

Report on the
PROBLEM OF MERCURY EMISSIONS INTO
THE ENVIRONMENT OF THE
UNITED STATES

to the
WORKING PARTY ON MERCURY,
SECTOR GROUP ON UNINTENDED
OCCURRENCE OF CHEMICALS IN
THE ENVIRONMENT,
OECD

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I. INTRODUCTION

Purpose of Report

At its second meeting in October 1971, the Sector Group on Unintended Occurrence of Chemicals in the Environment decided to investigate the problem of mercury emissions. A Working Party on Mercury was formed with representation from Canada, Japan, Sweden and the United States, the countries with experience in mercury control methods; Representatives from each country were requested to prepare a report on the environmental problems of mercury emissions in their country. From these submissions the Working Party is to prepare a single paper on the nature of the mercury problem and the consequence of present or anticipated actions to control mercury emissions. This report is the United States' Working Party member's contribution.

The Problem

Over the years mercury and its compounds have become useful to society in a host of applications. They have also developed a notorious reputation as toxic substances in the workplace, the home and the environment, with a long record of disabling sickness and fatality.

Mercury metal is a liquid at ordinary temperatures which expands uniformly, is a good electrical conductor, amalgamates readily with most other metals, and is quite volatile. It combines with many other elements to form a large variety of inorganic and organic compounds with a wide range of useful properties.

Metallic mercury is used in a variety of electrical and mechanical devices such as switches, light bulbs, batteries, flow meters, thermometers, and barometers. It is extensively used as an electrode in the manufacture of chlorine and caustic soda and as a catalyst in the production of vinyl chloride. An amalgam of mercury with tin/silver is widely used in the filling of teeth. Mercury compounds are used as biocides and preservatives in many industrial and agricultural applications. Mercury is also used in a variety of pharmaceuticals and cosmetics. In 1969 almost 80,000 flasks (a flask is 76 pounds) of mercury were consumed in the United States in all applications.

The primary source of mercury for industrial use is from the red sulfide ore, cinnabar. The ratio of domestic production to annual consumption is highly variable. U. S. producers have virtually no control over the price of mercury, and mines open and close frequently as the price changes. The

price of mercury is presently below \$270 per flask and the number of operating mines has decreased from 109 in 1969 to less than 10 today.

As an environmental contaminant mercury comes from many types of man-made and natural sources and enters the environment through direct discharges to the air, water and soil. The principal man-made discharges are through the various applications of pesticides, chlor-alkali production, fuel burning, catalytic processes, ore refining, sewage treatment waste incineration, phosphate rock processing, paint manufacture and use, and breakage of mercury-containing devices. Recent concern has focused on pesticides applications and water discharges from chlor-alkali plants since these sources have been closely associated with contamination of the food chain. A limited amount of mercury may be emitted from natural ore bodies, thus causing contamination of the local air and aquatic environment.

Because mercury can be introduced into the environment in many forms--some volatile, some water soluble, some stable, some unstable--it is readily transported throughout the environment. In addition it is easily transformed from one physical or chemical state to another depending on the environmental conditions. It also has a strong tendency to accumulate in sediments and in organisms, depending on its form.

In the air mercury may fall out or be rained out and be entrained in soil and water. It may also be transported great distances in the atmosphere. In soil and water, it may be transformed to a volatile or soluble form, taken up by organisms, deposited in bottom sediments or released to the air. The cycle is dynamic, but once mercury enters the environment its removal becomes a complex long-term problem.

Long before the present concern over methylmercury contamination in fish and poisonings due to treated seed, there were many documented cases of mercurialism, both acute and chronic, from a variety of industrial and domestic exposures. The mercury problem is not new, only society's awareness of it is. The forms of mercury vary in their relative toxicity to man and animals. The alkyl (methyl and ethyl) compounds are believed to be the most toxic followed by metal mercury vapor, which is more toxic than the inorganic salts and organic compounds other than alkyl. The affinity of the mercury alkyl compounds and metal vapor for nervous tissue, especially the brain, explains the rather bizarre neurological symptoms seen in man and other mammals suffering severe mercury intoxication. Although effects at these high levels are striking, the United States is just as concerned about the more subtle psychological effects at low levels. We are also very concerned over the likely teratogenic and mutagenic effects which may impact on future generations of men and animals.

Although present emphasis is on protecting the human population from ingesting excessive levels of methylmercury in diet, the concentrations of mercury recently observed in the air and water convince us that there must now be an effort to drastically reduce the human intake of mercury from all routes of entry.

The problem of mercury in the environment boils down to these salient features:

1. There are diverse and widespread man-made sources emitting mercury into the environment.
2. The many physical and chemical forms of mercury are such that it moves readily from one environmental medium to another, it is easily transformed physically and chemically, it tends to accumulate in sediment, soil and living organisms, and once in the environment, it may be impossible to remove.
3. Although methylmercury in the diet is believed to present the greatest environmental hazard to human health at present, due consideration must be given to the potential hazard now and to future generations from the intake of other forms of mercury through other routes such as metallic vapor from air. Our concern for the future reflects the as yet unknown, but likely teratogenic or mutagenic effects of mercury compounds
4. Even if all discharges to the environment were suddenly (and miraculously) stopped, the residual already produced by past activities, (e.g. the current accumulation in fish and the aquatic sediment), if not removed or permanently stabilized, will present a mercury problem for years to come.

II. PRODUCTION, USES AND CONSUMPTION OF MERCURY

The chief source of mercury is the red sulfide ore, cinnabar. Commercial deposits of cinnabar are located in the U.S., China, Italy, Mexico, the Phillipines, Peru, Spain, the U.S.S.R. and Yugoslavia. During 1969, there were 109 operating mercury mines in the U.S. which produced 29,360 flasks of mercury (West, 1970). An additional 31,924 flasks were imported into the United States (West, 1970). The ratio of domestic to annual consumption is highly variable. U. S. producers have virtually no control over the price of mercury, and mines open and close frequently as the price changes. The price of mercury is presently below \$270 per flask and the number of operating mines has decreased from 109 in 1969 to less than 10 today. World production of mercury during 1969 amounted to 285,343 flasks (West, 1970).

Mercury is used in a variety of ways, including electrolytic preparation of chlorine and caustic soda, paints, industrial and control instruments, pharmaceuticals and agriculture.

The mercury cell electrolytic method of producing chlorine and caustic soda and/or caustic potash requires large quantities of mercury for startup and to replace what is lost during operation. Mercury cells in the United States accounted for 27.9 percent of the total installed chlorine capacity in 1969 (West, 1970). The diaphragm-type cell electrolytic method, which utilizes no mercury, accounted for the rest of the installed capacity. Approximately 0.5 lb. of mercury is lost for each ton of chlorine produced (National Materials Advisory Board, 1970). The National Materials Advisory Board (1970) predicted that by 1975 the average loss of mercury per ton of chlorine produced would be reduced to about 1/4 lb.

There are a number of reasons why the use of mercury cells has become increasingly popular. The National Materials Advisory Board (1970) states that purity of products from these cells is superior to that available from diaphragm cells, an important consideration for certain markets. Mercury cells have higher power consumption than diaphragm types; however, mercury cell derived caustic requires no steam for evaporation, a necessity for diaphragm liquor (National Materials Advisory Board, 1970). Although economics have historically favored diaphragm cells, this increment has gradually narrowed and may be incidental within the near future (National Materials Advisory Board, 1970). A new dimensionally stable anode made of specially coated titanium has been devised that may increase the attractiveness of the use of the diaphragm-type cells (West, 1970).

All metals except iron can be amalgamated with mercury. Before the development of the cyanide process for gold and silver, mercury was widely used in the "patio" process to form amalgams for recovering silver and gold. Potassium, sodium and zinc amalgams are used as reducing agents. Sodium amalgam has been used in production of tetraethyl and tetramethyl

lead; however, a continuous electrolytic process suitable for their production threatens to curtail the use of sodium amalgam (National Materials Advisory Board, 1970).

One of the more important uses of mercury as a catalyst is in the production of vinyl chloride. However, the trend has been toward the use of ethylene, a less expensive feedstock than acetylene (National Materials Advisory Board, 1970). Approximately 0.074 lb. of mercury is lost per 1000 lbs. of vinyl chloride monomer in the acetylene process (National Materials Advisory Board, 1970). Mercury in the form of a mercuric oxide catalyst is used with sulfonated anthraquinone products as raw materials to make vat dyes and is expected to retain its place in the industry without rivalry from any replacement catalyst (National Materials Advisory Board, 1970). Organic mercurial salts are used in urethane elastomers for casting, sheeting and sealant applications. The National Materials Advisory Board, (1970) estimated that there would be an increase in the use of mercury for this purpose from 600 flasks in 1968 to 1,570 flasks in 1975. Mercury based catalysts in the production of vinyl acetate monomer and acetaldehyde have been replaced by other catalysts (National Materials Advisory Board, 1970).

During World War II considerable quantities of mercury were used in fulminate for munitions and blasting caps (including mercuric chloride for tracers as munitions other than fulminate). An average of 4,400 flasks was used annually for this purpose during World War II; however, this decreased to 420 flasks in the postwar period (Pennington, 1959).

An amalgam of mercury with a silver/tin alloy is used extensively for the filling of dental cavities. Under normal economic conditions there are no substitutes for the mercury amalgam; but if mercury were in short supply its use in dental amalgams could be sharply curtailed to practically zero (National Materials Advisory Board, 1970). Current substitutes would produce satisfactory restorations even though they would not be as permanent (National Materials Advisory Board, 1970).

Considerable amounts of mercury are used in electrical apparatus, instruments and for laboratories. Such uses include batteries, lamps, electron tubes, pressure sensory devices, thermometers, gauges, barometers, valves, pump seals, meters, electrical switches and relays.

An unusual use for mercury was the utilization of 290 flasks for a pool of the metal to provide a frictionless support for floating the optical assembly of a telescope (West, 1970).

Mercury is used in a variety of pharmaceuticals including diuretics, anti-septics, skin preparations and preservatives for cosmetics and soap. Substitutes are available in each of these areas of mercury use (National Materials Advisory Board, 1970).

A relatively unique application for mercury is its use for frozen mercury precision castings. Owing to its smooth surface and low uniform expansion, mercury is superior to wax or plastic pattern materials.

Possible new industrial uses of mercury include a technique for purifying aluminum. Mercury is added to form aluminum mercury crystals which are melted in a molten salt bath, the mercury vaporized and condensed and the purified aluminum separated from the bath (West, 1970). An electrolytic process for recovery of zinc from drosses or other by-products involves precipitation of elemental zinc at the cathode and alloying with mercury. Zinc of high purity can be recovered (National Materials Advisory Board, 1970).

Mercury is used as ballast as part of the emergency flotation system in some research and rescue submersibles. One such vehicle would jettison approximately 2500 lbs. of mercury into the water in case of an emergency in order to provide flotation or trimming capability.

Mercury based pesticides have been used in paints, paper and pulp manufacture, and agriculture. During 1970 there were 380 mercury based pesticides registered under the Federal Insecticide, Fungicide and Rodenticide Act (Table I).

The use of mercury based slimicides fell sharply in 1965 consequent to a U.S. Food and Drug Administration ruling that paper or cartons which came in contact with food must be free of mercury. In order to conform with the Food, Drug and Cosmetic Act of the United States where a large percentage of the Canadian pulp and paper products are being sold most of the Canadian companies have discontinued the use of mercury slimicides in the last 10 years (Finreite 1970). Finreite (1970) found only 9 mills in Canada recently using phenylmercury slimicides. Regardless, there was, at least until recently, some use of mercury based slimicides in the United States.

Mercury based pesticides are used in the paint industry for three general purposes: (1) preservation of interior paint formulations in the can (2) mildewproofing both oil and water based exterior paints and (3) as an antifouling agent in marine paints. Mercury based compounds have been or are being used as biocides in laundry products, industrial water systems, cooling towers, air conditioner filters, adhesives, starches, glues, floor waxes, sanitizing or disinfecting rinses, fabrics, textiles, fibers, logs, lumber, paper, broom corn, cellulose sponges, etc.

Table 1 Number of mercury based pesticides registered under the Federal Insecticide, Fungicide and Rodenticide Act during 1970.

USE	Phenyl Mercury	Alkyl Mercury	Other Organo- Mercury	Mercury as Elemental	Mercurous Chloride	Mercuric Chloride	Other Inorganic Salts	Total
Trees	6	1	-	-	-	-	-	7
Flowers	1	3	-	-	-	-	-	4
Ornamentals and Turf	39	16	2	-	25	23	-	105
Humans	1	-	-	13	-	-	3	17
Home	21	3	-	-	-	-	2	26
Wood preservatives	20	3	-	1	-	-	1	25
Antifouling paints	13	1	-	2	3	2	39	60
Hospital	36	1	-	-	-	-	2	39
Industrial maintenance	59	2	-	-	-	-	-	61
Crop	14	6	1	-	-	-	-	21
Restaurants	6	-	-	-	-	-	-	6
Others	10	1	-	-	-	2	-	13
Total	226	37	3	16	28	27	47	384

The primary agricultural use of mercury based pesticides is for the fungicidal treatment of cereal grains. Secondary agricultural uses are for the treatment of diseases of turf, fruits and trees.

U. S. consumption of mercury steadily increased from approximately 30,000 flasks per year from 1930 to 1939 to almost 80,000 flasks in 1969 (Table 2). However, the U. S. consumption of mercury decreased to approximately 60,000 flasks in 1970. In 1969 the primary consumers of mercury in order of importance were the electrolytic preparation of chlorine, electrical apparatus, paint, industrial and control instruments, dental preparations, and catalysts (Table 3). In 1970, the primary consumers in order of importance were electrical apparatus, electrolytic preparation of chlorine, paint, industrial and control instruments, catalysts, and agriculture.

The chlorine industry consumed approximately 4,000 flasks of mercury annually during the period 1955-59. Consumption increased steadily until 1969 when 20,720 flasks were consumed. However, in 1970, consumption decreased to 14,977 flasks. The consumption of mercury by the paper and pulp industry decreased from approximately 3000 flasks annually in the period 1960-64 to 316 flasks in 1970 (Table 3).

The National Materials Advisory Board (1970) estimated that there would be an increasing trend in the United States consumption of mercury from 73,855 flasks in 1968 to 84,544 flasks in 1975 (Table 4). The Board also estimated trends in consumption by uses (Table 4). These estimates were made before attention was called to environmental problems associated with mercury in the United States during 1970.

Table 2 Mercury consumed in United States, ^{3/}

	Flasks ^{1/}
1928	34,482
1933-37	29,900
1938-42	32,400
1943-47	45,400
1948-52	46,900
1953-57	51,900
1955-59	54,346
1960-64	66,564
1965	73,560
1966	71,509
1967	69,517
1968	75,422
1969	79,104
1970	61,490 ^{2/}

^{1/} Represents total consumption for indicated year or average yearly consumption for indicated years.

^{2/} Preliminary estimates.

^{3/} Source US Bureau of Mines.

Table 3 Mercury consumed in the United States by uses, ^{7/}
(Flasks)

	1955-59 (average)	1960-64 (average)	1965	1966	1967	1968	1969	1970 ^{4/}
Agriculture (includes fungicides and bactericides for industrial purposes)-----	7,500	3,096	3,116	2,374	3,732	3,430	2,689	1,812
Amalgamation-----	243	361	268	248	219	267	195	216
Catalysts-----	848	773	924	1,932	2,489	1,914	2,958	2,641
Dental preparations ^{1/} -----	1,489	2,186	1,619	1,334	1,359	2,089	3,053	1,799
Electrical apparatus ^{1/} -----	9,285	10,540	16,097	16,257	14,610	17,484	18,650	15,789
Electrolytic preparation of chlorine and caustic soda-----	4,172	7,430	8,753	11,541	14,306	17,453	20,720	14,977
General laboratory use:								
Commercial-----	986	1,459	1,119	1,563	1,133	1,246	2,041	1,513
Industrial and control instruments ^{1/} -----	5,998	5,450	4,628	4,097	3,865	3,935	6,981	4,035
Paint:								
Antifouling-----	1,213	639	255	140	154	392	244	193
Mildew proofing-----	5/	4,772	8,211	8,280	7,026	10,174	9,486	8,771
Paper and pulp manufacture-----	6/	2,831	619	612	446	417	558	316
Pharmaceuticals-----	1,615	3,350	418	232	283	424	724	571
Redistilled ^{1/} -----	9,509	9,662	12,131	7,267	7,334	8,252	2/	2/
Other ^{3/} -----	11,488	9,807	15,402	15,632	12,563	7,945	9,689	6,521
Total known uses-----	54,346	66,564	73,560	71,509	69,517	75,422	77,988	59,154
Total uses unknown-----	--	--	--	--	--	--	1,116	1,100
Grand Total-----	54,346	66,564	73,560	71,509	69,517	75,422	79,104	61,490

^{1/} A breakdown of the "redistilled" classification showed averages of 47 percent for instruments, 13 percent for dental preparations, 23 percent for electrical apparatus, 10 percent for general laboratory, and 7 percent for all other uses in 1965-68.

^{2/} In 1969-70 "redistilled" mercury is broken down and included in the categories for which it is used.

^{3/} Includes mercury used for installation and expansion of chlorine caustic soda plants.

^{4/} Preliminary estimates

^{5/} Unavailable

^{6/} Included with agriculture

^{7/} Source U. S. Bureau of Mines

Table 4 Estimates of trends in consumption of mercury made
by the National Materials Advisory Board (1970).

Use	Estimated Mercury Consumption flasks	
	1968	1974-75
Agriculture	3,480	2,650
Amalgams	259	250
Catalysts:		
Urethanes	800	1,580
Vinyl Chloride Monomer	500	250
Anthraquinone Derivatives	175	220
Miscellaneous	230	340
Dental Applications	3,500*	3,800*
Electrical Apparatus	17,200	22,700
Electrolytic Preparation of Chlorine & Soda	17,424	22,884
General Laboratory Use	2,075*	2,075*
Industrial & Control Instruments:		
Switches & Relays	2,500	2,650
Other Instruments	6,400*	6,000*
Paints	10,588	10,725
Paper & Pulp	375	250
Pharmaceuticals	600	650
Others**	7,815	5,960
Total	73,855	84,544

* Includes some redistilled mercury.

