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MERCURY in the ENVIRONMENT

THE HUMAN ELEMENT

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OAK RIDGE NATIONAL LABORATORY

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Robin A. Wallace • William Fulkerson
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all you may ever want to know about

MERCURY

wherein are provided brief but sundry accounts of adverse effects and unhappy circumstances relating to mercury

IN THE ENVIRONMENT,

and also containing much matter to exercise the judgment and reflection of the reader concerning the imperfections of our knowledge, the wisdom of our regulations, and particularly the role of

THE HUMAN ELEMENT

in the dissemination of mercury, and other grave and curious observations which those readers will like best who understand them most.

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(January 1971)

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CONTENTS

Abstract v

1. Foreword 1

2. Societal flow of mercury in the U.S. during 1968 2

3. Sources of environmental mercury 4
   - A. Man-made 4
   - B. Natural 6

4. Mercury levels 6
   - A. Environmental 6
     - i. Air 6
     - ii. Water 7
     - iii. Rocks, soils, sediments, and sludges 8
     - iv. Fossil fuels 9
   - B. Biological 9
     - i. Plants 9
     - ii. Animals and animal materials 10
     - iii. Man 13

5. Forms and transformation of mercury in the environment 15

6. Turnover, translocation, and accumulation of mercury in plants and animals 17
   - A. Plants 17
   - B. Aquatic organisms other than fish 18
   - C. Fish 18
   - D. Other animals and man 19

7. Criteria for evaluating mercury levels 21
   - A. Toxicity to aquatic organisms 21
   - B. Toxicity to man 21
     - i. Individual incidents 21
     - ii. Epidemiological incidents 22
     - iii. Industrial incidents 23
     - iv. Medical observations 23
   - C. Biological studies 24
   - D. Determination of an allowable daily intake (ADI) for mercury 25
   - E. Economic and social costs of mercury pollution 25

8. Existing standards and tolerance limits 27
   - A. Air 27
   - B. Water 28
   - C. Food 28

9. Appendices 29
   - A. Some physical and chemical properties of mercury 29
   - B. Analytical methodology for environmental mercury 31
     - i. Sampling and sample treatment 31
     - ii. Analytical methods for determining mercury 32

C. The mercury cell chlor-alkali process 35

D. Some recommendations for further work 37
i. Economic and social factors related to mercury usage 37
ii. Research in analytical measurements 37
iii. Restitution of mercury-polluted lakes and streams 38
iv. Evaluation and elimination of atmospheric mercury pollution 38
v. Mercury levels in water and fish 39
vi. Genetic and long-term effects of mercury exposure 39
vii. Site and mode of action of methylmercury 39
viii. A protective agent for methylmercury poisoning 40

10. References 41

A. Specific 41
B. General 47

* * * * *

Tables

1. Known mercury dischargers in the United States (as of September 1970) 48
2. Maximum mercury concentration in air measured at scattered mineralized and nonmineralized areas of the western United States 50
3. Mercury in selected rivers of the United States, 1970 51
4. Analysis of soils for mercury in ppm 52
5. Mercury levels in water sediments 53
6. Analyses of fossil fuels for mercury in ppm 53
7. Mercury levels in animals from various areas in the United States 54
8. State fishing restrictions because of mercury (as of September 1970) 55
9. Analysis of Canadian birds, fish, and animal products for mercury in ppm 56
10. Analysis of human tissues for mercury in ppm 57
11. Daily intake of mercury and mercury concentrations in blood and hair of man 58
12. Total mercury and methylmercury contents of Swedish foods in 1966 58
13. Total mercury and methylmercury content in the axial musculature of fish caught upstream and downstream from industrial outfalls 58
14. Effect of a paper mill on the mercury content in aquatic organisms 59
15. Content of mercury in organs of pike specimen in ppm 59
16. Lethal concentrations of mercury compounds for various aquatic organisms 60
17. Some physical properties of metallic mercury 60
18. Sources of mercury losses from the chlor-alkali process 61
19. Schemes for decontamination of mercury-polluted lakes 61

ABSTRACT

This review is directed toward an understanding of the uses, sources, distribution, and toxic effects of mercury, and the main emphasis is on man as the distributor and recipient of mercury in the environment. The societal flow of mercury is initially considered; the U.S. demand is divided into dissipative and potentially recyclable uses, and several important externalities to materials operations are identified. The main conclusion is that there is a need to pay more attention to the social implications of present use practices and to recycle much more effectively or develop alternatives to present mercury technology. The principal man-made sources of environmental mercury in the U.S. are chlor-alkali plants, smelting operations, and probably the burning of fossil fuels, whereas the principal natural sources are the land mass itself and geothermal processes.

Mercury concentrations found in environmental materials are very difficult to assess, and a discussion of the problems encountered is provided. Data for "background" and "contaminated" situations in air, water, rocks, soils, sediments, sludges, fossil fuels, plants, animals, foods, and man are drawn together and briefly evaluated. In a consideration of the transformation of mercury in the environment, it is concluded that mercury, in whatever form, is potentially exchangeable among the air, land, and water phases, and that it represents a unique pollutant because it is essentially indestructible. Swedish studies have indicated that "abnormal" concentrations of mercury in terrestrial animals appeared concomitantly with the introduction of alkylmercury seed dressings in agriculture during the 1940's, but that mercury levels in fish have steadily risen over the past 100 years in parallel with increased industrial activity. It has also been found that whatever the source and compound of mercury introduced into an aquatic system, essentially only the highly toxic methylmercury form is found in the flesh of fish. The formation of methylmercury appears to be promoted by bacterial processes occurring within contaminated bottom sediments, and such sediments can slowly release their mercury burden as methylmercury for many years to come. Thus, a major problem for the future is the decontamination of sediments even after overt pollution has ceased.

Details are provided regarding the toxicity of mercury and its compounds. Alkylmercurials primarily affect neurological processes because of their ability to cross the blood-brain barrier, and they appear to be several orders of magnitude more toxic than other mercury compounds. Mercury vapor is intermediate between the alkylmercurials and other mercury compounds in its effects, again because of its partial ability to cross biological membrane barriers and enter cells. The exact mechanism whereby alkylmercurials affect neural cells is not understood, but part of their pronounced toxicity is due to their lengthy retention by the body. No effective antidote is presently available for these compounds. The criteria for mercury levels are based almost entirely on relatively short-term effects, and virtually no information is available on the long-term results of sub-acute exposure to mercury. Nevertheless, tentative standards and guidelines have been set by several organizations and U.S. Government agencies for mercury in air, drinking water, and food, and these are provided along with some background information.

Finally, appendices are included which consider aspects of the physical and chemical properties of mercury, evaluate analytical procedures used for mercury determination, discuss the chlor-alkali manufacturing process and procedures for reducing mercury losses, and provide some present and future needs with respect to our understanding and dealing with mercury in the environment.

1. FOREWORD

Then it became apparent that we had a brand new ballgame. Consider an area like Silver Lake. It is located in the Green Mountain National Forest, atop a mountain and accessible only by four-wheel drive vehicles or by pack trip. It has no industry whatever -- but it does have mercury contamination.

When Joe's Pond in Danville, Vermont produces fish contaminated eight times beyond the level set by the FDA, *we are warned that the issue is far more complex than we had supposed.* Presently, fish from all bodies of water 20 acres or larger are being analyzed. In almost every case, the findings show substantial amounts of mercury poisoning.

Vermont is not a heavily industrial state, and has none of the chemical industries identified as the prime polluters. The orchards discontinued use of mercury years ago. So did most milk testing plants and paper processors. So where does the mercury come from?

What we have learned about mercury recently indicates that what we see and know about pollution is not as frightening perhaps as the unknown and unseen.

– Senator W. Prouty (1)

The surge of technological progress over recent years has provided many benefits for mankind and, albeit with some inequities, has permitted his number to grow, feed, clothe, and house themselves as never before. Technological progress set within the framework of an interacting environment has not been without mixed blessings, however, and we are just beginning to appreciate the potential effects of what Rene Dubos has described as our "ability to modify the earth and shape it, thus determining the evolution of our own social life through a continuous act of creation." Too often we have found that an advance in one sector of our lives has led to difficulties in another sector; too often our consumer-oriented technology has produced environmental dangers which were not anticipated. However, in a world in which individuals and nations are competing for trade, goods, economic security, and even the essential elements of human life, it has become impossible to turn back the clock to a simpler and perhaps less complicated time. Rather, we must place increased emphasis on our scientific and legislative abilities to find and to enact judicious solutions for many of our present difficulties.

This review is directed toward an understanding of one of those stepchildren of our advancing civilization: mercury in the environment. Its uses, sources, distribution, and toxic effects are presented, and the tolerance limits with which authorities have wrestled in order to cope with mercury contamination are briefly discussed. The emphasis throughout is primarily on man as the distributor and recipient of mercury in the environment. In writing the review, we have attempted (perhaps with only partial success) to avoid most of the jargon and tunnel vision of the specialist and yet to refrain from handwringing over birds, bunnies, and babies. The reader is assumed to be the intelligent layman, but appendices and references are provided to enable a professional pursuit of specifics. As will be seen, knowledge and understanding of many matters is still rudimentary, but we hope that this summation will help partially reveal "the unknown and unseen" and serve as a guide for action by indicating those efforts which must be undertaken to solve the problems.

We are indebted to the Environmental Mutagen Information Center, Biology Division, for much of the information contained in this report, and to those many librarians and individuals, especially Azolene Vest, John Wassom, and Arietta Wyrick, who have helped us obtain the source materials. We have relied particularly on the articles written by Berlin *et al.* (2), Greenspoon (3), Löfroth (4), Seagran (5), Smart (6), and members of the U.S. Geological Survey (7). Finally we are grateful for the many helpful comments and criticisms provided by John Gibbons (ORNL-NSF Environmental Program), John Gilbert (Biology Division), Irving Spiewak (Reactor Division), Richard Brown (Physics Department, State University of New York at Albany), and Anthony J. Smith (Tennessee Valley Authority at Muscle Shoals, Alabama).

The less time all of us spend talking among our colleagues in endless conferences and meetings and the more time we spend working with each other and the public, the better our prospects will be for a livable world.

– Senator E. S. Muskie (8)

2. SOCIETAL FLOW OF MERCURY IN THE U.S. DURING 1968

The quantity of mercury used each year in the world increased at a rate of about 1800 flasks per year for the ten-year period previous to 1968 (9) (a flask contains 76 lb of mercury and is the standard commercial unit of trade). Most of this increase was due to the U.S. demand, which grew at a rate of 1600 flasks per year over the same period (10) until in 1968 it represented about 30% of the world primary production of 257,000 flasks.

