differences were apparent when probabilistic sampling was used as an alternative.

Figure A.1 presents the general properties of these distributions, for sample sizes of 100 and 500. The ration of mean to median increases only modestly for GSDs less than about 2, but sharply thereafter. The ration of the 95th percentile to the 5th percentile was quite stable and conformed to theoretical expectations (note that t=1.645 for the 95th percentile and -1.645 for the 5th percentile. The exponent 0.04 is 1/2t for t=1.645. The ration of maximum to minimum values is less well behaved (right-hand scale), and increases with sample size.

Figure A.2 explores the ratios of mean to median, which is seen to be independent of sample size, although the data become somewhat erratic for GSD > 4. The ratios of maximum to mean values are plotted in Figure A.3, and it is seen that GSD plays a larger role than sample size. One might expect considerable sampling variance for sample sizes less than 100, however. For the n=20 case (1 gsd value only), 10 trials were averaged in order to give some stability to the result. Maximum-to-minimum ratios are plotted in Figure A.4, and this range increases by about two orders of magnitude for each doubling of GSD. For GSD's in the range 2-3, 2-3 orders of magnitude variation would be expected in the parameter being sampled, with a strong increase due to sample size as well.

Figure A.5 plots the distribution of the sums of 4 log normal distributions, with GSDs of 1.6, 2, 2.5, and 3, each with a median of 10. The mean of this sum is about 57, the median about 49, and that this distribution is neither normal nor log normal. The 5th percentile is about 50% of the median, while the 95th percentile is about 2.4 times the median. This results from the tendency towards the mean produced by adding 4 independent distributions together with the skewness towards higher values produced by the basic log-normality. Such a distribution could describe the combined distribution of dietary mercury produced by combining four different types of fish, for example, shellfish, canned fish, freshwater fin fish, and marine fin fish. The central limit theorem also reduces the maximum combined value observed relative to the sum of maximum individual constituent values.

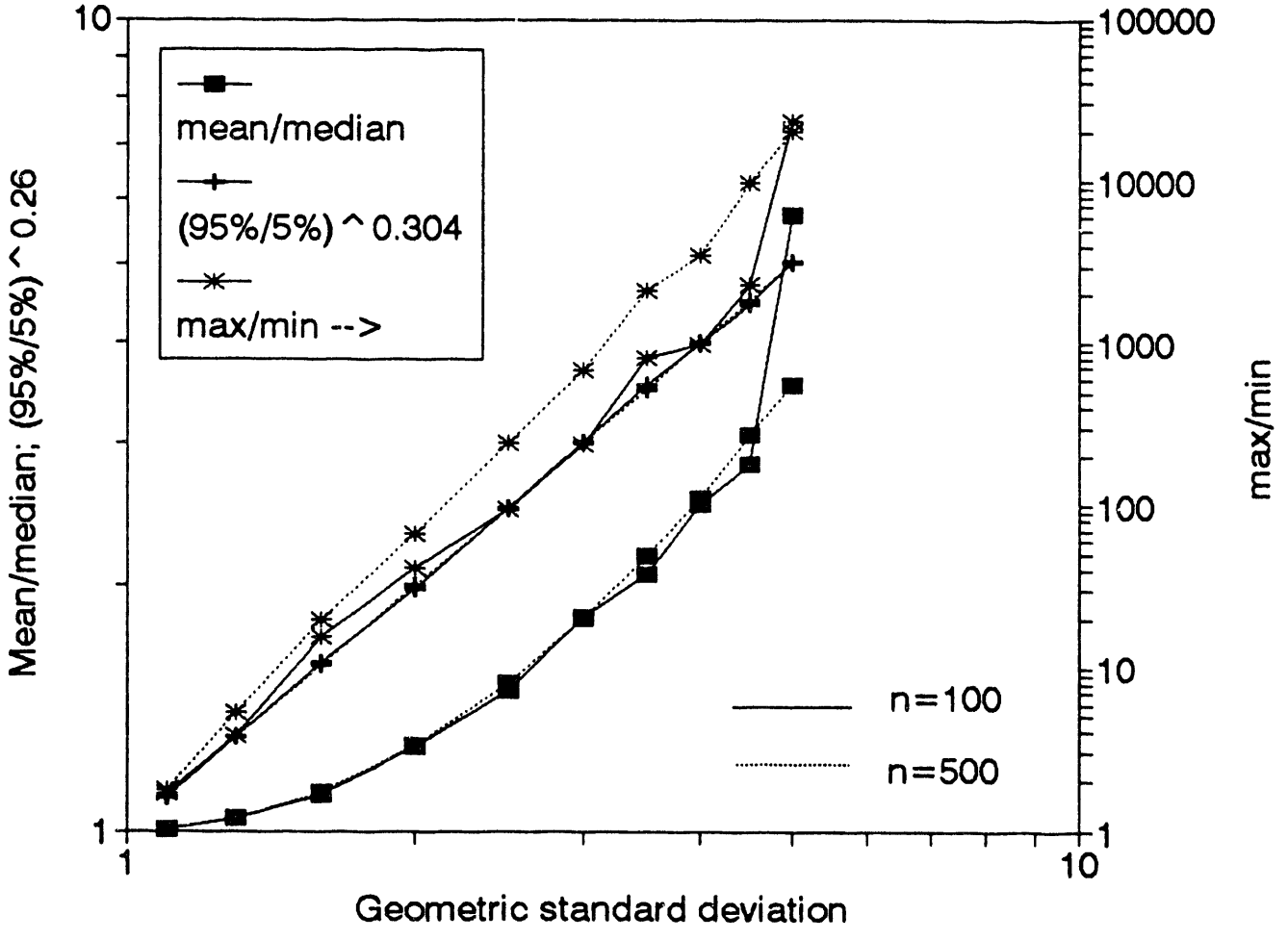


Figure A.1. General properties of distributions, for sample sizes of 100 and 500.

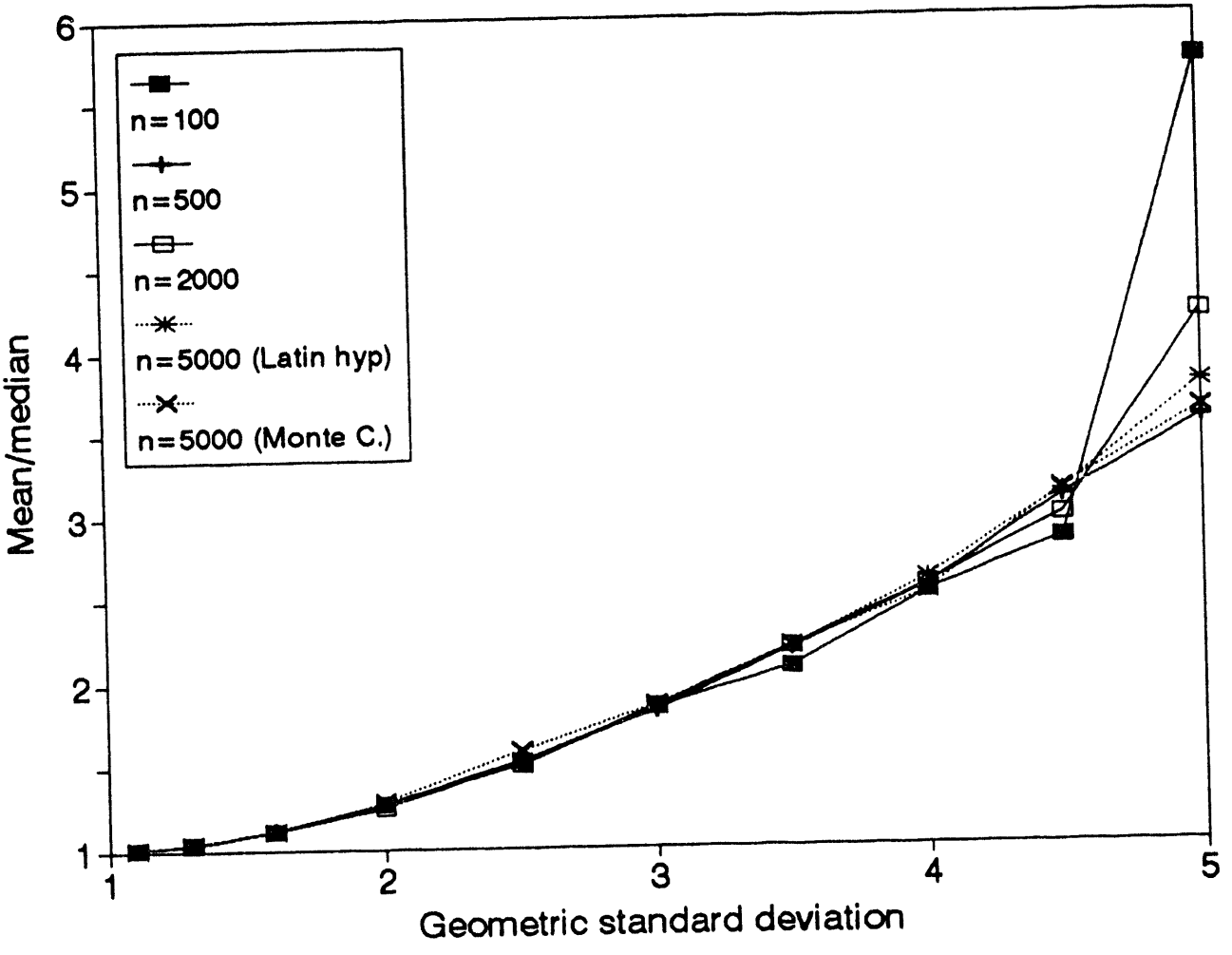


Figure A.2. Ratios of mean to meadian.