** Includes mercury requirements for start-up of new chlorine cells.

III. HAZARDS OF MERCURY

Hazards to Man

General

Mercury in many forms is toxic to man and other living things. In terms of toxicity mercury and its compounds may be divided into three categories: (1) alkyl (methyl and ethyl) mercury salts; (2) elemental mercury vapor; and (3) inorganic mercury salts and phenyl and methoxyethyl mercury compounds (Report of an International Committee, 1969).

Alkyl forms of mercury are generally considered to be the most toxic of the three categories, probably because of their relatively slow excretion rate and their tendency to concentrate in the brain and other central nervous system tissues. Elemental mercury vapor ranks second in toxicity. It concentrates in nervous tissue to a greater extent than the inorganic salts and non-alkyl organic compounds although it is excreted more rapidly than alkyl mercury.

It should be pointed out that no definitive studies have been conducted to compare the relative toxicities of the various forms of mercury during chronic exposure. During acute exposures there does not appear to be any great difference among the various forms (Berglund, et al., 1971).

In non-occupational exposures to mercury, the routes of entry into the human system are quite different for the different forms. The organic forms are more likely to be taken in from food and water (an exception is dimethylmercury which may occur as vapor in the air) while elemental mercury is mostly absorbed through inhalation of the vapor. The inorganic salts of mercury may be inhaled or ingested in environmental exposures.

Methylmercury

Mercury in many different forms can be toxic; however, methylmercury is one of the most hazardous to man. It is easily transported and persists in the natural environment. It is formed by the methylation process from any type of mercury in the aquatic habitat and is accumulated by organisms common in the human diet. It has a long retention time within the human body, has serious effects on the human nervous system and damages developing human tissue.

The extent of exposure of the general population to methylmercury appears to be chiefly through fish and possibly other foods and not directly through water or air (Study Group on Mercury Hazards, 1970). Methylmercury may be absorbed through the skin, respiratory tract and the alimentary

canal (Berglund et al., 1971). Data from rats and man indicate that more than 90 percent of the methylmercury in food is absorbed (Berglund and Berlin, 1969). Berglund and Berlin (1969) state that methylmercury salts if applied on the skin, e.g. dissolved in an ointment, may give rise to clinical toxicity in man. There is considerable risk of absorption of methylmercury in the respiratory tract in connection with the occupational handling of seed dressings.

Approximately 10 percent of the total body burden of methylmercury in man is found in the head, presumably most of it in the brain (Berglund, et al., 1971). Also, particularly high levels of methylmercury are found in liver and kidneys (Berglund et al., 1971). Methylmercury accumulates in red blood cells, and in man these cells contain over 90 percent of the methylmercury in the blood stream (Report of an International Committee, 1969). A direct relationship between levels of mercury in blood and hair was found for Swedish individuals who were considered to have reached an equilibrium state between dietary intake and body burden of mercury (Study Group on Mercury Hazards, 1970). In fatal cases of Minamata Disease, the ratio of mercury in brain, liver and kidney was of the order of 10:40:50 (Study Group on Mercury Hazards, 1971).

According to Berglund et al. (1971) the following can be stated relative to methylmercury poisoning. Methylmercury in organisms is relatively stable. Methylmercury administered to animals is present almost entirely as methylmercury in the brain and blood. The main route of excretion in man is by the feces, which is about ten times higher than the urine. Elimination from the body can be described as a monophased exponential course. The half life of methylmercury in man ranges from 70 to 90 days for the whole body; however, limited data indicate that it is somewhat shorter in the blood and somewhat longer in the brain than in the body as a whole. Because of the slow elimination rate in man the steady state between uptake and elimination is reached approximately one year after exposure has started. The organ which first shows injurious functional disorders from exposure to methylmercury is the nervous system. Postnatal poisoning has a symptom free latent period of about one month from the time of exposure to the onset of symptoms. Symptoms are sensory disorders, ataxia and concentric constrictions of the visual fields. The diagnosis is difficult to establish in the case of only mild or atypical symptoms. Besides raised mercury levels in blood and hair, no clinical laboratory investigations have given any clear and common positive finding. Blood levels in Japanese cases of methylmercury poisoning at Niigata were estimated to be in excess of 0.2 ug/g whole blood at the onset of the disease. Hair levels in all cases except one was about or exceeded 200 ug/g. The Japanese cases at Minamata who died all had levels of methylmercury in the brain equal to or more than 5 ug/g.

Methylmercury can effect genetic material. Methylmercury added to a medium in which plant cells are growing causes disturbances in the mechanisms of cells division and chromosome breakage (Berglund et al., 1971). It also causes disturbances in the mechanism of cell division in fruit flies and in tissue culture of mice and man (Berglund et al., 1971). In persons who have been exposed to methylmercury from the consumption of fish, a correlation has been found between the mercury levels of blood cells with the frequency of chromosome breakage in circulating lymphocytes cultured in vitro (Berglund et al., 1971; Larsson, 1970). There are grounds for assuming that the effects on genetic material in man are of the same character as those in lower animals. Berglund et al. (1971) states it must be assumed that exposure to methylmercury involves certain genetic risks; however, it is not possible with presently available data to estimate the extent of such risks and the relationship to different exposures of methylmercury.

In experiments with animals, methylmercury has been shown to cause teratogenic effects. There is also a risk that methylmercury accumulations in the fetus could cause disturbances in chromosome segregation during fetal development (Ramel, 1969). According to the Report of an International Committee (1969) data from the Minamato cases presented by Murakami indicate teratogenic effects occurring at an earlier stage of development than would be the case of central nervous system damage from methylmercury intoxication. This report states "Because of the experimental evidence of strong effects of methylmercury compounds on cell division and chromosome segregation it is conceivable that this early effect may have resulted from induced chromosomal alterations of humans".

Methylmercury readily crosses the placental barrier. Tejning (1970) found a 30 percent higher concentration in fetal red blood cells than in maternal red blood cells; however, the fetal plasma had a lower concentration than did the maternal plasma. The Report of an International Committee (1969) states "Studies in animals and man indicated that methylmercury easily penetrates to the fetus via the placenta. The concentration of mercury in the fetal blood is about 20 percent higher than in the mother and the same statement should apply to the brain of the fetus as well." In the Minamata Japan incident, 22 children (5.8 percent) born between 1955-58 had cerebral palsy. The expected frequency of cerebral palsy was 0.1-0.6 percent (Berglund et al., 1971). None of the mothers had clinical symptoms of methylmercury poisoning. However, an examination of 15 of the mothers several years after their pregnancy revealed that 11 of the 15 had neurological abnormalities consistent with methylmercury poisoning. The Study Group on Mercury Hazards (1970) states that the affected children did not eat contaminated fish or shellfish and the mothers apparently were not affected. They further state that the clinical symptoms were more difficult to elicit and more varied than in the case of methylmercury poisoning in adults and children without fetal exposure. Also, the disease in prenatally exposed children varied in severity; some children having mild to moderate spasticity and ataxia, and,

others having severe intellectual retardation, seizures, and evidence of more generalized brain damage. Prenatal methylmercury poisoning cannot be distinguished from other types of cerebral palsy and diagnosis would have to be done epidemiologically with the support of mercury levels in blood and hair (Berglund et al., 1971).

Berglund et al. (1971) concluded that of 230 cases of known methylmercury poisoning slightly over 20 were prenatal. A total of 20 cases were due to occupational exposures, three cases were from skin preparations containing methylmercury, and the rest were due to consumption of food containing methylmercury of which 150 were from eating fish and shellfish, while the rest were from eating seed treated with methylmercury or meat from animals that had been fed such seed. In the Japanese methylmercury poisoning incident at Minimata, one-third of the cases were lethal (Berglund et al., 1971). In serious cases of methylmercury poisoning a degree of disability persists for a considerable period of time. Hunter (1955) reported a sixteen year old boy exposed for only a few months to methylmercury compounds was unable to work after 20 years because of persistent ataxia tremors and inability to recognize objects by touch. In Minimata Japan, brain damage was largely accomplished by the time methylmercury poisoning diagnosis had been made and although chelating agents increased the rate of excretion of mercury in the urine they were clinically ineffectual (Study Group on Mercury Hazards, 1970). The Study Group on Mercury Hazards (1970) states that because of the delay in recognition of these outbreaks in Japan, it is likely that the number of persons who were truly effected was appreciably greater than reported. Furthermore, relatively early cases with less neurologic deterioration were recognized in Niigata, Japan suggesting that many mild cases were missed in Minimata. It appears that compensatory mechanisms of the nervous system can delay clinical recognition of methylmercury poisoning, even though the patient has partial brain damage (Study Group of Mercury Hazards, 1970).

Forty families in Minimata Japan affected with methylmercury poisoning had 61 cats of which 50 percent were affected with methylmercury poisoning (Kurland, 1960). A commonly observed syndrome in cats from the affected households included unsteadiness, frequent falls, circling movements and convulsions. The sensitivity of human beings to methylmercury is presumably higher than in the case of cats which is higher than in the case of rats (Takeuchi, 1970).

The mercury blood levels of a few individuals in Sweden who consumed high levels of methylmercury contaminated fish exceeded the low levels of the toxic cases reported in Japan. The Study Group on Mercury Hazards (1970) believes this is due to individual variation and sensitivity to methylmercury.

Berglund et al. (1971) made calculations of the degree of exposure which may cause intoxication in adults with the aid of data on radioactive trace doses of methylmercury in man. The lowest reported level in the brain of persons who had died of methylmercury poisoning was estimated to be about 5ug Hg/g at onset of the disease. Based on these data, they estimated the total body burden in cases of fatal poisoning to be about 80 mg mercury. Using a daily elimination rate of one percent of the total body burden, they calculated that the body burden of 80 mg is reached after prolonged exposure corresponding to 0.8 mg mercury per day. Berglund et al. (1971) state that uncertain trace doses data indicate that about one percent of the total body burden is found in 1,000 ml of whole blood and for a body burden of 80 mg this would correspond to about 0.8 ug/g mercury in whole blood. They further state that on the basis of available material it would appear justifiable to assume that clinically manifest poisoning of adults sensitive to methylmercury may occur for a level in whole blood of 0.2 ug/g mercury, which is reached on exposure to about 0.3 mg mercury per day or about 4 ug/kg body weight per day.

According to Berglund et al. (1971): "A safety factor must be applied between the lowest mercury level and the exposure that may be assumed to cause clinically manifest intoxication with neurologic symptoms in adults and an acceptable mercury level and exposure for the population." They were of the opinion that a factor of 10 gives a sufficient safety margin. Thus, acceptable levels of mercury would be as follows:

- (1) whole blood - 0.02 ug/g,
- (2) red blood - 0.04 ug/g, and
- (3) hair - 6 ug/g.

Based on the above levels, the acceptable daily intake of methylmercury would correspond to 0.03 mg mercury or approximately 0.4 ug/kg body weight (Berglund et al. 1971).

In view of the large number of persons exposed to a varying extent in the Japanese instances of methylmercury poisoning, it seems likely there was an overrepresentation of individuals especially sensitive among those who fell ill (Berglund et al., 1971). Levels of mercury have been found in Sweden and Finland higher than 0.2 ug/g in blood cells in some 20 persons and levels exceeding 0.4 ug/g in 4 persons; while levels have been found in hair of at least 130 persons in Japan exceeding 50 ug/g without clinical symptoms of methylmercury poisoning (Berglund et al., 1971).

The diagnosis of methylmercury poisoning is based on neurologic symptoms and theoretically it is conceivable that brain lesions may occur at lower exposures and levels than which cause neurologic symptoms and which could not be diagnosed by available methods (Berglund *et al.*, 1971). Lofroth (1969) noted that: "One of the observable effects of methylmercury poisoning in man is the impairment of the coordination of muscle movement, etc., resulting from damage to certain brain cells. Thus, Lofroth raises the question: "whether these effects are brought about only at and above some threshold value of methylmercury intake." He further states: "as to the gross clinical symptoms one can state that a threshold mechanism is operating. This threshold mechanism is, however, not due to a methylmercury threshold, but to a threshold in the number of damaged brain cells. After damage of one or a few cells, other cells may take over the net result showing up as no effect in the clinical investigation. When too many cells have been damaged during a short time, the clinical results do show up early. This type of mechanism can erroneously be classified as a methylmercury threshold mechanism." He also states: "however, even a low frequency of brain cell damage, above the natural inactivation rate of these cells, during a long time has an effect on the organism as the number of available cells for each brain function is limited. Such a damage may then, have serious effects in later stages of life."

The possible synergistic effects of methylmercury in combination with other neurotoxic chemicals, e.g. DDT, PCBs, lead, which are also found in food are not known. Chernoff and Courtney (1970) found that a combination of NTA and methylmercury resulted in a slight enhanced fetal toxicity and teratogenicity of methylmercury in rats.

No known drugs are effective for the treatment of methylmercury poisoning; however, the work of Dr. Clarkson of the University of Rochester School of Medicine and Dentistry, using non-absorbable polymer sulfhydryl-containing resins possessing a high affinity for methylmercury to increase fecal excretion offers promise.

Organic Mercury Compounds Other Than Alkyl

Organic mercury compounds may enter the body by inhalation, skin absorption or oral ingestion (Report of an International Committee, 1969). Goldwater (1964) states that in studying occupational exposure to mercurials it is virtually impossible to evaluate the role of skin absorption, since inhalation and possibly ingestion also occurs when skin absorption is taking place. The Report of an International Committee (1969) states that the efficiency of absorption by the respiratory tract is unknown but should be high. The Committee notes that when mercury is as aerosols, it is likely to dissolve quickly in body fluids and be distributed to the blood. Furthermore, the

Committee notes that exposure to mercury vapor is more dangerous than dust. Phenylmercurials are readily absorbed through the intact skin and measurable quantities of phenylmercurials can be absorbed through the skin from clothes (Goldwater 1964).

Phenylmercury and methoxyethylmercury show a distribution pattern in the body similar to inorganic mercury compounds, except that a higher concentration may be present in the liver, alimentary tract and red blood cells and a lower concentration in the renal cortex (Report of an International Committee, 1969).

Phenylmercury and methoxyethylmercury are degraded in the body to inorganic mercury (Report of an International Committee, 1969). Excretion is through the urine and as inorganic mercury in feces. Goldwater (1964) states that absorbed phenylmercury compounds leaves the blood in a matter of hours and much is excreted in the urine.

Phenylmercury has been shown to affect genetic material in a manner similar to methylmercury; however, since it is excreted more rapidly and is degraded in the body to inorganic mercury the genetic risk to man would seem to be much less than in the case of methylmercury. Compared to methylmercury, relatively little phenylmercury crosses the placental barrier (Berlin and Ulberg, 1963).

The toxicity of organic mercury compounds (including phenyl and methoxyethylmercury) which degrade to inorganic mercury is similar to that of mercuric salts (Report of an International Committee, 1969). No conclusive evidence of toxic effects have been shown from long-term exposure to phenylmercury salts (Report of an International Committee, 1969). The Report of an International Committee (1969) recommended a limit at which the average concentration in air should not be exceeded during the working day on a continuing basis of 0.10 mg Hg/m^3 for inorganic mercury salts and phenyl and methoxyethylmercury salts.

Inorganic Mercury

Absorption of inorganic mercury is mainly through inhalation of elemental mercury vapor or aerosols of mercury salts. Mercury also enters the body via the skin; however, the rate of penetration is slow (Report of an International Committee, 1969). Contamination of skin or workclothes with mercury could cause heavy exposure to mercury vapor by inhalation (Report of an International Committee, 1969). Furthermore, mercuric salts are absorbed in the gastrointestinal tract, so that either by direct absorption in the lungs or by clearance of the alimentary tract, aerosols or mercuric salts can be taken into the body. Man will absorb via the respiratory

tract 75 to 25 percent of the mercury vapor at concentrations ranging from 50 ug to 350 ug/m³ (Report of an International Committee, 1969). Magos (1967) exposed mice to different concentrations of elemental mercury vapors and found that the percent uptake of inhaled mercury was inversely related to the air concentration. At concentrations below 100 ug/m³ uptake exceeded 98 percent.

After acute administration of inorganic salts to animals and man, the highest levels of inorganic mercury are found in the kidneys and the second highest in the liver (Report of an International Committee, 1969). The brain accumulates mercury to a greater extent when man is exposed to vapor than when he is exposed to inorganic mercury salts (Holmstedt, 1967; Report of an International Committee, 1969).

Elemental mercury vapor produces its toxic effect after being oxidized to mercuric ions in the body and this oxidation occurs partly in the blood and tissues, but mainly in the erythrocytes (Report of an International Committee, 1969). After being inhaled, mercury occurs in blood, partly unchanged and partly oxidized. In man the placenta constitutes a barrier to the absorption of inorganic mercury by the fetus (Berlin and Ullberg, 1963).

In man excretion of inorganic mercury is by the kidney, by the liver in the bile, by the intestinal mucosa, by the sweat glands, and by the salivary glands; however, urinary and fecal routes of excretion are the most important (Report of an International Committee, 1969). Elimination from the brain, thyroid and testes is slow; thus, accumulation of mercury in these organs is possible (Report of an International Committee, 1969).

Intoxication from mercury vapor or from the absorption of mercuric salts is due to the action of mercuric ions (Report of an International Committee, 1969). The kidney is the critical organ after acute exposure to inorganic mercury salts; however, the central nervous system is the critical organ in long-term exposure to mercury vapor (Report of an International Committee, 1969). In the case of chronic exposure to mercury vapor it is not clear whether mercury levels in the brain or testes reach toxic concentrations before severe renal damage occurs (Report of an International Committee, 1969).

