

## An assessment of mercury emissions and health risks from a coal-fired power plant

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### Abstract

Title III of the 1990 Clean Air Act Amendments (CAAA) directed the US Environmental Protection Agency (EPA) to evaluate the rate and effect of mercury emissions in the atmosphere and technologies to control such emissions. In response to this and other forthcoming analyses the US DOE sponsored a risk assessment project at Brookhaven (BNL) to evaluate health risks of mercury emissions from coal combustion. Methylmercury (MeHg) is the compound predominantly responsible for human exposures to atmospheric mercury in the United States, through the fish ingestion pathway. In the BNL study, health risks to adults resulting from Hg emissions from a hypothetical coal-fired power plant were estimated using probabilistic risk assessment techniques. This study showed that the effects of emissions of a single large 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.

*Keywords:* Risk assessment; Mercury; Coal burning; Fish consumption; Health risks; Paresthesia

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### 1. Introduction

The US Environmental Protection Agency (EPA) is preparing, for the US Congress, a report evaluating the need to regulate mercury (Hg) emissions from all sources of emissions including the electric utility industry. This study, to be completed in 1995, will have important health and economic implications. Parallel with these efforts, the US Department of Energy, Office of Fossil Energy, sponsored a risk assessment

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project at Brookhaven National Laboratory (BNL) to evaluate methylmercury (MeHg) hazards independently. This work is described in a report by Lipfert et al. [1]. The current paper summarizes the basic conclusions of this assessment and highlights issues dealing with emissions control and environmental transport.

## 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 US, the main anthropogenic emissions of mercury to the atmosphere are attributed to burning of fossil fuels and municipal waste, whereas in the past the chlor-alkali 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 mercury 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 [2–6] and three risk assessments on mercury sources [7–9]. 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 mercury [2]. 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 [10, 11]. 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 [2] described above criticized a previous Federal Drug Administration effort by Tollefson and Cordle [3] 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 that 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 [6] who used the data on Iraqi grain poisoning of mothers and children to estimate the risks entailed in the current US EPA reference dose (0.3 µg/day per kg of body weight). Stern concluded that the current reference dose should be reduced to 0.07 µg(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 [7]. 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. [8] 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 helped us to establish important assumptions of the BNL risk analysis:

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. Considering equilibrium rather than instantaneous blood levels of Hg.

## 3. Framework of the present assessment

Our estimation of health risks associated with mercury emissions involves: (1) source term characterization (including efficiency of pollution control); (2) environmental

Table 1  
Features of the BNL mercury assessment model

Mercury emissions	Fraction of Hg emitted as Hg <sup>++</sup> is estimated based on Cl <sup>-</sup> content of coal. Effects of air pollution controls are estimated.
Transport, dispersion, and deposition	Gaussian plume transport and dispersion model for three classes of Hg compounds 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 the global pool. Global deposition is increased in proportion to (net) emissions.
Effects of local sources on fish Hg content	The Hg concentration 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 large hypothetical power plant.
Treatment of uncertainties	Uncertainties in Hg emissions, transport, chemical reactions, deposition, and uptake by fish are combined in an "impact factor" by which Hg in freshwater fish increases with local Hg deposition. Uncertainties in the distribution of baseline body burden of Hg are assessed by comparison with measurements.

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 Ref. [1].

#### 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. [12] indicated that recently measured Hg emissions are in the range of 4–8 lb/10<sup>12</sup> BTU, roughly corresponding to Hg contents