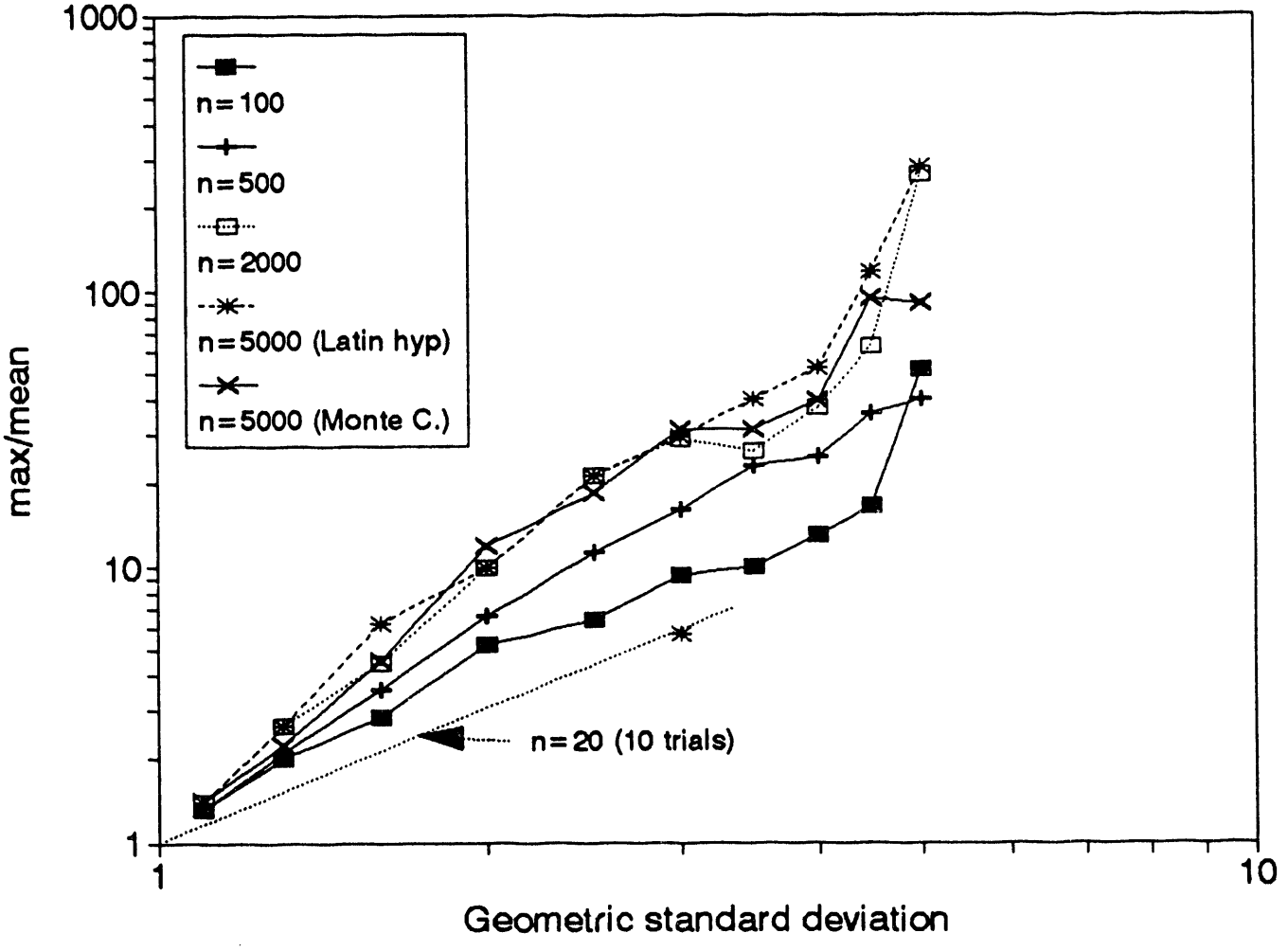


Figure A.3. Ratios of maximum to mean value.

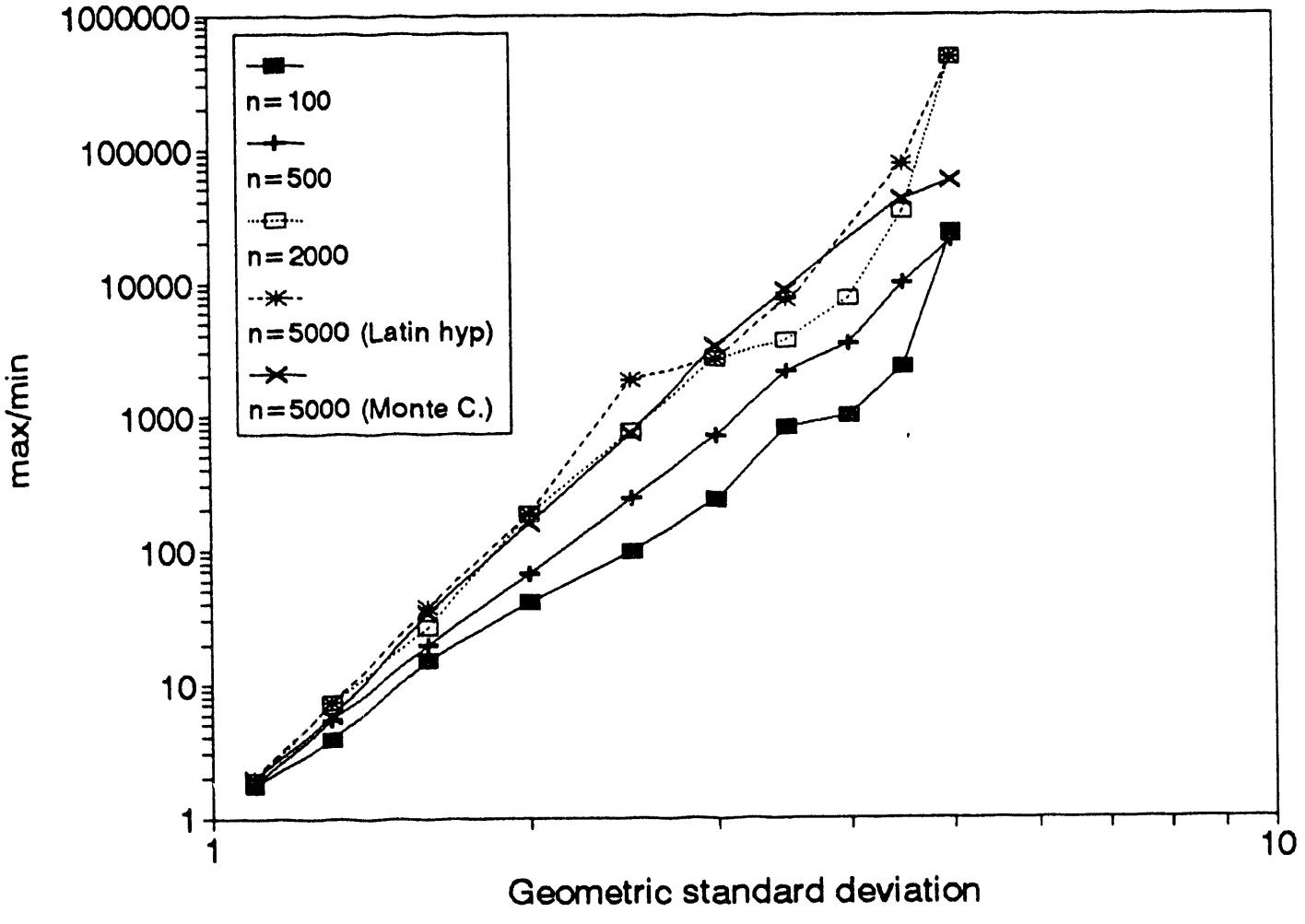


Figure A.4. Ratios of maximum to minimum values for various log-normal distributions.