Symptoms of acute poisoning from inorganic salts or mercury vapor are acute gastroenteritis, with abdominal pains, nausea, vomiting, and sometimes bloody diarrhea and severe kidney injury leading to anuria with uremia (Report of an International Committee, 1969). In cases of chronic exposure to mercury vapor, symptoms involving the central nervous system are most commonly seen, the principal features being tremor and psychological disturbances (Report of an International Committee, 1969).

The Report of an International Committee (1969) states that evidence from USSR indicates that increased excitability of the central and autonomic nervous system, together with increased frequency of slight anemia and hyperthyroidism occurred among workers exposed to 0.01 to 0.03 mg per m³ of mercury vapor. There is a positive correlation between the degree of exposure to inorganic mercury and mercury concentration in urine; however, this conclusion is valid only on a group basis and does not consistently apply to individual cases (Goldwater, 1963). Furthermore, there is a marked variation in urinary excretion of mercury in urine among similarly exposed persons and a lack of correlation between mercury in urine and clinical manifestations of poisoning. Evidence indicates that on a group basis mercury concentrations of 0.1 mg/m³ in air will correspond to 100 to 300 ug/l of urine and a decrease in exposure to 0.05 mg/m³ should result in a decrease of the concentration of mercury in the urine to about 50 percent of these levels (Report of an International Committee, 1969).

The Threshold Limit Value (TLV) for non-alkyl forms of mercury, including metal vapor has been 0.1 mg/m³ for an 8-hour occupational exposure over a 5-day work week. In 1970 the American Conference of Governmental Industrial Hygienists issued a notice of intended changes to reduce the TLV to 0.05 mg/m³ (ACGIH, 1970) in conformance with the recommendations of the Report of an International Committee (1969). It should be understood that these limits are for occupational exposures only and do not apply to environmental or general population exposures which would require much lower limits to protect the public health. The Environmental Protection Agency, in developing its National Emission Standards for mercury under Section 112 of the Clean Air Act, is using an allowable ambient concentration of 1.0 ug/m³ (0.011 mg/m³) for a 30 day average exposure as a working number for calculating emission standards for sources emitting mercury to the air. This number includes an allowance for continuous as opposed to occupational exposure and an ample margin of safety as required by law.

The Report of an International Committee (1969) recommended a limit for mercury vapor in which the average concentration in air should not be exceeded during the working day on a continuing basis of 0.05 mg/m³. The FAO/WHO guidelines for permissible concentration of mercury in foodstuffs other than fish is 0.05 ppm. A study of 642 workers from 21 chloralkali plants in the United States and Canada showed a strong correlation between mercury vapor concentrations in the workplace and nervous system symptoms such as loss of appetite, weight loss, tremors, and insomnia (Smith, et al., 1970). Timeweighted exposure concentrations ranged from near zero to 270 ug/m³ with approximately 85 percent of the workers exposed to 100 ug/m³ or less.

Hazards to Wildlife and Domestic Animals

Wildlife and domestic animals can absorb toxic doses of mercury from their food. The acute toxicity of methylmercury to birds is in the same order as that for laboratory animals, i.e., 12 to 20 mg/kg (Study Group on Mercury Hazards, 1970). According to the Study Group on Mercury Hazards (1970) mercury residues in liver-kidney composites of birds experimentally killed from eating methylmercury treated seeds ranged from 30 to 130 ppm for pheasants, 70 to 115 ppm for jackdaws and 50 to 200 ppm for magpies. They conclude that liver-kidney residues of 30 ppm in birds indicate critical exposure and that normal levels are less than 1 ppm mercury. However, Fimreite and Karstad (1971) found that the tolerance level to mercury may be lower in hawks than in pheasants and chickens. They found that hawks which died after experimental exposure to methylmercury had mercury residues in their livers of 17 to 20 ppm. Furthermore, they found that a steady diet of chicks containing 7 to 10 ppm in their liver was fatal to hawks. The signs of poisoning prior to death of the hawks were essentially neurological, consisting of weakness in the extremities, difficulty in walking and standing, and inability to control muscle movements.

Hens with mercury liver levels of 3 - 13 ppm were found to lay eggs with significantly lowered hatchability than controls (Fimreite, 1970). Birds excrete considerable amounts of mercury through molting and egg laying. Cats which had been fed fish and shellfish naturally contaminated with 5.7 ppm mercury were killed (Study Group on Mercury Hazards, 1970). A variety of wild and domestic animals are susceptible to methylmercury poisoning from eating seafood. Takeuchi (1970) reports that in the Japanese methylmercury poisoning of humans at Minamata caused by consumption of sea food; cats, crows and sea birds also fell ill presenting such symptoms as unsteadiness, frequent falling down to the ground and abnormal movement. Takeuchi also states that dogs and pigs were affected.

Hazards to Aquatic Life

McKee and Wolf (1963) have summarized the effects of mercuric chloride, mercuric cyanide, mercuric nitrate, mercurial - organic compounds and mercury on aquatic life and it will not be repeated here.

In natural waters it is methylmercury which is of primary concern from an environmental standpoint. It appears, that in most cases, levels of methylmercury in water, which will not result in unacceptable level of residues of mercury in fish from a human health standpoint, will protect aquatic life from acute toxic affects. For example, fish are believed to be able to concentrate mercury from water by a factor of 3000 or more. Thus, at a concentration factor of 3000, a level of mercury in water of 0.17 ppb would correspond to a residue level of 0.5 ppm in fish, while a level in water of 0.07 ppb would correspond to a residue level of 0.2 ppm mercury in fish.

The more subtle effects of mercury on fish e.g., effects upon reproduction and behavior have not been adequately evaluated. The Study Group on Mercury Hazards (1970) reports, that they were informed by Swedish investigators that when pike were reared for a season in water containing 0.1 ppb of methylmercury and then placed in clean water they underwent continuing mortality. Such low rates of mortality undoubtedly would not be detected in nature. They also reported that Swedish workers said that a population of fish from below a source of mercury pollution definitely weighed less at each age than those taken upstream from the source. They further reported that a Swedish worker had shown them data which showed behavior inadequacy of fish exposed to methylmercury, and that this effect increased with treatment level and with length of exposure.

Mercury compounds have been shown to interfere with primary production and to be toxic to phyto and zooplankton. Harris, White, and MacFarlane (1970) reported a significant reduction in photosynthesis and growth in marine and fresh water phytoplankton exposed to 1 ppb methylmercury compounds. At levels of 50 ppb photosynthesis was stopped. Ekeles (1962) found that methylmercury phosphate was lethal to species of marine phytoplankton at levels of 60 ppb.

IV. SOURCES OF MERCURY TO THE ENVIRONMENT

Man-Made Sources

General

No complete list of man-made sources of mercury to the environment exists. This is especially true for sources to the atmosphere. For example, emissions from cement plants have been detected by the Environmental Protection Agency source sampling teams. All extractive metallurgy may be suspected of emitting mercury to the atmosphere. Emission inventories published up to this time do not list all sources of mercury, thus the weight of mercury emitted to the atmosphere is substantially higher than tested in these inventories.

Chlorine Industry

The electrolytic preparation of chlorine and caustic soda and/or caustic potash utilizing the mercury cell process consumed during 1968 approximately 0.5 of mercury per ton of chlorine produced (National Materials Advisory Board, 1970). In 1967 the discharge from the mercury cell process in Sweden amounted to 51 to 85 g per ton of chlorine produced resulting in a discharge of 30 to 40 g to water; 5 to 10 g to hydrogen gas; 1 to 10 g to the caustic and 15 to 25 g to ventilation (Study Group on Mercury Hazards, 1970). In 1970 it was technically possible for the Swedish industry to reduce their total discharge of mercury to about 0.53 to 1.11 g per ton of chlorine produced consisting of a discharge of 0.01 to 0.1 g to water; 0.01 g to hydrogen gas; 0.01 to 0.5 g to caustic; and 0.5 g to ventilation (Study Group on Mercury Hazards, 1970). Fimrite (1970) reported a loss of 0.5 lb. per ton of chlorine produced in Canada by the mercury cell process. There has been a drastic reduction by the U. S. chlorine industry in the quantity of mercury discharged to the environment as evidenced by a reduction in the consumption of mercury by the industry from 20,770 flasks in 1969 to 14,977 flasks in 1970.

Caustic soda and potash produced by the mercury cell process may contain from 4 to 5 ppm mercury (Study Group on Mercury Hazards, 1970) and since it is used extensively in other industrial processes and products it is a potential source of mercury to the environment. Likewise, chlorine produced by the mercury cell process is a potential source of mercury to the environment. Also, the hydrogen gas produced during the preparation of chlorine by the mercury cell process contains mercury. Without controls, as much as 50 lbs. of mercury per 100 tons of chlorine produced would be emitted to the atmosphere. End box and cell room ventilation air could emit an additional 5 to 25 lbs. of mercury per 100 tons of chlorine produced. Controls are being applied which drastically reduce these emissions.

Pulp and Paper

Prior to 1965, organic mercury compounds (mainly phenylmercury) were used extensively in the pulp and paper industry for impregnating mechanical pulp and for slime control. At the present, the use of mercury compounds

by the pulp and paper industry is at a minimum. Registrations of mercury compounds under the Federal Insecticide, Fungicide and Rodenticide Act for pulp and paper were cancelled in 1970. Boureng (1967) found that approximately 5 to 20 percent of the mercury used is discharged to waterways, the rest remaining in the product. However, the mercury contained in the product would eventually be discharged to the environment e.g., by incineration of paper in trash.

The pulp and paper industry is a major consumer of caustic soda. Caustic soda produced by the mercury cell process may contain significant quantities of mercury which may result in contamination of paper products and/or result in wastewater discharges.

Laboratories, Hospitals, and Dental Clinics

Mercury is used in commercial laboratories, hospitals, and dental clinics for variety of purposes, including drugs, reagents and dental preparations, disinfectants and sterilizing solutions. These uses undoubtedly result in discharges to municipal sewage treatment plants; however, no accurate estimates of the total contribution of mercury from these sources are available.

The Bureau of Foods, Food and Drug Administration, Department of Health Education and Welfare has estimated that 25 percent of an approximate 200,000 pounds of mercury per year used in dental preparations or 50,000 pounds is lost in particles of amalgam which are scraped off the tooth or fall into the mouth and which are then spit out into dental bowls and hence to sewers (Mercurial Pesticides Registration Review Panel, 1971). The Bureau of Foods estimated that approximately 50 percent or 100,000 pounds of the total annual use of mercury in dental preparations is actually put into teeth. The Bureau of Foods states that this is not available for absorption to the blood stream until decay sets in and, as a rule, by this time the amalgam has been subject to leaching and has reached a state of disintegration. They further state that a substantial part of such a filling is inevitably swallowed and thus contributes inorganic mercury to the body load. Some of this mercury in fillings would eventually reach sewage treatment plants after being excreted by the body.

Paint

Mercury may be discharged to the environment from the manufacture and use of paints containing mercury by: (1) wastewater discharges from manufacturing, (2) volatilization of mercury from painted surfaces, (3) discharges to sewers or drains from the washing of paint brushes, rollers, containers, etc., used to apply paint, mainly latex, water-thinned paints, (4) the slow release to water of mercury in anti-fouling paints on ship bottoms and (5) the discharge to waterways of anti-fouling paints removed from ships. Data are not available to accurately estimate the amount of mercury discharge to the environment from these sources; however, essentially all mercury used in paint could enter the environment.

The slow release of mercury from anti-fouling paints to the water can be of significance in localized areas as evident by fish stored in cages in small boat harbors showing increased mercury levels (Hanson, 1971). Increased concentrations of mercury have been found in sediments in the vicinity of shipyards reconditioning ships previously painted with mercury based anti-fouling paints. For example, in the vicinity of a Dry Dock in Hampton Harbor, James River, Virginia, the Environmental Protection Agency has measured up to 5.2 ppm mercury, dry weight, in bottom sediments. The Massachusetts Department of Public Health (1971) found mercury levels up to 83 and 17 ppm in the sediments of Quisset Harbor, Falmouth and Sippican Harbor, Marin, Massachusetts, respectively. Their investigations indicated that the most probable source of contamination was the mercurial anti-fouling paint used in boatyards and marinas.

Catalysts

Industrial use of mercury catalysts can result in significant discharges of mercury to the environment, e.g., the source of mercury to the environment resulting in the Japanese instances of methylmercury poisoning was from the uses of mercury catalyst in the production of vinylchloride (Kurland et al., 1960). The Study Group on Mercury Hazards (1971) reported a Swedish vinylchloride plant discharged 300 kg per year. Hanson (1971) reports that prior to 1964, several tons of spent mercury catalysts were dumped in the sea off the Swedish East Coast. Kurland et al. (1960) found 12.5 ppm mercury in muds of Galveston Bay adjacent to a holding basin where spent catalyst used in the production vinylchloride had been dumped. They also reported the mercury lost from the reactors in the Texas vinylchloride plant was flared or incinerated and dissipated into the atmosphere. The Environmental Protection Agency has found a considerable number of discharges of mercury to waterways from manufactures using mercuric oxide catalysts with sulfonated anthraquinone products as raw materials to make vat dyes.

Mining and Refining

Considerable quantities of mercury were used in the early days of gold and silver mining to recover free gold and silver from placer and lode ores. Undoubtedly, large quantities were introduced into waterways which may still be present in some instances. The discharge of tailings from the mining of cinnabar and other metals sulfide ores, are a potential source of mercury to waterways. Johnsson (1970) reported that base metal deposits around the Great Lakes contain as much as several hundred ppm mercury.

Refining of sulfide ores where the ores are heated in retorts or furnaces can result in significant discharges of mercury to the air. Stahl (1969) states that in refining of mercury ores, stack losses of mercury should not exceed 2 or 3 percent, although much higher losses have recurred. He estimated that with a stack loss of only 3 percent in the United States mercury smelting would emit 50,000 lbs. of mercury into the atmosphere annually. Furthermore, Stahl points out that the refining of other sulfide ores emit mercury vapor into the atmosphere.

Fossil Fuels

The Environment Magazine (1971) reported that nine power plants in Illinois and the St. Louis area discharge large quantities of mercury into the atmosphere, presumably resulting from the burning of fossil fuels. A Springfield, Illinois power plant was cited as discharging 5,300 pounds of mercury into the air annually.

Petroleum, natural gas, and oil field brines all have been reported to contain significant quantities of mercury. White et al., (1970) reports that crude oils from the Cymric Oil Field, California contain from 1,900 to 21,000 ppb mercury and that the natural gases from the field are saturated with mercury vapor. Mercury vapor during transport of these gases in pipelines combines with hydrogen sulfide from "sour" gases of other oil fields and is precipitated in the pipelines (White et al., 1970).

According to White et al., (1970) mercury separates from the crude oil at the local pumping station in the Cymric Oil Field and the yield may be in the order of hundreds of tons. They report that light petroleum of the Abbott Mine California contain 100,000 ppb mercury and tarry petroleum from the mine contain 500,000 ppb. Tar from a Wilbur Spring District, California oil test was reported to contain 1,000 ppb mercury (White et al., 1970). Oil field brines from the Cymric Oil Field contain up to 200 ppb mercury (White et al., 1970). Hanson (1970) reports that Swedish fuel oil may contain 2-4 ppm mercury while gasoline may contain 2-17 ppm mercury. According to Larson (1970) Swedish fuel oils have an average content of 3 ppm mercury. There are insufficient data to evaluate the amount discharged into the environment from the mining and use of petroleum and natural gas; however, discharges to waterways of oil, oil field brines, the flaring of waste gas, and the use of natural gas, fuel oil and gasoline as a source of power and heat, may be emitting large quantities of mercury to the environment.

The Study Group on Mercury Hazards (1971) states that based on presently available information, coals in the United States may contain from a few ppb to several ppm mercury and that it is reasonable to estimate that the average concentration is at least 1 ppm. The Study Group further states that based on an annual consumption of 500 million tons of coal per year, one million pounds of mercury per year would be released to the environment. However, this estimate may be high, as Ruch et al. (1971) found that 55 raw coal samples from 10 coal seams in Illinois had a mean mercury concentration of 0.18 ppm and eleven coal samples from states other than Illinois had mercury concentrations within the range or slightly lower than Illinois coals. The United States Geological Survey report "Mercury in the Environment" (1970) states that typical samples of bituminous coal from the United States contain from 1 to 25 ppb and many anthracite coals contain from 1,200 to 2,700 ppb. Pit coals in Sweden have a mercury content ranging from 60 ppb to 400 ppb (Larsson, 1970). Presumably, when coal is burned, the mercury which it contains would be distilled off into the atmosphere.

Sewage Treatment Plants

The mercury content of sludge from sewage treatment plants give a good picture of mercury emission from urban areas. Larsson (1970) found that sludge from some 30 towns in Sweden had mercury concentrations ranging from 0.8 to 40 mg/Kg dry weight. The high levels were found in strongly urbanized areas with richly differentiated industry. Larsson (1970) states that purification of the sewage water may lead to a reduction of mercury up to 97 percent. One treatment plant in Stockholm, serving 600,000 inhabitants discharged to the receiving water about 350 kgs per year of mercury (Hanson, 1971). The Environmental Protection Agency measured mercury being discharged at the rate of approximately 2,700 lbs per year in the effluent of the Whites Point District, Los Angeles Sanitation Sewage Treatment Plant outfall based on a 24 hour composite sample taken during the fall of 1970. Also, mercury was measured being discharged at the rate of approximately 2,000 pounds per year from the Hyperion, Los Angeles City Sewage Treatment Plant outfall based on a 24 hour composite sample. The sludge produced by this plant, which is deposited at sea, was estimated to contain during a years time approximately 1100 pounds of mercury. The incineration of sewage sludge containing mercury can result in discharges of mercury to the atmosphere, e.g., the Environment Magazine (1971) reported 570 lbs. of mercury per year being emitted from Chicago's Stickney Sludge Dryer Incinerator.

