An Assessment of Mercury Emissions and Health Risks from a Coal-Fired Power Plant

V.M. Fthenakis, F. Lipfert, and P. Moskowitz
Analytical Sciences Division, Department of Applied Science
Brookhaven National Laboratory
Upton, NY 11973

Presented at the 1994 AIChE Summer National Meeting
Denver, Colorado, August 14-17, 1994

ABSTRACT

Title III of the 1990 Clean Air Act Amendments (CAAA) mandated that the U.S. Environmental Protection Agency (EPA) evaluate the need to regulate mercury emissions from electric utilities. In support of this forthcoming regulatory analysis the U.S. DOE, sponsored a risk assessment project at Brookhaven (BNL) to evaluate methylmercury (MeHg) hazards independently. In the U.S. MeHg is the predominant way of exposure to mercury originated in the atmosphere. In the BNL study, health risks to adults resulting from Hg emissions from a hypothetical 1000 MW coal-fired power plant were estimated using probabilistic risk assessment techniques. This 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.

1. INTRODUCTION

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

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

This research was performed under the auspices of the United States Department of Energy under Contract
No. DE-AC02-76CH00016

MASTER

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. This work is described in a report by Lipfert et al, 1994. The current paper summarizes the basic conclusions of this assessment and highlights issues dealing with emissions control and environmental transport which may be of special interest to this audience.

2. BACKGROUND

2.1 Mercury Sources

Natural mercury arises from the degassing of the earth's crust through volcanic gases and evaporation from the oceans. Currently, in the U.S. the main anthropogenic emissions of mercury to the atmosphere are attributed to burning of fossil fuels and municipal waste, whereas in the past the chloroalkali and the wood pulping industries were producing significant amounts of atmospheric mercury. Mercury has a special place in the list of environmental pollutants, since its adverse health effects have been convincingly demonstrated in several unfortunate poisoning incidents (e.g. Japan 1950s and 1960s, Iraq 1970s). Although Hg is distributed throughout the environment, its toxic effects result mainly from ingestion of contaminated seafood. Unlike many other air pollutants, mercury residence times in the atmosphere are long and it may eventually be dispersed over large areas. As a result adverse effects must be considered on both local and regional scales.

2.2 Mercury Risk 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. We reviewed five assessments of baseline risks (i.e., NAS, 1991; Tollefson and Cordle, 1986; FDA 1993; NIEHS, 1994; Stern, 1993) and three risk assessments on mercury sources (i.e., New Jersey

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

municipal solid waste incinerators, 1992; Constantinou et al, 1993; Rowe, 1993). 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. Some highlights from these studies are given below.

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 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 a grain poisoning incident in Iraq (Marsh et al., 1987; Bakir et al., 1973). 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 that "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 Federal Drug Administration 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. Tollefson and Cordle had concluded "The majority of fish consumers in the United States could easily double their intake and still remain below the ADI."

2.2.3 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 grain poisoning of mothers and children, to estimate the risks entailed in the current U.S. EPA reference dose (0.3 mg/day per kg of body weight). Stern concluded that the current reference dose should be reduced to 0.07 mg/kg/day to preclude effects on fetal development.

2.2.4 New Jersey Municipal Solid Waste Incinerators. In 1992, the New Jersey Task Force on Mercury Emissions Standard Setting released a study 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, run-off, rates of Hg accumulation in aquatic sediments and subsequent bioaccumulation in fish. Their assessment does not actually estimate health risks *per se*, since dose-response information underlying the reference dose levels is not considered. 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."

2.2.5 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.

This review of previous assessments help us determine important issues which were incorporated in the BNL analysis:

1. Use of probabilistic rather than deterministic methods.
2. Desegregation 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.