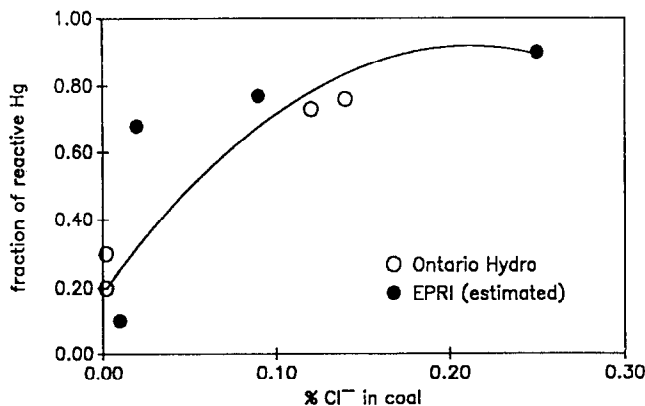


Fig. 1. Data on the fraction of total emitted Hg measured as reactive Hg, as a function of fuel chloride content. Graph from Felsvang et al. [16], additional data (+ symbols) from Ontario Hydro.

in coal of 0.05–0.10 ppm. Earlier data on mercury in coal [13] had indicated a higher mean and a much larger range of values (0.04–0.34 ppm). The difference between early and recent estimates may be due to improved methods of sampling and analysis. In this first-level analysis, we assume values in the upper end of these ranges (e.g., 0.10–0.25 ppm).

Almost all of the mercury in flue gas is in the gaseous state [14]. However, recent studies indicate that 50–90% of the mercury in flue gas from coal firing may be oxidized [12, 15]. Data for species distribution in the flue gas of coal-fired power plants are still somewhat uncertain. The conversion of elemental to oxidized Hg appears to depend on the interplay of several factors, including temperature of the combustion zone, residence times and temperatures in heat exchangers, and the chloride content ( $\text{Cl}^-$ ) in the coal. According to Felsvang et al. [16] the  $\text{Cl}^-$  content appears to have a strong influence in converting  $\text{Hg}^0$  to  $\text{Hg}^{++}$  (Fig. 1). As shown in this figure, in four of the five plants using coal with  $\text{Cl}^-$  content above 0.02%, the fraction of oxidized mercury was 0.70 or more. The estimated range of  $\text{Cl}^-$  in US coals is 0.04–0.2%, so that it is expected that a large fraction of the emitted mercury will be in oxidized form.

#### 4.2. Mercury emissions control

Mercury is difficult to remove because it is present mainly in vapor form and tends to pass through the particulate control devices 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  $\mu\text{g}/\text{m}^3$  versus 50–500  $\mu\text{g}/\text{m}^3$ ) [17]. These low concentrations pose 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 compounds present. As discussed above, most of the Hg in coal is vaporized, while about 10% is associated with 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  $\text{HgCl}_2$ , 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 ( $0.1\text{--}10\text{ mg/m}^3$ ) typically encountered in flue gases makes sampling and detection difficult.

*Electrostatic precipitators (ESPs).* All of today's US coal-burning power plants have some kind of particulate control device, usually ESPs. Preliminary measurements at plants burning bituminous coal indicate that conventional ESPs remove less than 20% of the Hg present in flue gases [12]. Since most of the Hg is in the vapor phase, ESPs cannot be effective control devices unless Hg condenses onto particulates at or before the device. Meij [18] suggests that Hg removal efficiency across an ESP improves with increasing chloride content in coal, suggesting that  $\text{Cl}^-$  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 [17]. 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  $\text{HgCl}_2$ , are very soluble. Field measurements of mercury removal efficiency at full-scale and pilot plant wet scrubbers have ranged from 10% to over 90%. Meij [18] indicates removal efficiencies from 10% to 70% for such systems. Nobblett et al. [19] also suggest that Hg removal depends on the chloride content in the coal, simply because of the oxidation to water-soluble  $\text{HgCl}_2$ . For plants burning western US 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 data are limited because they were obtained from only a few plants; they indicate, however, that Hg removal by a wet scrubber depends on the fraction of oxidized species present.