Uses of mercury at waste water treatment plants which may result in mercury being released to the system are: (1) mechanical shredders (comminutor) utilizing mercury seals, (2) flow meters and rate of flow controllers and (3) rotary sewage flow distributors utilizing mercury seals. The Environmental Protection Agency estimated that during 1971 approximately 2.5 tons of mercury were lost to the environment from rotary sewage flow distributors at municipal sewage treatment plants.

Mercury contained in the effluents of wastewater treatment plants, as well as the common practice in some places of disposing of sewage sludge at sea, will result in discharge of mercury to the aquatic environment. Incineration of sewage sludge will result in discharges of mercury to the atmosphere.

Incineration

Incineration of materials containing mercury, such as wood, paper, trash, batteries and used containers, will result in discharges of mercury to the atmosphere. During 1970 the Environmental Protection Agency measured an average of 2.4 ppm mercury in 11 samples of refuse collected from 6 different incinerators. The Environment Magazine (1971) reports that 5,400 pounds of mercury per year is being discharged to the air from Chicago's Calumet Incinerator.

Phosphate Rock Industry

The United States Geological Surveys' unpublished preliminary analysis of natural phosphate deposits reveals that Wyoming phosphate deposits contain from 90 to 2700 ppb mercury while Florida deposits range from 10 to 110 ppb mercury. Phosphate rock is utilized for the production of phosphorous and fertilizers. In the production of phosphorous by electrolytic furnaces, it is assumed that mercury contained in the phosphate rock would be volatilized and emitted to the atmosphere. In the production of fertilizers, waste water discharges could be a source of mercury to the water environment. Hanson (1970) found that in Sweden a fertilizer factory with a production rate of 50,000 tons of phosphorous pentoxide resulted in a loss of 220 kgs of mercury per year to the water.

Raw Materials and Basic Chemicals

As mercury is found as a trace element in rocks, sands, etc. it is possible that significant amounts of mercury are entering the air and water environment from the processing of raw materials. Hanson (1970) found significant amounts entering the environment from such sources in Sweden; however, such data are not available from the United States. He also found significant amounts, up to and exceeding 0.5 ppm in the basic chemical, sulphuric acid.

Agricultural Use of Pesticides

Agricultural use of mercury based pesticides can be a significant source of mercury to environment. The use of mercury based fungicides to treat seed grain flooded after planting (e.g. rice) can be a direct source of mercury to the water environment. The Environmental Protection Agency estimated that 9,550 pounds of mercury were applied to the rice growing area in Louisiana during 1969, including 1,560 pounds in the Calcasieu River watershed. Seed grain treated with mercury based fungicides can be a significant source of mercury to wildlife feeding on such grain. The drainage of vats used for the dipping of bulbs, corms, broom corn, logs and lumber in mercury based pesticides, is a potential source of mercury to soil and waterways.

Considerable quantities of mercury based fungicides are used on turf, including golf greens and tees fairways, parks, cemeteries and house lawns. It is expected that runoff from such areas under some conditions could result in the discharge of mercury to waterways; however, little data are available to determine how significant of a source this is. Frequent use of turf fungicides may lead to significant accumulation of mercury in soils, since as much as 15 lbs. of mercury is added to each acre in a single application (Boer, 1944). Considerable quantities of raw logs and lumber are treated with mercury based pesticides. After treatment, lumber is dressed prior to use and the shavings may be used in the manufacture of paper, wallboard, or as fuel which when burned would be a direct source of mercury to the atmosphere. The use of a mercury based preservative to treat downed timber after a hurricane has been reported as a possible source of mercury contamination of remote New England lakes and ponds.

Use of Pesticides in Water Systems

The use of mercury based biocides in water systems, with the subsequent drainage or spillage of such water can result in significant discharges of mercury to the water environment. Mercury based biocides have been used extensively in swimming pools, industrial water systems, cooling towers, heat exchangers, air conditioners, laundry systems, etc. Billings (1970) reported that discharges from a laundry using approximately 5 lbs. of PHAS-30 (phenylmercuric acetate) per day as a mildew inhibitor was sufficient to cause serious mercury contamination of fish in the Iron River, Michigan.

Containers

The disposal of empty containers previously used for the holding of mercury products are a potential source of mercury to the environment. Used phenylmercuric acetate drums were a source of mercury to Boone Reservoir, Tennessee, resulting in fish kills. During the period July 9-13, 1960, over 500,000 fish were killed, while during the period May 9-13, 1969, over 2,300,000 fish were killed (FWPCA, 1969). The drums were used as flotation devices at boat docks or were floating derelicts.

Miscellaneous Sources

The manufacture and formulation of mercury compounds (e.g. pesticides), mercury reclaiming, broken clinical thermometers, disposal of electrical apparatus containing mercury, etc., all are potential sources of mercury to the environment; however, data are not available as to how significant these sources are.

Natural Sources

Some geological formations contain significant quantities of mercury. Mercury from such sources can be emitted to the environment by leaching and volatilization, and this may account for high concentration of mercury in bodies of waters with no known man-made sources of mercury pollution (Study Group on Mercury Hazards, 1970). According to Jonasson (1970) it is highly probable that lake areas overlying mineralized clay sediments, or black shales which may contain up to 2 ppm of mercury will contain considerable mercury in their waters.

Areas naturally high in mercury are believed to be associated with areas of volcanic activity. Klein and Goldberg (1970) reports that Bostrom and Fisher (1969) found between 1-2 and 400 ppb mercury in pelagic sediments (on a calcium carbonate-free basis) on a traverse across the East Pacific Rise. The highest values occurred on the crest of the ridge in a zone of high heat flow and it was suggested that a degassing of the mantle through volcanic activity governs the mercury distribution pattern. White et al., (1970) reports water condensed from volcanic fumaroles have been found to

contain 0.3 to 6 ppb mercury. They also reported mercury content of thermal and mineral waters from the northern California coast range up to about 5 ppb and 20 ppb mercury. In the Aqua delley Spring of Siskiyou County, California, the mercury content of precipitates of thermal springs range up to 500,000 ppb (White et al., 1970). Shacklette (1970) points out that anomalous concentrations of mercury are found in air over mercury rock mineral deposits (up to 1,500 ng/m³ at ground level and 55 ng/m³ at 400 feet above ground) and that small amounts are found in air over nonmineralized areas.

V. ENVIRONMENTAL DATA FROM NORTH AMERICA

Poisonings

Man

Curley et al., (1971) has described the Alamogordo, New Mexico tragedy where a family was poisoned from eating meat from a hog fed mercury treated seed. In August of 1969, Mr. Huckleby and five of his neighbors obtained waste seed from a local granary which they used for hog feed. The grain had been treated with an alkylmercury fungicide, either Panogen or Ceresan. Mr. Huckleby began feeding his hogs the grain in late August or early September. After 2 to 3 weeks, one hog was slaughtered and the family ate the meat for the next 3 to 5 months. Three of the feeder pigs developed blindness, lack of coordination, and posterior paralysis by mid October and 12 of the 14 pigs died in the next three weeks.

During December three of Mr. Huckleby's children became seriously ill and mercury poisoning was suspected. They showed symptoms of ataxia, incoordination, blindness, and loss of consciousness. A level of 29.4 ppm mercury was measured in the muscle of the hog which Mr. Huckleby fed his family, while 32.8 ppm mercury was found in the grain feed to the hog. Urine samples obtained from Mr. Huckleby's children on January 3, 1970 had levels of mercury of 0.16, 0.21, 0.20 and 0.06 ppm. At the time the children became ill, Mrs. Huckleby was pregnant and three months later the baby was born. On January 3, Mrs. Huckleby's urine contained 0.09 ppm mercury and on February 11, it contained 0.18 ppm. Concentrations of mercury in the newborn baby's urine ranged from 2.70 ppm at delivery to 1.56 ppm several days later, indicating placental transfer to the fetus.

Curley et al., (1971) states; "these data clearly show that mercury accumulated in animal tissues and human body fluids and confirm that compounds containing organic mercury were, in fact, the causative agents in the poisoning incident." Curley et al. (1971) states that details of the epidemiology, symptomatology and diagnoses, and the clinical history and treatment of the patients for mercury poisoning will be reported by the Center for Disease Control in a report which is now in preparation.

Recently, at a Congressional hearing, Dr. Roger Herdman, Deputy Commissioner of the New York State Department of Health testified that a dieting woman who ate swordfish twice a day for more than a year had been diagnosed as a victim of mercury poisoning.

Domestic Animals

There are a few reported instances of poisoning of livestock from the misuse of mercury-treated seed. Most of a herd of 44 adult swine and 5 litters of pigs fed several weeks on a diet, one half of which consisted of seed grain treated with a methylmercuric compound, died within a three month period (Kahrs, 1968).

In June of 1970, a resident of the Washington, D. C. area, reported to the Federal Water Quality Administration, U.S. Department of Interior (now part of the Environmental Protection Agency) that his cat was showing symptoms of mercury poisoning. His description of the symptoms were almost identical to those which Kurland (1960) reported for methylmercury poisoned cats from Minamata, Japan, i. e., unsteadiness, frequent falls, circling movements and convulsions. This was of interest, as Takeuchi (1970) has pointed out, the sensitivity of human beings to methylmercury is presumed to be higher than in the case of cats. The owner of the cat reported that he had been feeding the animal an almost exclusive diet of smelt, which he purchased from a local food store and stored in his home freezer. The packages of smelt which he currently had on hand were marked as being from the town of Erie, Michigan which is in the vicinity of the Detroit River, Lake St. Clair and St. Clair River, all known to have mercury pollution problems.

The cat subsequently died and kidney and liver tissue samples were analysed for mercury by the United State Air Force Environmental Health Laboratory, Kelly Air Force Base, Texas. The lab report contained the notation that the samples were treated in a low temperature ashier and a possibility exists that at one time during the process the temperature might have gone higher causing a loss of mercury and, that if error exists, the reported results are low. The mercury residues measured in liver and kidney tissue samples were 63 ppm and 7 ppm, respectively. Takeuchi (1970) reported that cats affected with methylmercury poisoning from Minamata, Japan had mercury levels in liver samples ranging from 37.0 to 145.5 ppm for 15 cats, while mercury levels in kidney samples ranged from 12.2 to 36.1 ppm for 7 cats. He also reported those cats not affected with methylmercury poisoning had mercury levels in liver samples ranging from 0.6 ppm to 6.6 ppm for 13 cats while mercury levels in kidney samples ranged from 0.1 to 0.8 ppm for 10 cats. Based on these data, the cat is definitely suspect of being a victim of methylmercury poisoning.

Wildlife

According to the United States Fish and Wildlife Service (1970) two bald eagles found dead in Minnesota during 1970 had lethal amounts of mercury in their kidneys. Residues levels in their kidneys were reported to be 130 and 117 ppm mercury. The Study Group on Mercury Hazards (1970) concluded that liver-kidney residues of 30 ppm in birds indicate critical exposure. The Fish and Wildlife Service (1970) stated that they believed the eagles were picking up the mercury through ingestion of fish.

Fish

Fish kills due to mercury, even though not common, are not unknown. In Boone Reservoir, Tennessee over 500,000 fish were killed during the period from July 9-13, 1968, while over 2,300,000 fish were killed during the period from May 9-13, 1969 by phenylmercuric acetate (FMPA, 1969).

Environmental Levels

Air

Very little is known about the quantitative or qualitative aspects of mercury in the atmosphere. The Study Group on Mercury Hazards (1970) points out that mercury may occur in the atmosphere as a vapor or as an aerosol or both, and may be present as organic or inorganic mercury. Since dimethylmercury is volatile it is likely that some of the atmospheric mercury is in that form.

Williston (1963) found that in the San Francisco Bay area (Los Altos) concentrations of mercury in the atmosphere range from over 0.5 to 25 ng/m³ during the winter and from over 1 to 50 ng/m³ during the summer. He found that concentrations depended upon wind direction, wind speed, and seasonal temperature variations. High levels were found always to coincide with high smog levels. According to Fleischer (1970) unpolluted air contains 1 to perhaps 10 ng/m³ mercury while in the case of "natural pollution" over mineralized areas the air will contain up to 62 ng/m³ mercury. McCarthy *et al.* (1970) concluded that the naturally occurring mercury content of air is highest over areas where the rocks are richest in mercury, ranging from 2,000 to 20,000 ng/m³ at the surface and from 29 to 103 ng/m³ at 400 feet above ground. They state that the maximum content of mercury in air is found near midday; lesser amounts in the morning and evening; and minimum amounts near midnight. McCarthy *et al.* (1970) found that the background concentrations of mercury in air at 400 feet above ground in the Southwestern U.S. range from 3 to 9 ng/m³.

A recent study of airborne mercury concentrations by the Environmental Protection Agency in seven urban areas showed maximum 24-hour levels ranging from less than 300 ng/m³ (the minimum detectable value) in Altoona, Pennsylvania, to 310 ng/m³ in New York City. The report of this study has not yet been published, but should be in print in the near future. The State of Wisconsin conducted a study near a chlor-alkali plant and found levels as high as 3300 ng/m³ near the source (Public Intervenor's Report on Mercury Air Pollution, State of Wisconsin, 1971). The State of New York has measured airborne concentrations as high as 2300 ng/m³ for 24 hours in a non-industrial urban area (Personal communications, New York Department of Environmental Conservation, 1971).

Dr. D. J. Sibbet of Geomet, Inc. ran a test of mercury levels in a room after painting with an interior latex paint. The room had not been painted for over 4 years prior to the test. With doors and windows closed, he found a level of 0.3 ng/m³ before application of paint. After painting the initial readings were about 4 ng/m³; after 200 hours readings ranged from 1.3 to 1.7 ng/m³. The readings decreased rapidly the first few hours. Readings were decreasing very slowly the time the test

Table 5 Summary of total mercury measured in water samples from U.S. rivers and lakes obtained during October and November, 1970.^{1/}

State	Number of samples with ug/l							
	^{2/} <.5	.5-.9	1.0-1.9	2.0-2.9	3.0-3.9	4.0-4.9	5.0-5.9	6.0-6.9
Alabama	18	-	-	-	-	-	-	-
Alaska	8	-	1	-	-	-	-	-
Arizona	10	-	-	1	-	-	-	-
Arkansas	10	3	-	-	-	-	-	-
California	6	11	10	3	-	1	-	1
Colorado	17	2	-	-	-	-	-	-
Connecticut	24	1	-	-	-	-	-	-
Delaware	3	-	-	-	-	-	-	-
District of Columbia	1	-	-	-	-	-	-	-
Florida	8	4	3	1	-	-	-	-
Georgia	17	-	-	-	-	-	-	-
Hawaii	4	2	2	-	-	-	-	-
Idaho	5	1	1	-	-	-	-	1
Illinois	13	2	1	-	-	-	-	-
Indiana	19	2	-	-	-	-	-	-
Iowa	8	1	2	-	-	-	-	-
Kansas	-	4	4	2	1	1	-	-
Kentucky	3	3	1	-	-	-	-	-
Louisiana	11	1	1	-	-	-	-	-
Maine	6	1	-	-	-	-	-	-
Maryland	13	-	-	-	-	-	-	-
Massachusetts	8	6	-	-	-	-	-	-
Michigan	15	4	-	-	-	-	-	-
Minnesota	12	1	-	-	-	-	-	-
Mississippi	8	-	-	1	1	-	-	-
Missouri	9	-	2	1	1	-	-	-
Montana	8	-	-	-	-	-	-	-
Nebraska	7	2	1	-	-	-	-	-
Nevada	3	4	1	-	-	-	-	-
New Hampshire	3	1	-	-	-	-	-	-
New Jersey	18	-	-	-	-	-	-	-
New Mexico	15	-	-	-	-	-	-	-
New York	27	6	-	-	-	-	-	-
North Carolina	21	-	-	-	-	-	-	-
North Dakota	5	1	-	1	-	-	-	-
Ohio	9	8	7	-	-	-	-	-
Oklahoma	9	2	1	-	-	-	-	-
Oregon	13	-	-	-	-	-	-	-
Pennsylvania	42	1	-	-	-	-	-	-
Rhode Island	4	-	-	-	-	-	-	-
South Carolina	16	-	-	-	-	-	-	-
South Dakota	5	1	-	-	-	-	-	-
Tennessee	10	2	-	-	-	-	-	-
Texas	27	-	3	-	-	-	-	-
Utah	8	2	1	-	-	-	-	-
Vermont	3	-	-	-	-	-	-	-
Virginia	11	-	-	-	-	-	-	-
Washington	13	-	1	-	-	-	-	-
West Virginia	12	-	-	-	-	-	-	-
Wisconsin	15	-	1	-	-	-	-	-
Wyoming	9	-	-	-	-	-	-	-
Puerto Rico	10	-	-	-	-	-	-	-
Total	579	79	44	10	3	2	0	2

^{1/} Summarized from Durum *et al.*, (1970).

^{2/} Below detection limit.

was discontinued after 200 hours. The room was occasionally ventilated and the readings dropped to about $.3 \text{ ng/m}^3$. When the room was again closed the reading approached the before ventilation readings after approximately three hours.

Water

The United States Geological Survey, Department of Interior analyzed water samples obtained during October and November 1970 from U.S. rivers and lakes for both dissolved and total mercury (Durum et al., 1971). Sampling sites were: (1) surface water sources of public water supplies for cities of more than 100,000 population, or for some States, the largest city in the State, (2) water sources downstream from major municipal and/or industrial complexes in each State, and (3) U.S. Geological Survey bench mark stations established for measuring long-term natural trends in stream flow and water quality. Dissolved mercury was found to range from below the lower limit of detection of 0.1 to 4.3 ug/l and was found in 7 percent of the samples. Concentration of total mercury above the detection limit of 0.5 ug/l were measured in 140 out of 719 samples (Table 5). Concentrations of mercury ranging from 0.5 to 0.9 ug/l were measured in 79 samples while concentrations from 1.0-1.9 ug/l; 2.0-2.9 ug/l; 3.0-3.9 ug/l; 4.0-4.9 ug/l; 5.0-5.9 ug/l; and 6.0 to 6.9 ug were measured in 44, 10, 3, 2, 0, and 2 samples, respectively (Table 5).