In order to illustrate the sources, consumption, and factors which would enter into the recycle potential of this relatively large amount of valuable yet hazardous metal, a highly simplified flow diagram for the U.S. in 1968 is provided in Fig. 1. The flows are given as percentages of the total demand of 75, 400 flasks (3,11). The rectangular boxes in Fig. 1 represent processes, material manipulations or summations, and direct purchased inputs to or outputs from materials operations. The ovals, on the other hand, represent unpurchased inputs or unsalable outputs or residuals. Although by no means a complete set, they do represent an initial attempt to identify important externalities to the various materials manipulations which can have a significant effect on the flow and especially on recycle. For example, a 23% depletion allowance and/or prospecting subsidies are unpurchased inputs to the U.S. mining operation which tend to keep the price of mercury produced from domestic virgin ore down and consequently have a negative effect on recycle. Similarly, the effluents from the mining operation (tailings, overburden, mercury vapor and sulfur dioxide air pollution) are unsalable outputs, the cost of which is borne by society in general. The low import duty of about 3% [scheduled for further reduction extending into 1972 (3)] again favors the use of mercury from low-priced foreign ore in preference to recycled material. Also involved is the economic-political issue of balance of payments deficit, which is an indirect cost to the importing operation.

In 1968, then, the U.S. supply of mercury came from four sources: 18% from recycled material; 36% from U.S. mining; 22% from net imports; and 24% from releases from Government stockpiles.* The U.S. demand for mercury has been broken down into recyclable and dissipative uses, which account for 74 and 26% of the total respectively. Recyclable uses are defined as those for which it is technically feasible to recycle, whereas for dissipative uses it is not.

The major use of mercury is as a cathode in the electrolytic preparation of chlorine and caustic soda (see section 9C). Actual consumption in this manufacturing process is small for each unit, although large quantities are required for new installations (10% of 1968 demand). Because of the many plants now in operation, however, the requirement to replace losses has become a major use (13) (23% of 1968 demand, indicated as unpurchased output in Fig. 1). Mercury cells account for 30 to 35% of the present chlorine production, and the U.S. Department of Commerce has estimated an annual 7% growth rate in chlorine production through 1974, with approximately 50% of the increased production capacity based on mercury cell technology (14). Technological innovations in a new make of mercury cell, however, will reportedly decrease the amount of needed start-up mercury by about 40% (15). Other potentially recyclable uses of mercury are for electrical apparatus (lamps, arc rectifiers, and particularly mercury battery cells), industrial and control instruments (switches, thermometers, barometers), and in general laboratory applications.

The largest dissipative use of mercury is for antifouling and mildewproofing paints. Mercury compounds are also widely used in agriculture as a result of their broad antifungal capabilities. Many fungus diseases of seeds, bulbs, plants, and vegetation are controlled by mercury formulations (16) (Table I in ref. 6). Mercury

*The General Services Administration strategic stockpile is set at 200,000 flasks, although this is expected to be reduced to 125,000 (12); the Atomic Energy Commission stockpile quantity is classified, but releases are made from time to time.

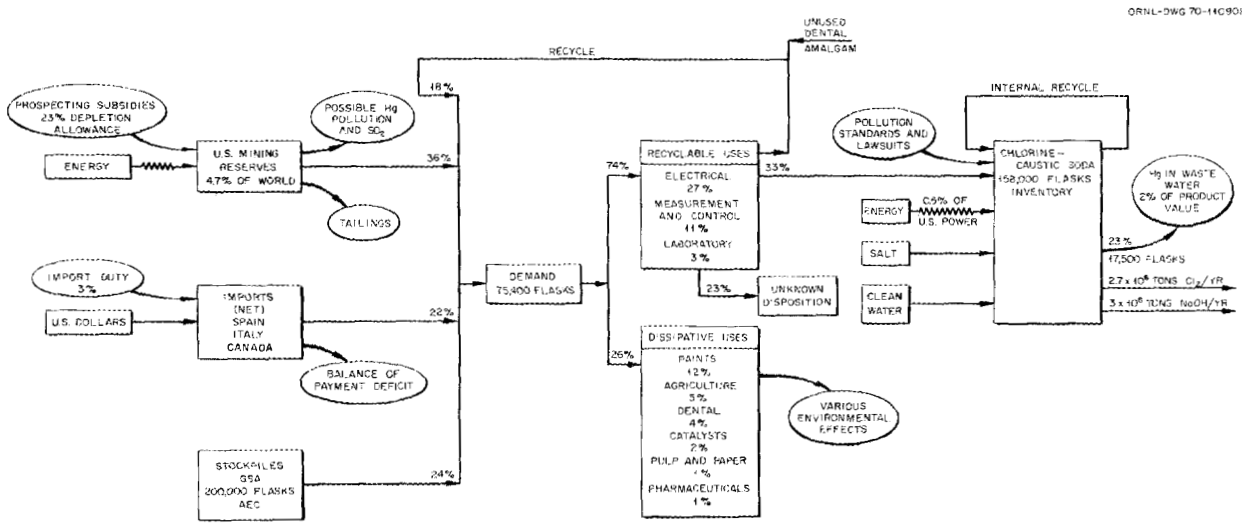

Fig. 1. Mercury flow through U.S. society (1968).

is extensively used in dental amalgams, for catalytic purposes, and, to a decreasing extent, for paper and pulp manufacture. Mercury compounds are also formulated into many of the over-the-counter cosmetics (creams and lotions, hair preparations, facial make-up) and patent medicines which are available. Some of the latter mercury-containing products are presently sold as antacids, astringents, eye drops, laxatives, nasal sprays, skin antiseptics, contraceptives, vaginal douches and suppositories, and rectal suppositories (see ref. 17 for an old listing of approximately 160 products).

* * * * *

As a final consideration, it should be pointed out that the United States resources of mercury are about 4.7% of the total world resources, based on ore which contains more than about 0.05% mercury (18). In 1968, the ore produced in the U.S. contained 0.275% mercury by weight on the average (3). Fig. 2 gives the estimates of U.S. supply of virgin mercury as a function of price. The 0.05% ore can be mined economically at about $800 per flask. Even if the price per flask were $1500, however, U.S. resources would only be about 1,500,000 flasks. Assuming our yearly demand for mercury does not change over the next 30 years, and assuming 20% recycle, we will still require a total of 1,600,000 flasks of mercury from virgin ore by the year 2000. The necessity of our dependence on foreign supply thus seems obvious *unless we can recycle much more efficiently or develop alternatives to present mercury uses and practices.*

As examples of alternate technology (19), other processes, including the use of diaphragm cells, are available to replace those using mercury as a cathode in the electrolytic preparation of chlorine and caustic soda. For other industrial and agricultural purposes, copper and numerous organic chemicals may be substituted for mercury compounds. For some dental uses metal powders, porcelain, and plastic materials are preferred to mercury amalgams. In the pharmaceutical field, sulfa drugs, iodine, and various antiseptics, disinfectants, and contraceptives may be used instead of mercury chemicals. However, there are few satisfactory substitutes for those applications in electrical apparatus and in industrial and control instruments that depend on mercury's high specific gravity, fluidity at normal temperature, or electrical conductivity.

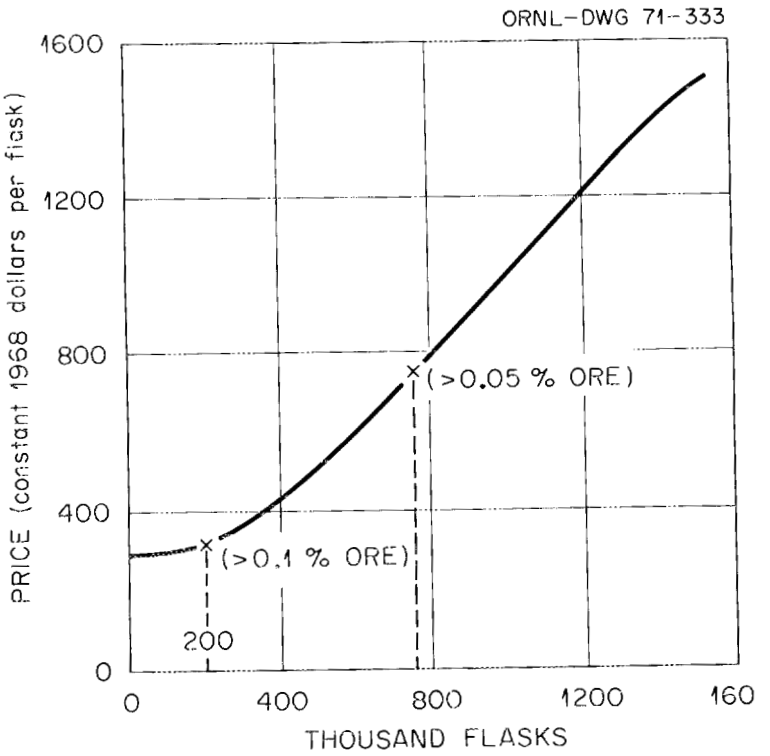

Fig. 2. Projected resources of mercury. From Greenspoon (3).

3. SOURCES OF ENVIRONMENTAL MERCURY

A. Man-Made

Commercial discharges of mercury into the environment can be deduced from data on use and inventory losses and are incorporated into the societal flow diagram (Fig. 1). By far the single largest source during 1968 was derived from inventory losses suffered by the chlor-alkali plants (20). During the summer of 1970, however, lawsuit threats and tentative water standards ("unpurchased inputs") caused many of these manufacturers to reduce their discharge of mercury by a factor of 10 to 100. These reductions were monitored by the Department of Interior, which found that overall the level of mercury emission dropped 86% from 287 lb per day in July to 40 lb per day in September (21). In this survey of 129 companies of all types that were "known to use mercury," 79 were found to emit no detectable mercury discharges, while the remaining 50 firms (Table 1) were classified as polluters of rivers, streams, and lakes.

The societal flow diagram (Fig. 1) also indicates that of the mercury used in 1968 for other potentially recyclable uses, such as electrical equipment, measurement and control apparatus, and general laboratory use, 520 tons were recycled and 660 tons had an unknown disposition. Some of the latter went to increased inventories, but probably much of it (in batteries, fluorescent tubes, switches, etc.) ended up in landfills, dumps, and incinerators, and some of it was discovered in the survey by the Department of Interior (Table 1). Known users of mercury which were not covered by this survey were laboratories (including hospitals). These institutions may be another but so far overlooked source of mercury pollution (22).

