

"UNDERSTANDING THE SOURCES, TRENDS, AND IMPACTS OF MERCURY IN THE ENVIRONMENT"

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Abstract

Over the past decade, the presence of mercury in the environment has become a major public concern because, at high levels of exposure, mercury can cause adverse health effects in humans and animals. Specifically, mercury concentrations in fish have been of interest to many groups because of the potential for adverse human health effects. The purpose of this paper is to examine the numerous man-made and natural sources of mercury in the environment, to review mercury's movement in the environment, and to discuss the potential health effects associated with exposures to mercury.

In response to concerns regarding the mercury problem, federal and some state regulatory agencies have scrutinized municipal waste combustion (MWC) facilities (which includes waste-to-energy [WTE] and non-energy recovering incinerators) and are in the process of appropriately requiring strict controls to minimize mercury emissions from these plants. As a result, the MWC industry has devoted its resources into providing a more effective way of controlling chemical emissions via technological measures. However, similar strict controls have yet to be required for any other mercury emissions sources (e.g., power plants, smelters, etc.). Although MWC facilities do emit mercury as a result of combusting waste materials containing mercury, it should be recognized that the industry has taken many measures to control mercury emissions, and has committed itself to attacking the problem of mercury accumulation in the environment. Due to these efforts, mercury emissions associated with MWC facilities have been declining rapidly, and are anticipated to decline even more in the future.

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Introduction

Over the past decade, the presence of mercury in the environment has become a major public concern because, at high levels of exposure, mercury can cause adverse health effects in humans and animals. Specifically, mercury concentrations in fish have been of interest to many groups because of the potential for adverse human health effects. The purpose of this paper is to examine the different sources of mercury in the environment, to review its movement in the environment, and to discuss the potential health effects associated with exposures to mercury.

A review of the scientific literature, in addition to recent findings from international experts on mercury, indicates that the mercury issue is much more complex than is often conveyed. There are, in fact, numerous man-made and natural sources of mercury emissions into the environment. Research scientists, particularly in Sweden, the United States, Canada and Germany, recognizing the unique and complex factors affecting mercury in the environment, have been involved in broad-ranging studies in an attempt to characterize sources of mercury in the environment and its fate and transport once it is released, and to assist in developing methods to minimize mercury emissions from man-made sources.

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While addressing the issues described above, this paper will focus on the role that MWC facilities play.

Forms of Mercury

Mercury occurs in various physical and chemical forms in the environment, which are known as chemical species. The type of mercury species dictates its toxicity and its movement through the environment. There are three general forms of mercury: elemental mercury, inorganic mercury compounds, and organic mercury compounds. Of all mercury forms, organic methylmercury is of greatest concern with respect to human health, aquatic life, and the food chain because of its toxicity and its ability to accumulate as it travels up the food chain.

Elemental mercury (often abbreviated as Hg^0) is usually referred to as mercury vapor, when present in air. It is typically referred to as metallic mercury when present in liquid form, such as in a thermometer (Von Burg and Greenwood 1991). Most of the mercury present in the atmosphere is in the form of elemental mercury vapor (Lindqvist 1991).

Inorganic mercury compounds are formed between the ionic species of mercury, Hg^{2+} (mercuric salts) or Hg^+ (mercurous salts), and molecules or ions with an unshared pair of electrons. Examples of inorganic mercury compounds include mercuric chloride ($HgCl_2$), mercurous chloride (Hg_2Cl_2) and mercuric sulfide (HgS) in cinnabar, the most common natural mercury ore (Von Burg and Greenwood 1991).

Organic mercury compounds can be formed as a result of either bacterial or chemical processes. These compounds include monomethylmercury (CH_3Hg) and dimethylmercury [$(CH_3)_2Hg$]. In lakes, for example, organic mercury compounds can be formed by bacteria present in sediments (Von Burg and Greenwood 1991). Organic mercury compounds have also been widely used as pesticides and preservatives in paint.

Sources of Mercury in the Environment

Mercury in the environment comes from both natural and man-made sources. Although it is difficult to accurately quantify global mercury emissions, it has been estimated that natural sources account for between 40-65% of the total global mercury released annually (NAS 1978, Von Burg and Greenwood 1991, EPRI 1991, Nriagu 1989, Fergusson 1990, Watson 1979, Fitzgerald et al. 1984, Lindqvist 1991). The remaining mercury that is released annually comes from man-made sources.

Mercury emissions from natural sources have been occurring ever since the earth was formed, with some of the greatest releases coming from volcanic eruptions. Man-made emissions have been occurring for centuries, and have included emissions and discharges from gold extraction and mining activities, and more recently (over the past century) agricultural uses and industrial practices. These emissions sources have resulted in an accumulation of mercury in surface soils on land, and in sediment in aquatic systems.

Natural Sources. Natural sources of mercury in the atmosphere are the oceans and the earth's crust and soils. Releases of mercury result from natural degassing of mineral mercury from the earth's crust and oceans; evaporation of mercury from sea spray; geologic weathering and wind erosion of soils; microbial decomposition of surface soils and sediment; and emissions from volcanoes and fires. Estimates of the amounts of mercury in the global environment contributed by natural sources range from 60 tons/year to 17,400 tons/year, with an average of about 5,500 tons/year (Nriagu 1989, Fergusson 1990, Watson 1979, Fitzgerald et al. 1984, USEPA 1992a). An example of the magnitude of natural mercury emissions can be seen from the amount of mercury emitted during a volcanic eruption. It has been estimated that about 250 tons per year of mercury emissions were associated with the recent eruptions of and subsequent offgassing from Mt. Pinatubo.²

Man-Made Sources. Elemental and inorganic mercury compounds are used in the manufacture of many commercial products including thermometers, thermostats, barometers, other pressure-sensing devices, batteries (for hearing aids, cameras, toys, radios, calculators, etc.), energy-efficient and fluorescent lamps (for outdoor lighting, motion picture projection, health treatment, and photography), and dental preparations. Military explosives also contain mercury. Mercury has been used in anti-fouling and mildew-resistant paints, and in agriculture to control fungal diseases. However, the use of mercury-containing fungicides has led to a number of large-scale poisonings, such as the incident in Iraq in 1971, where many people, accidentally exposed to bread made from wheat seed or wheat grown from seed that had been treated with a methylmercurial fungicide, died or developed severe neurological disorders. Mercury is no longer registered for agricultural use (Eisler 1987) or use in indoor paint in the United States, but it is still permitted to be used in outdoor paint, although many manufacturers have voluntarily removed all mercury from paint. Other consumer products from which mercury has been phased out include mirrors, glass, textiles, and paper (USEPA 1992a). The mercury content in consumer batteries has been declining, and is expected to continue decreasing in the near future (USEPA 1992a).

Mercury-containing products are often disposed of as MSW, and are typically landfilled or combusted at MWC facilities. Sources of mercury in the MSW stream

²Dr. B. Siegel, School of Public Health, University of Hawaii. Personal communication, October 30, 1992.