Mershaw (1970) reported that 73 samples from United States waters analyzed for dissolved mercury during May - July 1970, ranged in concentration from less than 0.1 to 17 ppb. Of the total, 34 were below the detection limit of 0.1 ppb while of the remainder, 27 were from 0.1 to 1.0 ppb; 10 were from 1.0 to 5.0 ppb and two were over 5.0 ppb. He concluded that the mercury content in unpolluted rivers or where mercury deposits are not known are less than 0.1 ppb. Jenne (1971) also concluded that mercury concentrations in major U.S. streams are commonly less than 0.1 ppb but may reach 0.5 ppb or more below sources of contamination. Pierce et al., (1970) tabulated statistics on the mercury content of stream sediments. From these data he concluded that any stream sediment containing more than 1 ppm mercury is possibly from mercury mineralization or surface contamination by mercury-bearing wastes. Klein and Goldberg (1970) reported that concentrations of mercury in dried sediments off the coast of California varied from .02 to 1.0 ppm with the highest levels close to sewer outfalls.

Hammond (1970), states that measurements of the oceans made 35 years ago indicate a range in mercury content of 0.03 to 2.0 ppb, but recent measurements with modern techniques seem to average close to 0.1 ppb. The form in which mercury is found in the oceans is unknown.

Land

Green (1959) estimates that the average abundance of mercury in the earth's

crust is 60 ppb. Mercury content of rocks in the earth's crust range from 10 ppb to 20,000 ppb (U.S. Geological Survey, 1970). Igneous rocks *i.e.*, those formed by melting and cooling, are the basic source of mercury and these contain less than 200 ppb mercury and average 100 ppb (U.S. Geological Survey, 1970). The United States Geological Survey (1970) reports that soil averages 100 ppb mercury and varies within relatively narrow limits; while sedimentary rocks average less than 100 ppb mercury and seldom exceed 200 ppb except for certain organic-rock shales which may contain 10,000 ppb or more mercury (U.S. Geological Survey, 1970). A more recent United States Geological Survey report (Shacklette *et al.*, 1971) reported that the average concentration of mercury in soils throughout the United States is 71 ppb, with the average in the western States being 55 ppb and the average in the eastern States being 96 ppb. The maximum concentration found was 4,600 ppb. Weiss *et al.* (1971) measured the mercury content of glacial ice from Antarctica and Greenland. Samples of ice deposited prior to 1952 had an average mercury concentration of 60 ng/kg ranging from 30 to 75 ng/kg while samples of ice deposited between 1952 and 1965 had an average mercury concentration of 125 ng/kg ranging from 37 to 230 ng/kg.

Residue Levels

Man

Goldwater (1964) reported ranges of mercury concentration in blood for 4 States in the United States. He reported that mercury in all of 10 samples from California were below the detectable limits of .005 ppm, while 34 out of 40 blood samples from Ohio, 5 out of 14 blood samples from Missouri, and 38 out of 65 blood samples from New York were below detectable limits. Mercury concentration in the blood samples from Ohio ranged up to 0.008 ppm, while blood samples from Missouri and New York ranged up to 0.07 and 0.045 ppm, respectively. Thus, at least some of the blood levels were above the 0.02 ug/g (approximately 0.02 ppm) considered as an acceptable level in whole blood by Berglund *et al.* (1971). Goldwater (1964) does not present data as to what form of mercury was present in the blood samples; however, he did state the few high values strongly suggest some unrecognized unusual exposure to mercury.

Jervis *et al.* (1970) analysed hair samples from residents of the Kenora-Dryden District and the Lake St. Clair area in Canada which had a known history on the frequency of fish consumption from contaminated water. They state that nearly 75 percent of the persons tested had abnormal levels in their hair and several had concentrations of 50 to 100 ppm corresponding to their practice of having one to five meals per week of fish during the previous 10 months, while those who had less than 1 meal per two weeks during this period had essentially normal concentrations of mercury in the

hair. An examination of the data they presented in their report showed that 26 out of 37 hair measurements exceeded the 6 ug/g level of mercury in hair considered as acceptable by Berglund et al. (1971).

Martz et al. (1971) reported on the mercury content of hair from residents of Angwin, California. They found that concentration of mercury in hair of approximately half of the local elementary school students was high. Serial analyses of the hair established that mercury concentration in the hair of the high school subjects reflected an exposure dating back approximately eighteen months earlier. They subsequently traced the exposure of the children with high mercury levels to swimming in an institutional pool treated with a phenylmercuric acetate algicide. On January 11, 1971, they measured 270 ppb of mercury in the pool, after a regular schedule of treatment with the algicide for 10 weeks. An average of 39.6 ppm mercury was found in the hair of 22 children which had been exposed to mercury in the pool. Since some of the hair in the children had grown since the children were exposed to mercury from swimming, the hair was not subjected directly to the mercury in the water. Since such hair still contained mercury, the authors concluded that it must reflect an internal exposure to mercury.

Aquatic Life

For all practical purposes no measurements of mercury residues in United States fish existed prior to April of 1970. No overall compilation has been made of mercury residues found in aquatic life by various State, Federal, public and private investigators; however, the occurrence of such residues has been widespread.

Buhler et al. (1971) found that the mercury content of muscle tissue exceeded the 0.5 ppm maximum guidelines set by the Food and Drug Administration for approximately 85 percent of the brown bullhead from various rivers, lakes, and bays in California, Idaho, Oregon, and Washington. The guideline was exceeded in 74 percent of the northern squawfish, 54 percent of the channel catfish, 47 percent of the largemouth bass, and 11 percent of the white sturgeon. Buhler et al. (1971) also reported that crayfish from the Columbia River downstream from Longview, Washington contained an average mercury concentration of 2.18 ppm. Gebhards (1971) found that of the 160 fish samples analyzed for mercury content from Idaho waters, 19.3 percent were at or exceeded the 0.5 ppm Food and Drug Administration guideline. He also reported that the mercury content of 46 percent of the channel catfish and 39 percent of the yellow perch exceeded the 0.5 ppm guideline. Henderson and Shanks (1971) found mercury concentrations above a detectable limit of 0.05 ppm in 16 of 146 composite samples of fish collected by the Bureau of Sport Fisheries and Wildlife in the fall of 1969 as part of the National Pesticide Monitoring Program. Values ranged from 0.06 ppm to 1.25 ppm mercury net weight whole fish.

Anas (1971) measured mercury in fur seals collected on the Pribilof Island, Alaska and off Washington in 1970 and found all samples contained mercury. He found mean mercury values for liver tissue to be 0.20 ppm for 10 pups,

10.8 ppm for 29 two and three year old males and 67.2 ppm for 29 females in ages 5-19. The highest value obtained was 172 ppm mercury in the liver of a 19-year-old female.

In August of 1970, the Federal Water Quality Administration (1970) made a survey of State Directors of Fish, Game and Conservation Departments for information on the mercury problem as it is related to the natural resources under their jurisdiction. Eighteen out of the 48 states responding reported that residues of mercury in excess of 0.5 ppm were present in fish in their States; however, a considerable number of the States reported that they were only getting started in a mercury testing program. Texas also reported that in Lavaca Bay oysters they found up to 5 ppm mercury in an oyster bed below an industrial discharge of mercury containing wastes. Low mercury levels were found in oysters from beds in the outer estuary.

Aquatic Birds

Following the Canadian discovery of high mercury levels in the fish of Lake St. Clair, the Bureau of Sport Fisheries and Wildlife, U.S. Department of Interior undertook a preliminary assessment of mercury residues in birds and their eggs collected on the St. Clair Flats Public Hunting Grounds during the period of May 21 to 23, 1970 (Dustman et al., 1970). Mercury residues were found in liver of great blue herons up to 175 ppm while 23 ppm were found in their carcasses. They also found mercury residues in liver of common terns up to 39 ppm while 75 ppm were found in their carcasses. Dustman et al. (1970) pointed out that these residues are comparable to those in birds from Sweden that died under experimental dosages with methylmercury and in birds that died under field conditions in several Scandinavian countries.

Dustman et al. (1970) also reported that mercury residues in the eggs of 3 of 5 terns, a grebe and a mallard ranged from 1.3 to 2.0 ppm. Mercury in breast muscle of 4 out of 8 mallards, one out of 4 blue-winged teal, and in all of four lesser scaup exceeded 0.5 ppm. Also mercury in the breast muscle of one bird of each species exceeded 1.0 ppm. The maximum mercury concentration in the liver of each species was 5 to 6 ppm. Dustman et al. (1970) concluded that many birds which depended upon water areas in the Lake St. Clair region have high residues of mercury in their tissue and eggs, demonstrating the extension of the water pollution problem into the marshland environment.

Baskett (1970) reported that mercury residues in fulvous tree ducks and mottled ducks collected in 1967 and 1968 from the rice fields of the Texas Gulf Coast exceeded 0.5 ppm in 2 of 3 adults as did whole-body readings for 2 ducklings. Only one of the eight adult or yearling pintails collected during the breeding season in North Dakota had more than 0.5 ppm mercury in breast muscle and ranged from 0.033 to 0.0 ppm (Baskett, 1970). Five of eight shovelers collected from the same location had more than 0.5 ppm

in their breast muscle ranging from 0.17 to 2.26 ppm. Baskett (1970) also stated that 39 scaup taken at Lake St. Clair in the winter by the Michigan Department of Natural Resources averaged 0.6 ppm in breast muscle.

Vermeer (1971) reported on a survey of mercury residues in composite egg samples of aquatic bird species collected from Alberta, Saskatchewan and Manitoba, Canada. Mercury residues were reported for California gull ranging from 0.1 to 0.4 ppm while ring-billed gull ranged from 0.1 to 0.5 ppm; Franklin's gull ranged from 0.10 to 0.13 ppm, Herring gull ranged 0.3 to 1.0 ppm; common tern ranged from 0.1 to 0.72 ppm; double-crested cormorant ranged from 0.3 to 0.7 ppm; white pelican ranged from 0.1 to 0.4 ppm; great blue heron ranged from 0.2 to 0.4 ppm; Western grebe ranged from 0.03 to 0.17 ppm; American avocet ranged from 0.15 to 0.16; Canada goose ranged from 0.03 to 0.05 ppm; mallard ranged from 0.02 to 0.06 ppm; gadwall ranged from 0.1 to 0.3 ppm; and lesser scaup ranged from 0.03 to 0.2 ppm. The highest mercury levels were found in herring gulls which may be related to its fish eating habits. Mercury residues in livers of California gull females were reported to average 5.5 times higher than in their eggs.

Other Birds

Dale (1970) reported that pheasant tissues collected in 1968 at five locations in California had mercury levels ranging up to 6.60 ppm. Of the 23 pheasant tissue samples analyzed 10 were above 0.15 ppm. In 1969, 13 pheasant tissue samples collected in the Tule Lake area, California ranged from 1.6 to 6.76 ppm, while Montana pheasant and Hungarian partridge had mercury residues in tissue ranging from 0.05 to 0.38 ppm and from 0.27 to 0.3 ppm, respectively. Buhler et al. (1971) reported on mercury residue of 137 ring-necked pheasants collected from agricultural areas of Oregon during 1970. They found mercury residues exceeding 0.5 ppm in breast muscle in 2 out of 59 birds from Umatilla County, 5 out of 64 birds from Willamette Valley and 7 out of 64 birds from Malheur County. A majority of mercury present in the tissue was in the form of methylmercury. Also, they found that the concentration of mercury in pheasant liver was normally about 2 1/2 times as great as that found in muscle. Griffith (1971) reported that laboratory findings revealed that virtually all of the mercury in California pheasants was in the form of methylmercury.

The August 1970 survey of State Directors of Fish, Game and Conservation, Departments conducted by the Federal Water Quality Administration revealed: (1) that California had found up to 4.7 ppm mercury in pheasant flesh; (2) Colorado had found from 0.04 to 0.6 mercury in pheasants, 0.4 to 2 ppm in pheasant eggs, 0.04 to 15 ppm in sage grouse and 0.05 to 0.4 ppm in band tailed pigeons, (3) Idaho had found from 0 to 7 ppm mercury in pheasants, and (4) Oregon had found that 3 of 94 pheasants analyzed contained mercury levels in breast tissue in excess of 0.5 ppm.

The Alberta Interdepartmental Committee on Pesticides (1970) reported that pheasants and Hungarian partridge collected from southern Alberta in 1959 contained an average concentration of mercury in the flesh of 0.45 ppm, ranging from 0.24 to 0.79 ppm. The Committee stated that background levels in biological material appears to be from 0.002 to 0.035 ppm mercury and, presumably higher concentrations in tissue samples are from some kind of contamination. Fikreite *et al.* (1970) reported on mercury residues in birds collected from southern Alberta and Southern Saskatchewan during 1968 and 1969. They found elevated mercury residues levels in the livers of ring-necked pheasant and gray partridge. The ring-necked pheasant livers had a mean of 2.2 ppm mercury ranging from 0.4 to 5.9 ppm while the gray partridge livers had a mean of 1.1 ppm mercury ranging from 0.4 to 2.7 ppm. These were the two species of resident upland game birds most frequently associated with farm yards, roadsides and cultivated fields where mercury treated seed grain could be found. Lower levels were found in other species of birds except for the Horned Lark which had a mean of 1.5 ppm in their livers ranging from 0.02 to 10.2 ppm. Elevated mercury residue levels were found in the egg contents of the majority of the predatory birds samples and were most pronounced in the eggs of the great horned owl and red-tailed hawk. The eggs of the Great Horned Owl had a mean mercury concentration of 1.9 ppm while the mean in the eggs of the red-tailed hawk had a mean mercury concentration of 0.3 ppm, ranging from 0.03 to 1.6 ppm.

Food

There are only limited data on mercury residues in food. The Food and Drug Administration conducted a limited study of mercury residues in foods as part of their Pesticide Total Diet Study and this indicated background levels of mercury in order of 0.002 to 0.005 ppm. Jarvis *et al.* (1970) reported on the mercury content of selected Canadian foods (Table 6). They found levels of mercury averaging 254 ppb in wheat; 250 ppb in flour; 95 ppb in milk powder, 118 ppb in beef-muscle, 102 ppb in beef liver, 236 ppb in perch, 275 ppb in pike, and 415 ppb in walleye. However, the Mercurial Pesticide Registration Review Panel (1971) points out that at the International Conference on Mercury, held in Ann Arbor, Michigan, September 30 to October 2, 1970, questions were raised as to the validity of the results. The Mercurial Pesticide Registration Review Panel (1971) states that studies should be repeated to confirm their potential significance from a human health hazard.

The misuse of mercury treated seed is a potential mechanism by which grain containing excessive levels of residues of mercury could show up in the human food supply. The Food and Drug Administration reported that 140 cars of grain at 100,000 lbs. each and one hundred fifty thousand pounds of potatoes were seized during 1970 because of mercury contamination (Mercurial Pesticide Registration Review Panel, 1971). Also, during the period July 1967 through February 1970, the Food and Drug Administration obtained 12 judgments for voluntary destruction of mercury contaminated

Table 6 Summary of mercury content of selected food products in Canada. ^{1/}

Commodity	No. of Samples	Range, ppb	Average
Wheat	5	79-400	254
Flour	6	140-380	260
Milk Powder	5	60-180	95
Tea	8	10-110	64
Beef - Muscle	3	10-310	118
- liver	5	14-220	102
-kidney	3	22-130	70
Pork	3	18-170	96
Turkey - muscle	3	12- 33	22
Chicken - muscle	14	25- 61	37
- liver	4	22- 59	46
Salmon	2	8- 32	20
Halibut	2	14- 31	23
Cod	4	27- 80	54
Flounder	2	62-170	116
Scallops	3	9- 68	30
Pickering	3	150-420	152
Perch	6	200-260	236
Pike	6	62-630	275
Whitefish	5	11-400	189
Walleye	26	80-1540	415
Sucker	3	50- 50	
Hungarian Partridge	18	20- 75	33
Pheasant - muscle	38	6-460	43
- liver	5	5-220	55

^{1/} From Jervis et al. (1970).

grain. An additional party voluntarily followed suit bring the quantity of destroyed grain to about 14 million lbs. (Mercurial Pesticide Registration Review Panel, 1971).

Fishing and Hunting Restrictions

Fishing

A survey of State Fish and Game Departments conducted by the United States Bureau of Sport Fisheries and Wildlife revealed that 17 states had imposed state fishing restrictions or warning of some type because of excessive levels of mercury in aquatic life as of September 1, 1970 (Table 7). The August 1970 survey of State Directors of Fish, Game and Conservation Departments conducted by the Federal Water Quality Administration listed one additional State which had imposed fishing restrictions, i.e., New Mexico. Since these surveys there have been additional States issuing fishing restrictions or warnings of some type e.g., California issued a warning that large striped bass and catfish from the Sacramento-San Joaquin Delta and San Francisco Bay area should not be eaten more than once a week and pregnant women should not eat them at all (Griffith, 1971). Current data on State fishing restrictions or warning currently in effect are not available.

There has been no consistency in the type of fishing restriction or warning issued by the various states or for that matter the criteria on which the restrictions or warnings are based. For example, actions have varied from a warning by California for people not to eat fish more than once a week and pregnant women not to eat them at all, to complete closures of certain commercial and sport fisheries by Michigan (FWSA, 1970). Other examples of the types of actions taken by States are: (1) Mississippi: warnings to sport fisherman to release fish caught from certain waters, (2) New Hampshire: advise to fishermen not to eat large quantities of yellow perch, chain pickerel, and smallmouth bass, (3) New Mexico: advise to fisherman not to eat more than two pounds per week of brown trout or black bullhead from a particular reservoir, (4) Wisconsin: warnings to fishermen not to eat more than 1 meal per week from the Wisconsin River, and (5) Tennessee: a public statement by a Task Force that citizens wanting to take a very conservative approach to the consumption of bass species from certain areas might temporarily limit themselves to no more than 3 meals of bass per week.