3.0 FRAMEWORK OF THE PRESENT ASSESSMENT

The estimation of health risks associated with mercury emissions involves: 1) source term characterization (including efficiency of pollution control); 2) environmental fate and transport; 3) human exposure; 4) dose response analysis; and 5) risk characterization. This study is based on a hypothetical coal-fired power plant, as a paradigm. The analysis explicitly recognizes that the general public's wide range of fish consumption patterns and that health effects are keyed to equilibrium levels of Hg body burden, not to the acute effects of individual doses. The features of the BNL analysis are summarized in Table 1 and the following sections. More details can be found in Lipfert et al., 1994.

Table 1 Features of the BNL 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 are considered.
Accumulation in surface waters	All Hg deposited within the watershed is assumed to enter the water body. Incremental wet and 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	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 three different types of fish meals (i.e., canned tuna fish, marine shell and fin fish, and freshwater fish) 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 and power plant contributions to MeHg in freshwater 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.

4. SOURCE TERM CHARACTERIZATION

4.1 Emissions of Mercury from Power Plants

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/10¹² 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 change is probably due to improved methods of sampling and analysis.

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 Hg⁰ to Hg⁺⁺ (Figure 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%, so that it is expected that a large fraction of the emitted mercury will be reactive.

The coal -burning plant considered in this analysis is assumed to have a rating of 1000 MW_e, a capacity factor of 80%, a stack height of 200 m, and to be located in the upper midwest in flat terrain in the vicinity of sport fishing lakes. Average mercury content in the coal is assumed (0.08 ppm).

4.2 Mercury Emissions Control

Mercury seems to be the most difficult to remove trace metal because it is present mainly in vapor form and tends to pass through particulate control devices common in fossil fuel plants with which most fossil fuel plants are equipped. Several methods of mercury control are being used at waste incineration plants, especially in Europe. Mercury concentrations in flue gas from utility boilers, however, are about two orders of magnitude lower than those in flue gas from waste incineration plants (e.g., 1-10 ug/m³ versus 50-500 ug/m³) (Chang and Owens, 1994). This difference poses both detection and removal challenges.

The up-to-date work on mercury control technologies shows mercury removal effectiveness to be strongly dependent on the mercury species present. 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 HgCl₂, 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 mg/m³) typically encountered in flue gases present a difficult challenge for sampling and detection methods.

Electrostatic Precipitators (ESPs). All of today's U.S. coal-burning power plants have some kind of particulate control device, usually 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. 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.

Wet Scrubbing. About 20% of the 325 GW coal-fired capacity in the US is equipped with wet flue gas desulfurization (FGD) systems (Chang and Owens, 1994). A typical FGD system uses an alkaline slurry sprayed in the flue gas over a liquid-gas contact medium. Obviously, mercury solubility in the scrubbing slurry depends on the mercury's form. Elemental mercury is essentially insoluble in FGD scrubbing slurry, while some of the oxidized species, such as HgCl_2 , are very soluble. Field measurements of mercury removal efficiency at full scale wet scrubbers have ranged from 10% to over 90%. Meij (1991) 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%. These tests show that Hg removal by a wet scrubber depends on the fraction of oxidized species present.

Dry Scrubbing. Dry scrubbing systems may be of the spray dryer absorption (SDA) or of the dry injection type. In a spray dryer, the sorbent solution is atomized into the incoming flue gas streams to increase the liquid-gas interface. 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. Dry injection generally involves pneumatic introduction of a dry, powdery material into the flue gas. In either system the powder-carrying gas passes through a fabric filter or ESP where the dry product is collected. High efficiency removal of both elemental and oxidized mercury may require a combination of SDA and dry solvent injection. 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 1,

these efficiencies range from 6% to 96%. Also shown in this table is a strong influence of the coal Cl^- content on the removal efficiency.