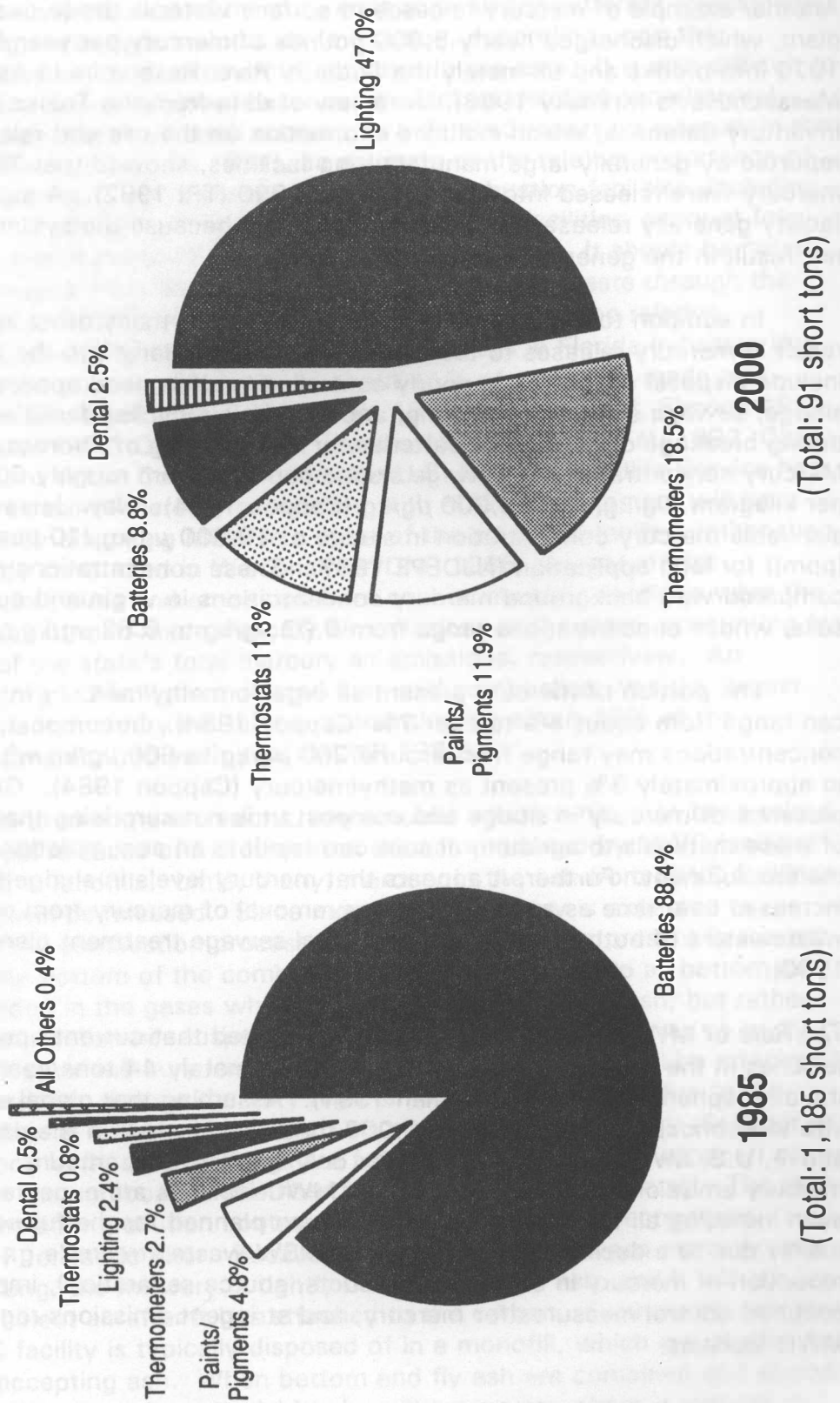
are provided in Figure 1. As shown in Figure 1, batteries accounted for most of the nearly 1,200 tons of mercury in the U.S. waste stream in 1985. Between 1985 and 2000, the total amount of mercury in the waste stream is expected to decrease to approximately 90 tons per year, primarily due to a decrease in mercury from consumer batteries. For all other mercury sources, with the exception of light bulbs (primarily fluorescent and energy-efficient light bulbs that contain higher concentrations of mercury than normal light bulbs), the total amount of mercury for each source will also decrease. This is largely due to the removal or reduction of mercury in consumer products, recycling, and source separation of mercury-containing materials.

Even though mercury emissions are decreasing, mercury is still present in MSW, and each method of MSW management results in some form of mercury release. For example, mercury associated with MSW can be emitted from materials recovery operations (including those that recycle mercury), from the stack of an MWC facility, through landfill gas emissions, or from leachate released from an MSW landfill. Mercury emissions can also occur from landfills containing industrial and hazardous waste.

Man-made, airborne sources of mercury have been estimated to contribute from 2,200 tons/year to 8,300 tons/year into the earth's atmosphere, with an average of about 4,400 tons/year (Nriagu 1989, Watson 1979, Lindberg 1987, Lindqvist 1991). The two types of man-made sources of mercury to the atmosphere are point sources (i.e., distinct sources) and diffuse sources (i.e., widely scattered sources). Point sources include chlor-alkali facilities (which produce chlorine and caustic soda), utilities that burn fossil fuels (i.e., coal, oil, and natural gas) or peat, metal smelters, recycling facilities, geothermal plants (Ferrara et al. 1992), mining drainage, and sewage and waste combustion plants. Emissions from diffuse sources include landfills, dental activities, hospital and chemical laboratories, production of electrical and electronic equipment, agricultural burning, and paint applications (Lindqvist 1991). On a global scale, Lindqvist (1991) estimates that roughly one fifth of the mercury emitted from man-made sources is from diffuse sources, while approximately four fifths is emitted from point sources.

There are other important man-made sources of mercury in the environment besides airborne releases, including discharges of mercury-containing wastewater into surface water. Continuous discharges of mercury into water bodies, even from decades ago, can result in a persistent source of contamination for aquatic life. Health impacts caused by eating fish containing high levels of mercury first became known after a poisoning outbreak in Minamata Bay, Japan in the 1950's (WHO 1976). This epidemic was caused by many years of industrial releases of metallic mercury, which was used as a catalyst at an acetaldehyde plant near Minamata Bay, a small fishing bay used by subsistence fishermen. The metallic mercury was discharged into the bay as waste sludge, and was converted to organic mercury compounds, especially methylmercury (Klaasen et al. 1986).

FIGURE 1
SOURCES OF MERCURY IN MUNICIPAL SOLID WASTE
IN 1985 AND 2000 (a)



(a) Data obtained from USEPA (1992a) and Malcolm Pirnie (1992)

Another example of mercury releases to surface water is the Nyanza manufacturing plant, which discharged nearly 5,000 pounds of mercury per year from 1940 to 1970 into brooks and ultimately the Sudbury River Reservoirs in Ashland, Massachusetts (Krimsky 1988). A review of data from the Toxics Release Inventory database, which includes information on the use and release of chemicals reported by generally large manufacturing facilities, showed that 751 pounds of mercury were released into U.S. waters in 1990 (TRI 1992). A modern MWC facility generally releases no mercury into water because the system design does not result in the generation of wastewater.

In addition to industrial discharges of mercury, many other activities can result in mercury releases to the environment, particularly into the air. These include disposal of wastes in poorly controlled landfills, land application of sewage sludge, sewage sludge composting, and materials recycling facilities (e.g., releases during breakage or storage of batteries, or the crushing of fluorescent light bulbs). Mercury concentrations in sewage sludge can vary from roughly 600 micrograms per kilogram ($\mu\text{g}/\text{kg}$) to 100,000 $\mu\text{g}/\text{kg}$ (Cappon 1984). New Jersey has an allowable mercury concentration in sludge of 10,000 $\mu\text{g}/\text{kg}$ (10 parts per million [ppm]) for land application (NJDEPE 1987). These concentrations may be compared with background mercury concentrations in virgin and cultivated U.S. soils, whose concentrations range from 0.02 $\mu\text{g}/\text{kg}$ to 0.63 $\mu\text{g}/\text{kg}$ (ATSDR 1989).

The portion of mercury present as organic methylmercury in sewage sludge can range from about 1% to over 7% (Cappon 1984). In compost, mercury concentrations may range from around 200 $\mu\text{g}/\text{kg}$ to 600 $\mu\text{g}/\text{kg}$ with less than 1% to approximately 3% present as methylmercury (Cappon 1984). Given the presence of mercury in sludge and compost, it is not surprising that the application of these materials to agricultural soils can result in an accumulation of mercury in the environment. Further, it appears that mercury levels in sludge residues have increased over time as a result of better removal of mercury from process wastewaters of both industry and municipal sewage treatment plants (USEPA 1990).