Hunting

In Canada all adult pheasants and Hungarian partridge collected in Southern Alberta in June and July of 1969 showed levels of mercury in their tissues above the 0.1 ppm tolerance level established by the Canadian Federal Food and Drug Directorate for food products. Therefore, the 1969 hunting season in Alberta on these two species was closed by Alberta authorities with the

Table 7 State fishing restrictions because of mercury -- September 1, 1970.

State	Closure of sport fishery	Closure of commercial fishery	Warning or catch and release for sport fishery	Embargo or warning to commercial fishery
Texas		Oysters, 19,900 acres Lavaca Bay		
Michigan	So. L. Huron, West L. Erie take no walleye, drum, white bass	Detroit R., L. St. Clair, St. Clair R. closed So. L. Huron, West L. Erie closed to walleye, drum, white bass	Detroit R., L. St. Clair, St. Clair R. catch and release only	Embargo on species other than walleye, drum, white bass
Wisconsin			Wisconsin R., catch and release recommended; no more than 1 meal per week	
Ohio		L. Erie, closed to walleye	Lake Erie - warning released via news	Embargo on white bass
New York	L. Onondago		L. Champlain, Erie, Ontario Oswego R., Niagara R., St. Lawrence R., danger warnings	
New Hampshire			Merrimac R., Connecticut R. danger warnings for pickerel, yellow perch, smallmouth bass	
Vermont			L. Champlain, L. Memphremagog, danger warning	L. Champlain, L. Memphremagog, embargo on sales
Pennsylvania			L. Erie, danger warning for walleye, drum, smallmouth bass, white bass	
West Virginia			Ohio R., danger warning	Ohio R., request to stop operations
Alabama		Tombigbee R. closed. Mobile R., Tensaw R., Mobile-Tensaw system, Tennessee R. and impoundments, closed	Tombigbee R. up to Jackson Dam, warning Mobile R., Tensaw R., Mobile-Tensaw system, Tennessee R. and impoundments, warning	
Georgia	Savannah R., New Savannah Dam to Highway 12 closed Brunswick Estuary, closed	Brunswick Estuary, closed		
Louisiana			Calcasieu R., warning	
Mississippi		Pickwick L., closed	Pickwick L., warning	
North Carolina			Danger warning (general)	
South Carolina	Savannah R., Augusta to coast, closed	Savannah R., Augusta to coast, closed		
Tennessee		Tennessee R., Pickwick L. closed	Tennessee R., Pickwick L., warning, catch and release	
Virginia			N. Fork Holston R. below Saltville, warning	

statement that the closure would remain in effect until data indicates mercury residues have dropped to a safe level in the birds (Alberta Inter-departmental Committee on Pesticides, 1970).

On September 23, 1970, the Idaho Fish and Game Department (1970) announced that about one fourth of 300 wild pheasants tested contained over 1 ppm mercury and recommended that: (1) persons not eat more than one meal per week of pheasant, (2) pregnant women avoid food with a known or suspected mercury content and (3) the backs and all giblets of pheasant be discarded. The announcement further stated that the hunting season would go on as scheduled.

Removal of Fish from the Market

The Food and Drug Administration found that during the period from December 1970 through February 1971 almost 4 percent of the canned tuna on the wholesale market contained residues of mercury over the Food and Drug Administration guideline of 0.5 ppm, ranging up to approximately 1 ppm. Species of tuna primarily involved were yellowfin, albacore, bigeye and bluefin. Approximately 12,500,000 cans of domestic tuna were voluntarily removed from the United States market. On May 6, 1971, the Food and Drug Administration announced that a three-month study showed that all but 42 of 853 samples of swordfish contained mercury at or above their guideline of 0.5 ppm. The announcement also stated that the average level was double the guideline and some samples were 3 times the acceptable levels. The Food and Drug Administration advised the public to stop eating swordfish. Previous to this, approximately 4,000,000 lbs. of swordfish had been seized by Food and Drug Administration or voluntarily withheld from the market because of excessive levels of mercury.

VI MOVEMENT OF MERCURY IN THE ENVIRONMENT

Air

Significant quantities of mercury are emitted to the atmosphere from the manufacture of chlorine, industrial use of catalysts, mining and burning of fossil fuels, mining and refining of ores, incineration of sewage sludge, incineration of trash (including paper, wood, containers etc.), and the processing of phosphate rock (See Chapter IV, Sources of Mercury to the Environment). Mercury is also emitted into the atmosphere from soil and mineralized land areas and by volcanic activity (See Chapter IV, Sources of Mercury to the Environment). The form of mercury in emissions from industrial sources has not been quantified to any extent; however, undoubtedly much of it is in the elemental form, as a vapor or aerosol (Study Group on Mercury Hazards, 1970). Some of the atmospheric mercury is probably in the form of vaporized dimethylmercury which reaches the air primarily through evaporation from soil or water (Study Group on Mercury Hazards, 1970). Jonasson (1970) states that most mercury compounds will degrade to mercury metal under the action of sunlight.

The vaporization rate of mercury will approximately double for every 10°C increase in temperature (Stahl, 1960). Likewise, the saturation level of mercury in air in equilibrium with metallic mercury, increases logarithmically with increasing temperature (Vaughn, 1967). These characteristics may explain the correlation of mercury atmospheric levels with season and time of day. For example, during 1969 and 1970 the monthly concentrations of total mercury in precipitation samples at Stromsardt in Central Sweden showed seasonal changes relating to evaporation during warm summer months (Study Group on Mercury Hazards, 1970). A low of 0.03 ug/l mercury was measured in the samples during December while a high of 1.45 ug/l was measured in July. Williston (1968) found that atmospheric mercury concentrations were the highest in the San Francisco Bay Area in the summer. He also found that prolonged cool wet weather would lower the average readings while prolonged warm dry temperature would increase the minimum readings.

Williston (1968) states that because of the ability of mercury to adsorb on any and all surfaces, dust particles in air will normally carry more than the expected amount of mercury and that mercury evaporated into the air will tend to concentrate on dust particles. He found that high atmospheric mercury levels always coincided with high smog levels in the San Francisco Bay area. Williston (1968) states that it is merely speculative whether the high mercury content of smog is merely coincidental or whether mercury vapor's catalytic effects under the influence of ultra-violet light may be contributory to the smog.

Air currents will transport mercury in the atmosphere as evidenced by the fact that Williston (1968) found that the mercury content over the San Francisco area was dependent primarily on wind direction and speed. In still air, inasmuch as metallic mercury vapor is approximately 8 to 9 times heavier than air, mercury will tend to collect near the surface of the ground (Stahl, 1969).

Mercury is removed from the atmosphere by dry fallout as well as by precipitation. Jenne (1970) states that more mercury may possibly be deposited by dry fallout than by rainfall during dry seasons. In an industrial area of Chicago, 4.8 ng/m^3 or two and 1/2 times as much particulate mercury was found as was found in a rural area where the concentration was 1.0 ng/m^3 (Jenne, 1970).

Rainfall may remove most if not all of the mercury in the atmosphere. McCarthy et al. (1970) measured 20 ng/m^3 of mercury in the air near a mine the day before a rain-storm while several hours after the rain no mercury was detected in the air. Snow also removes mercury from the atmosphere as evident by the measurement of mercury in the permanent snow fields of Greenland by Weiss et al. (1971).

Land

Soils

Significant quantities of mercury can enter soils from the disposal of sewage sludge; agricultural use of mercury based pesticides; dry fallout from the atmosphere; and rainfall (see Chapter IV, Sources of Mercury to the Environment). Jonasson (1970) states that a considerable amount of mercury in soils is present as elemental vapor, probably adsorbed.

Jarren (1966) found that soil horizons with either a high clay content or a high organic content carried a significantly greater quantity of mercury than did the soil profile as a whole. Jonasson (1970) quotes Anderson (1967) as finding that a high humus content is needed in soils for the mercury content to exceed 150 ppb, assuming no previous pollution, say from seed dressing. The affinity of certain soils for mercury is illustrated by Ross and Stewart (1962) finding that phenylmercuric acetate applied as an orchard spray failed to migrate below the surface 2 inches which contained 500 to 1,000 ppb mercury, depending on the number of sprays applied. According to Jonasson (1970), clays can play a key role in the collection and retention of mercury ions and at a pH of 5 the adsorption of mercury by the clays tested was at a maximum.

Leaching or soil erosion can move mercury from soils to waterways. The ability of certain soils to retain mercury close to their surfaces should

facilitate the movement of mercury from such soils by erosion. Mercury is more easily washed free from mineral soils than from soils containing humus matter (Jonasson, 1971). Humates contain sulphur sites upon which mercury may absorb very strongly (Jonasson, 1971).

A slow breakdown of phenylmercuric acetate, ethylmercuric compounds and methylmercuric compounds to mercury ion and mercury vapor in soils has been demonstrated (Kimura and Miller, 1964). Jonasson (1970) states that most mercury compounds will degrade to mercury metal under the action of sunlight.

Tomomura, Nakagami, et al. (1968) have isolated a bacterial strain of Pseudomonas from soil heavily contaminated with phenylmercuric acetate, which was found to be very resistant to mercury compounds. Its growth was inhibited only at concentrations of mercury 1000 times that inhibiting the growth of Escherichia coli and P. aeruginosa. The organism was also found to be capable of splitting the linkage between mercury and carbon in organic mercurials, including phenylmercuric acetate, ethylmercuric phosphate and methylmercuric chloride, to form metallic mercury (Furukawa et al., 1968). Furthermore, the organism was able to stimulate vaporization of phenylmercuric acetate and mercuric chloride from the media in which it was cultured and the evidence suggested that the vaporized compounds were different from the original compound (Tomomura, Maeda et al., 1968; Tomomura, Maeda, and Futai, 1968).

What significance such organisms play in the movement of mercury compounds from soil to air is not known. Regardless, significant quantities of mercury are emitted from soil to the atmosphere as is indicated by the presence of high mercury concentrations in the air over mercury enriched soils.

Meiss et al. (1971) states that permanent snow fields record the introduction of matter into that atmosphere. They were able to show an increase in the content of glacial ice from Antarctica and Greenland. Samples of ice deposited prior to 1952 had an average mercury concentration of 60 ng/kg ranging from 30 to 75 ng/kg while samples of ice deposited between 1952 and 1965 had an average mercury concentration of 125 ng/kg ranging from 87 to 230 ng/kg. They concluded that the increased fluxes of mercury to the atmosphere was due to the activities of man causing greater exposure of terrestrial surfaces which allowed more mercury vapor and gaseous compounds to enter the atmosphere.

Biota

Some mercury is translocated to fruits, tubers or seeds in plants following the foliar applications of mercury fungicides (Smart, 1968). Foliar applications of phenylmercuric acetate to rice resulted in the translocation of mercury to the grain (Lindberg, 1961). There is some translocation of mercury from treated seed to the plant and harvested seed; however, the amount is relatively small (Mercurial Pesticide Registration Review Panel, 1971).

Terrestrial animals accumulate mercury mainly from their food (Johnels and Westermarck, 1969). Man is also occupationally exposed to mercury. Birds accumulate mercury mainly from eating mercury treated seed or mercury contaminated fish (Johnels and Westermarck, 1969).

As a rule, mercury compounds are not methylated in higher animals (Larson, 1970); however, Westoo (1969) reported that there was a very small conversion of mercury compounds to methylmercury in hens. The eggs of hens fed methylmercury contained only methylmercury in their eggs (Westoo, 1969). Buhler, Claeys and Rayner (1971) reported that the majority of mercury present in wild Oregon pheasant tissue was methylmercury and Griffith (1971) reported that the same was true for California pheasants.

The tendency for mercury to be deposited in growing feathers, claws, and beaks of birds is so strong that feathers and other keratinous structures will eventually contain all the whole body load of mercury (Study Group on Mercury Hazards, 1970). Since birds molt once or twice a year, large amounts of mercury will be excreted. Wild pheasants show rapid seasonal changes in mercury level, depending on the availability of mercury treated seed; however, the fish eating osprey loses methylmercury much slower. The half life of methylmercury in the osprey is in the order of 2 to 3 months (Study Group on Mercury Hazards, 1971).

Water

Waterways

Significant quantities of mercury are discharged to waterways from manufacture of chlorine, manufacture of pulp and paper, laboratories, hospitals, dental clinics, manufacture and use of paint, industrial use of catalysts, mining and refining of ores, disposal of used mercury containers, sewage treatment plants, processing of phosphate rock, industrial utilization of raw materials and basic chemicals, and the use of mercury based pesticides (see Chapter IV, Sources of Mercury to the Environment). In some areas, natural waters overlying geological formations naturally enriched in mercury may pick up mercury by leachings or volatilization (See Chapter IV, Sources of Mercury to the Environment). Mercury enters waterways directly from dry fallout and precipitation. Soils, especially those rich in organic matter, absorb much of the mercury contained in rainfall. However, in urban areas where much of land surface consists of roofs, pavement and other nonsoil surfaces, there is a likelihood that a considerable portion of the mercury contained in precipitation would enter waterways directly or indirectly through sewage treatment plants. Erosion of mercury enriched soils would contribute mercury to waterways.

Because mercury is strongly sorbed on particulate matter and forms complexes with particulate organic matter, mercury entering into streams

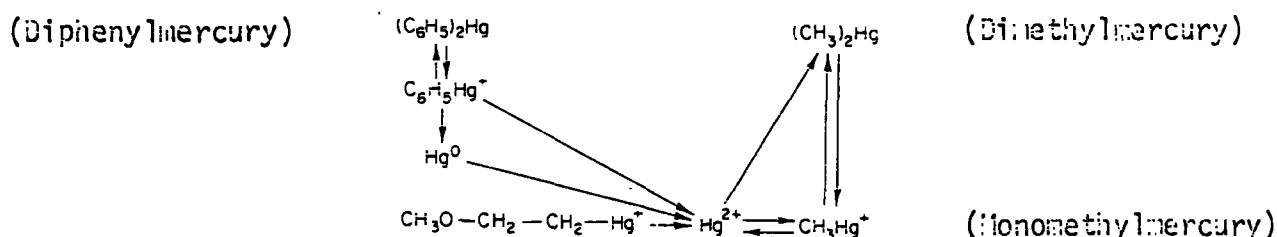
will quickly be removed from the water itself. Hannerz (1969) found that suspended solids in pond water act as scavengers of mercury carrying the absorbed mercury to the bottom. The ability of mercury to become absorbed is shown by the fact that water solutions at a pH of 5 to 8 and containing less than 500 ppb mercury, lose mercury to walls of glass or polyethylene containers to the extent of about 70 percent after 5 to 10 days (Jonasson, 1970). According to Jonasson (1970), mercury will probably be very tightly bound and concentrated by sediments containing high concentrations of metal oxides. Jenne (1970) states that the available evidence indicate that stream sediments and related fine grained materials remove a high percentage of any slugs of mercury introduced into streams, within a distance of few to several miles. The water movement of bottom sediments would transport any mercury in association with such sediments.

Transformations

It is necessary to understand the transformation reactions between the different compounds of mercury in nature if the ecological effects of the different kinds of discharges and risk involved are to be evaluated (Jernelov, 1969). Mercury is usually discharged to the water environment in one of the following forms:

- (1) As inorganic divalent mercury, Hg^{2+}
- (2) As metallic mercury, Hg^0
- (3) As phenylmercury, $\text{C}_6\text{H}_5\text{Hg}^+$
- (4) As methylmercury, CH_3Hg^+ and
- (5) As alkoxiyalkylmercury, $\text{CH}_3\text{O}-\text{CH}_2-\text{CH}_2-\text{Hg}^+$

Jernelov (1969) presented the following diagram showing some of the steps by which mercury and its compounds are converted to methylmercury:



Mildly reducing conditions, which occur in many lake and stream sediments, can cause mercury to precipitate as the sulfide cinnabar which has an extremely low solubility; however, very strong reducing conditions may increase the solubility somewhat by converting the mercuric ion to free metal (Hem, 1970). Cinnabar can be slowly oxidized in ferric ion charged water derived from pyrite freeing the mercury into solution (Jonasson, 1970). Oxygen deficient bottoms are often rich in hydrogen sulfide. Under such conditions hydrogen sulfide will react with inorganic mercury ions to form

mercury sulfide (Study Group on Mercury Hazards, 1970). Jernelov (1969) found that when mercury was added to mud as a sulfide it was not methylated under permanent anaerobic conditions. However, he found that mercury bound to organic substances and subsequently subjected to anaerobic conditions could be methylated in the presence of hydrogen sulfide and the process under such conditions could be very fast.

If the water bottoms become aerobic the mercury sulfide can be oxidized to the sulfate and the methylation process can proceed (Jernelov, 1969). Thus, if waters which have anoxic bottoms are oxygenated through decreased supply of nutrients, i.e., by decreasing the BOD load from pollution, or by other efforts at reversing eutrophication, the methylation process can proceed and methylmercury can be released to the water (Larsson, 1970; Study Group on Mercury Hazards, 1970; Mercurial Pesticide Registration Review Panel, 1971).

Metallic mercury can be oxidized readily to divalent mercury ions under conditions present at the bottoms of lakes and rivers, and this has been shown to occur experimentally as well (Jernelov, 1969). The divalent inorganic mercury produced has an extremely strong affinity for organic muds and experimentally it has been shown that it can be biologically methylated in the bottom sediments both naturally and in aquarium (Jensen and Jernelov, 1969). Jernelov (1969) states that in Sweden investigations have been made of sediments from a large number of lakes and rivers regarding the occurrence and rate of methylation of mercury and in all cases, microorganisms capable of methylating mercury were found to be present in the sediments. Wood *et al.*, (1969) showed methylmercury could be produced from divalent mercury by enzymatic and non-enzymatic reactions of methanogenic bacteria. The methylation by biological activity produces both mono and dimethylmercury (Wood *et al.*, 1969; Jensen and Jernelov, 1969; Jernelov, 1969). Monomethylmercury has a strong tendency to remain in water solution while dimethylmercury has a strong tendency to evaporate into the atmosphere (Larsson, 1970).