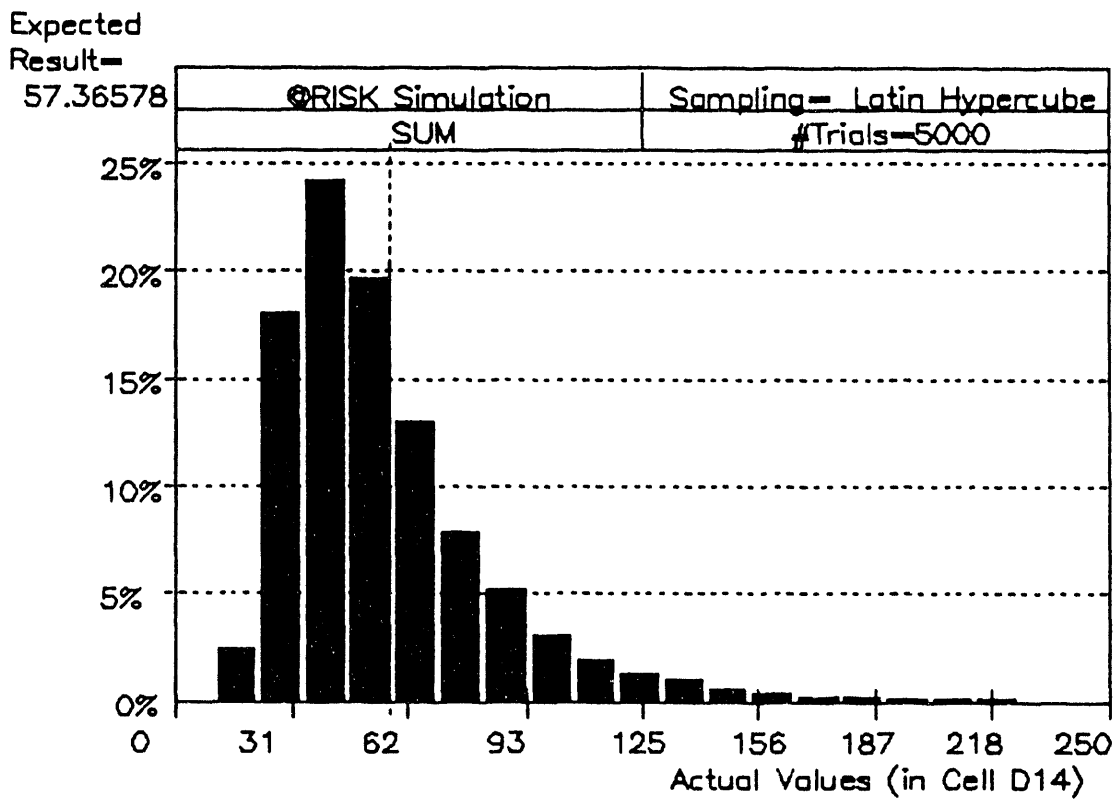


Figure A.5. Distribution of sums of four individual log-normal distributions.

Appendix B - Data on Mercury in Fish from Upper Michigan Lakes

Gloss et al. (1990) and Cusimano et al. (1989 report data from 49 lakes in the Upper Peninsula of Michigan which were sampled as part of the National Surface Water Survey. These lakes were thought to be remote from local sources of mercury pollution. A variety of species were caught, and detailed data were presented for 864 fish taken from 37 lakes. These data were made available to BNL for this analysis. The species included for mercury analysis are given in Table B.1, and are dominated by yellow perch in terms of numbers of fish examined. The data on each fish include age, length, weight, and mercury content. The mercury data reported are total mercury from muscle specimens; the MeHg content was reported to be about 99%.

Table B.1 Average Fish Characteristics

Species	Number	Age(y)	Length(mm)	Weight(g)	Hg(μg/g)	Hg Dose(μg)
brook trout (BT) *Salvelinus fontinalis*	28	2.2	283	382	0.15	91
large mouth bass (LMB) *Micropterus dolomieui*	72	3.4	235	258	0.35	125
small mouth bass (SMB) *Micropterus salmoides*	4	4.0	289	353	0.30	100
northern pike (NP) *Esox lucius*	86	4.2	526	1000	0.44	593
walleye (WE) *Stizostedion vitreum*	8	4.2	388	578	0.28	194
white sucker (WS) *Castostous commersoni*	110	4.7	323	426	0.12	71
yellow perch (YP) *Perca flavescens*	540	4.2	158	59	0.30	31

The lake data available in Cusimano et al. (1989) included pH, DOC, certain chemical parameters, lake and watershed areas, sampling depth, and hydrological type. Data on the fish caught included counts of species by method of capture (angling, gill nets, trap nets and beach seines; the last three categories were combined to distinguish them from normal modes of sport fishing) and the time spent in each method. The catch of all species combined was divided by the time spent to provide an index of productivity; an additional index was devised by dividing by the lake surface area. A few lakes were sampled twice as a check on repeatability; these catches were added and divided by the total time. Average properties of the lakes are given in Table B.2. Deducing the importance of lake properties on mercury levels can be important for a national assessment of the effects of coal burning because lake properties (especially acidity) vary regionally and also affect fish abundance.

Table B.2. Physical Characteristics of the Lakes sampled by Gloss et al. (1990)

parameter	mean	median	std. dev.	range
surface area (ha)	18	9	37	4-262
sampling site depth (m)	6.5	4.3	5.3	1.5-20.1
Secchi depth (m)	2.6	2.3	1.3	0.85-7.6
elevation (m)	332	282	100	220-546
watershed area (ha)	1376	60	7794	10-54500
watershed/lake ratio (logs)	8.6	7.0	(GSD) 3.16	2.3-1687
pH	5.96	5.8	1.4	4.43-8.5
DOC (mg/L)	5.36	4.55	3.2	0.26-17.7

This analysis was intended to explore the dependence of Hg levels on fish characteristics vis-a-vis lake characteristics. This analysis was conducted at two levels. First, data on the individual fish caught were used, in order to test for the dependence of mercury levels on various physical and chemical parameters. Then averages for each lake and major species (large mouth bass, northern pike, white sucker, and yellow perch) were used to examine the influence of lake characteristics in more detail; data on mercury levels in the lake waters were not available. Finally, the fish catch statistics were used to examine the probabilities of sport fishermen catching high-mercury content fish by angling.

Analysis of Individual Fish. The average mercury concentration of the 7 species in Table B.1 is 0.27 μg/g, which is considerable higher than the grand average of fresh-water species in the EPA database (0.19 μg/g). However, the distributions of mercury were seen to be approximately log normal, so that the medians may be a better measure. These values were: BT, 0.10; LMB, 0.27; SMB, 0.28; NP, 0.36; WE, 0.26;, WS, 0.07; and YP, 0.18 μg/g, for a 7-species mean of 0.22 μg/g, which compares with 0.14 μg/g in the EPA data base. Figure B.1 presents these average data as a function of weight (for plotting convenience). Although all species fall on a common length-weight characteristic (filled symbols), average mercury concentrations do not relate to average weight; brook trout and white suckers have considerably lower Hg for their weights, on average. The last column in Table B.1 gives average mercury dose received per fish (assuming consumption of the whole fish, which corresponds to an overestimate of about a factor of 2), does not necessarily correspond to the product of the average weight and the average Hg concentration, since the two factors are positively correlated for each fish species. Figure B.1 shows that only northern pike provide above average mercury doses.

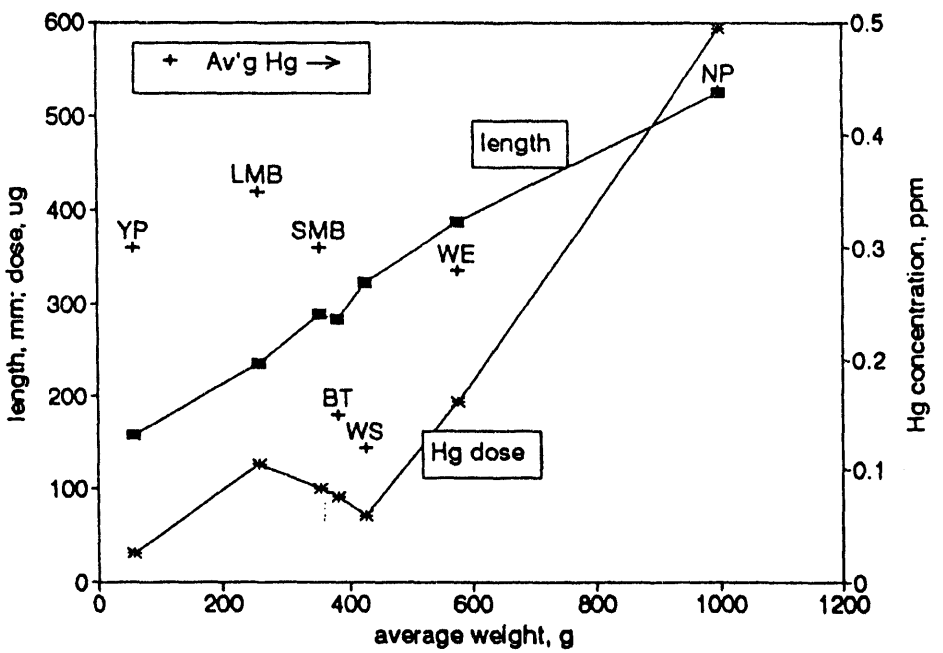


Figure B.1. Average fish length (connected squares) and mercury dose (individual crosses) vs. fish weight, by major species, for 49 Upper Michigan lakes. Data from Gloss et al. (1990). Plotting symbols are: BT = brook trout, LMB = large mouth bass, NP = northern pike, SMB = small mouth bass, WE = walleye, WS = white sucker, YP = yellow perch.

Individual fish weight was seen to be proportional to (length)^3 with only a few outliers; this relationship provided a data quality check. Mercury concentrations were considerably more scattered, however, suggesting the influence of environmental factors. Multiple regression analysis was used to try to separate the effects of various factors. Since yellow perch were so numerous in the dataset and are not considered a desirable game fish, they were set aside as a first step in the analysis. Also, Gloss et al. (1990) noted the importance of lake hydrology in their original analysis of the data (which was centered on yellow perch); the lake population was therefore divided into two groups: drainage lakes and all others. This gave four subsets for analysis. The independent variables were fish weight (wt) as an overall index of growth and lake pH.