Table 1. Effect of Chlorine Content in Coal on Removal of Hg from Flue Gas

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

The efficiency of the SDA system in Plant H increased from 96% to more than 99% when a carbon injection system was added. Felsvang et al. (1993) reports a pilot plant study which further quantifies the effect of adding sorbent injection to the SDA. Two types of coal of different chloride content (i.e., 0.019% Cl^- , and 0.094% Cl^-) were used in this study. With SDA alone, the mercury removal efficiency from the low Cl^- emissions was 57%, whereas from the high Cl^- was 78%. Combining SDA with activated carbon increased the efficiency for both types of coal to 90%. However, it was found that the low Cl^- coal required five times more carbon than the other. This is probably due to the fact that higher Cl^- content produces a higher fraction of soluble Hg compounds (mainly as HgCl_2 , which is also more reactive). In a SDA system without carbon injection, the inherent removal of elemental Hg is practically zero due to its lack of solubility, whereas the inherent removal of water-soluble oxidized Hg is 95%. With carbon injection, removal of elemental Hg can increase from up to about 60% when sufficient quantity of carbon is injected (Figure 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 3).

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.

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 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, char filters may be a relatively expensive option to implement in power plants, given the high flow rates of stack gases.

In summary, overall rates of Hg removal range from a low of 10% to a high of 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, corresponding to the use of ESPs.

5. ENVIRONMENTAL FATE AND TRANSPORT

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 $(\text{CH}_3)_2\text{Hg}$ 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 and dimethyl

mercury ($\text{CH}_3)_2\text{Hg}$ 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. 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.

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).

Mercury is removed from the atmosphere by dry and wet deposition. 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. Extension to downwind distances greater than 50 km is only a matter of applicability of the dispersion model.

5.1 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 80% 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, according to Figure 1.

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 (C/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	0.75
(Hg species)	Hg^0 Hg^{++} $\text{Hg}_{\text{part.}}$
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. (1985) 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. 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

$$R_{\text{airborne}} = Q - R_{\text{td}}$$

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.

$$R_{\text{td}} = 2\pi \int_0^{50} F_{\text{td}}(r) r \, dr$$

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

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

where $DD_i(r)$ and $WW_i(r)$ are the fluxes of dry and wet deposition. These fluxes are calculated by

$$DD_i(r) = V_d a_i C(r,0) I_d$$

$$WW_i(r) = WR a_i C(r,0) P$$

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 4; 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 $17 \text{ mg/m}^2\text{y}$, 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. 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 actual deposition measurements shown in Figure 5 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%. Sensitivity studies would be useful to identify those elements of the local impact calculations that have the largest effect.

6. HUMAN EXPOSURE

Fish ingestion is assumed to be the only important pathway for exposure to MeHg (Gunderson, 1988). 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 versus 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.

7. DOSE-RESPONSE ANALYSIS

7.1 Fish consumption rates

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. The average daily (baseline) MeHg dose for consumers of freshwater fish in the upper Midwestern U.S. was estimated to be about 4.6 ug/d. The 99th percentile was about 34 ug/d, which is only about 10% of the U.S. EPA estimated dose threshold for adult health effects. As discussed above, local increases in mercury deposition are assumed to have proportional effects on the mercury dose to the population.

7.2 The steady-state diet-blood-body burden MeHg relationship

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. The half-life expressing the differential effect of MeHg uptake and extraction controls the magnitude of the response expected from a given rate of daily input together with body weight. A lognormal representation of half-lives determined on 48 Iraqi MeHg poisoning victims was used as input to the risk assessment. It is necessary to account for the effective dose averaging that takes place as equilibrium body burdens of MeHg are reached and maintained. The way to accumulate a lot of MeHg is to eat fish often rather than eating large quantities of fish. 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.

7.3 Health Effects

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 assessment, we use data from the Iraqi grain poisoning incident to develop a dose-response 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.

8. RISK CHARACTERIZATION

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. 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 we estimated risk using probabilistic sampling techniques (i.e., Latin Hypercube and Monte Carlo). 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, which for the baseline case 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.