The pH of water will effect the methylation process. A high pH will favor a higher production of the more volatile dimethyl mercury and a much faster discharge of the mercury into the atmosphere (Larsson, 1970). This effect may be due to a high pH favoring organisms which produce more dimethylmercury or the transformation of monomethylmercury into dimethylmercury (Larsson, 1970). A more acid pH will result in the production of a high proportion of the less volatile monomethylmercury (Study Group on Mercury Hazards, 1970). Also, dimethylmercury decomposes to monomethylmercury at a low pH. Thus, under acid conditions, the total amount of mercury, as monomethylmercury, dissolved in water should be greater (Larsson, 1970; Study Group on Mercury Hazards, 1970).

Although methylation will occur anaerobically, the process appears to be more efficient in aerobic systems (Study Group on Mercury Hazards, 1970). Jernelov (1969) states that the speed of the methylation is higher when the bottom sediments contain more organic matter and conversion rates can be high in sewage treatment plants.

Jernelov (1970) found that the mucus on pike is able to convert inorganic divalent mercury almost completely into methylmercury within a short period of time of 2 to 4 hours. However, in later tests he did not find the methylation rates to be as fast. At the International Conference on Environmental Mercury Contamination at Ann Arbor, Michigan, September 30-October 2, 1970, Jernelov indicated that subsequent tests showed that there was a seasonal trend in the methylation rate by mucus and that this trend was associated with microorganisms occurring in the fish mucus during the winter.

Jernelov (1969) raised the question as to whether or not the methylation of mercury would be faster for certain concentrations of inorganic mercury. He found that in aquarium experiments that there was a steep increase in the amount of monomethylmercury observed as the concentration of added inorganic mercury in mud reached 1 to 10 ppm. However, Jernelov points out that in the experiments conducted there are several other possibilities other than a threshold value to explain the observed relationship.

Jernelov (1969) states that the conversion of phenylmercury to methylmercury has been studied and shown to occur in nature. He further states that it seems plausible that the formation of mono and dimethylmercury from phenylmercury proceeds along more than one synthetic pathway. Observations in nature repeatedly indicate that discharge of phenylmercury has a stronger and faster effect on the mercury concentration in fish than the discharge of a similar amount of inorganic mercury (Jernelov, 1969).

Jonasson (1970) states that most mercury compounds will degrade to mercury metal under the action of sunlight whether in soil or water surfaces or in the atmosphere. According to Jernelov (1969), the conversion of alkoxylalkylmercury to inorganic divalent mercury is well known to occur.

In summary, it appears that no matter what form mercury is introduced into the aquatic environment, it eventually can be converted to methylmercury.

Accumulation by Fish

Fish accumulate mercury directly from the water as well as their food. However, available data indicates that probably they obtain most of it directly from the water. Jernelov (1971) found that by comparing mercury concentration in predatory fish and food fish that the predatory fish could absorb no more than 20 percent of the mercury present in the food fish. Johnels et al. (1967) demonstrated that the concentration factor from water to pike is in the order of 3,000 or more. A linear relation between

age or weight of pike and the mercury content in axial musculature or other organs was demonstrated by Johnels and Westermark (1967). For low levels of mercury in fish (below 0.2 ppm) no increase or a very moderate increase in mercury content was found to occur with increasing weight of the fish. As the mean level of mercury increased they found that the mercury level in relation to weight increased noticeably. For extremely high levels of mercury, caused by manifest contamination, they found no relation to age or weight. Wallace et al. (1971) interprets this to indicate that there is a threshold level of mercury in the environment, above which fish cannot eliminate mercury from their muscular tissues faster than it is incorporated and above which accumulation thus occurs. Lofroth (1969) states that this relationship is an indication that fish are adapted to a mercury concentration of less than 0.2 ppm. According to Lofroth (1969) all data which have accumulated regarding the natural concentration of mercury in fish indicate that the maximum natural concentration is 0.2 ppm fresh weight or less.

Larsson (1970) states because of the relation between size and mercury level in fish, conclusions of the mercury content of the "standard pike" should only be drawn from fish with a weight lying between 500-1600 g. Sweden, when comparing residues of mercury in fish samples for restricting fishing, adjusts the level of mercury measured in pike to fit a size fish weighing 1 kg or about 2.2 lbs. (Study Group on Mercury Hazards, 1970).

Hannerz (1968) found that in brackish water uptake of methylmercury by pike is less than in fresh water; however, he found that in saltwater, cod concentrate methylmercury faster than in brackish water. According to Larsson (1970), cod in seawater accumulate mercury to higher levels than in brackish water, presumably because cod in 100 percent seawater swallow more water than they do in brackish water.

Hannerz (1968) found that in exposing pike to methylmercury for 70 to 90 days, that the concentration factors were the greatest in kidneys followed by liver, spleen, stomach, heart, gills, brain, fins, gonads, muscles, scales, eyes and bone. Salmon retained in cages in rivers for up to two months first showed an increase in the mercury content of the blood corpuscles followed by the liver, while the increase in muscular tissue progressed considerably more slowly (Hasselrot, 1968). Jernelov (1970) states that according to experiments the ratio between the mercury content of liver and body muscles varies between approximately 0.1 to 50 and that with a ratio greater than 1 (liver content/muscle content) the content in the fish muscle will rise; while if on the other hand the ratio is clearly lower than 1 (0.2) then the content in the fish muscles can be expected to decrease or at least remain constant.

Nietinen and his associates of the Institute of Radiochemistry, Helsinki University, Finland, have shown in a series of unpublished papers, that the

loss of methylmercury from fishes has two components, fast and slow (Study Group on Mercury Hazards, 1970). The fast loss occurs early and lasts only a few weeks while mercury is being redistributed through the body. The subsequent loss from established binding sites follows slowly; indeed, the half life estimated from this component is in hundreds of days--on the order of 2 years. They also have shown extremely low rates of loss from aquatic mollusks and crayfish, and they have noted distinct species differences.

Mercury is present in Swedish fish almost entirely as methylmercury (Berglund et al., 1971). This is further substantiated by Lofroth (1969) who in his review of the health hazards and side effects associated with the emission of mercury compounds states that mercury is almost 100 percent in the form of methylmercury in all types of fish investigated-- fresh water, Baltic, and Atlantic. The fact that practically all mercury in fish is methylmercury is also substantiated by Moren and Westoo (1970); Johnels and Westermarck (1969); and Westoo (1969). Furthermore unpublished analysis of a variety of salt and fresh water fish by the United States Food and Drug Administration have showed that practically all mercury in fish from the United States is methylmercury. This is not surprising in light of the fact that all forms of mercury introduced into the aquatic environment can be converted into methylmercury.

Tillander et al. (1970) found that excretion rates of methylmercury in a species of seal (Pusa hispida) was slower than any other mammal and about like fish.

Decontamination

The continuing supply of mercury from bottom sediments to the water and the slow rates of excretion of mercury by fish give little hope for quick improvement in levels of mercury residue in fish. The Swedish experience confirms this. In Sweden mercury in pike in most lakes has dropped little if at all since mercury bans became effective in early 1966. These lakes where the fish residues have not dropped tend to be biologically poor and acid. Only about three lakes apparently have had mercury levels in pike drop to a demonstrable extent. Rivers have a better chance due to continual flushing action (Study Group on Mercury Hazards, 1970).

Jernelov (1969) calculated that it would take from 10 to 100 years for the methylation process to remove the mercury from the bottom of lakes. These calculations were based on the yield over a period lasting from 1 week to 2 months of mono and dimethylmercury from bottom sediments taken from contaminated lakes and rivers and kept under natural conditions. In Minamata Bay, Japan, once the cause of the pollution was determined and eliminated, mercury levels in shellfish dropped from 85 ppm to 10 ppm over a two year period and remained constant for at least a five

year period (Irukayama, 1966). Rivers should have a better chance of being decontaminated because of the flushing action of currents moving sediments downstream. Mercury levels of salmon placed in cages below former sources of mercury in some Swedish rivers showed considerable improvement within 3 years (Study Group on Mercury Hazards, 1970).

Swedish workers have considered the following approaches to the decontamination of mercury contaminated waterways: (1) introduce oxygen-consuming materials to create continuous anaerobic conditions in the sediments, thereby reducing methylation, (2) increase the pH of the sediments to favor dimethylation and increased volatilization, (3) cover the sediments with fresh finely divided materials with high adsorptive affinity (e.g., quartz and silicates), (4) cover the sediments with inorganic inert materials of any type, i.e., bury them, and (5) remove mercury-bearing sediments by dredging or pumping (Study Group on Mercury Hazards, 1970). The first two approaches appear to be impractical, however Sweden is evaluating the other approaches (Study Group on Mercury Hazards, 1970).

Experiments have been conducted in Sweden to evaluate covering sediments by layers of inorganic sediment of varying thicknesses (0-20 cm), with and without Tubificidae (oligochaete worms) and Anodonta (a bivalve) (Study Group on Mercury Hazards, 1970). These studies have revealed that: (1) in the absence of Tubificidae, methylmercury accumulated in fish only when the sediments were uncovered, (2) in the presence of large populations of these worms, fish accumulated methylmercury when the covering layer was less than 2 cm, and (3) in the presence of Anodonta, which stirs the sediments, leakage of methylmercury occurred if the covering layer was less than 9 cm.

Swedish workers have conducted tests to evaluate the effectiveness of ground silicate, on the uptake of mercury by fish from sediments contaminated with metallic mercury, ionic mercury, and phenylmercury (Study Group on Mercury Hazards, 1970). These tests have revealed that there was no reduction in uptake when the pollutant was phenylmercury; however, a decrease in uptake by a factor of two occurred when inorganic mercury was the pollutant.

The removal of mercury contaminated sediments by dredging appears to have some serious shortcomings. For one thing, the cost to dredge any extensive area may be excessive. The dredging of a Finnish port increased the soluble mercury concentration in the water from a level of 0.5 to approximately 10 ug/l (Stephan, 1971). This increase took "some weeks" to reach a peak; however, it returned to background in a "few more weeks" (Stephan, 1971). Swedish workers were of the opinion that by dredging there was a considerable risk of increasing the rate of methylation of mercury in the sediments (Stephan, 1971). Measurements taken on sludges dredged from mercury sludge banks in Sweden indicated that while some 95 percent of the suspended solids can be retained in the sludge, only 50-60 percent of the

mercury will remain in the sludge, the remaining 40-45 percent being discharged with the supernatant (Stenhans, 1971).

Environmental Budget

In order to fully evaluate the ecological consequences of mercury in the total environment and to insure the identification of all discharges of mercury to the environment, there should be available a total budget for the environmental distribution of mercury, including sources, transfer, transportation, deposition and fate. Sufficient data are not presently available to produce a model of the flow of mercury from origin to disposal.

VII. FORMULATION OF POLICIES AND STANDARDS

General

In the formulation of policies and standards to control the introduction of mercury into the environment, the United States Environmental Protection Agency is using a total environmental approach. Historically monitoring and research has been inadequate in tracing the environmental dangers of ultra-deleterious elements in small quantities. Moreover, normally abatement has been sought only after the pollutant has gotten into the environment, damage has occurred, and can be conclusively shown. Typically, only one source or effect of a pollutant has been traced, rather than considering the effects on the total environment. We believe that regulatory actions should be focused upon prohibiting, restricting, and preventing the introduction of toxic substances before damages to human health and welfare have occurred.

The United States Environmental Protection Agency in determining the need to control the introduction of all toxic substances into the environment weighs the following general considerations: (1) the nature and magnitude of the foreseeable hazards associated with use of a particular substance and (2) concurrently, the nature of the benefit conferred by the direct and indirect use of a given substance.

In approaching the problem of taking regulatory action to abate pollution for toxic substances, such as mercury, the United States Environmental Protection Agency proceeds in as far as is practical, by:

1. Identifying all significant sources and applying equitable reduction requirements to them,
2. Bringing in other State and Federal agencies and utilizing all information available to them,
3. Keeping the public informed, and
4. Giving the dischargers a reasonable opportunity to state their point-of-view and being sure of the most current information relating to each discharge and the sources program for abatement before regulatory action is commenced.

However, it must be recognized that because of statutory limitations, some sources of pollution are not controllable, while others may not at this particular time be identifiable. Therefore, the United States Environmental Protection Agency does not permit the facts that it is not possible for us to abate all sources of pollution or that it has not identified every significant source of a pollutant to be used as justification for not taking regulatory action against known and controllable sources. It is obvious, that where the total amount of

a toxic substance being introduced into the environment must be reduced, first consideration must be given to restricting the most significant and controllable sources when taking regulatory actions.

Residues in Aquatic Life

Standards for allowable quantities of mercury in aquatic life are primarily for the protection of human health. All residues of mercury in fish must be considered to be methylmercury (See Chapter VI, Movement of Mercury in the Environment). Furthermore, as Westoo (1969) has pointed out, broiling, boiling or frying fish does not remove methylmercury. In fact, since fish lose water by these processes, a corresponding increase in the concentration of methylmercury in fish is observed (Westoo, 1969). Also, for the most part, concentrations of mercury in the aquatic habitat, which will result in acceptable residue levels of mercury in fish, will also be acceptable for the protection of aquatic life (See Chapter III, Hazards of Mercury). Thus, in formulating standards for allowable quantities of mercury in fish, the prime consideration is the toxicity of methylmercury to man.

The following salient points relative to the toxicity of methylmercury to man are summarized from Chapter III, Hazards of Mercury:

1. Man absorbs from ingested food practically all of the methylmercury present,
2. Methylmercury in man is relatively stable, i.e., it remains in man as methylmercury,
3. The half life of methylmercury in man is from 70 to 90 days;
4. Because of the slow elimination rate in man the steady state between uptake and elimination is reached approximately one year after exposure has started,
5. Methylmercury has a propensity for the human nervous system and about 10 percent of the total body burden is in the head, presumably most of it in the brain,
6. Methylmercury is neurotoxic,
7. Postnatal methylmercury poisoning is not easy to diagnose, especially in the case of only mild or atypical symptoms,

8. Besides raised mercury levels in blood and hair, no clinical laboratory investigations have given any clear and common positive findings for methylmercury poisoning,
9. Diagnosis of methylmercury poisoning is based on neurological symptoms and it is conceivable that brain lesions may occur at lower exposures and levels than those which cause neurologic symptoms and which could not be diagnosed by available methods,
10. It is not possible, with presently available data, to estimate what the long term effects of subclinical methylmercury poisoning are on man,
11. In non-fatal cases of methylmercury poisoning disability can persist for an extended period of time,
12. Compensatory mechanisms of the nervous system can delay clinical recognition of methylmercury poisoning, even though brain damage has already occurred,
13. There are presently no known drugs effective for the treatment of methylmercury poisoning,
14. There probably are individual variations in sensitivity to methylmercury,
15. Methylmercury is teratogenic,
16. Prenatal methylmercury poisoning cannot be distinguished from other types of cerebral palsy and diagnosis would have to be done epidemiologically with the support of mercury levels in blood and hair,
17. In man, concentrations of methylmercury in fetal blood are about 20 percent higher than in the mothers blood,
18. There is a greater risk of methylmercury poisoning to the fetus than to the mother and affected children can be born to mothers showing no clinical symptoms of methylmercury poisoning,
19. Methylmercury has been shown to be mutagenic to test organisms,
20. A correlation in man between the frequency of chromosome breakage in lymphocytes and mercury level in blood cells has been shown,

21. It must be assumed that exposure to methylmercury by man involves certain genetic risks; however, it is not possible with presently available data to estimate the extent of such risks,
22. An intake by man of 0.8 mg per day of mercury (as methylmercury) corresponding to a level of 0.8 ug/g of mercury (as methylmercury) in whole blood may be fatal,
23. Clinically manifest poisoning of adults sensitive to methylmercury may occur at a level in whole blood of 0.2 ug/g mercury (as methylmercury) which can be reached on exposure to about 0.3 mg mercury (as methylmercury) per day,
24. Based on item 23 and using a safety factor of 10, acceptable levels of mercury (as methylmercury) would be 0.02 ug/g in whole blood and 6 ug/g in hair, and
25. Based on item 24, an acceptable daily intake of mercury (as methylmercury) would be 0.03 mg per day.

The conclusion that an acceptable level of mercury (as methylmercury) in human blood is 0.02 ug/g, corresponding to a daily intake of mercury (as methylmercury) of 0.03 mg, was reached by a prestigious Expert Group appointed by the Swedish Board of the National Institute of Public Health, in consultation with the Swedish National Board of Health and Welfare and the Swedish National Veterinary Board to make a toxicologic-epidemiologic evaluation of the risks involved in the presence of mercury in fish intended for consumption (Berglund et al., 1971). The United States has no basis at present to disagree with their conclusion. However, in light of the susceptibility of the human fetus to methylmercury and our inability to fully evaluate possible unmeasured effects of exposure to methylmercury, we are of the opinion that it is desirable to minimize the intake of methylmercury by man to as low a level as possible.

The Study Group on Mercury Hazards (1970) asserts that the human intake of mercury appears to be mainly through fish. If all drinking water contained the maximum recommended allowable concentration of 5 ppb and with an average daily intake of 2 liters of water, a person would ingest 0.01 mg of mercury per day. However, it appears that mercury in water supplies for the most part are much below this level. Also, present indications are that mercury in most other foods, with the exception of contaminated wildlife, grain, etc., are at a relatively low level. Thus, in formulating an acceptable level of mercury in fish, the assumption has been made that fish is the only significant source of human intake of mercury. However, recent measurements of mercury in ambient air suggest that intake of mercury from inhalation may be significant.