The following relationships were found for individual fish (all terms statistically significant):

For drainage lakes:

yellow perch:	Hg = 0.916 + 0.0035 wt - 0.12 pH	(R^2 = 0.72, n=206)
all other species:	Hg = 0.221 + 0.00025 wt - 0.011 pH	(R^2 = 0.36, n=200)

For all other lakes:

yellow perch:	Hg = 0.703 + 0.0009 wt - 0.092 pH	(R^2 = 0.18, n=345)
all other species:	Hg = 0.613 + 0.00016 wt - 0.070 pH	(R^2 = 0.23, n=113)

Figure B.2 displays these relationships; the interactions among species and lake type and pH are apparent. The pH relationship found by Gloss et al. (1990) is given in Figure B.3; their mercury-pH coefficient is seen to be 0.06, which is reasonably consistent with the regression equations presented above. Also, in Figure B.3, excepting one outlier, the pH relationship for drainage lakes is seen to be less important than for seepage lakes. It is also apparent from Figure B.3 that dissolved organic carbon (DOC) is an important parameter. Gloss et al. did not specify the meaning of the regression line on Figure B.3; it appears to pertain to seepage lakes with DOC levels.

In addition, the legal size limits (vertical dotted lines on Figure B.2 impose a bias on the actual average mercury levels in fish likely to be consumed. The average Hg concentrations in fish above legal length limits (the limit for yellow perch was based on "likely consumption levels" [Gloss et al., 1990]) were:

brook trout:	0.20 μg/g
walleye	0.35 μg/g
large mouth bass	0.54 μg/g
small mouth bass	0.19 μg/g (based on a sample of 1)
northern pike	0.56 μg/g
yellow perch	0.39 μg/g

These values imply that consumption of fish containing mercury above the advisory limits is highly likely; estimated consumption rates of freshwater fish are discussed below.

Analysis by Lake Averages. The analysis of average Hg levels by major species and lake is presented in Figures B.4 to B.8. The stratification by hydrological type was dropped at this point, although Figure B.4 shows that the variables interact: most of the seepage lakes have relatively low watershed/lake area ratios (WLARs) and lower values of DOC; there are more high pH drainage lakes than seepage lakes. Aside from one lake with pH < 6 and DOC > 17, DOC tends to be positively correlated with pH and with WLAR.

The characteristics of the four major sportfishing species are seen to differ considerably. The plotting parameters used here were:

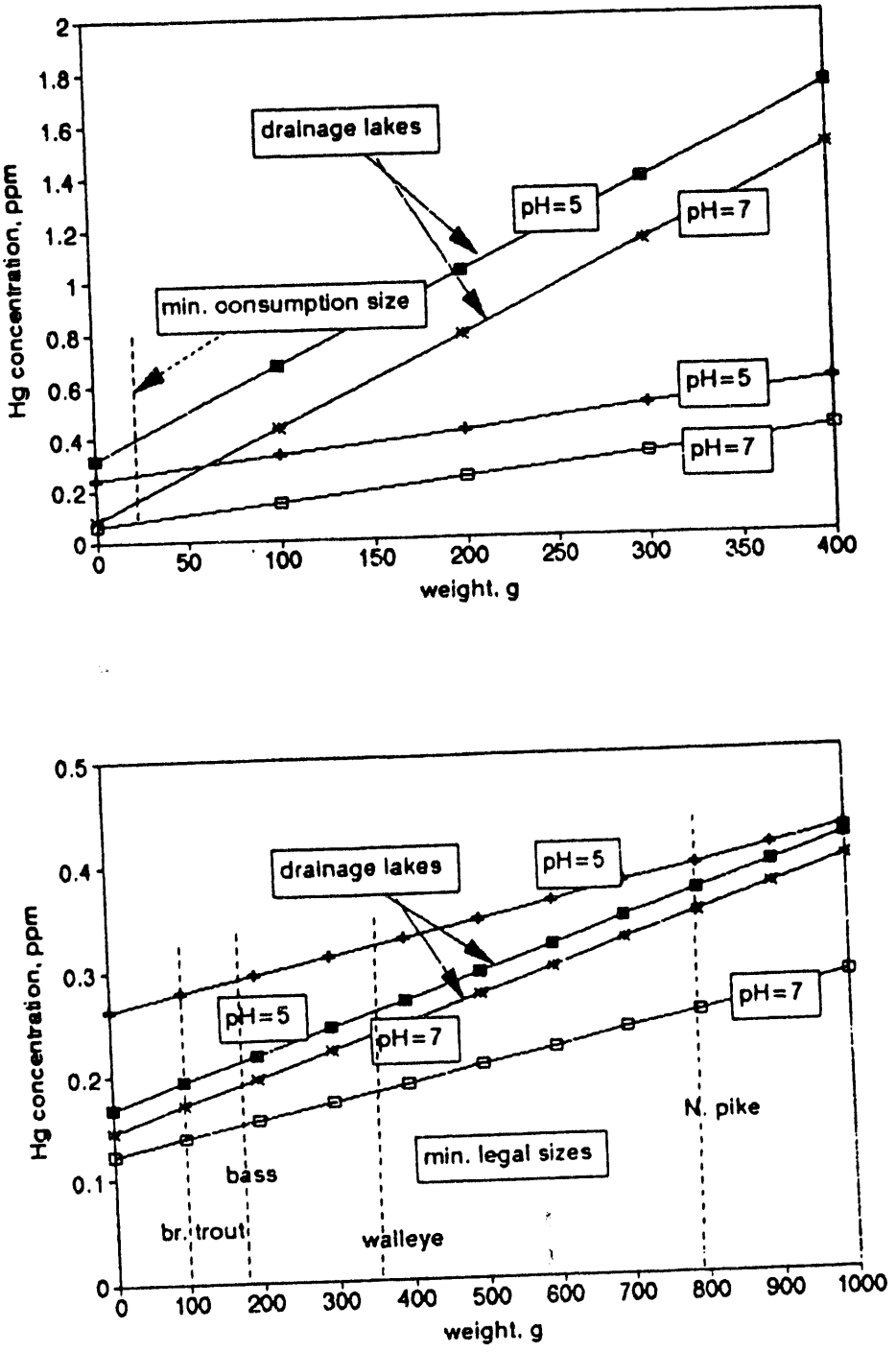


Figure B.2. Results of regression analysis of individual fish mercury content vs. fish weight and lake pH, for 49 Upper Michigan lakes, by lake type. Data from Gloss et al. (1990).
(a) yellow perch. (b) all other species.

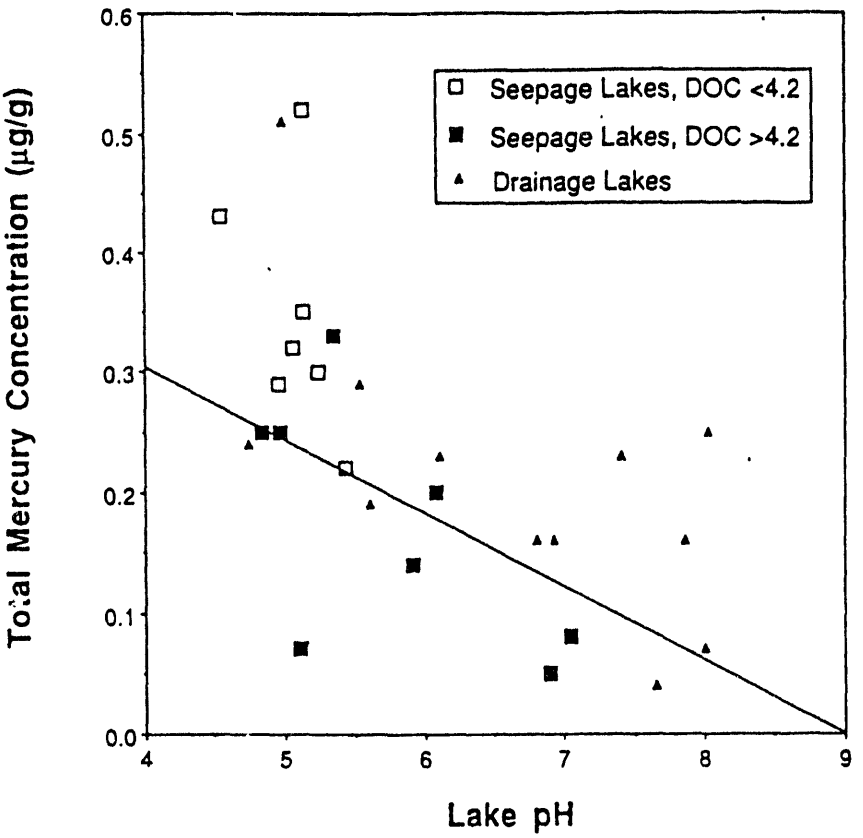


Figure B.3. Relationship between lake pH and total Hg concentration for yellow perch ages 2-4 in 27 lakes. DOC value of 4.2 mg/L is mean value for all seepage lakes. Source: Gloss et al. (1990).

1. Lake pH (average of 2 determinations in different seasons): thought to influence the metabolic uptake of mercury by fish.

2. DOC (average of 2 determinations in different seasons): thought to affect the residence time of mercury in lake water.