*Dry scrubbing.* Dry scrubbing systems are of either the spray dryer absorption (SDA) or 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. [16] report mercury removal efficiency of several SDA systems in coal-fired power stations operating in the US and Europe. As shown in Table 2, these efficiencies range from 6% to 96%. Also shown in this table is a strong influence of the coal  $\text{Cl}^-$  content on the removal efficiency. 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. [16] also report 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%  $\text{Cl}^-$ , and 0.094%  $\text{Cl}^-$ ) were used in this study. With SDA alone, the mercury removal efficiency from the low  $\text{Cl}^-$  emissions was 57%, whereas from the high  $\text{Cl}^-$  it was 78%. Combining SDA with activated carbon increased the efficiency for both types of coal to 90%. However, it was found that the low  $\text{Cl}^-$  coal required five times more carbon than the other. This is probably due to the fact that higher  $\text{Cl}^-$  content produces a higher fraction of soluble Hg compounds (mainly as  $\text{HgCl}_2$ , 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 about 95%. With carbon injection, removal of elemental Hg up to 60% can be accomplished when sufficient quantity of carbon is injected (Fig. 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. [20] and Felsvang et al. [16] 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 (Fig. 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 [21], mercury is effectively removed by these devices.

Table 2  
Effect of chlorine content in coal on removal of Hg from flue gas

Plant	Fly ash loading	Coal $\text{Cl}^-$ content	% Hg removal
A	High	Low (0.01%)	14
B	High	Low (0.01%)	23
C	High	Low (0.01%)	6
D	High	Low (0.01%)	16
E	Low	High (0.1–0.3%)	55
F	Low	High (0.1–0.3%)	44
G	Medium	High (0.1–0.3%)	89
H	High	High (0.1–0.3%)	96

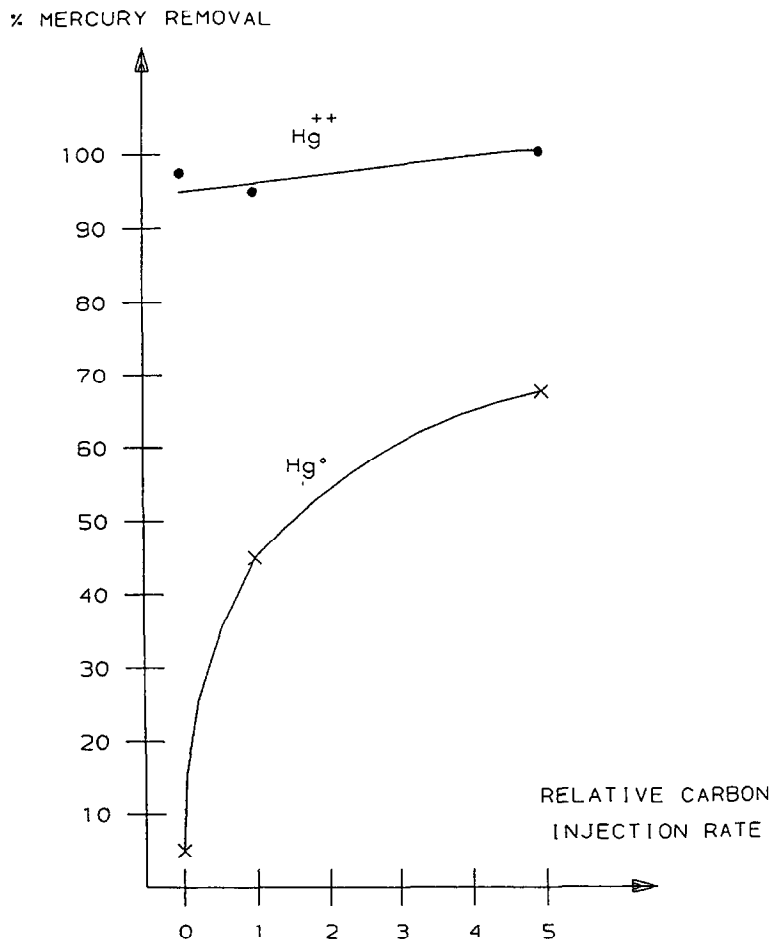


Fig. 2. Effect of carbon injection on Hg removal by spray-dryer absorption systems,  $T = 160^{\circ}\text{F}$  (from Felsvang et al. [16]).