In order to evaluate the risks of mercury in fish, it is necessary to know how much fish people eat. If all fish contained 0.5 ppm mercury, the daily limit of 0.03 mg mercury intake could be reached by eating 60 gm of fish per day. If all fish contained 1.0 ppm, the daily limit could be reached by eating 30 gm of fish. Kolbye (1970) states that the average daily intake of fish in the United States is 40 gm per day. However, according to a survey of household food consumption by the Agriculture Research Service, United States Department of Agriculture (1965), the average daily intake of fish is approximately 24 gms. The Food and Drug Administration, United States Department of Health, Education and Welfare has established a "0.5 ppm mercury interim guideline for fish" (Kolbye, 1970). Based on the United States average daily consumption of fish, the daily intake of methylmercury by persons in this country should be below 0.03 mg.

According to Berglund (1971), the average consumption of fish in Sweden is about 30 g of fish flesh per day; however, consumption varies considerably, with a few percent of the population never eating fish, whereas some individuals may consume up to 500 g per day. Likewise, fish consumption in the United States varies according to personal preferences, as well as regional, ethnic and cultural patterns.

Sweden has adopted a limit of 1 ppm mercury in fish; however, this is combined with the recommendation that fish from non-banned waters be eaten only once a week (Lofroth, 1969). Berglund et al. (1970) states relative to the Swedish situation that a level of 1 ppm in fish might result in some high consumers reaching the lowest level assumed to be present in persons sensitive to methylmercury poisoning, with clinically manifest poisoning and neurologic symptoms. They further state that with this level in fish, one third of the population could reach levels above the acceptable level; with a level in fish corresponding to 0.5 ppm some persons with extremely high fish-consumption might reach the lowest toxic level, and about one tenth of the population might reach the highest acceptable level; with a level of 0.2 ppm with free consumption, or 1 ppm in combination with a restriction of the consumption of contaminated fish to one meal a week, exposure would be within the acceptable level.

Tejning (1967) estimates that the average meal of fish in Sweden consists of 150 gm, ranging from 100 to 200 gm. It is believed that in the United States very few people on the average eat more than 2 or 3 meals of fish per week. Assuming, that the average meal of fish in the United States is 150 gm containing 0.5 ppm of mercury, then an average weekly intake of 0.22 mg would be reached by eating 450 gm of fish per week. Taken on a daily basis, this would mean an average daily intake of 0.032 mg of mercury. However, all fish eaten would not necessarily contain residues of mercury at or near the 0.5 ppm level. Therefore, assuming no other intake of mercury the Food and Drug Administration interim guideline of 0.5 ppm mercury in fish is a reasonable basis for the protection of public health.

However, it is recognized that as recommended by the Study Group on Mercury Hazards (1970), the normal and extreme patterns of fish consumption in this country need better documentation.

A Food and Drug Administration Ad Hoc Committee of Scientific and Medical Experts from this country and Canada expressed consensus support for the 0.5 guideline (Food and Drug Administration, 1971). Likewise, the Study Group on Mercury Hazards (1970) concluded "on the basis of their examination of their experience in Sweden and Finland that the interim Food and Drug Administration guideline of 0.5 ppm in fish is, for the present, a sound basis for the protection of public health. However, the margin of safety may not be large, and residual uncertainties remain in regard to possible hazards where large amounts of contaminated fish are eaten."

The purpose of the Food and Drug Administration guidelines of 0.5 ppm for the maximum level of mercury in fish is to remove from the market or prevent fish from reaching the market with excessive levels of mercury. However, we are also faced with another problem, i.e., levels of mercury in fish which are indicative of pollution. We consider that when a range of fish species in an inland or estuarine body of water have residue levels of mercury equal to or greater than 0.5 ppm that there is an indication of gross mercury pollution. Background levels of mercury in fish would normally be below 0.2 ppm. Therefore, when a range of fish species in an inland or estuarine body of water contain between 0.2 and 0.5 ppm mercury, while consumption might not represent a hazard, it is indicative of mercury pollution, and should lead to investigative activities to find the mercury source.

Drinking Water

The Bureau of Water Hygiene of the United States Public Health Service (now part of the United States Environmental Protection Agency) has proposed a standard of 0.005 ppm for mercury in drinking water (Bureau of Water Hygiene, 1970).

Residues in Bottom Sediments of Waterways

The presently available data indicate under many conditions, a total mercury content of at least 1 ppm dry weight in bottom sediments of waterways can result in gross contamination of aquatic life (See Chapter VI-Movement of Mercury in the Environment).

Discharges to the Total Environment

As mercury moves from soil to both the atmosphere and water, as it also moves from the atmosphere to both land and water; and since all forms of

mercury in the aquatic environment can be converted to methylmercury, it is the policy of the United States Environmental Protection Agency that the discharge of mercury to all environmental media should be reduced to the lowest levels possible.

Discharges to the Atmosphere

Emissions of mercury to the atmosphere, and the effects of this mercury, are not well documented. Neither the sources of mercury emissions, nor the effects of these emissions on human health or welfare, has been studied in sufficient detail to form final conclusions as to their hazard to man's environment. It is believed that annual emissions to the United States atmosphere approach 1000 tons, and without control will increase at least 5 percent a year.

Much of the earth's mercury is in the sulfide form and buried in the earth. Very little of this mercury can enter the environment through natural processes. Man, by removing fossil fuels and metallic ores from underground, and burning or heating them breaks down the sulfide compound and introduces elemental mercury to the environment. Relatively little is known about the movement of mercury through the environment. However, it is certain that mercury once released to the environment may move between air, water, land and living organisms. Emissions of mercury to the atmosphere contribute to methylmercury contamination of the aquatic environment.

Recent limited measurements by the United States Environmental Protection Agency of mercury levels in the ambient air of a few large cities showed 24 hour average mercury levels of 110 to 810 ng/m³. The Department of Environmental Conservation in New York found mercury concentrations in non-industrial urban areas ambient air as high as 2300 ng/m³.

Techniques and devices to control emissions of mercury into the atmosphere are not available at the present time for most sources. An immediate attempt to control all sources of mercury emissions to the United States atmosphere could have a severe impact on our way of life. A comprehensive program to identify sources of mercury emissions to the atmosphere, mercury's movement through the environment once emitted, and means to control these emissions if necessary, is essential. As knowledge of sources and control technology becomes available it is believed that all man-made emissions of mercury should be controlled.

Waste Water Discharges to Public Waters

It is the policy of the United States Environmental Protection Agency that all man made discharges of mercury to public waters should be prohibited. This policy is based on the facts that: (1) all forms of mercury in the aquatic environment can be converted to methylmercury, (2) fish

and other aquatic life concentrate methylmercury from the aquatic environment, (3) the health of man is adversely affected by eating mercury contaminated aquatic life, (4) mercury contaminated waterways may remain contaminated for a period of possibly up to 100 years, (5) there are no proven methods of decontaminating mercury polluted waterways, (6) there are many mercury polluted waterways in this country, and, (7) the imposition of fishing restrictions, because of mercury pollution, has had a significant economic impact on recreational and commercial fishing industries.

VIII. ACTIONS TAKEN

Pesticides

Under authority of the United States Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA) all pesticides used in interstate commerce must be registered with the Administrator of the Environmental Protection Agency. Under FIFRA two procedures are available for withdrawing registration, cancellation and suspension.

Cancellation is the milder of the two procedures and is used when there is substantial question as to safety of the product. Cancellation is effective thirty days from receipt of official notice unless challenged by the registrant. If the cancellation is challenged then either a statutory scientific advisory committee is convened or a public hearing is held, or both, in order to decide whether or not to affirm the cancellation. Meanwhile the registrant is allowed to market the product in interstate commerce until a decision is reached.

Suspension is a more drastic procedure and is warranted whenever the Administrator determines that continued use of the product presents an "imminent hazard to the public." Immediately upon notice of suspension of registration the registrant must cease all interstate shipments of the product.

During the period 1969-1970 four notices of cancellation were issued:

1. February 1, 1969 - Cancellation of phenylmercuric monoethanol ammonium acetate compounds for use on apples, and strawberries; phenylmercury nitrate compounds for use on almonds and prunes; and phenylmercury urea compounds for use on barley, corn, cottonseed, flax, oats, peas, rice, rye, sorghum, and wheat,
2. February 26, 1970 - Cancellation of phenylmercuric acetate or phenylmercuric ammonium acetate compounds for use on millet, rye, and sugarcane, and
3. March 12, 1971 - Cancellation of hydroxymercurichlorophenol compounds for use on snap beans, sweet corn, cowpeas, flax, peanuts, peas, potatoes, seedlings (transplant bed), soybeans, sweet potatoes, and velvet beans; hydroxymercurinitrophenol-hydroxymercurichlorophenol compounds for use on potatoes and sweet potatoes; methylmercury 8-quinolinolate compounds for use on apples; and phenylmercuric acetate or phenylmercuric ammonium acetate compounds for use on apples, cherries, peaches, strawberries and sugarcane.
4. On August 8, 1970, all mercury products bearing claims and/or directions for use as slimicides or algicides, and for use

in laundering were cancelled. This action was taken because the use of mercury compound which results in water contamination is potentially injurious to man and his environment.

On March 9, 1970 all pesticide products containing alkyl mercury for use as seed treatments were suspended, while on October 7, 1971, algicides for use in swimming and wading pools and for industrial uses were suspended.

Water

Background

On April 2, 1970, the American Embassy at Ottawa, Canada transmitted to the Secretary of State a telegram which contained the text of a note drawing attention to mercury contamination in certain boundary waters, principally Lake Saint Clair. The Federal Water Quality Administration, Great Lakes Regional Office, U. S. Department of the Interior (now part of the Environmental Protection Agency) immediately initiated an investigation of mercury contamination in the Saint Clair River, Lake Saint Clair, Detroit River, and western Lake Erie.

During April of 1970, it became evident that the mercury pollution problem was not limited to the Great Lakes area, but was of national scope. Therefore, on May 1, 1970, the Regional Directors of the Federal Water Quality Administration were directed to identify and measure the existing and potential threat from mercury contamination on a top priority basis. Work was immediately initiated and concentrations of mercury in effluents sediments and biological materials were measured. Known users of mercury were systematically checked to ascertain if they discharged mercury to the aquatic environment. Also natural waters, where suspect, were checked to determine if they were contaminated with mercury. Priority was given to the determination of (1) the extent and intensity of the contamination, (2) whether unacceptable residues in fish might be present, and (3) whether water supplies might be endangered.

Water Supplies

During the period, May 1970 through June 1971, the Division of Water Hygiene of the Environmental Protection Agency, analyzed 698 samples of raw and finished waters collected from the sources and/or treated waters of 273 community, recreational area, and Federal installation water supplies in 31 States, the Virgin Islands, and Puerto Rico. Over ninety percent of the water supplies analyzed were surface or combined surface and ground water supplies.

Table 8 Summary of findings of mercury discharges to waters as of April 26, 1971.

		Number of Dischargers or Users								
Effective Date		Sept. 3	Sept. 17	Oct. 1	Oct. 15	Oct. 29	Nov. 12	Nov. 25	Dec 18 70	Apr 26 71
I.	Known Mercury Dischargers 51	. . . 52	. . 53	. . . 53	. . . 54	. . . 60	. . . 62	. . . 64	. . . 73
	Analysis Positive Discharges greater than									
	sampled for which most recent 1.0 lb/day . . .	14. . .	14. . .	14. . .	14. . .	14. . .	15 . . .	11. . .	13. . .	10. . .
	analysis indicates the pres- 0.5-1.0 lb/day . .	9. . .	9. . .	9. . .	9. . .	9. . .	10 . . .	8. . .	7. . .	9. . .
	ence of mercury. Most recent 0.25-0.5 lb/day . .	2. . .	2. . .	2. . .	2. . .	3. . .	5 . . .	5. . .	6. . .	7. . .
	data shown as well as compar- less than									
	ative findings where there 0.25 lb/day . . .	26. . .	27. . .	28. . .	28. . .	28. . .	30 . . .	38. . .	38. . .	47. . .
	have been reductions in the									
	discharge of mercury.									
II.	Mercury Users - Discharge 36	. . . 36	. . 36	. . . 36	. . . 36	. . . 43	. . . 48	. . . 57	. . . 64
	Analysis Negative Discharges									
	samples for which most recent									
	analysis did not detect the									
	presence of mercury above the									
	concentration of 1 part per									
	billion.									
III.	Mercury Users - No Discharge 43	. . . 86	. . 114	. . 153	. . 173	. . 183	. . . 183	. . . 192	. . . 647
	Mercury users (or potential									
	users) determined not to be									
	discharging mercury on the									
	basis of an on-site inspection.									
Total Analyzed (I & II)		87	88	89	89	90	103	110	121	137
Total Investigated (I, II & III)		130	174	203	242	263	286	293	313	884

Of the 273 community, recreational area, and Federal installation water supplies examined, 261 showed either no detectable quantities of mercury present or concentrations of less than 0.001 ppm in the raw and finished waters. In 11 of the water supplies examined, the mercury concentration ranged from 0.0010 ppm to 0.0048 ppm. One of several samples collected from a large community water supply exceeded the tentative drinking water standard of 0.005 ppm.

Industrial Discharges

In previous programs of the Federal Water Quality Administration of the United States Department of Interior top priority on compiling a list of potential mercury uses and systematically checking them to ascertain if they were discharging mercury into public waters. Investigations and sampling of the potentially serious discharges were initiated in early May of 1970, and by the end of June and in early July, sound evidence had been developed to support regulatory actions. On July 14, 1970, the Secretary of the Interior acting under authorities of the Federal Water Pollution Act forwarded telegrams to the Governors of 17 States in which mercury pollution was suspected and urged the Governors to act vigorously in eliminating known discharges of the metal. The Secretary also announced that he would notify all industries across the nation which are shown by investigations and data to be responsible for mercury pollution and that court action will be sought in any confirmed cases of mercury pollution if corrected measures are not taken swiftly on local levels. On July 22, 1970 the Secretary of the Interior announced that he was submitting the names of United States industrial firms to the Justice Department for possible prosecution on the charge of discharging mercury into the Nation's waterways in sufficient quantities to constitute a serious hazard to public health.

On July 24, 1970, it was announced that the Justice Department was filing charges against 10 United States industrial plants which are discharging mercury into the Nation's waterways. Subsequently, stipulations to reduce mercury discharges to less than 1/2 lbs per day and requiring plans for further reductions by December 1, 1970 were entered in the courts in 9 cases.

In August of 1970, the Federal Water Quality Administration, Department of the Interior, (now part of the Environmental Protection Agency) initiated a series of administrative meetings with mercury discharges at both the Headquarters and Regional level to establish abatement programs. Most of these meetings have produced agreements at least equivalent to those achieved by the Justice Department in the courts. High intensity investigation of newly discovered potential discharges and a continuing process of resurvey of known discharges have continued to the present.

As of April 26, 1971 the Environmental Protection Agency has investigated 834 potential mercury discharges (Table 8). The most recent analyses

available indicates that 73 sources were discharging mercury to the aquatic environment while 64 sources the presence of mercury above the concentration of 1 ppb were not detected (Table 3). On the basis of an on-site inspection, 647 potential mercury discharges were determined not to be discharging mercury to the aquatic environment. These investigations clearly show that one of the major sources of mercury to the aquatic environment is the chlorine industry. Of the 73 sources presently discharging mercury to waterways, 31 are from chlorine manufacture (Table 9). Other major sources of mercury to the aquatic environment are chemical and pesticide manufacture, dye manufacture, and mercury reclaiming (Table 9).

Wastewater Treatment Plants

Because it was determined that mercury seals on rotary-type trickling filters could be a source of mercury to wastewater treatment plants, the United States Environmental Protection Agency initiated a program on October 29, 1970, in cooperation with State water pollution control agencies to achieve the total replacement of such seals in municipal and industrial sewage plants. The Federal Agency also notified state water pollution control agencies that the use of mercury seals in trickling filters was no longer considered acceptable for any reason. In order to eliminate this hazard at Federal installations, the United States Environmental Protection Agency requested on February 26, 1971 that each Federal agency conduct a survey of its sewage treatment plants to identify rotary distributor trickling filters equipped with mercury seals and develop a schedule for early elimination of these seals in both Federal and State agencies.

Air

Pursuant to the authority contained in the Clean Air Act, the United States Environmental Protection Agency on March 31, 1971, designated mercury as a hazardous pollutant for which hazardous emissions standards will be promulgated. Proposed emission standards for chlorine production and primary processing of mercury - bearing materials were published on December 7, 1971 and are scheduled for promulgation within 180 days thereafter. The proposed regulations would limit mercury emissions to 5 lbs. per days.

In order to intelligently reduce the emissions of mercury to air under the United States Clean Air Act the Environmental Protection Agency will take the following actions; 1) conduct a more thorough and detailed identification of emission sources; 2) develop accurate emission factors for each source category based on actual source sampling measurements; and 3) adapt or develop control systems adequate to reduce emissions below a level which would be hazardous.

Table 9 Types of industrial discharges to the aquatic environment for which the most recent analyses indicates the presence of mercury as of April 26, 1971.

Source of discharge	Number
Chlorine manufacture	31
Chemical & pesticide manufacture	6
Dye manufacture	5
Mercury reclaiming	4
Pulp and paper manufacture	3
Instrument manufacture	3
Laboratories	3
Aluminum industry	2
Lamp & light bulb manufacture	2
Plastic manufacture	2
Paint manufacture	2
Gold mining	1
Manufacture of switches	1
Manufacture of meters & pumps	1
Pulp and paperboard manufacture	1
Manufacture of catalysts	1
Manufacture of batteries	1
Manufacture of ammunition	1
Manufacture of magnets	1
Plastic fabrication	1
Sugar mill	1
Total	73

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