The statistical distributions of the Hg content of 3 different seafood categories, their rates of consumption and numbers of meals to reach the equilibrium dose were combined to define the dose distributions. The distribution of equilibrium body burdens was based on the distributions of doses, body weights, and biological half-lives. The distribution or responses was defined by the distribution of doses, after considering the uncertainties in the dose-response function. The effects of power plant contributions on marine species were found 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.

9. CONCLUSIONS

This analysis showed that the effects of emissions of a hypothetical power plant may double the extreme case (upper percentile) exposure to MeHg resulting from consuming fish from freshwater in the vicinity of the plant. However, even at these more elevated exposure levels the incidence of mild neurological symptoms (paresthesia) to adults is estimated to be quite small, especially when compared with the estimated background incidence. For example, in a population of 10,000 heavy fish eaters, ~1 case of paresthesia due to fish consumption would be expected in the absence of a power plant, <3 cases with a plant, in a total of ~ 220 cases due to all other causes.

Some issues of uncertainty still remain; these include but are not limited to: fish consumption patterns of sensitive population elements (e.g., pregnant women) ; accumulative effects from several local or regional sources; long-term impacts.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

13. REFERENCES

- Ahmed, F.E. (1991), Seafood Safety, National Academy Press, Washington, DC. 446 pp.
- Bakir, F., et al. (1973), Methylmercury poisoning in Iraq, *Science* 181:230-241.
- Bloom, N.S., Lupsina, V., and Prestbo, E. (1993). Fluegas Mercury Emissions and Speciation from Fossil Fuel Combustion, presented at the Second International Conference on Managing Hazardous Air Pollutants, Washington, DC, July 13-15, 1993.
- Burmester, D.E. et al. (1990), Monte Carlo Techniques for Quantitative Uncertainty Analysis in Public Health Risk Assessments, pp. 215-221, in SUPERFUND '90, HMCRI's 11th Annual National Conference and Exhibition, November 26-28, Washington, DC.
- Chang R. And Owens D., Developing Mercury Removal Methods for Power Plants, EPRI Journal, pp. 46-49, July/August 1994.
- Chu P., B. Nott, and W. Chow, Results and Issues from the PISCES Field Tests, presented at the Second International Conference on Managing Hazardous Air Pollutants, Washington, DC, July 13-15, 1993.
- Constantinou, E., Brown, S., Wu, X., and Seigneur, C. (1993), Uncertainty of Mercury Health Risk Estimates, AWMA paper 93-TA-36A.06, presented at the Annual meeting of the Air&Waste Management Association, Denver, CO.
- FDA, 1993: Carrington, C., personal communications.
- Felsvang, K., Gleiser, R., Juip, G., and Nielsen, K.K., Air Toxics Control by Spray Dryer Absorption Systems, presented at the Second International Conference on Managing Hazardous Air Pollutants, Washington. DC, July 13-15, 1993.

Greenberg, A. et al. (1992), Mercury in Air and Rainwater in the Vicinity of a Municipal Resource recovery Facility in Northwestern New Jersey, Proc. 1992 Int. Symp. on Measurement of Toxic and Related Air Pollutants, Durham, NC. Air & Waste Management Assoc., Pittsburgh, PA.

Greenberg, A., personal communication, January 10, 1994.

Gunderson, E.L. (1988), FDA Total Diet Study, April 1982-April 1984, Dietary Intakes of Pesticides, Selected Elements, and Other Chemicals, J. Assoc. Off. Anal. Chemists, 71:1200-9. Detailed data listings provided separately.

Hall, E.T. (1974), Mercury in Commercial Canned Seafood, J. AOAC 57:1068-73.

Iverfeldt, A., and Lindqvist, O. (1986), Atmospheric oxidation of elemental mercury by ozone in the aqueous phase, Atmospheric Environment 20:1567-73.

Jozewicz, W., Krishnan, S.V., and Gullett, B.K., Bench-scale investigation of mechanisms of elemental mercury capture by activated carbon, presented at the Second International Conference on Managing Hazardous Air Pollutants, Washington, DC, July 13-15, 1993.