The Role of MWC Facilities. It has been estimated that current operating MWC facilities in the United States contribute approximately 44 tons/year of mercury into the atmosphere (Kiser and Sussman 1991). Assuming that global mercury releases into the atmosphere are roughly 10,000 tons/year, based on the data summarized above, U.S. MWC facilities account for only 0.4% of this amount. Overall, mercury emissions associated with U.S. MWC facilities are expected to decrease, even including all the facilities that have been planned for the future. This is mainly due to a decline of mercury in the MSW waste stream (e.g., from a reduction of mercury in consumer products, source separation), improved air pollution control measures for mercury, and stringent emissions regulations for MWC facilities.

The relative contribution of mercury emissions among different atmospheric sources varies significantly from location to location, depending upon the distribution of industries and commercial activities in the area. It is also difficult to accurately quantify this breakdown because of a lack of detailed experimental emissions measurements from each source. In a detailed report on mercury in the Swedish environment (Lindqvist 1991), an estimate of the relative importance of emission point sources in 1987 shows that waste combustion facilities, including those that burn hazardous and medical waste, and MWC facilities, account for roughly 15% of total man-made Swedish mercury emissions. It should be noted that Sweden manages more than 50% of its municipal solid waste through the combustion process (SAPCSWM 1988). A rough estimate of the relative importance of various airborne mercury emission sources in Florida indicates that MWC facilities account for a maximum of about 11% of total man-made and natural emissions combined (KBN 1992, Coale 1992, Snyder 1992, Simons 1991, Walker and Cooper 1992, Ogden 1992a, Ferraro 1992, Patrick et al. 1992, Oke 1978). In addition, in Lee County, Florida, the U.S. Fish and Wildlife Service has stated that the release of mercury associated with MSW management will be reduced by 70% by disposing MSW in a state-of-the-art MWC facility, rather than a landfill. A study conducted in Wisconsin (WDNR 1986) estimated that application of latex paints and emissions from coal combustion facilities were the two major sources of man-made mercury air emissions in the state, accounting for 48% and 32% of the state's total mercury air emissions, respectively. An emissions inventory in Michigan indicated that coal combustion was the largest source of man-made mercury emissions, accounting for about 42% of the estimated 15.9 tons emitted each year (MDNR 1989).

In addition to airborne emissions, some public concern has also been raised regarding potential releases of mercury from the ash produced by MWC facilities that is deposited in landfills. Little, if any, mercury in the ash from MWC facilities that is landfilled will be released. Since most of the mercury is expected to volatilize during the combustion process, little or no mercury is present in the ash that falls from the bottom of the combustion chamber (referred to as bottom ash). Particles suspended in the gases which do not fall out in bottom ash, but rather exit the combustion chamber suspended in exhaust gases, are referred to as fly ash. Some fraction of mercury leaving the combustion chamber will be attached to these fly ash particles, despite the very high temperatures. This is due to the presence of carbon in the particles, which strongly binds the mercury (Schager et al. 1992). Carbon in fly ash may originate from the MSW (e.g., newspaper) or may be injected after the combustion chamber for mercury pollution control. The fly ash is then collected in pollution control equipment such as an electrostatic precipitator (ESP) or fabric filter. Because the association between mercury and carbon is so strong, the mercury is tightly bound to the fly ash, and it will not be released into ambient air after it has attached to the carbon. Further, the ash from a modern MWC facility is typically disposed of in a monofill, which is a special type of landfill only accepting ash. When bottom and fly ash are combined and placed in an ash monofill, where the material hardens like concrete, there is virtually no

opportunity for either the leaching of mercury to water or the volatilization of mercury into air. This is also the case for many co-disposal sites, where ash and raw trash are separated, but disposed at the same location. In contrast, mercury captured in ash from other combustion sources that is not disposed of in a specially designed monofill may be subsequently released into the environment.

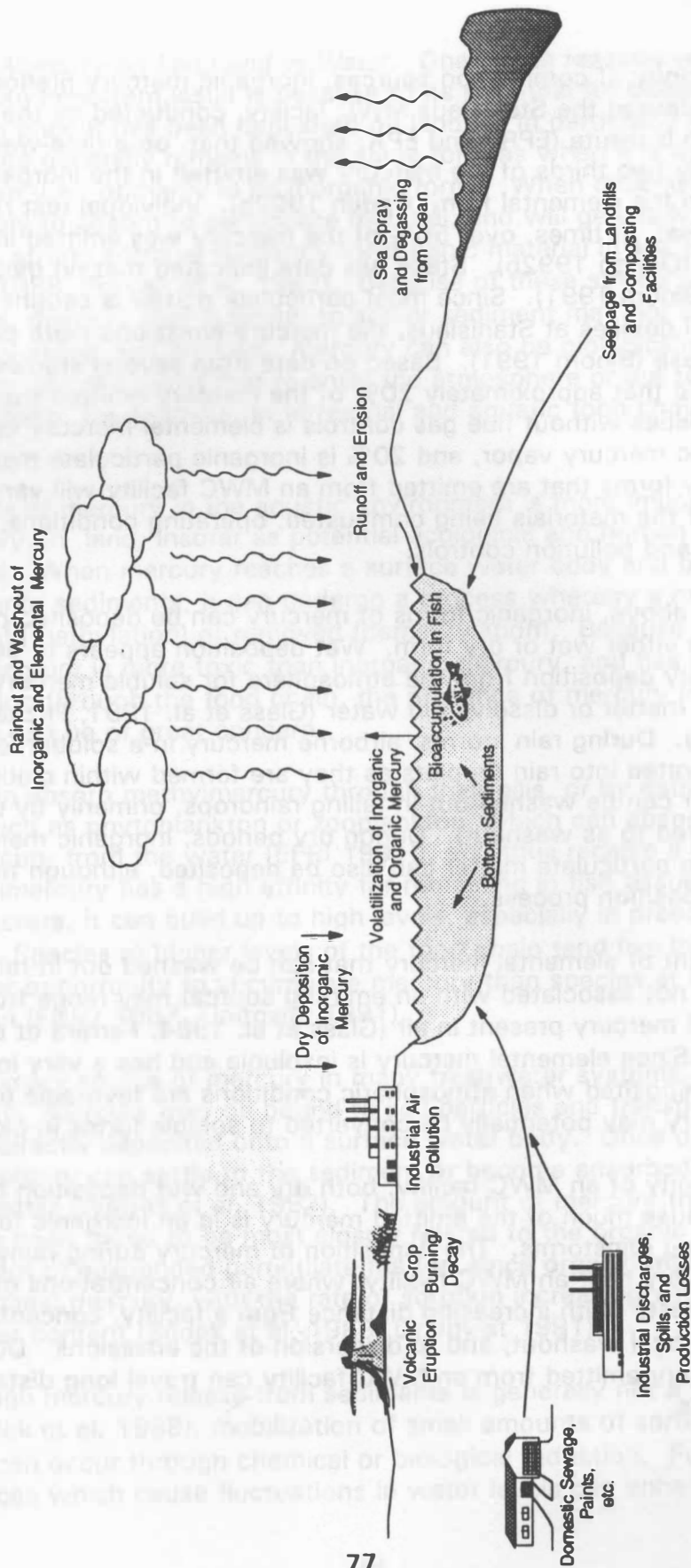
For the reasons presented above, including a decline of mercury in the waste stream and better mercury control measures at modern MWC facilities, mercury emissions associated with these facilities are not a significant source of total mercury in the environment today. Further, it should be noted that there is no conclusive correlation establishing a link between mercury emissions associated with modern MWC facilities and the accumulation of mercury in fish. In contrast, direct correlations have been established between elevated fish tissue concentrations and discrete point sources (e.g., industrial discharge, releases due to mobilization of mercury in areas with significant peat deposits). Although many states do have fish advisories due to the presence of mercury in fish above levels of concern, there is no evidence that modern MWC facilities are responsible for the elevated fish tissue levels.