3. Ratio of watershed to lake surface areas (WLAR) (a logarithmic distribution was assumed, because of the very large value for the reservoir): a larger catchment area should concentrate more atmospheric mercury into the water.

4. The dependent variable used to characterize average mercury levels in this portion of the analysis was the average mercury concentration divided by the average weight, for each species and lake. There were three high outliers that were not considered, apparently resulting from inordinately low weights (from 10 to 23 g for large mouth bass, for example). These values may have resulted from miscoding the species in the original dataset.

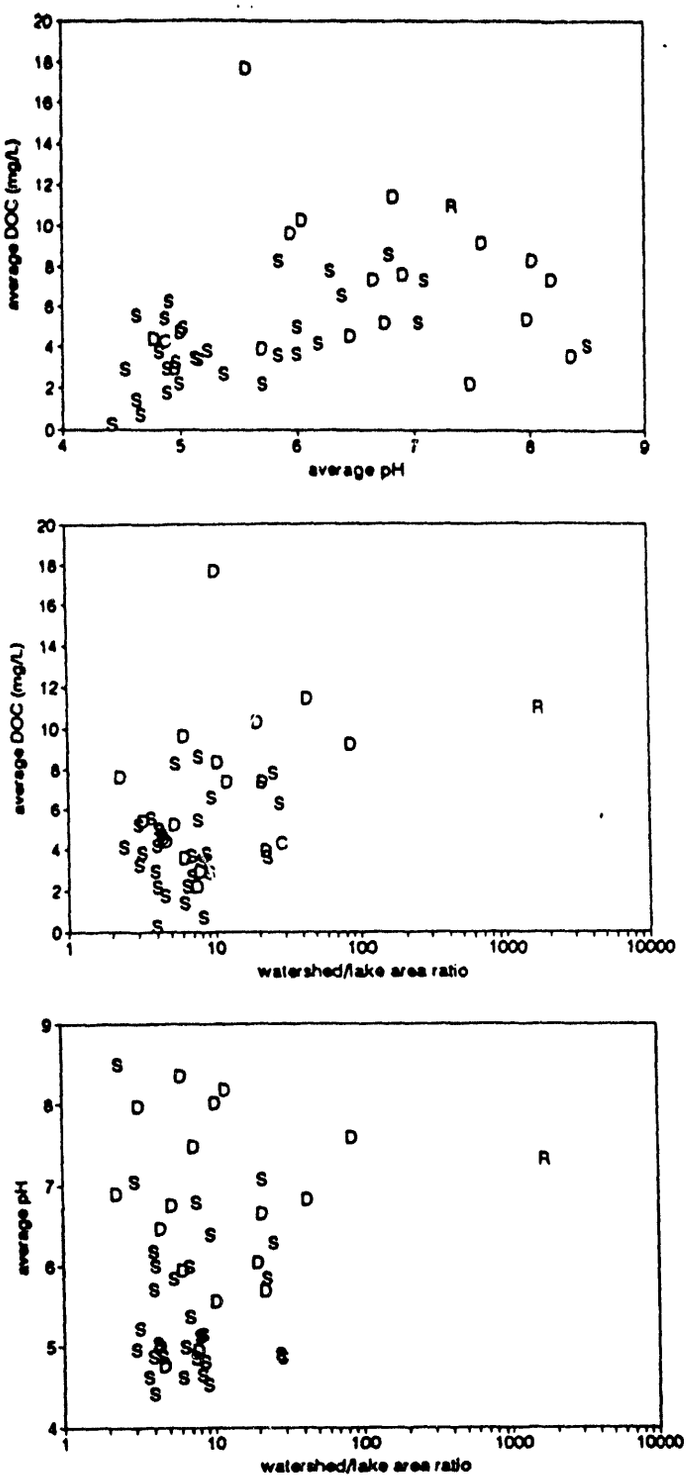


Figure B.4. Properties of 49 Upper Michigan lakes. Data from Gloss et al. (1990).
(a) DOC vs. pH. (b) DOC vs. watershed/lake area ratio. (c) pH vs. watershed/lake area ratio.

The data for northern pike (Figure B.5; n=8 [relatively few were caught]) suggest a positive influence of DOC, and indeterminate effect of WLAR, and no effect of pH. However, it should be noted that no northern pike were caught in low-pH lakes. The best multiple regression of northern pike mercury levels explained 44% of the variance, using DOC and WLAR (which had a negative effect), but neither parameter was statistically significant ($p > 0.05$). For large mouth bass (Figure B.6, n=9 after dropping one outlier), the important parameters were pH (negative) and WLAR (positive but not significant). 60% of the mercury variance was explained. For yellow perch (Figure B-7, n=31 after dropping one outlier), the important parameters were pH (negative) and WLAR (negative but not quite significant). 34% of the variance was explained. For white suckers (Figure B.8, n=12 after dropping one outlier), the important parameters were DOC (positive) and WLAR (negative but not significant).

The preceding analysis of mercury in Upper Michigan fish is limited by the relatively few observations, but suggests the following conclusions:

1. The effects of lake characteristics on fish mercury levels vary by species; maximum levels for different species (after accounting for weight) will not occur in the same lake, in general. This in turn suggests that assessments should be specific to defined fish species, and that generic bioaccumulation factors should not be used.

2. The effects of DOC and pH have the expected signs, when statistically significant, which reinforces the conventional wisdom about the relevant mechanisms for bioaccumulation of Hg.

3. The relative size of the catchment area for atmospheric mercury was never a statistically significant parameter, in spite of its large range, and was negative in 3 of the 4 cases. This finding suggests the influence of other factors with regard to terrestrial transport of deposited Hg, which could include the nature of the watershed terrain and ground cover and lake volume. It would appear from these data to be problematic to assume that Hg deposited in the watershed will end up in fish.

Analysis of Fish Capture Rates. Another element to be considered in estimating mercury doses from recreational fishing is fish capture rates. For example, if acidity in lakes increases the bioavailability of mercury but also limits production rates, the increase in the mercury dose actually experienced by recreational fishermen may be limited.

Data on the fish caught included counts of species by method of capture (angling, gill nets, trap nets and beach seines; the last three categories were combined to distinguish them from normal modes of sport fishing) and the time spent in each method. The total catch (all species combined) was divided by the time spent to provide an index of productivity; an additional index was devised by dividing by the lake surface area. A few lakes were sampled twice as a check on repeatability; these catches were summed and divided by the total time.

Figure B.9 presents data on fish catch rates, based on numbers of fish caught. For angling, Deep Lake stands out with a high catch rate; large mouth bass and bluegills were caught here; mercury levels for the bass in this lake were near average. This datum helps support a trend towards higher catches with increasing DOC and with near-neutral pH. No fish were caught by rod in 31 of the 49 lakes; 11 of these were drainage lakes and 20 were seepage lakes. These data suggest that there is about a 60% chance of coming up empty after 2 hours of fishing in a randomly selected lake in this area.

For net fishing (all modes combined), the most numerous species were shiners, brown bullheads, and yellow perch (Elevenmile Lake), none of which are desirable game fish. Figure B.9 also suggests the best harvests were obtained at pH values between 6 and 7.