**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 [21], in a literature review of several controls, reports that char filters are the most efficient devices for mercury removal. However, he does not refer to Hg speciation and does not consider the variation of  $\text{Cl}^-$  in the flue gas streams. 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 [12]; in the interim, we assume a range of Hg in coal (0.10–0.25 ppm) and a 10% removal efficiency, corresponding to the use of ESPs.



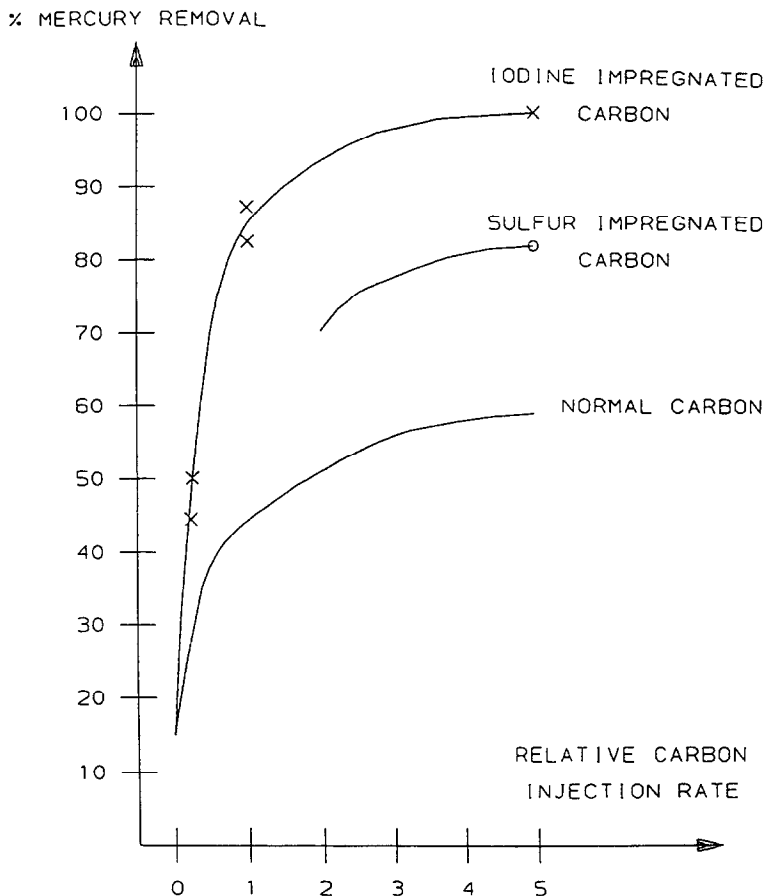


Fig. 3. Effect of chemical carbon activation agents on Hg removal (Felsvang et al. [16]).

## 5. Environmental fate and transport

Mercury is present in the atmosphere in three oxidation states: elemental,  $\text{Hg}^0$ ; monovalent,  $\text{Hg}^+$ ; and divalent mercury,  $\text{Hg}^{++}$ . 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.,  $\text{HgCl}_2$ ) are extremely water-soluble, methyl chlorides (e.g.,  $\text{HgCH}_3\text{Cl}$ ) 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  $\text{HgCl}_2$ , at least initially. Background ambient air concentrations in remote areas are in the range of 1–3  $\text{ng}/\text{m}^3$ , and as high as 10  $\text{ng}/\text{m}^3$  in

urban areas. Lindberg [14] measured concentrations of total gaseous mercury in excess of 1000 ng/m<sup>3</sup> 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 [22], before it is removed from the atmosphere by dry and wet deposition. Elemental mercury has a residence time between 0.5 and 1.5 year, whereas most oxidized species have lifetimes of hours or days in the atmosphere [23]. 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).