Mercury Movement Through the Environment

There is a natural cycling of mercury in the environment. It is characterized by emissions of mercury from both natural sources (e.g., oceans, soils, volcanoes, forest fires, and the earth's crust) and man-made sources (e.g., industry) into the atmosphere, followed by atmospheric transport, then the deposition of mercury back on land and surface water, and finally the adsorption of mercury compounds to soil or sediment particles. Mercury cycling in hydrological systems is characterized by runoff into surface water bodies, subsequent settling onto the bottom of the water body, and to a lesser degree, volatilization. Bioaccumulation of mercury at increasingly higher levels of the foodchain may be important in these surface water bodies, especially for predatory species that consume smaller species that have been exposed to mercury. Figure 2 illustrates the movement of mercury through the environment.

Atmospheric Transport and Deposition of Mercury. The main form of mercury in the atmosphere is elemental (Lindqvist 1991). Because elemental mercury is insoluble, it remains in the atmosphere for a long period of time (i.e., it has a long residence time), whereas the inorganic form of mercury has a much shorter residence time because it more readily dissolves into rain or snow and can thereby be deposited onto land or incorporated into cloud droplets. Inorganic mercury accounts for a small fraction of the total mercury in the atmosphere -- no more than 5%. It is also possible that a very small fraction of airborne mercury may be organic methylmercury (Lindqvist 1991).

FIGURE 2 MOVEMENT OF MERCURY THROUGH THE ENVIRONMENT



In the vicinity of combustion sources, inorganic mercury predominates in air. Recent testing data at the Stanislaus MWC facility, conducted by the Electric Power Research Institute (EPRI) and EPA, showed that, on a time-weighted average, roughly two thirds of the mercury was emitted in the inorganic form, with the remainder in the elemental form (Ogden 1992b). Individual test runs demonstrated that, at times, over 50% of the mercury was emitted in the elemental form (Ogden 1992b). Stack gas data indicated that no methylmercury was detected (Bloom 1991). Since most particulate matter is caught in the air pollution control devices at Stanislaus, the mercury emissions were predominantly in the vapor phase (Bloom 1991). Based on data from several studies, Lindqvist (1991) estimated that approximately 20% of the mercury emitted from waste combustion facilities without flue gas controls is elemental mercury vapor, about 60% is inorganic mercury vapor, and 20% is inorganic particulate mercury. The specific mercury forms that are emitted from an MWC facility will vary, depending on the nature of the materials being combusted, operating conditions, flue gas characteristics, and pollution controls.

As noted above, inorganic forms of mercury can be deposited onto land and surface water in either wet or dry form. Wet deposition appears to be the principal means of mercury deposition from the atmosphere for soluble mercury adsorbed onto particulate matter or dissolved in water (Glass et al. 1991, Fitzgerald 1986, Matheson 1979). During rain storms, airborne mercury in a soluble form can become incorporated into rain droplets as they are formed within clouds (referred to as rainout), or can be washed out in falling raindrops, primarily by dissolving into the water (referred to as washout). During dry periods, inorganic mercury that is incorporated into particulate matter can also be deposited, although this is not the predominant deposition process.

The amount of elemental mercury that can be washed out in rainfall events (mercury that is not associated with an emitting source) may range from 1% to 20% of the total mercury present in air (Glass et al. 1984, Ferrara et al. 1986, Brosset 1987). Since elemental mercury is insoluble and has a very long residence time, it is only deposited when atmospheric conditions are favorable (e.g., insoluble elemental mercury may potentially be converted to soluble forms in cloud droplets).

In the vicinity of an MWC facility, both dry and wet deposition of mercury can occur. Because much of the emitted mercury is in an inorganic form, it can be washed out during rainstorms. The deposition of mercury during rainy periods is most important very near an MWC facility, where air concentrations of emitted mercury are highest. With increasing distance from a facility, concentrations decrease due to local washout, and to dispersion of the emissions. During dry conditions, mercury emitted from an MWC facility can travel long distances before deposition occurs.

Movement of Mercury on Land and in Water. One of the reasons why mercury has become such an important issue is because of its potential for bioaccumulation in the food chain once it has been deposited on land, or in particular, in water. Mercury on land generally remains in the same form as when it is deposited from the atmosphere (i.e., primarily in its inorganic form). When deposited on land, the mercury is likely to adsorb to particulate material, and will generally remain as such because of its strong sorptive tendency for organic material in soil and sediment (Bodek et al. 1988, Porvari et al. 1992). Because of these strong sorptive characteristics, mercury that is bound to soil or sediment may not be available to be taken up by plants and animals. Mercury can also be biologically converted into more volatile or soluble forms that re-enter the atmosphere or are taken up by plants and animals, for example, in terrestrial and aquatic food chains (USEPA 1984).

The fate of mercury in the aquatic environment is more important than the fate of mercury on land, insofar as potential ecological and human health impacts are concerned. When mercury reaches a surface water body and becomes incorporated into sediments, it can undergo a process whereby a methyl group (CH_3) is added (methylation) or removed (demethylation). Because organic methylated mercury is more toxic than inorganic mercury, and has the potential for bioaccumulation through the food chain, the presence of mercury in aquatic environments can be of great concern.

Fish can absorb methylmercury through their gills, or by eating smaller organisms (such as phytoplankton or zooplankton, which can absorb high amounts of methylmercury from the water [EPRI 1991]), and thus receive a concentrated dose. Methylmercury has a high affinity for remaining in fish tissue, and since it is difficult to excrete, it can build up to high levels, especially in predatory organisms (EPRI 1991). Species at higher levels of the food chain tend live longer and thus have a greater opportunity to accumulate mercury than species at lower levels of the food chain (Eisler 1987, Lindqvist 1991).

The primary source of mercury in many freshwater systems is in the inorganic form. Mercury may be bound to soil particles and transported via runoff, or it may be directly deposited onto a surface water body. Once deposited in water, the mercury can settle to the sediment or become adsorbed to suspended particulate matter (Watras et al. 1992). The amount of mercury that becomes adsorbed has been found to be most closely related to the organic matter content of the sediment or suspended particulate matter, since organic matter readily adsorbs inorganic mercury, with the rate of sorption increasing with increasing organic matter content (Bodek et al. 1988, Lindqvist 1991).

Although mercury release from sediments is generally not a predominant process (Bodek et al. 1988), mobilization of small amounts of sorbed mercury from particulates can occur through chemical or biological reduction. Further, in some areas, practices which cause fluctuations in water levels can enhance the

movement of mercury. This is particularly true in South Florida, where dredge and fill operations, lake drawdowns, and flood control practices have apparently resulted in an increased mobilization of mercury, especially in areas with significant peat deposits (FDER 1990a).

Inorganic mercury that is present in surface waters can be converted to methylmercury under favorable conditions. Methylation can occur under both aerobic (with oxygen) and anaerobic (without oxygen) conditions, although this process seems to occur to a greater extent under anaerobic conditions, and primarily through chemical processes (ICMGP 1992). Demethylation appears to be a predominantly biological process (ICMGP 1992). The rate of methylmercury formation depends on many interrelated factors, including mercury loadings, the amount and type of bacteria in sediments, sediment nutrient content, pH, reduction-oxidation conditions, suspended particulate matter levels, and the rate of particulate matter sedimentation (Eisler 1987). In Southern Florida, where draining and flooding of land occurs frequently, mercury methylation rates have been shown to increase in newly flooded impoundments, followed by an increased rate of uptake into the food chain (FDER 1990a).

Ultimately, a level is reached in most water bodies referred to as "steady-state" in which the rates of methylation and demethylation are balanced. In general, this steady-state level of methylmercury in sediment is on the order of about 1%, but can typically range from less than 0.01% to as high as 18% (Von Burg and Greenwood 1991, Lindqvist 1991, EPRI 1987, Stary et al. 1980, Jackson 1986). Under some unique circumstances, higher proportions of methylmercury can occur (Eganhouse 1975, Craig and Moreton 1983).