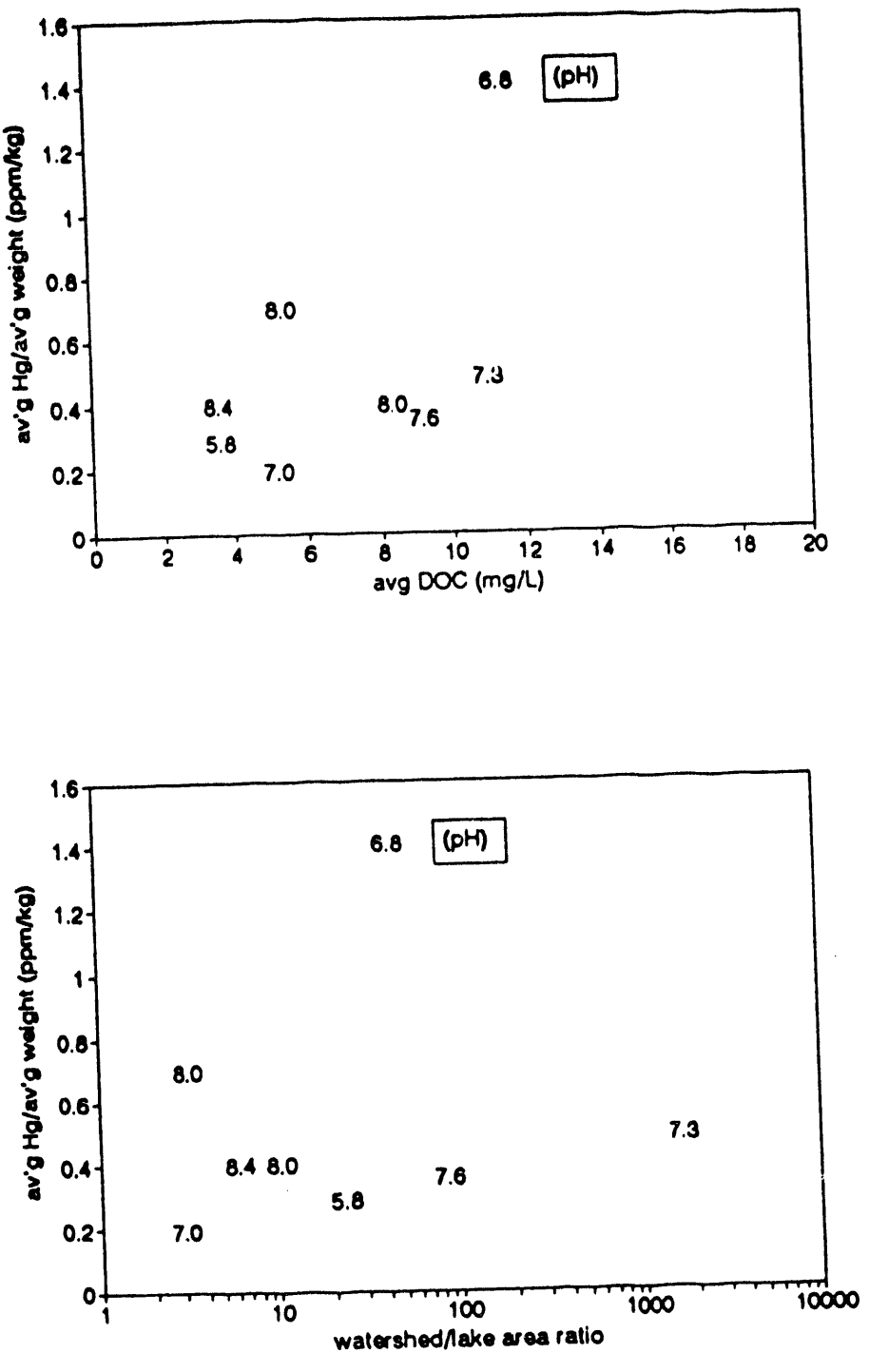
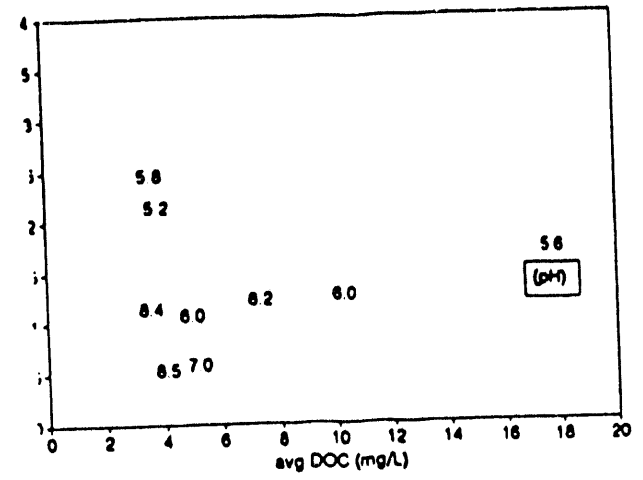
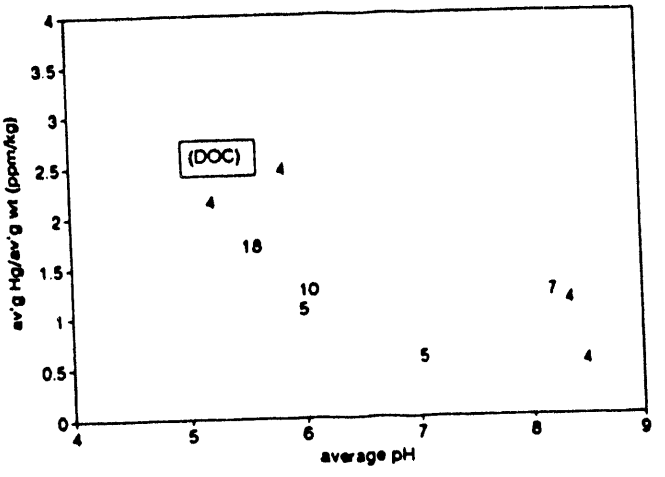


Figure B.5. Average mercury concentration per unit fish weight for northern pike in 49 Upper Michigan lakes. Lake pH values are shown for reference. Data from Gloss et al. (1990).
(a) as a function of DOC. (b) as a function of watershed/lake area ratio.





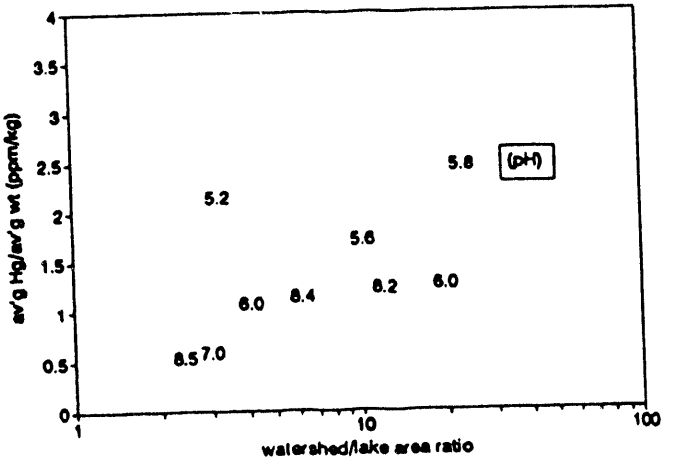
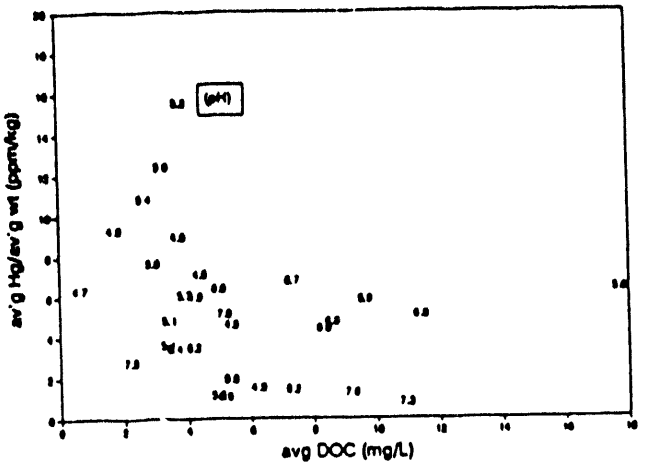
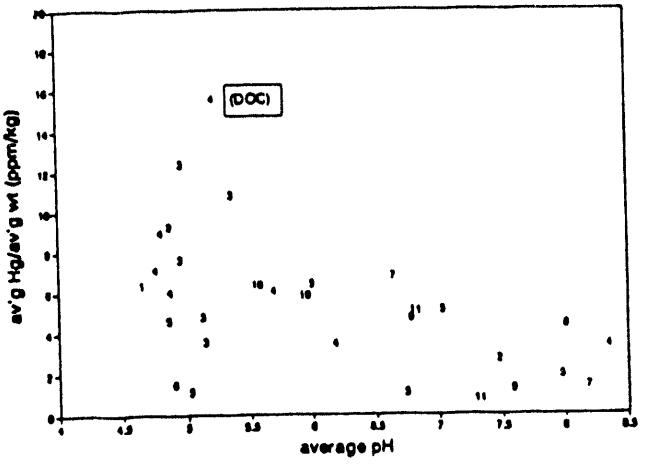


Figure B.6. Average mercury concentration per unit fish weight for large mouth bass in 49 Upper Michigan lakes. Data from Gloss et al. (1990). (a) as a function of DOC, lake pH values shown for reference. (b) as a function of PH, DOC values shown for reference. (c) as a function of watershed/lake area ratio, lake pH values shown for reference.





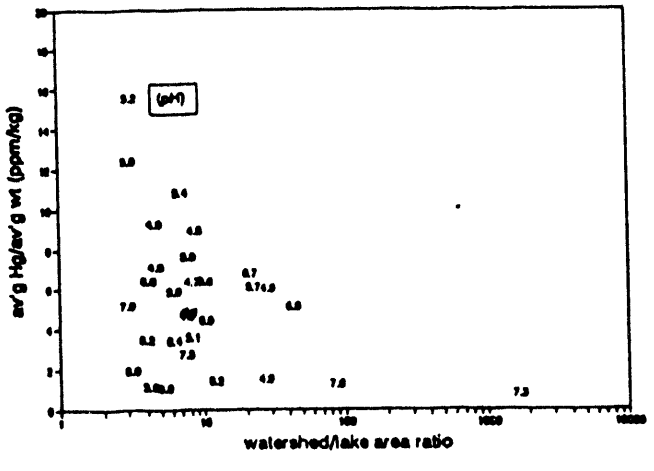
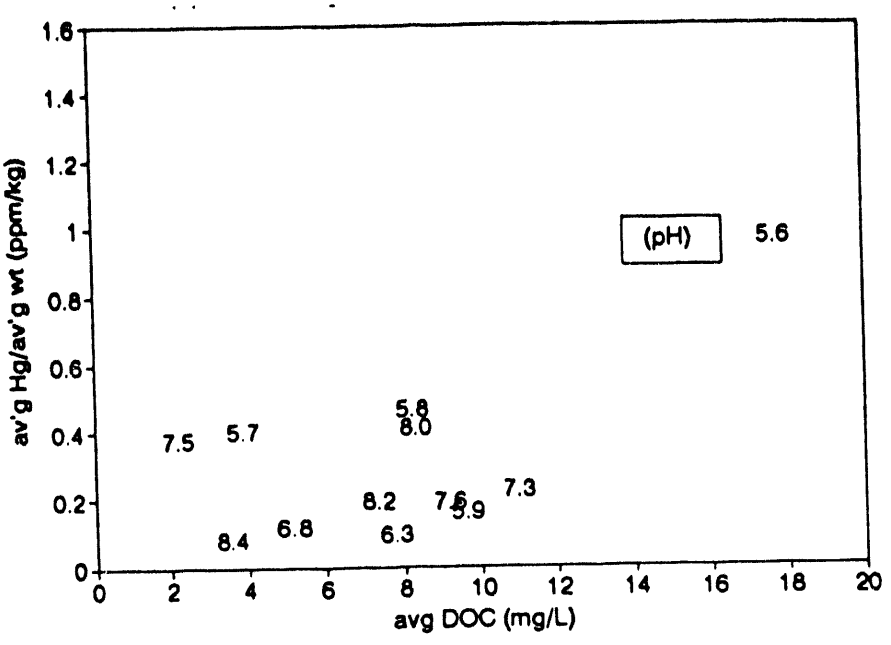


Figure B.7. As in Figure B.6, for yellow perch.