The dissipative uses of mercury listed in Fig. 1 include paints, agriculture, dental fillings, catalysts, paper and pulp manufacture, and pharmaceuticals, a total of 26% (745 tons) of the mercury demand in

1968. Mercury from these various uses enters the environment in a variety of ways and at different rates, and not all will produce the same consequences. For example, although mercury-formulated paints constitute a considerable source, their release into the environment is rather slow and probably has a minimal effect. Dental amalgams also apparently have no measurable effect on the user (23). On the other hand, mercury catalysts employed for acetaldehyde and vinyl chloride manufacture and mercury formulations used as seed dressings have promoted serious consequences (sections 4Bii and 7Bii), even though such usage represents a small percent of the total consumption. In February–March 1970, however, the U.S. Department of Agriculture suspended 42 alkylmercury compounds from interstate commerce, and, after subsequent litigation, a U.S. Court of Appeals upheld the suspension in November (24). Barring further appeals, the full environmental impact of these actions will not occur until the 1971 growing season.

Mercury compounds employed as slimicides in the pulp and paper industry have historically polluted lakes and rivers around the world, and in several countries they are still extensively used. In the U.S., fortunately, less toxic substitutes caused this use to decrease drastically in the early 1960's so that it now amounts to well less than 1% (14 tons in 1968) of the total demand. This decreased use was spurred by a Food and Drug Administration ruling concerning paper products which come in contact with food (11), another "unpurchased input" which significantly reduced pollution even though it was not the purpose of the government action in this case.

Other man-made sources of environmental mercury not covered or adequately measured by societal flow considerations may be equally or even more important. This would include first of all such "unsalable outputs" as the mine tailings and vapor released by the mining and smelting of mercury. Although these sources are difficult to quantify, nevertheless, high stream and lake water levels of mercury have been attributed to dump material and tailings (25,26). Also during the refining process, ore is heated to liberate mercury as a vapor, which is then collected in condensers. Stack losses during the process should not exceed 2–3%, although much higher losses have occurred (27). A stack loss of 3% would mean that about 31 tons were emitted into the atmosphere from smelting during 1968.

It is now well established that ore deposits of heavy metals are generally surrounded by aureoles in which a notable enrichment in mercury has occurred (7,26). Although significant amounts of mercury have occasionally been recovered as a by-product of tin, zinc, copper, and gold production (28,29), it would appear that considerable mercury generally escapes from stacks during the smelting of such metals (7). As an example, zinc concentrates from Tennessee contain an average of 5–10 ppm (parts per million) mercury (30); thus a single smelter handling 500 tons per day might emit daily as much as 10 lb mercury vapor.

Another source of airborne mercury is the combustion of paper products and fossil fuels (31,32). The ground wood pulp used for paper manufacture exported by the U.S., Canada, Sweden, and Norway has been found at various times to contain high levels of mercury (6). At the recent International Conference on Environmental Mercury Contamination (33), the question of mercury emission from the burning of fossil fuels was raised. The general consensus was that fly-ash from coal-burning plants contained negligible amounts of mercury, so that any mercury present in coal must be released to the atmosphere during the combustion process. The actual amounts of mercury in fossil fuels are quite variable (see section 4Aiv), but preliminary values for U.S. coals average from 0.5 to 3.3 ppm mercury. Since about 550 million tons of coal is burned in the U.S. every year, such a level would correspond to an annual release of anywhere from 275 to 1800 tons of mercury. No information is available on the mercury released during petroleum processing and oil burning. It is clear, however, that even though the mercury content of fossil fuels may be low (which frequently it is not), the enormous amounts of these substances consumed and burned in the U.S. each year contribute a substantial amount to the environmental mercury burden.

B. Natural

Mercury has a tendency to vaporize, so that the air over mercury ore deposits and precious metal or copper ore deposits generally contains elevated levels of mercury (7). Prospecting methods for these metals have even been developed which take advantage of this "natural" air pollution (34,35). Small amounts of mercury are also found over non-mineralized land areas (7), while insignificant amounts are found over the ocean (36); this would suggest that the land area is a major source of airborne mercury.

Rain washes mercury from the atmosphere, and calculations for the Swedish land mass have indicated that rain annually returns to the soil approximately 0.5 grams mercury per acre, which is about the same amount as that contributed by the agricultural use of seed-dressing formulations (37). Most of the mercury deposited by rainfall apparently becomes bound to the upper several inches of soil, from which it can again be released into the air. However, erosion of mercury-containing soil and leaching of soluble mercuric salts by rainfall can carry mercury to streams by runoff and groundwater discharge (7).

Relatively high concentrations of mercury are likely to occur in underground waters because of the longer and more intimate contact with mineral grains and other chemical factors. Oil field brines, hot springs, and geothermal stream fields have been associated with high mercury levels (7). Hot vapors which stream up through fine-grained muds produce mud volcanoes or "mud pots" and deposit considerable quantities of mercury during condensation (7). Many thermal waters, such as Sulphur Bank, California, Amedee Springs, California, Steamboat Springs, Nevada, and Boiling Springs, Idaho, are depositing mercury-rich sediments (7). Sulphur Bank is the most remarkable of the four, having produced more than 5,000 tons of mercury before mining operations ceased (38). The contribution of volcanic eruptions to environmental mercury are unknown, although this may represent a significant source (7).

4. MERCURY LEVELS

A persistent problem in evaluating mercury levels has been the difficulty in handling, storage, and sampling of materials (see section 9B), and many values reported below may be inaccurate by as much as a factor of 10. Also, in the data provided, no attempt has been made to distinguish the form of mercury present. Generally, however, air values refer to metallic mercury vapor, levels for animal tissues relate primarily to methylmercury-bonded materials, and values for other substances (water, earth materials, plants) refer to a variable mixture of metallic and ionic mercury.

A. Environmental

i. Air. Data on atmospheric mercury levels collected on clear days by the U.S. Geological Survey are given in Table 2 (7). Concentrations in air over non-mineralized areas varied from 0.003 to 0.009 $\mu g/m^3$ (micrograms per cubic meter of air, approximately equivalent to parts per billion), while those over copper, precious metal, and mercury mines were considerably higher, ranging up to 20 $\mu g/m^3$ at ground level. Concentrations found over oceans generally measure less than 0.001 $\mu g/m^3$ (35). The land surface sources of airborne mercury are apparently subject to meteorological controls; i.e., mercury release into the air is a function of barometric pressure, time of day, and season (7), with the latter two primarily being temperature functions. Immediately after a rainfall, mercury concentrations are negligible, even near ore deposits (7).

Air over urban industrial areas contains mercury levels higher than background, and several measurements (39,40) have indicated average values of 0.01 (Chicago, Illinois), 0.10 (Cincinnati, Ohio), and 0.17 (Charleston, West Virginia) $\mu g/m^3$. Concentrations of 1–14 $\mu g/m^3$ have been cited for New York City

(41). Some of the most careful recent measurements have been made at a continuous monitoring station located on the west side of San Francisco Bay in California (36). During days in which southwesterly winds brought air in from over the Pacific, levels averaged 0.002 $\mu g/m^3$ (Fig. 3*a*) or even less if breezes were strong enough. Southeasterly winds coming from over generally non-industrial land masses contained higher concentrations (Fig. 3*b*), whereas northeasterly winds from industrial areas provided high and erratic readings, averaging 0.008 $\mu g/m^3$ but with peaks over 0.020 $\mu g/m^3$ (Fig. 3*c*). Mercury rapidly adsorbs onto most surfaces, and much of the atmospheric mercury reported above has been found to be associated with dust and other particulate matter.

Air concentrations of 100 $\mu g/m^3$ or larger have been reported within industrial buildings such as chlor-alkali plants (2,42), workshops for thermometers and the repair of direct current meters (43,44), and, in the past, around work stations in the hat-felt industry (45,46). Concentrations in mines may range up to 20,000 $\mu g/m^3$ and in the air of ore processing plants up to 970 $\mu g/m^3$ (47).

ii. Water. Sea water contains anywhere from 0.03 to 2.0 ppb (parts per billion) mercury, depending on area, depth, and the analyst (48–53). In a generally accepted study of Pacific waters (50,51), mercury concentrations were found to increase from surface values around 0.10 ppb to 0.15–0.27 ppb at greater depths. The depletion of mercury in surface waters was attributed to its uptake by plankton and subsequent conveyance to depths by the biological activities of the marine food web. In an area seriously affected by pollution (Minamata Bay, Japan), values ranged from 1.6 to 3.6 ppb (54). Oceanic mercury is

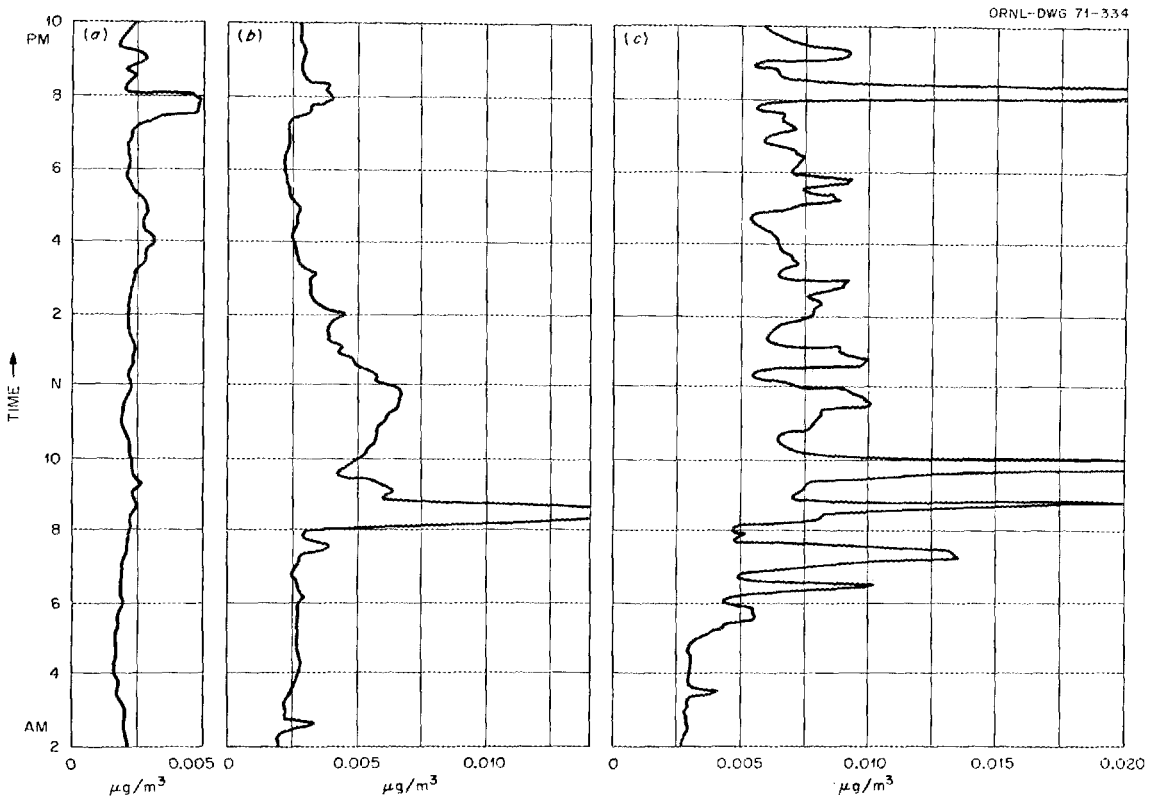

Fig. 3. Mercury vapor concentrations in the air over Los Altos, California, when the prevailing wind current is (*a*) coming from the ocean, (*b*) coming from the southeastern land mass, (*c*) coming from the northeastern industrial area. From Williston (36).

generally present as an anionic complex ($HgCl_4^{2-}$), which does not have as pronounced a tendency to bind to particulate substances and subsequently settle out as do mercury compounds found in freshwater situations (26).