Trends of Atmospheric Mercury Emissions from Man-Made Sources

Federal and State Emissions Legislation. Although there are numerous man-made sources of mercury emissions, most regulatory agencies have primarily focused on controlling mercury emissions from MWC facilities, and have spent little effort regulating emissions from other man-made sources. As noted above, mercury emissions from fossil fuel combustion and smelting can contribute significantly to overall mercury emissions in the environment. Electric utilities, in particular coal burning plants, are the largest man-made mercury emission source globally. In the absence of mercury emission reduction methods, mercury from electric utilities is projected to grow by 10-35% over the next two decades, and will likely account for over half of the U.S. man-made mercury emissions (CCAP 1991). To date, however, the USEPA and state regulatory agencies have not adopted requirements for the control of mercury emissions from these plants. Under the new Clean Air Act Amendments (CAAA) of 1990, USEPA is required to set emission standards for most major stationary sources of over 180 toxic air pollutants, including mercury. However, under the CAAA, Congress is only required to review in 1993 a USEPA study, involving the status of emissions regulations and control technology for electric utility steam generating facilities that burn fossil fuels, in order to determine

the need for additional controls. Although MWC facilities and electric utilities are specifically addressed in the CAAA, other important man-made sources of atmospheric mercury, including smelters and sewage sludge incinerators are not specifically mentioned.

The existing and anticipated regulation of mercury for MWC facilities stands in contrast to that for other man-made emission sources. The USEPA New Source Performance Standards (NSPS), which will specify standards as required by the 1990 CAAA, are expected to be proposed in late 1992 and promulgated one year later. These are expected to impose a stringent stack gas concentration limit (based on actual test results) that would be consistently achieved, resulting in the requirement for the use of mercury control methods at all new MWC facilities with a capacity exceeding 250 tons/day. USEPA is also developing emission guidelines for existing MWC facilities, emission limits for new medical waste combustors and emission guidelines for existing medical waste combustors (USEPA 1992b). In addition to USEPA's upcoming CAAA regulations that are expected to specifically address mercury emissions from MWC facilities, many states are beginning to require increasingly stringent mercury controls on these plants. The State of Florida, because of its concerns regarding mercury in the environment, is considering establishing stringent mercury emission limits for a number of stationary air pollution sources (FDER 1992). For example, the State of Florida recently imposed an emission limit of 140 $\mu\text{g}/\text{dry standard cubic meter (dscm)}$ for the MWC facility in Lee County, Florida. The State of New Jersey is also in the process of developing specific mercury regulations for MWC facilities.

Measures for Controlling Mercury Emissions. Increasingly effective mercury pollution controls have been considered for use at WTE facilities to achieve greater reductions in mercury emissions. Two types of state-of-the-art mercury controls at MWC facilities that have been proposed for use in the United States are the injection of either powdered activated carbon or sodium sulfide into the flue gas stream before the air pollution control system. When activated carbon is used, mercury bonds with the carbon and is collected in the pollution control equipment. Studies with activated carbon at MWC facilities, primarily in Europe³, have shown that from 50% to 90% of the mercury in the gases can be removed (Malcolm Pirnie et al. 1992, USEPA 1992a). Recent studies at the Stanislaus, California MWC facility, whose pollution controls include a dry scrubber and fabric filter, indicate that injection of activated carbon is effective in controlling from 50% to 95% of its mercury emissions (Malcolm Pirnie et al. 1992). Recent test results from New Jersey's Camden County MWC facility using injection of powdered activated carbon, in addition to a dry scrubber and ESP, indicate a similar mercury capture

³It should be noted, however, that differences exist between U.S. and European MWC facilities, including process conditions during testing (i.e., flows, waste feeds, etc.), differences in standard or reference conditions (temperature, pressure, percent oxygen or carbon dioxide, etc.), and differences in the test methodologies. Therefore, care should be exercised when comparing test results (Malcolm Pirnie et al. 1992).

efficiency. However, more carbon was required to achieve the same mercury capture efficiency, due to the type of existing control technology (i.e., ESP).

Using the reagent sodium sulfide, the mercury is converted to solid mercuric sulfide (HgS) in the flue gas, which is subsequently collected in the air pollution control equipment. Studies with sodium sulfide injection at several European MWC facilities have shown a similar effectiveness for controlling mercury emissions, ranging from 60% to 90%. Compared to activated carbon, however, sodium sulfide injection systems have shown a somewhat lower effectiveness for controlling emissions, and they can experience more operational problems (Malcolm Pirnie et al. 1992).

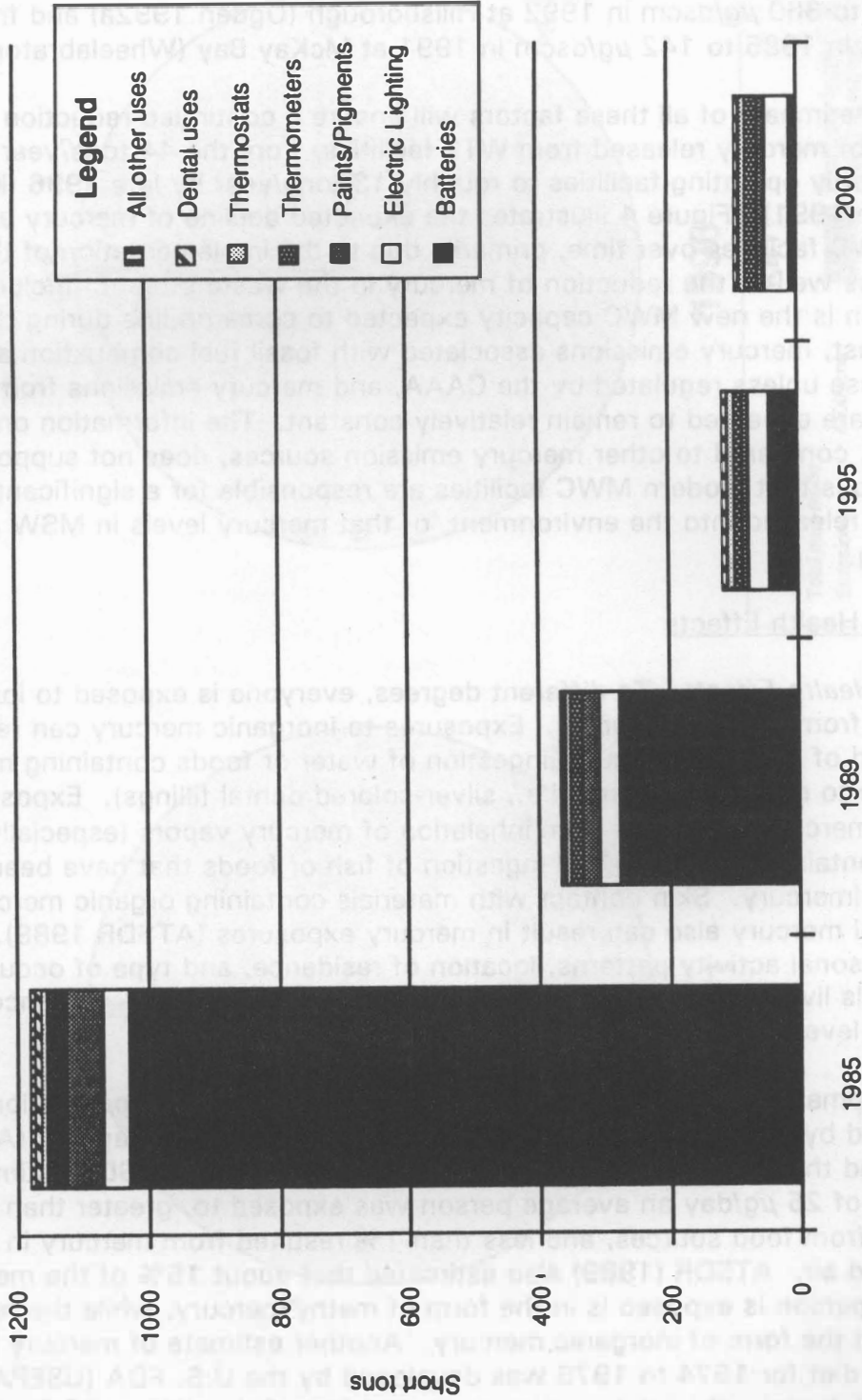
Trends of Mercury in the Waste Stream. In addition to a decline in mercury emissions due to increasingly stringent emissions limits, mercury emissions from MWC facilities are declining due to a phasing out of mercury use in consumer products, and a decrease in the presence of mercury in the MSW stream in general (see Figure 3). The principal source of mercury in MSW treated in MWC facilities is from commercial products such as household batteries, electrical lighting equipment, paints and pigments, and thermostats. Historically, batteries have accounted for most of the mercury in the waste stream (see Figure 1 for a breakdown of mercury in the waste stream). An estimate for 1985 indicates that 88% of the mercury in U.S. MSW resulted from household batteries, with the total amount of mercury in the waste stream amounting to roughly 1,200 tons (USEPA 1992a, Malcolm Pirnie et al. 1992).