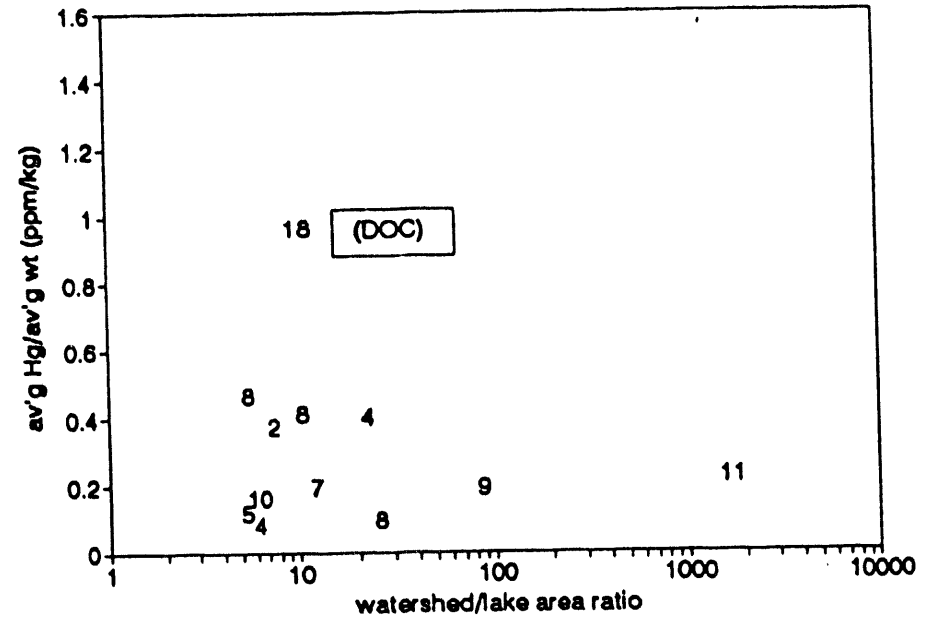


Figure B.8. As in Figure B.6, for white suckers.

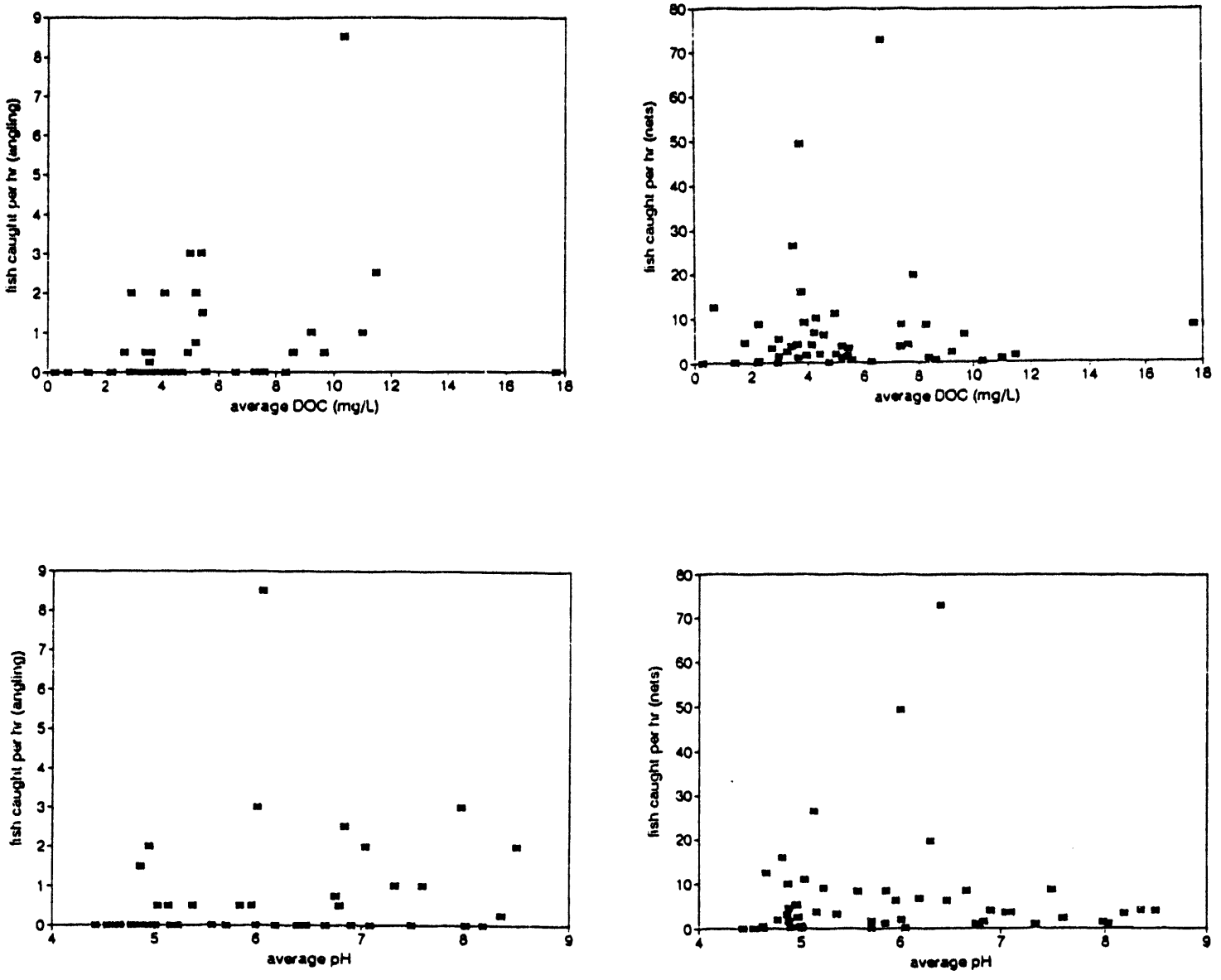


Figure B.9. Fish catch rates in 49 Upper Michigan lakes. Data from Gloss et al. (1990).
(a) fish caught angling vs. DOC. (b) fish caught angling vs. pH. (c) fish caught with nets vs. DOC. (d) fish caught with nets vs. pH.

Application to Subsistence Fishermen. Subsistence fishing populations are likely to use all available means to harvest fish from lakes, and probably to select the most productive lakes for their efforts. Sport fishermen are generally restricted to angling. Approximately the same fishing effort was expended in each of the 37 lakes with Hg data studied by Gloss et al. (1990), about 72 hours of net fishing and 2 hours of angling. The total mass of fish harvested in each lake varied greatly, from less than 1 kg to about 30 kg (based on the total weight of the species analyzed for mercury, of which only about half may be edible). However, a catch of 10/kg per day would not feed many people on a subsistence basis, so that the application of these data to a true subsistence situation seems problematic, especially since fish harvests are seasonal with higher catches in summer. The median catch per lake was 2.5 kg, or less than 1 kg/day. Richardson and Currie (1993) report a citation by Burkes (1990) of a mean fishery harvest of 115 g/d/capita (edible weight) among Canadian subsistence populations, but points out that not all of this fish is consumed by humans, locally.

To assess the use of freshwater fish as subsistence protein, the capture rates of the Upper Michigan lakes were combined with the geographic distributions of northern Minnesota lakes given by Sorensen et al. (1990). Using Cook County, MN, as an example, there are 12 lakes (there may be additional lakes not sampled by Sorensen et al.) and an estimated total surface area of 39 km^2 (1% of the land area). Scaling up the median fish harvesting rate from Cusimano et al. (1989) and estimating that half of the catch is edible yields an edible production rate of about 180 kg/day; for comparison, the median apparent per capita fish consumption rate for 98 Alaska subsistence communities was about 0.22 kg/day. The population of Cook County was 4092 in 1980, with 273 categorized as "American Indians." If the entire fish catch were assigned to the American Indian portion of the population, and if it were assumed that they lived on a reservation and were free to use any fishing method, this catch would provide about 3 meals per day (660 g/d). However, if the catch were restricted to angling and applied to the entire population of Cook County, the yield would be much smaller and probably less than the average national or regional fish consumption rate for the general population. Of course, additional fish could be obtained from Lake Superior, in this case, and there are probably other lakes in Cook County not included in this analysis. The conclusion thus follows that the rates of fish harvesting found in upper Michigan are probably adequate for subsistence by small groups of native American populations.