Normal ground water is considered to contain 0.02–0.07 ppb mercury (48,55). Data on surface waters in 31 states of the U.S. were collected during the summer of 1970 by the Geological Survey in cooperation with the Federal Water Quality Administration and are presented in Table 3 (7). Of the total samples from 73 areas, 34 contained less than the detectable (0.1 ppb) limit. Of the remainder, 27 samples contained 0.1–1.0 ppb, 10 samples ranged from 1.0 to 5.0 ppb, and only two samples contained more than 5.0 ppb. The fact that many of the samples were taken in areas of suspected mercury contamination would indicate that mercury concentrations in surface waters generally do not exceed "tolerable" limits (5 ppb) except perhaps in the immediate vicinity of waste outfalls. Other rivers draining basins having worked or unworked mercury deposits are known to have mercury levels exceeding 5 ppb (25,56), and values typically range from 10 to 80 ppb (25). Mercury was generally found associated with suspended particles, however, and levels were found to decrease markedly downstream from the deposit.

Rain, which serves to scrub mercury from the air, contains about 0.2 ppb mercury (48). Hot spring waters have frequently been shown to deposit mercury-rich sediments even though water levels are not particularly high. As examples, the waters of Sulphur Bank and Amedee Springs in California contain 1.5 and 2 ppb, respectively (7). The chemistry of a particular water body plays an important role in the leaching of mercury and its compounds and their subsequent solubility (7,26). Notably high levels (6–400 ppm) have been found in brines associated with petroleum or from geothermal wells (57,58).

In general, it would seem that the concentration of mercury in water is a poor indication of contamination, since both the solubility of mercury and many of its compounds (particularly mercuric sulfide) is low and sorption to particulates with subsequent settling out is the general rule. On the other hand, the influence on overlying water of various kinds of bedrock which naturally contain minute amounts of mercury has not been sufficiently studied, and for the moment there is no evidence to prove or disprove that mercury levels in large water bodies may be influenced by such factors. This is still thought to be a distinct possibility in lake areas overlying mineralization, clay sediments, or black shale (26).

iii. Rocks, soils, sediments, and sludges. The average concentration of mercury in the earth's crust is only about 0.07 ppm (59). It is therefore less abundant than such metals as platinum, uranium, and silver, but since it can be found in highly concentrated ores, it is more readily obtainable (27). The enormous amount of data on the relative mercury contents of various earth materials has been recently gathered together and published by the U.S. Geological Survey (7). Mercury concentrations in broad categories of rocks range from 0.01 to 20 ppm. Igneous rocks (those formed by melting and cooling) generally contain less than 0.20 ppm and average 0.10 ppm. Sedimentary rocks deposited by physical, chemical, and biological processes generally average less than 0.10 ppm and seldom exceed 0.20 ppm except for organic-rich shales, which may reach concentrations of 10 ppm or more. The general picture is complicated by the fact that certain areas of the world (notably the Donets Basin, Kerch-Taman area, and Crimea, U.S.S.R.) show extremely high mercury levels in nearly all rocks analyzed (100 times normal contents or more). The Colorado Plateau area in the U.S. is somewhat similar in this respect (7).

Available analyses on natural soils are provided in Table 4. Background levels in many soils appear to range under 0.10 ppm except in areas of mineralization, the anomalous Russian areas previously indicated, and some English garden soils. The actual content of mercury in soils is influenced by a variety of natural and artificial factors. Soils with a high humus content will tend to bind and retain mercury more avidly (60). Pollution from smelters (32) and from the agricultural use of mercurials also occurs. As examples of the latter, rice paddy field soils where organic mercury compounds are applied as dusts contain elevated

levels of mercury (61), and 2.6 ppm mercury was found in soil treated once with mercuric oxide to control potato root eelworm (62).

Because of mercury's tendency to adsorb readily onto a variety of materials, particulate matter suspended in water and bottom sediments of streams are more likely to contain higher concentrations of mercury than the water itself, whatever the source may be. Sediments immediately downstream of ore deposits or contaminated industrial discharges may contain from 0.2 to 200 ppm mercury (7). Concentrations recently found by the Federal Water Quality Administration in sediments around Lake Erie as well as other places are presented in Table 5. It has become apparent that literally tons of mercury have accumulated in river and lake sediments within the U.S.

As a final note of discouragement to those who have urged the agricultural use of sludge from sewage treatment plants, such materials also appear to be frequently contaminated with mercury. Several sets of data from Sweden indicate concentrations up to 120 ppm (63) and from 0.8 to nearly 40 ppm (64), with high levels being associated with urbanized industrial areas. Comparable findings have been made in Michigan (33), although testing in this case was performed on mud samples below sewage outfalls. The mercury concentration in marine sediments near the Los Angeles sewer outfall has also been recently found to be as much as fortyfold higher (0.82 ppm) than presumably uncontaminated sediments further away (0.02 ppm) (65). Some of the contaminating mercury may be derived from the trickling seals, flowmeters, and switches that many of the sewage plants use (33). However, high mercury levels in the air over urban areas (section 4Ai) would suggest that much of the contamination is derived from industrial sources.

iv. Fossil fuels. Throughout eons of time, the products and residues of geochemical processes and the life cycles of countless terrestrial and aquatic organisms have combined to yield the fossil fuel deposits upon which the world depends for much of its energy. The data on mercury levels in such products are meager and unrepresentative, but what are available have been culled from a variety of sources and listed in Table 6. Values for most coal range all the way from 0.001 to 33.0 ppm; extraordinary values are again found for products of the anomalous Russian areas. The data on petroleum products indicate that they may also occasionally contain high levels of mercury, with the Cymric crude oils from California (Santa Barbara area) ranging from 1.9 to 21 ppm. This would be in the range of elemental mercury solubility in hydrocarbons (66). The natural gas from the Cymric field is also saturated with elemental mercury. During transport in the pipeline, however, mercury vapor evidently combines with hydrogen sulfide from the "sour" gases of other fields and is precipitated in the pipelines (7).

B. Biological

i. Plants. All vegetable materials naturally contain traces of mercury, the actual amount depending on the locality from which the sample was taken, its species, and other factors. In pome fruit, the background levels are normally 0.04 ppm or less; in tomatoes, up to 0.02 ppm; in potatoes, up to 0.01 ppm; and in most wheat and barley, up to 0.08 ppm (6,67,68). However, exceptional values (68,69) have been found for wheat from Japan (0.15 ppm), France (0.25 ppm), and Canada (0.15–0.40 ppm). Rice from most parts of the world contains 0.015 ppm mercury or less (70), but higher values (68,69,71,72) have been found in samples from the U.S. (0.05 ppm), Japan (0.06–0.24), and Canada (0.21 ppm).

Plants take up and concentrate mercury to a variable extent. Marine algae, for instance, have been found to range from 0.023 to 0.037 ppm (73), which is several hundred times the accepted concentration for sea water (50,51). Trees and shrubs growing near known cinnabar veins contain up to 3.5 ppm mercury (7). Rice has also been shown to reflect the paddy field soil in which it grows; for example, in an experimental study a well drained soil containing 0.3 ppm mercury grew rice containing 0.3 ppm, while a poorly drained soil containing 1.4 ppm yielded rice containing 0.8 ppm (61). When a crop or foodstuff is

treated with a mercury fungicide in accordance with good agricultural practice, residues of mercury will also range higher than "normal" but are generally not greater than the following examples: apples, 0.1 ppm; tomatoes, 0.1 ppm; potatoes, 0.05 ppm (6). It would thus appear that mercury concentrations in plant materials generally range from 0.10 ppm down to 0.01 ppm or even less, but higher levels have been found, and such levels may be caused by naturally high concentrations of mercury in the soil or by the heavy application of mercury fungicides.

Mercury residues in seed grains dry-dressed and liquid-dressed with fungicides according to standard procedures have been found to range from 23 to 34 ppm (74,75). Washing of the dressed grain only removed from 10 to 40%, so it appears that even when dust dressings are used appreciable amounts of mercury are absorbed into the seed coat. Such dressed grain would thus be particularly hazardous if it ever found its way directly into human food sources or if eaten by wild animal populations after planting.

ii. Animals and animal materials. "Background" levels of mercury for animals and animal materials are difficult to assess, particularly for terrestrial samples, since the agricultural use of mercury products is so widespread and completely uncontaminated food sources are rare. Nevertheless, a literature survey suggests that normal values for eggs and the flesh of birds and animals are generally less than 0.02 ppm (69,76,77). Marine fish have mercury concentrations usually below 0.10 ppm and nearly always below 0.15 ppm (76,78,79), whereas mercury levels of 0.20 ppm or less are assumed to be normal for fresh water fish (4,31,80). The higher background levels found for fish when compared to other animals or fruits and vegetables is due to the marked ability of fish to accumulate mercury (48,81) (see section 6C).