Due to the decrease in mercury from consumer batteries, by the year 2000, batteries are estimated to account for only 9% of the MSW, with a total of roughly 90 tons of mercury in the waste stream (USEPA 1992a, Malcolm Pirnie et al. 1992). This translates to a 90% reduction in the amount of mercury in MSW in only 15 years, largely due to source separation of batteries, recycling of mercury-containing materials, and the discontinuation or reduction of the use of mercury in batteries, indoor paints, special paper coatings, film packs, and other commercial products. The use of mercury in paints, for example, is estimated to decline by 35% in the U.S. between 1980 and 1989 (CCAP 1991). In the State of New Jersey, the Dry Cell Battery Management Act and Toxic Packaging Reduction Act is estimated to result in a greater than 80% reduction of mercury in MSW by 1995. In contrast, however, some sources of mercury are expected to increase in the future, including the energy-efficient fluorescent light bulbs that contain higher concentrations of mercury than normal light bulbs, for which sales are likely to increase in the future. Although the percent contribution of remaining mercury sources may increase from 1985 to 2000 (as shown in Figure 1), the total amount for each source (in tons) will decrease.

Not surprisingly, the decline in mercury in MSW and the implementation of mercury emission controls has already had a significant impact on mercury emissions from MWC facilities. The effect of reducing the amount of mercury in

FIGURE 3

THE DECLINE OF MERCURY IN MUNICIPAL SOLID WASTE (a)



(a) Data obtained from USEPA (1992a) and Malcolm Pirnie (1992).

the waste stream alone can be illustrated by examining mercury stack gas concentrations measured at the Hillsborough and McKay Bay MWC facilities in Florida. Without the implementation of any specific source separation or mercury emission control programs, stack gas mercury levels decreased from 840 $\mu\text{g}/\text{dscm}$ in 1987 to 360 $\mu\text{g}/\text{dscm}$ in 1992 at Hillsborough (Ogden 1992a) and from 973 $\mu\text{g}/\text{dscm}$ in 1985 to 142 $\mu\text{g}/\text{dscm}$ in 1991 at McKay Bay (Wheelabrator 1992).

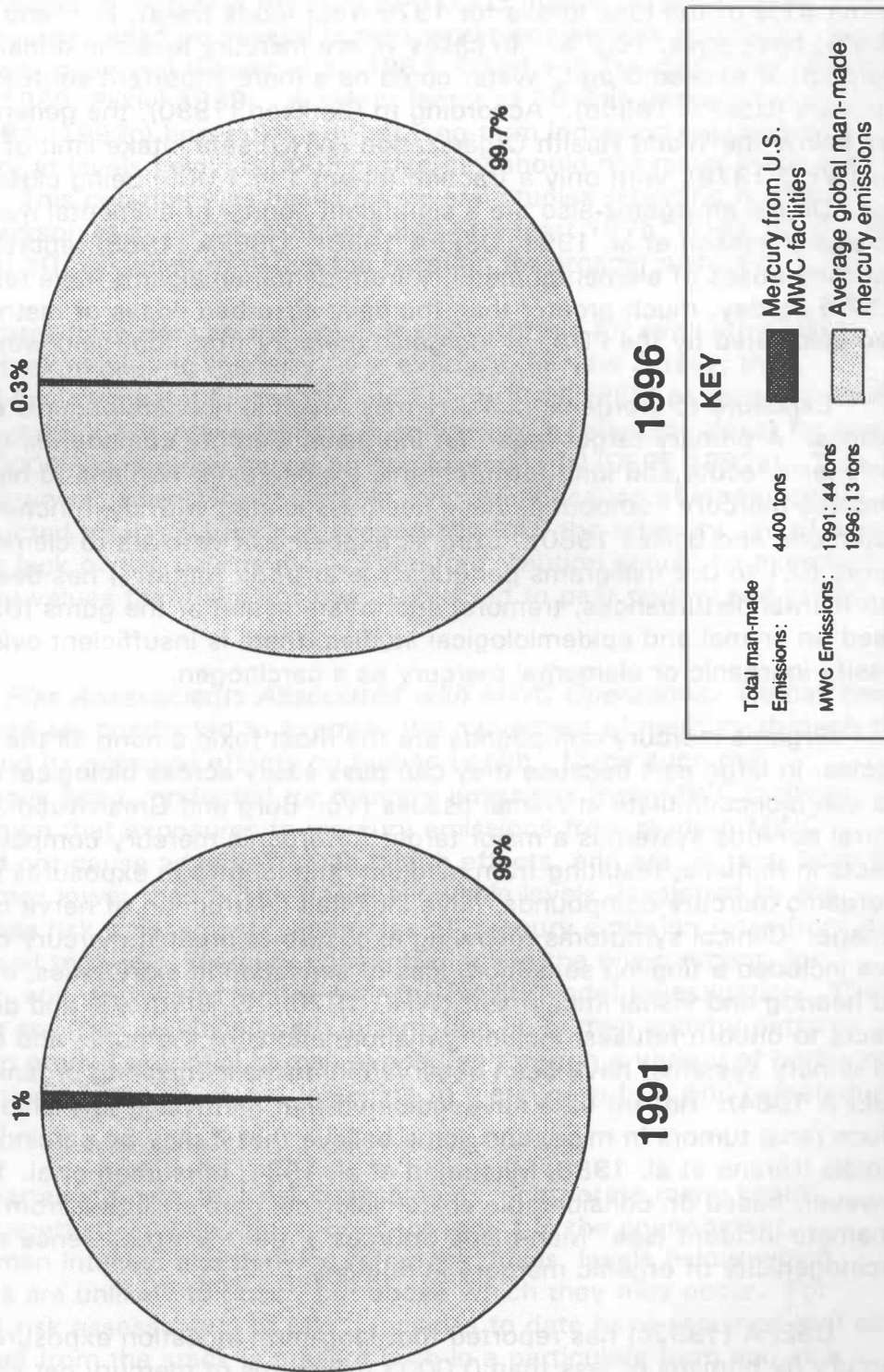
The impact of all these factors will ensure a continued reduction in the amount of mercury released from WTE facilities, from the 44 tons/year estimated for currently operating facilities to roughly 13 tons/year by late 1996 (Kiser and Sussman 1991). Figure 4 illustrates the expected decline of mercury emissions from MWC facilities over time, primarily due to the implementation of the 1990 CAAA, as well as the reduction of mercury in the waste stream. Included in this projection is the new MWC capacity expected to come on line during this period. In contrast, mercury emissions associated with fossil fuel combustion are expected to increase unless regulated by the CAAA, and mercury emissions from natural sources are expected to remain relatively constant. The information on MWC facilities, compared to other mercury emission sources, does not support the contentions that modern MWC facilities are responsible for a significant portion of mercury released into the environment, or that mercury levels in MSW are not changing.