Assessment of the Overall Distribution of Hg in Upper Michigan Lakes. Figure B.10 plots the frequency distributions of Hg, for individual species, and Figure B.11 plots Hg frequency distributions among lakes, averaged over all fish species in each lake. Not only are the distributions of Hg within each lake positively skewed, suggesting a log-normal distribution, the distributions of the lake averages and medians are similarly skewed. The average of these medians is 0.21 μg/g, with a GSD of 2.8, and the median lake had a median Hg level of 0.17 μg/g. Weighting by the number of fish caught (species analyzed for Hg), the median Hg level was 0.28 μg/g (15 fish caught). There was no relationship between median mercury level and total mass of fish harvested (Figure B.12). However, the most productive lakes had mercury levels slightly below the median for all lakes in this data set.

The distribution of median mercury levels in individual lakes is somewhat more variable than the distributions within lakes. Thus, assuming that a subsistence fishing population may be utilizing any randomly selected lake, a GSD of about 3.0 seems appropriate, with a median Hg concentration of about 0.17 μg/g. This implies 5% and 95% lake medians of about 0.03 and 1.05 μg/g, respectively. However, if more than one lake is utilized, as would seem likely for lakes of this size, the variability in mercury levels averaged over the population's total fish harvest would likely decrease. The larger the population at risk, the closer their mercury intakes will approach the average for the region. This may explain why epidemiological surveys of indigenous populations find so few cases of adverse health effects: sampling variability dominates studies of small populations, and the tendency toward more nearly average consumption levels affects large populations.

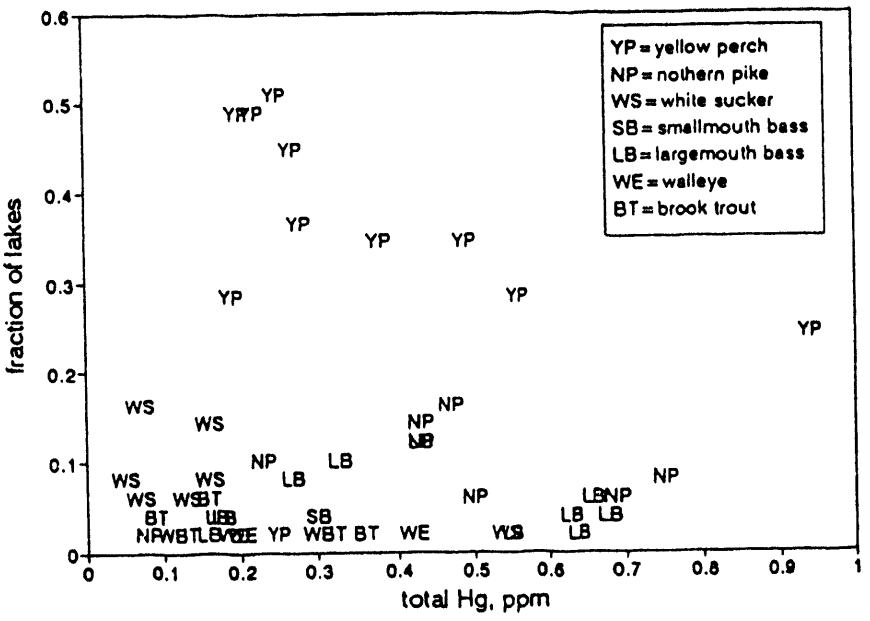


Figure B.10. Probability of fishing a lake with specified average fish Hg levels, by species. Data from Gloss et al. (1990). Plotting symbols are: BT = brook trout, LMB = large mouth bass, NP = northern pike, SMB = small mouth bass, WE = walleye, WS = white sucker, YP = yellow perch.

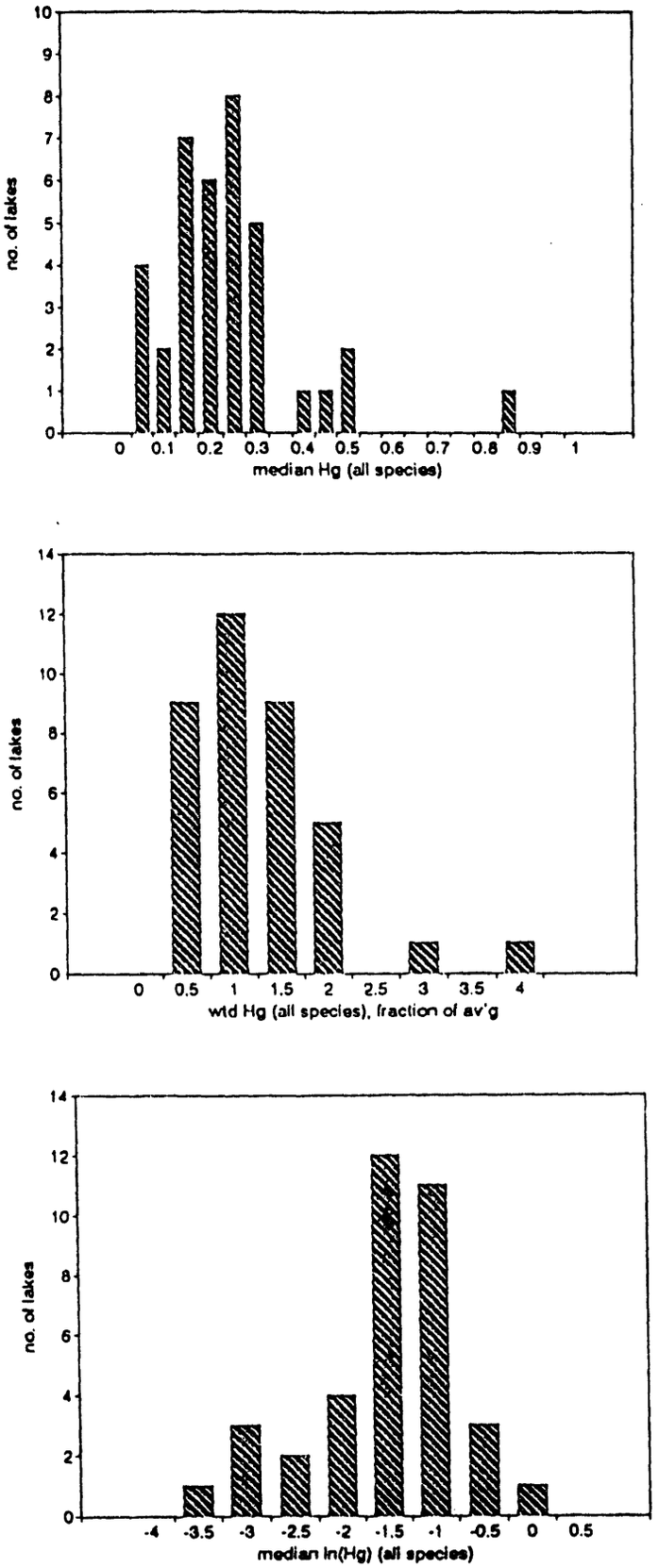


Figure B.11. Frequency distribution of median Hg levels for all fish species among 49 Upper Michigan lakes. Data from Gloss et al. (1990). (a) plotted against median Hg. (b) plotted against weighted average Hg as a fraction of the overall average. (c) plotted against the median log of Hg concentration.

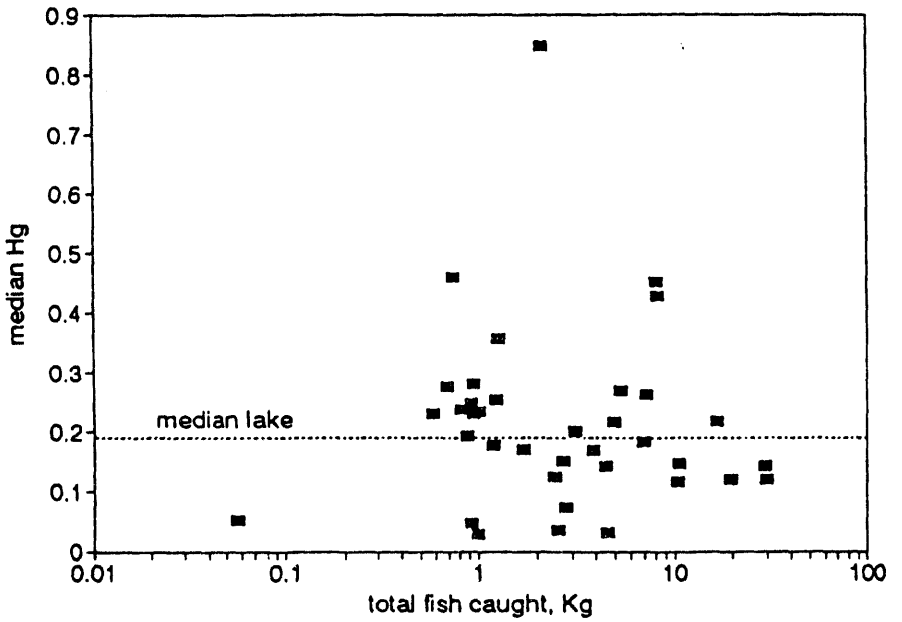


Figure B.12. Median Hg concentration by weight of fish caught, for each of 49 Upper Michigan lakes. Data from Gloss et al. (1990) and Cusimano et al. (1989).

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