* * * * *

About 1955, ornithologists in various parts of Sweden observed a decrease in populations of some seed-eating and predatory birds. In 1958, attention was drawn to the fact that birds found dead and sent for examination to the state veterinary medical institute had remarkably high residues of mercury (4–200 ppm) in their livers and kidneys (82). In quick succession, studies on shot and trapped birds also revealed remarkable levels (1–53 ppm), with 51% in one sample of 200 normal seed-eating birds having more than 2 ppm mercury in their livers (6,83); high residues of mercury were found in dead, dying, or trapped birds in other European countries during the early 1960's (84–86); seed-eating rodents and predatory animals (foxes) in Sweden were observed to contain levels of mercury up to 2.0 ppm (80,87); true herbivores (field-mons, deer, horses, rabbits), on the other hand, generally had levels ranging from 0.006 to 0.055 ppm (88). The conclusion drawn was that large numbers of seed-eating and predatory animals displayed symptoms and indications of mercury intoxication, and a source of mercury at the time was attributed to the use of mercury-containing compounds as seed-dressing in agriculture (83).

During the subsequent debate concerning the effects of mercury compounds from agricultural sources, analyses indicating mercury levels up to 9.8 ppm in Swedish fish were also made available (4,80), and elevated levels in fish from other Scandinavian countries were also observed (88–90). It then became apparent that mercury contamination of the Swedish environment was even more widespread than had been previously believed. In order to assess the historical development of this contamination, neutron activation analyses for mercury content of bird feathers from museum specimens collected over the previous century were performed (80,91). These studies revealed that in birds with a terrestrial food chain, a nearly constant level of mercury was maintained from the middle of the previous century until 1940. Subsequently, an increase in the mercury concentration of feathers occurred amounting to at least 10 to 20 times the previous level. Shortly after 1940, liquid treatments for seed grain using alkylmercury (methylmercury, ethylmercury) ingredients replaced dusting with other types of organic mercury compounds in many of the developed nations (including Sweden) because of the reduced hazards and

inconvenience to operators dressing the grain with specially designed machines (6). The simultaneous appearance of increased mercury accumulations in birds with a terrestrial food chain thus suggests that the alkylmercury seed-dressing agents are the main source of contamination of the terrestrial environment. In contrast, fish-eating birds showed a gradual increase in mercury content since the previous century, suggesting that the mercury contamination of water follows the general increase in industrial activity. Typical data found for the goshawk (diet of insects, rodents, and small birds) and crested grebe (diet primarily of fish) are provided in Fig. 4 (80).

As a final note on the Swedish experience, methoxyethylmercury compounds were substituted for alkylmercury compounds in seed-dressing formulations after February 1966, and the relative amount of dressed seed used for sowing was also reduced from 80% (prior to 1966) to 12% (1967). These changes apparently had no deleterious effect on crop yield (4). In 1966 and 1967, the mercury levels in the feathers of predatory birds with a terrestrial food chain abruptly dropped to levels about 50% above those typical for the previous century (80), and by 1969, population increases in several decimated species were noted

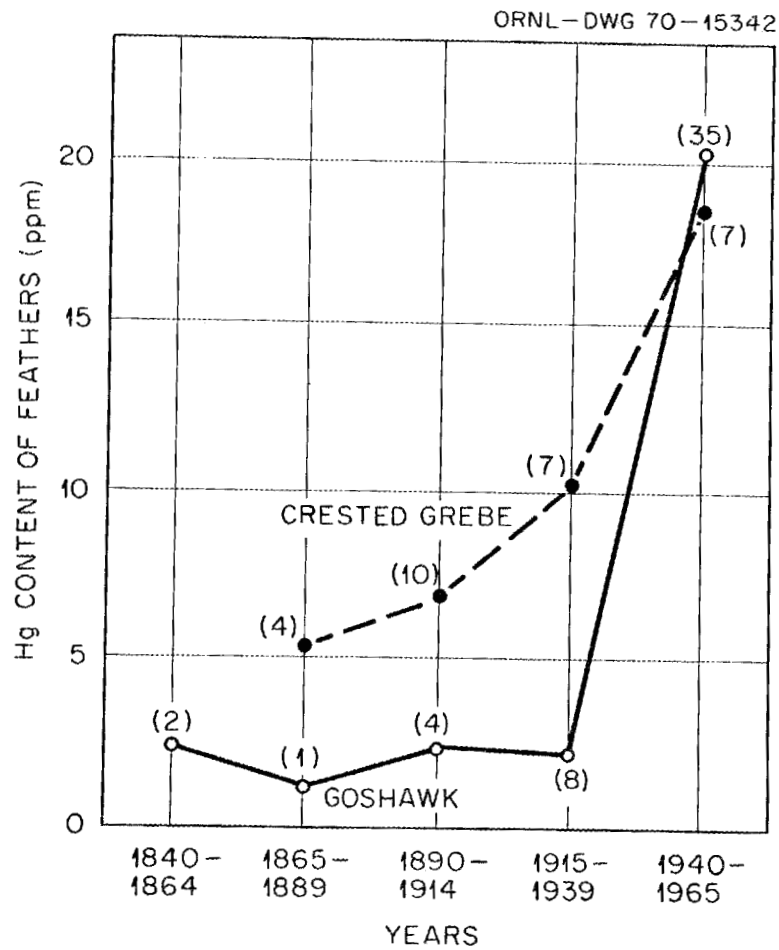

Fig. 4. Mercury levels in feathers of the crested grebe (closed circles) and goshawk (open circles) from Sweden during 1840–1965. The numbers of samples are indicated in parentheses. The ratio of mercury content in feathers relative to breast muscle is about 7:1. Drawn from data provided by Johnels and Westermark (80).

(64). The mercury contents of meats and eggs analyzed in 1967–68 were also significantly lower than in 1965–66 (Fig. 5) (77). Fish, however, generally continued to maintain high levels of mercury.

* * * * *

In 1967, a limited study of mercury residues in U.S. foods was conducted by the Food and Drug Administration as part of the Pesticide Total Diet Study (92). Six food classes were analyzed by neutron activation analysis, and the results indicated background levels of mercury in the order of 0.002–0.050 ppm. No further testing was performed in this country until 1970. In 1969, following warnings of significant mercury pollution in the central provinces, studies were quietly initiated by the Canadian Wildlife Services to define the situation. Shortly thereafter, several commercial catches of fish (walleye, northern pike, bass, and jackfish) taken from Lake Winnipeg, Cedar Lake, Saskatchewan River, and Red River in the Province of Manitoba were detained by the Canadian Federal Department of Fisheries and Forestries because they contained mercury residues ranging from 5 to 10 ppm (5). As a result of concurrent testing by Ontario officials, the Canadian government publicly embargoed all commercial fish taken from Lake Saint Clair effective March 23, 1970. Probably the most interesting revelation at this time, however, was the depth of information that had been developed in Canada on this matter over the previous 18-month period, with apparently no awareness in this country as to the seriousness of the situation until mid-March 1970, when the matter was made public (5). A flurry of analytical data on fish and wildlife populations

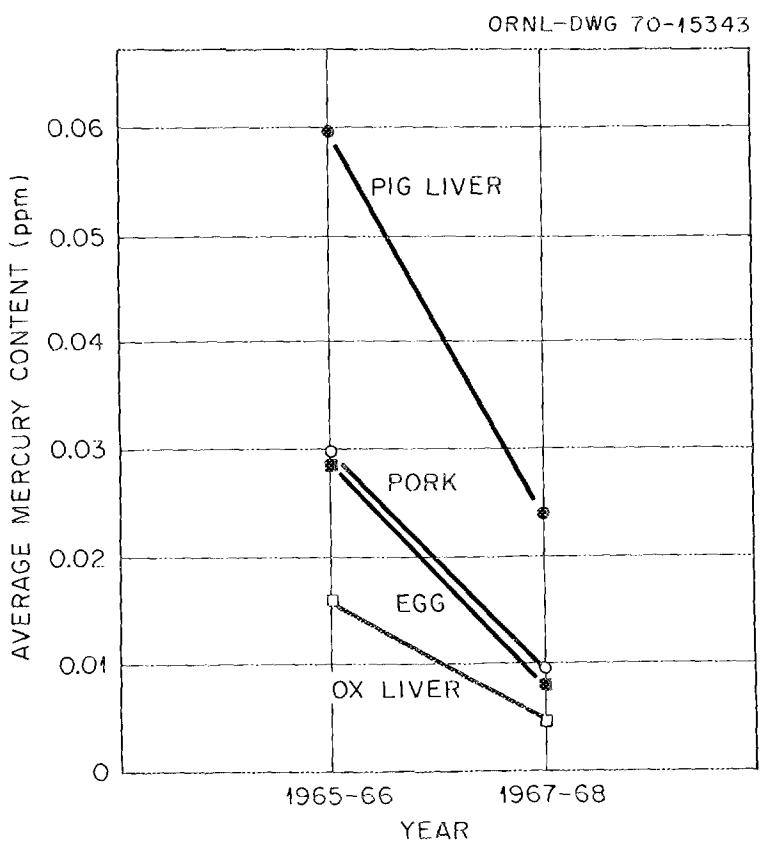

Fig. 5. Decrease of mercury content of animal foods in Sweden, due to changes in seed disinfection practices. From Westöö (77).

within the U.S. subsequently found its way into magazine articles, the popular press, and congressional testimony, indicating mercury levels up to 117 ppm in eagles from Minnesota and up to 4.4 ppm in fish collected from the North Fork of the Holston River (Table 7). As a result of this information, restrictions on sport and commercial fishing have been placed on waters within at least 18 states as of September, 1970 (Table 8) (93).

A systematic survey of wildlife and animal food products (including freshwater and marine fish) has been underway in Canada, and the initial results were made available in the summer of 1970 (69). In this study using neutron activation analysis, elevated levels of mercury were found in several pheasants from Alberta and in pike, pickerel, and whitefish from the Great Lakes area (Table 9). Avian products generally contained less than 0.05 ppm, and most fish (except those from heavily contaminated areas) averaged 0.2 ppm or less. The levels of mercury found in fish were somewhat lower than those found in other laboratories, and the explanation given was that smaller fish were sampled for analysis.

In the fall of 1970, the U.S. Food and Drug Administration fould that canned tuna sold in the U.S. contained up to 1.2 ppm mercury (average 0.37 ppm) and that frozen swordfish ranged from 0.18 to 2.4 ppm (average 0.93 ppm) (94). Surprisingly high levels (25–170 ppm) were also found in the livers of fur seals and sea lions off the western coast of North America (95,96). Even allowing for analytical and methodological errors, these results on large pelagic animals feeding at the top of the ocean food chain appear to be truly singular and so far unexplainable.