### 5.1. The BNL model for incremental Hg deposition from point sources

The model plant is assumed to be located near a lake in the upper Midwest and to have a capacity factor of 75%. In order to allow for a range of power plant capacities and Hg content in the coal, we assumed that the product of nameplate rating and Hg concentration was 250 ppm-MW (e.g., a 1000 MW plant and 0.25 ppm of Hg in coal, or a 2500 MW plant and 0.10 ppm in coal), which corresponds to annual Hg emissions of about 550 kg from a single plant. The emission controls (electrostatic precipitator) were assumed to reduce the mercury emissions by about 10%; this results in an estimated total Hg emission rate of 500 kg/yr comprising mainly elemental and reactive Hg compounds. The total deposition of mercury on ground level surfaces can be estimated once the distribution of ground level ambient concentration is known. A Gaussian plume dispersion model (the EPA ISCLT-2 model) was used to estimate the local annual average air concentrations of three types of mercury compounds (i.e., elemental, water-soluble, and particulate) for flat terrain and distances 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.

The calculations for the hypothetical power plant are based on the following parameters:

Stack height:	200 m		
Total Hg emission rate	17 mg/s (500 kg/yr)		
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 <sup>0</sup>	Hg <sup>++</sup>	Hg <sub>part</sub>
Dry deposition velocities (cm/s)	0.02	1.9	~0
Washout ratios	~ 0	10 <sup>5</sup>	10 <sup>6</sup>

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  $R_{\text{td}}$  is the rate of total deposition within the specified radius, calculated by integrating the product of deposition flux ( $F_{\text{td}}$ ) 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,

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

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

$$DD_i(r) = V_d a_i C(r, 0) I_d, \quad WD_i(r) = WR a_i C(r, 0) P,$$

where  $V_d$  is the deposition velocity (m/s),  $C(r, 0)$  is the ambient ground-level concentration of total mercury at distance  $r$  from the plant [ $\text{g}/\text{m}^3$ ],  $a_i$  are the fractions of concentrations of soluble, non-soluble and particulate forms of mercury,  $I_d$  is the fraction of time it does not rain heavily in a particular location,  $WR$  is the washout ratio, and  $P$  is the annual average precipitation in same location [ $\text{m}/\text{yr}$ ].

The results of these calculations are shown in Fig. 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 approximately  $18 \text{ mg}/(\text{m}^2 \text{ yr})$ , 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. The percentage of total Hg deposited out to 50 km is estimated to be in the range of 5–8%. This percentage would be increased considerably if a shorter stack height had been assumed, since maximum concentrations increase approximately inversely 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 near the stack is thus very uncertain, since some Hg could wash out of the plume while it is still aloft. The peak cumulative ambient concentration occurs in the

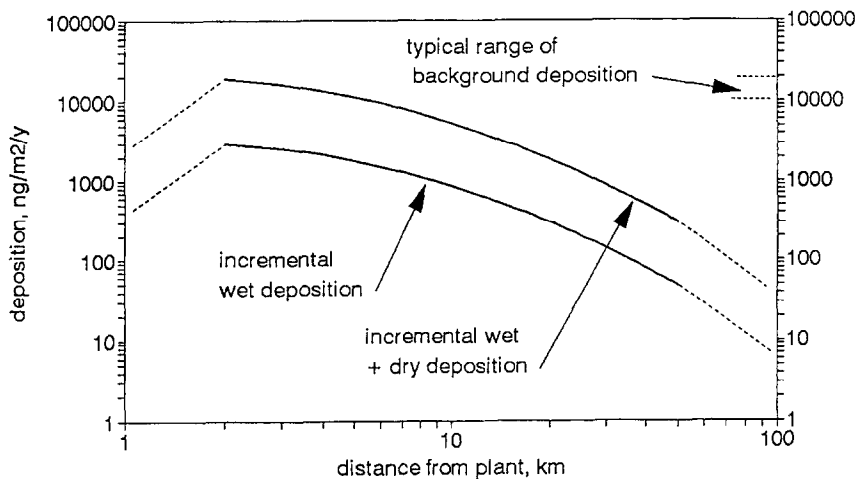


Fig. 4. Estimated rates of total and wet deposition of mercury from a hypothetical 3000 MW<sub>e</sub> power plant burning coal with average Hg content.