Mercury Health Effects

Human Health Effects. To different degrees, everyone is exposed to low levels of mercury from different sources. Exposures to inorganic mercury can result from inhalation of airborne mercury, ingestion of water or foods containing mercury, and exposure to dental amalgams (i.e., silver-colored dental fillings). Exposures to organic mercury can result from inhalation of mercury vapors (especially from paints containing mercury) and ingestion of fish or foods that have been exposed to methylmercury. Skin contact with materials containing organic mercury and elemental mercury also can result in mercury exposures (ATSDR 1989). Depending upon personal activity patterns, location of residence, and type of occupation, even individuals living in the same household or neighborhood can experience very different levels of mercury exposures (USEPA 1985a).

Estimates of the intake of mercury for the general U.S. population have been developed by the Agency for Toxic Substances and Disease Registry (ATSDR 1989) and the U.S. Food and Drug Administration (FDA). ATSDR estimated that of the total of 25 $\mu\text{g}/\text{day}$ an average person was exposed to, greater than 99% resulted from food sources, and less than 1% resulted from mercury in drinking water and air. ATSDR (1989) also estimated that about 15% of the mercury to which a person is exposed is in the form of methylmercury, while the remaining 85% is in the form of inorganic mercury. Another estimate of mercury intakes from the diet for 1974 to 1979 was developed by the U.S. FDA (USEPA 1985b). For an adult male, the intake estimate ranged from 2.4 $\mu\text{g}/\text{day}$ to 6.5 $\mu\text{g}/\text{day}$, with

FIGURE 4
THE RELATIVE CONTRIBUTION OF MERCURY EMISSIONS
FROM MWC FACILITIES IN THE U.S. (a)



Source for MWC emissions: Kiser & Sussman 1991.
 Source for global man-made mercury emissions: Nriagu 1989, Watson 1979, Linberg 1987, Lindqvist 1991.

almost 91% of the total intake for 1979 from foods (meat, fish, and poultry, 73.6%; beverages, 16.9%). In cases where mercury levels in drinking water approach or exceed 5 $\mu\text{g/L}$, water could be a more important source of mercury exposure (USEPA 1985b). According to Clarkson (1990), the general population is well below the World Health Organization (WHO) safe intake limit of 0.71 $\mu\text{g/kg-day}$ (WHO 1976), with only a fraction (about 1 in 1,000) being close to the safe limit. Dental amalgams also are a significant source of elemental mercury to humans (Akesson et al. 1991, USEPA 1988). USEPA (1988) reported that daily absorbed doses of elemental mercury from dental amalgams have ranged from 2.9 to 17.5 $\mu\text{g/day}$, much greater than the daily absorbed doses of methylmercury from food calculated by the FDA, or inorganic mercury from food and water.

Exposure to inorganic mercury may result in noncarcinogenic effects in humans. A primary target organ for inorganic mercury compounds is the kidney. Short-term, acute and long-term, chronic exposures of humans to high levels of inorganic mercury compounds have been associated with dysfunction of the kidney (Hammond and Beliles 1980). Chronic exposure of workers to elemental mercury vapors (0.1 to 0.2 milligrams per cubic meter of air [mg/m^3]) has been associated with mental disturbances, tremors, and inflammation of the gums (USEPA 1984). Based on animal and epidemiological studies, there is insufficient evidence to classify inorganic or elemental mercury as a carcinogen.

Organic mercury compounds are the most toxic among all the mercury species, in large part because they can pass easily across biological membranes and can bioaccumulate in animal tissues (Von Burg and Greenwood 1991). The central nervous system is a major target for organic mercury compounds. Adverse effects in humans, resulting from medium- and long-term exposures to high levels of organic mercury compounds, have included destruction of nerve cells and brain damage. Clinical symptoms following exposure to organic mercury compounds have included a tingling sensation, loss of sensation in extremities, incoordination, and hearing and visual impairment (WHO 1976). Embryotoxic and developmental effects to unborn fetuses, including malformations of the bone, and reproductive and urinary systems, have been observed in animals ingesting organic mercury (USEPA 1984). Recent data have indicated that methylmercury chloride can induce renal tumors in mice, and some believe that it may be carcinogenic to animals (Hirano et al. 1986, Mitsumori et al. 1981, Mitsumori et al. 1990). However, based on considerable epidemiological data collected from victims of the Minamata incident (see "Man-made Sources"), there is no evidence of carcinogenicity of organic mercury in humans.

USEPA (1992c) has reported that long-term ingestion exposures to inorganic mercury by humans of less than 0.0003 milligrams of mercury per kilogram body weight per day (mg/kg-day) would not result in adverse health effects. This criterion, which includes a safety factor of 1,000, was based on several studies conducted with rats in which kidney effects were observed (Andres 1984, Druet et al. 1978, Bernaudin et al. 1981). USEPA (1992c) has also derived a criterion for

inhalation exposures to inorganic mercury of 0.0003 mg/m³ for both medium- and long-term exposures based on several human workplace studies in which neurotoxicity was observed (Fawer et al. 1983, Piikivi and Tolonen 1989, Piikivi and Hanninen 1989, Piikivi 1989). A safety factor of 30 was used to derive this criterion. USEPA (1992c) has indicated that long-term ingestion exposures to organic mercury at levels below 0.0003 mg/kg-day should not result in adverse health effects. This criterion was based on several studies reporting human poisonings (Clarkson et al. 1976, Nordberg and Strangert 1975, WHO 1976). A safety factor of 10 was used to derive the criterion for organic mercury.

Some states have derived their own toxicity criteria for evaluating risks associated with exposures to mercury. For example, in New Jersey, the Department of Environmental Protection and Energy (NJDEPE) has recently issued a proposed acceptable daily intake (similar in concept to a reference dose) for organic mercury of 0.00007 mg/kg-day based on fish ingestion (NJDEPE 1992a). This value is not, however, scientifically credible, primarily because of inappropriate research conducted by the NJDEPE to support the RfD, the arbitrary use of a safety factor, and the lack of any peer review. Therefore, caution should be exercised when using any values that have not been subjected to peer review and public comment.

Human Health Risk Assessments Associated with MWC Operations. Human health risk assessments are conducted to examine the movement of mercury through the environment and its potential effects on human health. Many such risk assessments have been conducted for mercury emissions from MWC facilities; these have shown that exposures to mercury emissions from modern MWC facilities would not cause adverse human health effects, and are, in fact, from 10 to over 100 times lower than health-based allowable levels developed by the USEPA. In these risk assessments, estimates of mercury emission rates from the facilities are used to predict mercury concentrations in the environment, for example, in air, soil, and water bodies near the facility under investigation. These concentrations are then combined with information on human activity patterns near each facility, to predict potential human exposures through a variety of pathways, such as inhalation of mercury in air, ingestion of mercury in fish, and ingestion of mercury in soil.

It is important to note that risk assessments incorporate many health protective assumptions in determining concentrations in the environment, calculating human intakes, and deriving reference doses, levels below which adverse effects are unlikely to occur, but above which they may occur. For example, most risk assessments of MWC facilities to date have assumed that all mercury emitted from the stack is present both in a particulate form and as a vapor, thereby double-counting mercury concentrations in the environment. Risk assessments also often assume that emitted mercury accumulates for a period of 70 years (as opposed to a more realistic facility lifespan of 20-40 years). Further, the reference doses used in risk assessments are health protective due to the use

of safety factors. In a risk assessment, the use of health protective approaches results in an overestimation of risks, meaning that potential health impacts would actually be less than those that are predicted.