As a footnote to the subject of mercury levels in fish, it has been observed that mercury is not removed by boiling or frying the fish; rather, since the fish loses water by these processes, a corresponding increase in mercury level occurs (77,97).

iii. Man. The scanty information concerning mercury levels in various tissues of man (exclusive of blood and hair) has been collected from several sources and is summarized in Table 10. "Normal" subjects were generally selected on the basis of a high probability that they had had no unusual occupational or therapeutic exposure to mercury, but this was never really certain since the samples were autopsy specimens. There is a fairly wide spread of values, both among tissues and among analysts. In general, however, the highest levels were found in the kidney and liver, which are the two organs primarily responsible for the elimination of mercury from the body (see section 6D). Mercury levels did not appear to be a function of age (98), indicating that mercury does not accumulate in body tissues under normal circumstances. Also included in Table 10 are data concomitantly collected on subjects known to have been exposed to mercury. Again the kidney and liver contained high values, being anywhere from 2 to 50 times that of the "controls." Tissues derived from the brain (pituitary, olfactory lobe) also appeared to accumulate mercury upon exposure. One study indicated that a known total exposure of up to 400 mg mercury had little measurable effect on tissue levels, whereas higher exposures resulted in elevated levels (99).

Data on blood and hair derived from Scandinavian and Japanese sources is summarized in Table 11 (4). The ratio between the concentrations of mercury in blood cells and plasma seems to be, qualitatively, a measure of mercury intoxication, whereas the mercury concentration in hair reflects the mercury concentrations in the body at some time prior to sampling (4). Normal values of mercury concentration in blood and hair of Swedish adults who had no known excessive exposure to mercury range up to 0.01 ppm and about 1.35 ppm respectively. Values for normal adults from Finland, which also has a widespread contamination problem, were somewhat higher. Fish eaters show a definite elevation in mercury levels over control values, and there appears to be a linear relationship between fish consumption and the mercury concentration of blood (100). In severely contaminated individuals, values for mercury can average as high as 2.4 and 500 ppm for blood cells and hair, respectively [but see also section 7Bi(3)].

A widescale study of mercury in the head hair of "normal" Canadians has also been performed, indicating values from 0.2 to 6.0 ppm with an average around 1.5–3.0 ppm (69,101). Analyses were additionally performed on fish eaters from areas near contaminated bodies of water (Kenora-Dryden, Lake Saint Clair) and indicated that 75% of such individuals had abnormally high concentrations of mercury (>6 ppm) in their hair (69). Several who had consistently eaten fish at least once a week during the previous ten months had levels ranging from 50 to 100 ppm, whereas those who had consumed less than ten pounds of fish during this period (i.e., less than one meal per two weeks) had essentially normal concentrations in their hair.

Finally, in one brief report mercury levels were measured in the blood of five randomly chosen pairs of mother and child at the birth of the child (100,102). The average mercury concentration in the blood cells of the newborn child was found to be 28% above that of the mother. If the data for the children are plotted *vs* those for the mother, a relationship is obtained suggesting that both the relative and absolute accumulation of mercury from the mother-to-be into the human fetus may increase hazardously at higher mercury concentrations (Fig. 6).

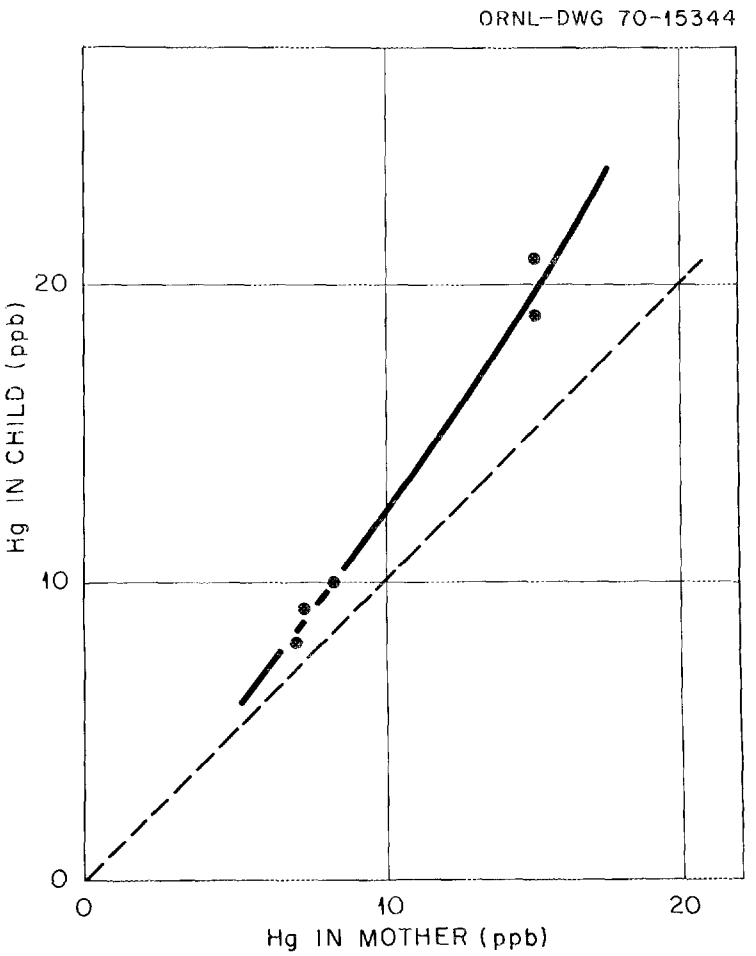

Fig. 6. Mercury content in the blood cells of the child *vs* that of the mother at the birth of the child. Dashed line has a slope of 1. From Löfroth (4), based on data of Tejning (102).

5. FORMS AND TRANSFORMATION OF MERCURY IN THE ENVIRONMENT

The principal mercury compounds commercially discharged into the environment and the primary sources of these compounds are the following (103):

(1) Metallic mercury, Hg^0 (chlor-alkali and instrument plants);

(2) Inorganic divalent mercury, Hg^{2+} (chlor-alkali plants);

(3) Phenylmercury, C_6H_5-Hg^+ (paints, pulp and paper plants);

(4) Methylmercury, CH_3-Hg^+ (agriculture);

(5) Methoxyethylmercury, CH_3O-CH_2-CH_2-Hg^+ (agriculture).

Countries in which methylmercury compounds were used as seed dressing agents have experienced a general contamination of terrestrial animals by mercury (Norway, Finland, Sweden before 1966), whereas those countries in which methoxyethylmercury compounds were used have not (Denmark, Sweden after 1966). Tests have shown, moreover, that most of the mercury present in food products from contaminated countries is in the form of methylmercury (Table 12) (77). This led to the initial assumption that methylmercury compounds released into the environment were primarily responsible for the mercury contamination of plants and animals, whereas other mercury compounds were relatively innocuous.

When fish specimens were also found with elevated levels of mercury, this mercury was initially assumed to be in the forms primarily released into industrial waters, i.e., inorganic mercury and phenylmercury (104). It was subsequently shown, however, that regardless of the nature of the mercury pollutant, essentially only methylmercury is present in fish flesh (Table 13) (77). The inevitable conclusion, therefore, is that transformation reactions between different compounds of mercury not only must occur, but are of central significance (105).

As an initial consideration for such transformations, the geochemistry of metallic and inorganic mercury involves numerous interconversions and transformations (7,26). Simply stated, mercury is generally present either as a variety of inorganic or humic complexes (which can interchange with one another) or as the relatively unionized and stable mercuric sulfide (HgS), the principal component of cinnabar. Under oxidizing conditions, however, mercuric sulfide is gradually converted to mercuric sulfate ($HgSO_4$), which dissociates much more readily and thus releases inorganic mercury into the web of geochemical interactions. Oxidizing conditions will also promote the conversion of metallic mercury (Hg^0) to inorganic mercury, particularly if there is material present (soil, alluvia, etc.) with which the resulting inorganic mercury can firmly bind. The mercuric compounds which find their way to the earth's surface will generally degrade to metallic mercury and volatilize under the action of sunlight (26). On the other hand, most of the mercury compounds which are washed into river and lake bottoms will, if anaerobic conditions prevail (i.e., hydrogen sulfide is present), be ultimately converted into the insoluble mercuric sulfide.

Into this general scheme must now be superimposed the recent and important discovery that decomposing fish and sediments from a large number of lakes and rivers contain microorganisms or biologically derived alkylating systems which are capable of forming methylmercury and dimethylmercury from inorganic mercury, even under anaerobic conditions (Fig. 7) (104,106). Such transformations would contribute to the release of mercury from sediments into water since methylmercury has a lower binding affinity than inorganic mercury and dimethylmercury is uncharged, volatile (bp = 94°C), and can be released into the atmosphere. However, both methylmercury and dimethylmercury would appear to be rapidly taken up and retained by larger aquatic organisms such as fish (see section 6C). In solution, dimethylmercury is rather unstable under acid conditions, and under such circumstances substantial

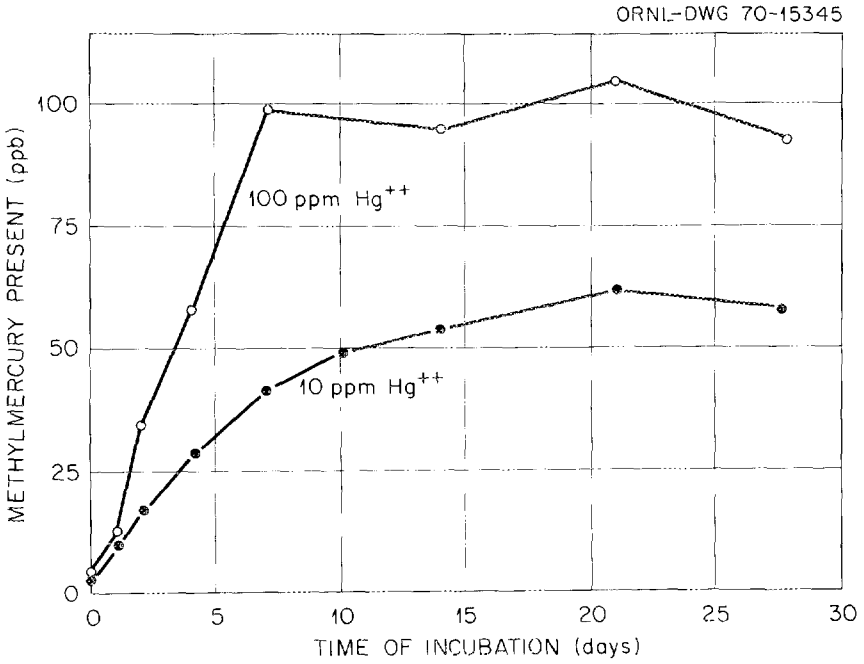

Fig. 7. Concentration of methylmercury in bottom sediment after addition of 10 (closed circles) or 100 (open circles) ppm inorganic mercury followed by variable times of incubation. From Jensen and Jernelöv (104).

decomposition to methylmercury can occur (106). Whatever dimethylmercury is released into the atmosphere is most likely converted to metallic mercury vapor together with methane and ethane. The photolysis of dimethylmercury has been a much studied reaction, being a classic method used by chemists to generate ethyl and methyl radicals (107). The conversion of phenylmercury and methoxyethylmercury to other forms of mercury also occurs, although less information on such transformations is available (103). Certain bacteria, moreover, are apparently capable of converting just about all types of mercury compounds into metallic mercury vapor (108,109).