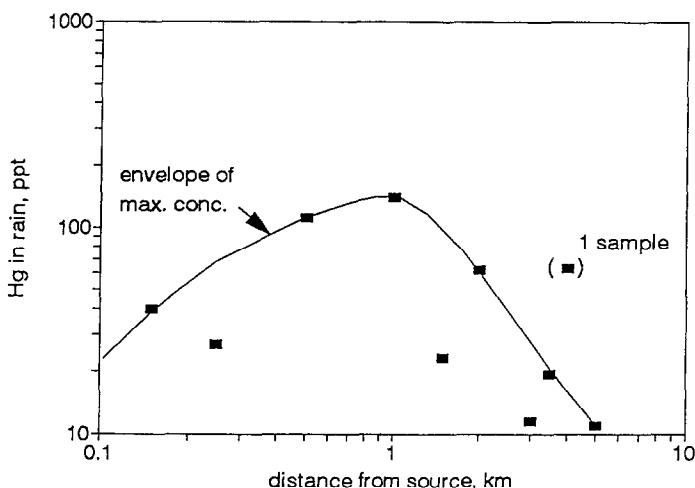


Fig. 5. Average concentration of Hg in precipitation, measured for six rain events, near a New Jersey incinerator. Data from Greenberg et al. [24].

range of 2–10 km, depending on the stack height and meteorological conditions. Ambient air concentrations, and, therefore, deposition rates decrease rapidly with distance; concentrations at 50 km are about one order of magnitude lower than the peak concentrations. The predicted average deposition rate and the percentage of Hg emissions deposited within an area of 50 km radius around the model plant are about 10 kg/yr and 5–8%, respectively. We assume that the remainder of the emitted Hg becomes part of the global background. The predictions of our model of peaking

deposition near the plant and rapid decrease with distance farther downwind, agree with measurements of wet deposition of Hg downwind of a municipal waste incinerator (Fig. 5) reported by Greenberg [24]. No data related to power plant plumes were available for the model verification exercise.

Probabilistic consideration of all the variations of power plant design, location, Hg emissions, atmospheric transport, deposition, and bioaccumulation may entail cumulative uncertainties of several orders of magnitude. We have allowed for some measure of this variability, by using an impact factor, representing the effect of local Hg deposition averaged over a nearby lake and the surrounding watershed, in comparison to deposition of background Hg on the same area. This factor was assigned a uniform probability distribution from 1.5 to 3.0 (50–200% increase over the background).

## 6. Human exposure

Fish ingestion is assumed to be the only important pathway for exposure to MeHg [25]. 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 bio-concentration 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 US was estimated to be about 4.6 µg/d. The 99th percentile was about 34 µg/d, which is only about 10% of the US 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 sporadically. 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 [26]. Effects of MeHg on the developing fetus are thought to be more critical than on adults. According to WHO [26], “it affects normal neurological 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. [27], 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 endpoint in question (i.e., adult paresthesia). First, the expected individual risk averaged over the entire population being considered, which for the baseline case is the entire US 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, a highly 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 three 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 of 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 US utility industry contributes less than 1% of the existing global pool each year [28]. The risk analysis was performed for several different assumptions involved in the metabolic model and predicted a baseline average risk level of about  $2 \times 10^{-5}$ – $3 \times 10^{-5}$  (2–3 chances in 100 000), with a 95th percentile risk range of  $6 \times 10^{-5}$ – $12 \times 10^{-5}$ . When power plant increments were added, the expected average risk level increased to  $4 \times 10^{-5}$ – $7 \times 10^{-5}$  with an upper 95th percentile risk of  $13 \times 10^{-5}$ – $27 \times 10^{-5}$ . 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

A simple model of Hg deposition from point sources was derived and used to predict the impact of Hg emissions from a hypothetical coal-fired power plant. Our study showed that these emissions 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, about 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 approximately 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) [29] accumulative effects from several local or regional sources, and long-term impacts.

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