For example, in human health risk assessments for several proposed and existing MWC facilities, fish tissue concentrations associated with mercury emissions were calculated. The concentrations of total mercury were 0.0006-0.01 mg/kg (Clement 1989, 1990a, 1990b). Mercury concentrations in fish inhabiting remote U.S. lakes, or lakes considered to be unpolluted, have ranged from 0.04-2.7 mg/kg (Rudd et al. 1980, FDER 1990b), while mercury concentrations in fish inhabiting polluted lakes have ranged from 0.26-5.1 mg/kg (Renfro et al. 1974, Rudd et al. 1980). A comparison of the measured and predicted fish tissue concentrations shows that concentrations of mercury associated with emissions from a modern MWC facility are lower than even those in fish inhabiting unpolluted surface water bodies. Each of the predicted fish tissue concentrations was also lower than the U.S. Food and Drug Administration (FDA) action level of 1 mg/kg for mercury in fish intended for human consumption (USFDA 1984a,b). Further, the estimated exposures resulting from fish ingestion were at least 100 times lower than the reference doses, indicating that adverse health effects would be unlikely to occur.

The impact of mercury emissions from MWC facilities can also be assessed by examining the resulting concentrations in soil. Many health protective assumptions are used to calculate these concentrations, including the assumption that all the emitted mercury from the stack is in a particulate form, available for deposition (whereas in reality a significant portion is in the vapor form, and will likely remain airborne). Further, it is often assumed that mercury emitted from the MWC facility will be deposited for 70 years, and retained in the top 1 centimeter of soil, without any losses (e.g., due to runoff, or mixing to greater depths).

An additional conservatism that is incorporated into health risk assessments is the assumption that a significant portion of the mercury emitted from an MWC facility is deposited within 10 kilometers of the facility. In reality, most of the mercury emitted from an MWC facility is in the vapor phase (since most particulate matter is caught in air pollution control devices), and is therefore not readily available for dry particle deposition. An uncertainty associated with deposition modeling is that wet deposition, such as washout, is typically not accounted for when modeling the deposition of mercury. Thus it is likely that deposition rates associated with soluble mercury forms generally within several hundred yards from the facility's stack are underestimated since washout is a predominant process near an emitting source during rain events (NJDEPE 1992b).

Predicted mercury soil concentrations associated with several modern MWC facilities (Clement 1989, 1990a, 1990b, 1992a) have ranged from 0.00005 mg/kg to 0.07 mg/kg. These are all much lower than the 14 mg/kg New Jersey clean-up level for total mercury in residential soil (24 NJR 373, Feb. 3, 1992), and

considerably lower than the average naturally occurring mercury concentration of 0.1 mg/kg in U.S. soils (USEPA 1985c).

Ecological Health Effects. As for humans, organic mercury is likely to pose the greatest threat to ecological receptors, since the methylated form of mercury is more soluble and can more easily penetrate the membranes of living organisms (Beijer and Jernelov 1979). Further, mercury can travel upwards through the food chain, with increasing concentrations at higher levels of the food chain. The bioconcentration factor for organic mercury in fish muscle, which measures the potential for a chemical to be accumulated in fish from water, is very high. One study reported a fish muscle BCF of 33,000 (McKim et al. 1976, as referenced in USEPA 1985b), indicating that this form of mercury accumulates readily in fish tissue. Ribeyre and Boudou (1984) have reported a similar BCF of 30,000 for fish muscle in rainbow trout.

At high levels of exposure, short-term, acute mercury poisoning can occur in fish, indicated by flaring of gill covers, increased frequency of respiratory movements, loss of equilibrium, and sluggishness (Eisler 1987). Long-term, chronic poisoning can cause emaciation, brain lesions, cataracts, and abnormal motor coordination.

Mercury is taken up by plants via the roots and above-ground parts; however, there seems to be little translocation of mercury within the plant (Godbold 1992). Symptoms of toxicity include stunting of seedling growth and root development, and an inhibition of photosynthesis which can cause yield reductions in plants (Kabata-Pendias and Pendias 1984).

In birds, violent neurological dysfunction in quail has been reported by Eisler (1987) following ingestion of inorganic mercury. Mercury poisoning in mammals is manifested by a variety of effects. At comparatively low concentrations, it can adversely affect reproduction, growth and development, behavior, blood density, histology and metabolism (Eisler 1987).

Ecological Risk Assessments Associated with MWC Operations. Ecological risk assessments for MWC facilities can be conducted to determine potential ecological impacts associated with stack emissions. Ecological risk assessments conducted for MWC facilities have found that environmental concentrations of mercury associated with MWC emissions are much lower than those that would be expected to adversely affect terrestrial or aquatic life. As with human health risk assessments, estimates of mercury emissions are used to evaluate the movement of the chemical through the environment, and to predict its concentrations in soil and surface water bodies. Because the ecological impacts associated with mercury emissions are most important for aquatic life, due to the potential for bioaccumulation and the toxicity of methylmercury, this is the focus of many risk assessments.

To determine the potential for adverse effects, predicted soil concentrations can be compared to plant toxicity criteria, while predicted surface water concentrations can be compared to Ambient Water Quality Criteria (AWQC) which have been established by the USEPA (1986) for the protection of aquatic life. The AWQC are chemical concentrations in water to which aquatic life can be exposed without experiencing adverse impacts. If the predicted concentrations are less than the toxicity criteria or AWQC, then adverse effects would be unlikely to occur.

A review of several ecological risk assessments (Clement 1990a, 1990b, 1992a) indicates that predicted concentrations of total mercury in water have ranged from 0.000000004 $\mu\text{g/L}$ to 0.0015 $\mu\text{g/L}$. All these concentrations are much lower than the USEPA (1986) four-day AWQC of 0.012 $\mu\text{g/L}$ and the 1-hour AWQC of 2.4 $\mu\text{g/L}$, indicating that mercury emissions from a modern MWC facility would be unlikely to adversely affect aquatic life.

Public concerns also have been raised regarding the potential accumulation of mercury emitted from MWC facilities in the food chain. For example, in Florida, some have suggested that mercury emissions from the Lee County MWC facility, even though it would be equipped with state-of-the-art mercury controls, would result in elevated fish tissue concentrations, and subsequent adverse impacts to animals high on the food chain (e.g., panthers) that could ingest concentrated doses of mercury in the fish. However, a more important factor contributing to elevated mercury concentrations in the South Florida environment results from land management practices (i.e., draining and flooding of land with peat deposits having high mercury concentrations that can increase the mobilization and methylation of mercury, followed by an increased biomagnification up the food chain [FDER 1990, Ware et al. 1990]). A human health and ecological assessment (Clement 1992b) that evaluated the movement of mercury in the environment associated with emissions from the Lee County MWC facility concluded, however, that environmental concentrations of mercury associated with the facility would not adversely affect humans or threatened or endangered species. The conclusions of the risk assessment were supported by the Florida Fish and Wildlife Service, which stated that mercury emissions associated with the Lee County MWC facility would not be likely to adversely affect the endangered Florida panther or other federally-listed threatened or endangered species (USFWS 1992). Further, the Florida Fish and Wildlife Service (USFWS 1992) noted that the facility would actually reduce the emission levels of mercury into the environment by at least 70%, by more efficiently and effectively removing mercury that would otherwise be released if deposited in a landfill.

Summary and Recommendations

In the past few years, mercury has become a focus of concern because of its potential effects to human health and the environment. Many have singled out MWC facilities as one of the primary sources of mercury in the environment today. As shown in this paper, however, mercury emissions associated with MWC facilities have been decreasing, and will continue to decrease, due to a decline of mercury in the municipal solid waste stream, the use of advanced air pollution control devices, and implementation of strict federal and state regulations.

In tandem with the scrutiny that has been aimed at MWC facilities, similar efforts should be made for other mercury emitters. Mercury emission limits should be implemented and enforced for all major sources, including, for example, utilities, smelters, and recycling facilities, as well as industries that discharge mercury into water bodies. State and municipal governments should establish incentives for industries to reduce the amount of mercury in their products, and to recover mercury from source separated materials. Public education programs should also be increased, so the public can understand how they can contribute to solving the mercury problem. These should highlight the importance of source separation efforts, and explain the costs and benefits of using mercury in industrial and consumer products. In this manner, consumers can make more informed decisions when purchasing and disposing of different products that may contain mercury. Such issues should be recognized by state and federal agencies, as well as environmental groups and industry, so that all involved parties can attack the mercury problem working together.

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