The most important presently known steps by which inorganic mercury and some mercury-containing compounds are interconverted in nature are thus summarized in Fig. 8. Some of the generalizations which emerge from this diagram and from what has previously been said are the following:

(1) mercury, in whatever form, is potentially exchangeable among the air, land, and water phases;

(2) mercury, in whatever form and from whatever phase, is potentially capable of being taken up by aquatic animals in the form of methylmercury or dimethylmercury;

(3) in an aquatic system, methylmercury can be formed directly from inorganic mercury (Hg^{2+}) under anaerobic conditions; but

(4) under *permanently* anaerobic conditions, mercury will tend to accumulate in bottom sediments either as HgS or Hg^0, and little methylmercury or dimethylmercury will be formed (110);

(5) methylmercury and dimethylmercury can be formed indirectly from either HgS or Hg^0 in the presence of oxygen or under oxidizing conditions (111);

(6) alkaline conditions will tend to promote the release of mercury from aquatic ecosystems via dimethylmercury.

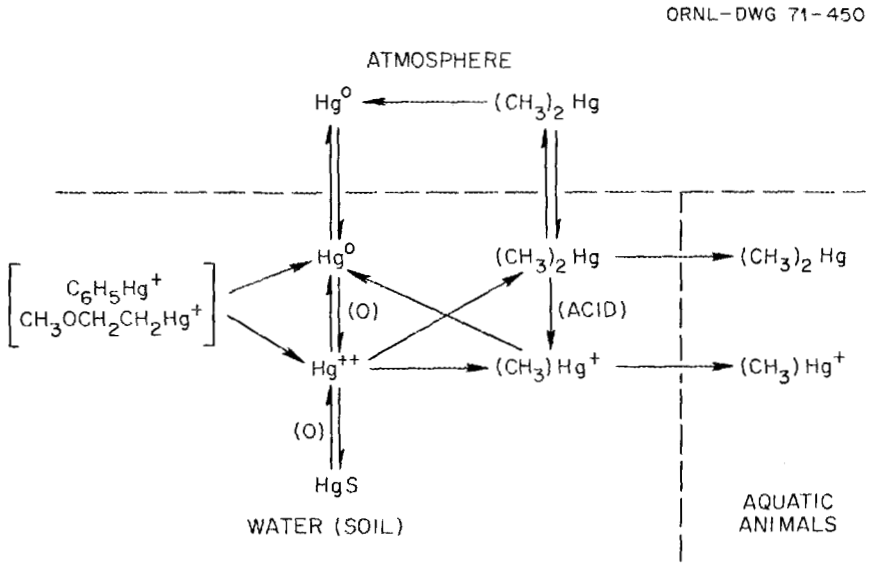

Fig. 8. Some of the transformation pathways of mercury and its compounds in nature.

More information is certainly needed on the interconvertibility of each of these steps; nevertheless, the overall equilibrium in nature tends to proceed towards the right, since the methylmercury and dimethylmercury compounds are not tightly bound to substrates, are water soluble or volatile, and are rapidly taken up by living organisms in which they are relatively stable. Assuming that the rate of conversion of inorganic mercury to methylmercury is the rate-determining step in the removal of mercury from contaminated bottom sediments, it has been calculated that this removal would normally take from 10 to 100 years unless the mercury is made inactive by its physical removal or by the elimination of its biological availability (103,112).

6. TURNOVER, TRANSLOCATION, AND ACCUMULATION OF MERCURY IN PLANTS AND ANIMALS

A. Plants

In general, it has been found that various plants (carrots, potatoes, turnips, lettuce, beans) grown in soil treated with inorganic mercury compounds accumulated little or no mercury in the edible parts of the plant (6). Conflicting evidence has been obtained as to whether or not the mercury from seeds dressed with methylmercury compounds eventually enters the growing plant and subsequent crop. In one investigation (6), the mercury level of wheat or barley was found to be 0.008–0.012 ppm whether or not they were grown from dressed grain. However, other studies have shown that mercury moved from the seed coating on rice grains up into the developing plant (113) and that the mercury content of harvested barley and oats was about 0.030 ppm when the seed corn was dressed, but only about 0.014 ppm when the seed corn was nondressed (114). Further indirect evidence that methylmercury dressings may translocate to other parts of the new plant are provided from data demonstrating that the average mercury content of eggs from hens fed grains grown from untreated seed and from methylmercury-treated seed were 0.012 and 0.027 ppm respectively (77). In the same experiment, eggs from hens fed on grains grown from methoxyethylmercury-treated seed contained 0.010 ppm, suggesting that different forms of mercury translocate to different extents.

When phenylmercury (PMA) compounds are applied to the foliage or bark of plants or trees, most of the mercury penetrates beyond the reach of surface washing within a few days and subsequently translocates to other parts of the plant (115). As a typical example of the problem, investigations (116) on apple trees receiving PMA cover sprays in late spring or early summer indicated that mercury residues in apples declined over a period of one month to approximately one third of the initial deposit. At this time, a large part of the residue was concentrated in a small portion of the apple at the calyx end. This stage was followed by a rapid accumulation of mercury in the fruit throughout the remainder of the growing season, until at harvest the residues were greater than the initial deposit. Fruit bagged to prevent direct contact with the PMA spray also accumulated mercury. Further studies (117) showed that mercury moved into the fruit by translocation rather than volatilization and that there was little or no uptake by the roots.

B. Aquatic Organisms Other than Fish

Little information exists on the turnover and translocation of mercury in aquatic organisms other than fish. However, one study (80) has shown how the effluent from a paper and pulp mill can dramatically increase the mercury levels in various organisms downstream from the plant (Table 14). The accumulation of mercury may occur primarily by passive surface adsorption, as has been shown for marine diatoms (118), or by a combination or surface adsorption with active metabolic uptake and ingestion of contaminated food organisms and debris, as is probably the case for animals such as insects. Regardless of the mechanism, mercury accumulated by lower aquatic organisms is ultimately passed on to animals at higher trophic levels such as fish.

C. Fish

Analyses made on fish caught above and below industrial outfalls are provided in Table 13 (77; see also ref. 119). These data demonstrate the effect that mercury discharge can have on fish and also indicate that whatever the natural or man-made source, mercury in fish muscle accumulates almost exclusively as methylmercury. The overall concentration factor from water to pike can be as high as 3000 (32), but an absolute value for this factor will depend upon such considerations as the species, the length of exposure, and the age and size of the fish. Fish can accumulate methylmercury released from bottom sediments either directly from the surrounding water (81,120) or indirectly through the food chain (5,80).

As a typical example of subacute exposure to alkylmercury in the surrounding water, trout were placed for one hour in a water bath containing 0.125 ppm ethylmercury and the mercury levels in various tissues followed thereafter (121). The conditions of exposure represent the presently recommended procedure for the control of external bacterial and parasitic infections in fish. The major finding was that absorption in the gills first occurred, followed by successive peaks of mercury concentration in the blood (2 days) and liver (4 to 5 days). Levels in the kidney, however, rose slowly and were maintained at about 1.6 ppm even after 7 months (Fig. 9). Most of the mercury in the blood was associated with the cellular rather than the plasma fraction. Muscle tissue generally remained at a low level (<0.5 ppm). In the same study, it was found that salmon fed a 30 day diet of ethylmercury-treated fingerlings also accumulated large amounts of mercury (9.1 and 1.9 ppm in kidney and muscle, respectively). Uptake of mercury is thus more rapid in the liver and kidney than in muscle, and higher levels have been found in the former tissues if the total amount of mercury in the fish is increasing (122).

Under stable environmental conditions involving the chronic exposure of the fish to very low levels of mercury, however, the amount of mercury in muscle tissue is greater than that in the liver or kidney (Table 15)(80,122). In one study, aimed at determining whether chronic mercury exposure produced an

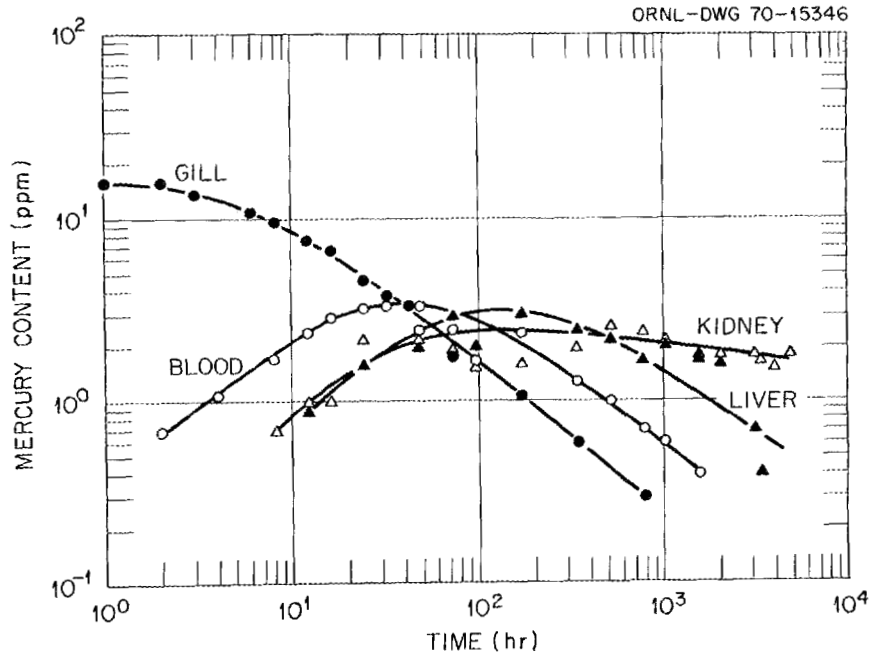

Fig. 9. Absorption and distribution of mercury in various tissues of trout following a 1-hour exposure to water containing 0.125 ppm ethylmercury phosphate (double log plot). Drawn from data provided by Rucker and Amend (121).

accumulative effect, the mercury content in the muscle of pike collected in three variously polluted waters was measured as a function of fish weight, the latter being the best indication of age (80). The results showed that there was a linear relationship between the weight and age of the fish and the mercury content in axial musculature, except when the concentration was less than about 0.2 ppm (Fig. 10). This can be taken as an indication that there is a threshold level of mercury in the environment, above which fish cannot eliminate mercury from their muscle tissues faster than it is incorporated and above which accumulation thus occurs.