Lindberg, S.E. (1980), Mercury partitioning in a power plant plume and its influence on atmospheric removal mechanics, Atmospheric Environment 14:227-31.

Lindqvist, O. (1985), Atmospheric mercury - a review, Tellus 37B:136-159.

Lipfert F.W., Moskowitz P.D., Fthenakis V.M., DePhillips, Viren J., and L. Saroff, Assessment of Mercury Health Risks to Adults from Coal Combustion, BNL-60435, Brookhaven National Laboratory, May 1994.

Lipfert, F.W., Dupuis, L.R., and Schaedler, J.S. (1985), Methods for Mesoscale Modeling for Materials Damage Assessment, Brookhaven National Laboratory Report to US Environmental Protection Agency, BNL 37508. Also see EPA/600/S8-85/028 (NTIS PB 86-144862/AS).

Marsh, D.O., et al. (1987), Fetal methylmercury poisoning. Relationship between concentration in single strands of maternal hair and child effects, Arch.Neurol. 44:1017-22.

McIlvaine R. W., Removal of heavy metals and other utility air toxics, presented at the Second International Conference on Managing Hazardous Air Pollutants, Washington, DC, July 13-15, 1993.

Meij, R. (1991), The fate of mercury in coal fired power plants and the influence of wet flue-gas desulphurization, Water, Air, and Soil Poll. 56:21-33.

NIEHS, 1994: Fouts J. Personal communication.

Nobblett, J.G., Meserole, F., and Owens, D. (1993), Control of Air Toxics from Coal-Fired Power Plants Using FGD Technology, presented at the Second International Conference on Managing Hazardous Air Pollutants, Wash. DC, July 13-15, 1993.

Obermiller, E.L., Conrad, V.B., and Lengyel, J. (1991), Trace Element Contents of Commercial Coals, presented at Managing Hazardous Air Pollutants: State of the Art, Washington, DC. Electric Power Research Institute, Palo Alto, CA.

Rowe, R.D., et al. (1993), Task 3 Methodology Report, Part II of III, Air Emissions, prepared for Empire State Electric Energy Research Corporation, New York, NY.

Stern, A.H. (1993), Re-evaluation of the References Dose for Methylmercury and Assessment of Current Exposure Levels, Risk Analysis 13:355-64, (1993).

Tollefson, L., and Cordle, F., Methylmercury in Fish: A Review of Residue levels, Fish Consumption and Regulatory Action in the United States, *Envir. Health Perspect.* 68:203-8, (1986).

Wolfe, R.J., and Walker, R.J. (1987), Subsistence Economies in Alaska: Productivity, Geography, and Developmental Impacts, *Arctic Anthropology* 24:56-81.

World Health Organization (WHO), Mercury Environmental Aspects, Environmental Health Criteria 86, Geneva, 1989.

World Health Organization (WHO), Methylmercury, Environmental Health Criteria 101, Geneva, 1990.

World Health Organization (WHO), Inorganic Mercury, Environmental Health Criteria 118, Geneva, 1991.

Figure Legends

Figure 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.

Figure 2. Effect of carbon injection on Hg removal by spray-dryer absorption systems

Figure 3. Effect of chemical carbon activation agents on Hg removal.

Figure 4. Estimated rates of total and wet deposition of mercury from a hypothetical 1000 MW_e power plant burning coal with average Hg content.

Figure 5. Average concentration of Hg in precipitation, measured for 6 rain events, near a New Jersey incinerator. Data from Greenberg et al. (1992)

Fig. 1.

REACTIVE Hg EMISSIONS FROM POWER PLANTS

% MERCURY REMOVAL

INFLUENCE OF ACTIVE CARBON TYPE
ON MERCURY REMOVAL

ESTIMATED Hg DEPOSITION FROM HYPOTHETICAL 1000 MW PLANT

Hg IN RAIN NEAR A WASTE INCINERATOR