D. Other Animals and Man

Inorganic mercury compounds are all absorbed readily into the lungs and gastrointestinal tract but poorly through the skin. After acute administration of inorganic mercury salts to animals and man, the highest level of mercury is found in the kidneys and the second highest in the liver. Elimination from brain, thyroid, and testis is slow, however, so accumulation of mercury in these organs with chronic exposure is possible (123,124). Organic mercury compounds may also enter the body by inhalation or oral ingestion and, in addition, are more readily absorbed through the skin than inorganic materials. Once inside the body, however, phenylmercury and methoxyethylmercury are degraded to inorganic mercury (125–127). These compounds thus generally undergo the same turnover patterns as inorganic mercury; i.e., they disappear fairly rapidly from the blood and accumulate in the kidney prior to excretion. They are then excreted at a moderate rate as long as the dose given is not large enough to injure the excretory organs. All of the above substances may be converted to methylmercury within the body to a slight extent. As examples, liver homogenates are able to form methylmercury from inorganic mercury (128,129), and hens fed seeds treated with inorganic, methoxyethyl-, or phenylmercury lay eggs containing methylmercury in the egg white (128).

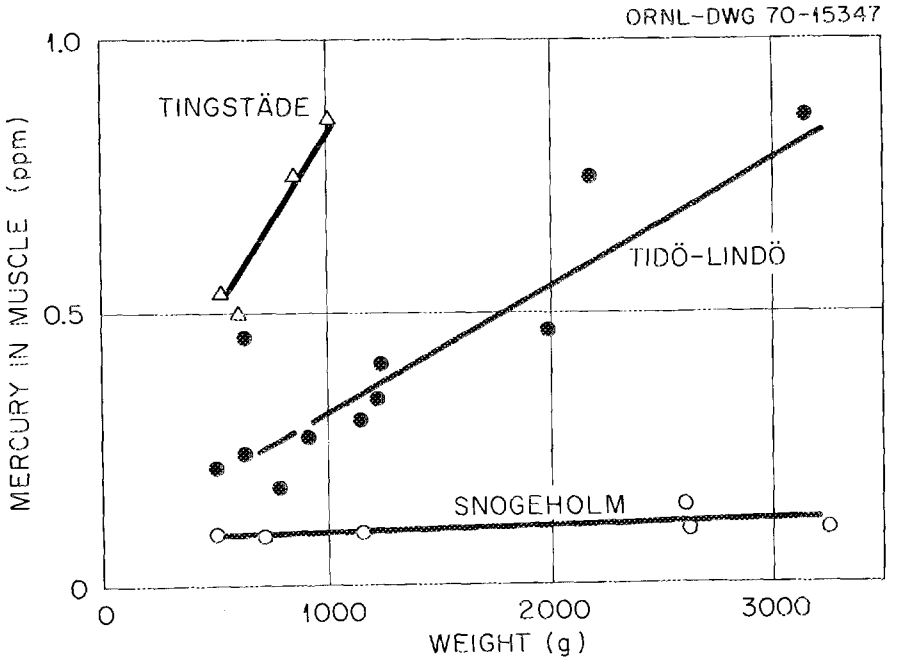

Fig. 10. Relation between mercury content of axial muscle and total weight in pikes collected from three different lakes in Sweden. From Löfroth (4), based on data of Johnels and Westermark (80).

Methyl- and ethylmercury (alkylmercury) compounds, on the other hand, are much more stable in the body and circulate for a long time unchanged in the blood (127,130). In the blood, 90% or more becomes associated with the blood cells rather than the plasma as in the case of inorganic mercurials (127,131–133). Because of the relatively low plasma concentration of alkylmercury compounds, they are taken up to a lesser extent by the kidneys, and there is little rise in urinary excretion with rising blood levels (131). Thus, the urinary output of mercury is not a very useful index of exposure to alkylmercury compounds. Alkylmercurials also accumulate in various body tissues and particularly the brain to a much greater extent than other mercury compounds (134,135) [more than 98% of the total mercury in the brain is in the form of methylmercury (2)]. Once inside the brain, a redistribution of mercury to cortical regions in the cerebellar and calcarine cortex occurs concomitantly, after severe exposure, with the appearance of neurological symptoms (4,136). The main route of excretion for alkylmercurials appears to be by means of the liver into the bile and thence to the feces; more than 90% can be resorbed again during passage through the gastrointestinal tract, however, so that considerable recirculation occurs and retention by the body is marked (136–138) [see section 7D(2)].

Mercury vapor enters the body primarily through the respiratory tract, and its subsequent distribution pattern is intermediate between the alkylmercurials on the one hand and inorganic mercury, phenylmercury, and methoxyethylmercury on the other. It is poorly soluble in plasma, where it is partly converted to inorganic mercury and subsequently excreted through the kidneys (139). Because of its lipid solubility and lack of charge, however, it can penetrate cell membranes to some extent and becomes generally diffused throughout the body (139–141), showing an affinity for brain and nervous tissue (140–143), particularly in localized areas (123,141,142). It also accumulates to a considerable extent in the mucous membranes of the intestinal tract and in the salivary glands (123).

7. CRITERIA FOR EVALUATING MERCURY LEVELS

Criteria are descriptive; they [are] an expression of the scientific knowledge of the . . . effects that can be expected to occur whenever the level of a pollutant reaches or exceeds a specific figure.

— Birmingham. Federal Government and Air and Water Pollution, 23 Business Lawyer 467.

"What do you know about this business?" the king said to Alice.
"Nothing," said Alice.
"Nothing *whatever*?" persisted the king.
"Nothing whatever," said Alice.
"That's very important," the king said, turning to the jury.

– Carroll. Alice in Wonderland.

A. Toxicity to Aquatic Organisms

Information on the lethal concentration of mercury compounds found for various aquatic organisms has been collected from a number of sources and is presented in Table 16 (7). While such information indicates that mercury compounds are remarkably toxic at relatively low concentrations, it unfortunately does not help to define what maximal levels should be avoided in order to maintain a healthy aquatic ecosystem. For instance, although 60 ppb ethylmercury has been found lethal to marine phytoplankton, as little as 0.1–0.6 ppb alkylmercury introduced into sea water will produce a measurable inhibition of photosynthesis and growth (144,145), and 1 ppb methylmercury or phenylmercury will reduce photosynthesis by fresh water phytoplankton to 40–50% of control values (145). In another study, half of the goldfish (a very hardy fish) continuously exposed to a concentration of 820 ppb mercuric chloride died within seven days, yet an exposure for only two days to 3 ppb mercuric chloride produced a measurable impairment in "learning" behavior (146).

As an added difficulty, experiments done under laboratory conditions in order to determine a tolerance level for a given substance may not apply to natural situations where synergistic effects play a role. The presence of copper, for instance, can appreciably increase the toxic effects of mercury (147). Chronic exposures must also be considered, since a species may eventually die in the stream because sublethal levels of mercury could interrupt its normal ability to feed or avoid predators. Even though the literature on chronic and synergistic exposure is inadequate, it is nevertheless apparent that concentrations of mercury below the proposed drinking water quality standard of 5 ppb (see section 8B) can have a detrimental effect on aquatic organisms.

B. Toxicity to Man

i. Individual incidents. The medical literature is replete with examples of acute and chronic exposures of individuals to mercury, both intentional and unintentional. A few examples which indicate the variety of experiences are provided below:

(1) (Ref. 148). A student nurse broke a clinical thermometer and wounded her hand. The healing of the wound was uneventful, but four years later the lower lobe of her left lung was removed after repeated bronchitis and pneumonia, and disseminated mercury emboli were found in the pulmonary arteries. The patient died six years after the original injury from empyema following the removal of the remainder of the lung, in which an abscess had formed.

(2) (Ref. 149). A 17-year-old farmer treated stored oats with a fungicide containing 1.5% mercury as methoxyethylmercury silicate. This work lasted for three hours and caused much dust. Abdominal pains,

diarrhea, and vomiting made him call a doctor seven days later. Among the symptoms were albuminuria with the presence of 0.9% blood in the urine and a light pulmonary edema. Two years later, a control check revealed no edema, the cardiovascular system was intact, and there was no neurological abnormality. Three years after spraying, the patient lost weight and became debilitated to the extent that he was unable to work. He also developed states of anxiety, rapid heart beat, headaches, insommia, and "tics" of the face, larynx, and pharynx. Urinary analysis for mercury at this time still showed an elevated level.

(3) (Ref. 150). In September 1969, a granary in New Mexico gave away floor sweepings, and a custodial worker collected some 3000 pounds to feed his hogs. About three weeks after he began feeding the seeds, he noticed that a number of the hogs appeared sick. He subsequently butchered the largest one that appeared well and began feeding the meat to his family. About three months later, three of his children developed impaired vision and eventually became comatose. Urine analyses (151) revealed high mercury contents, and hair samples (69) from two of the children contained 1,397 and 2,436 ppm mercury (these being the highest levels ever measured in a human). Six months later, two of the children were still blind (one in a coma), and the third appeared to be recovering, although with some impairment of vision. Checks revealed that the seed at the granary had been treated with Panogen, a methylmercury formulation.

ii. Epidemiological incidents. Examples of community poisonings in which mercury has been found as the causative agent have been reported in Iraq (152), Pakistan (153), Japan (154,155), Guatemala (1,156), and what is now Yugoslavia (157). In the latter case a community of 900 inhabitants along with cows and domestic animals developed muscular tremors and other symptoms of mercury intoxication following a fire in a mercury mine. Probably many other incidents involving mercury posioning have occurred, but either mercury was not identified as the causative agent or the general malaise of the population never reached alarming proportions. Details for two of the incidents are provided:

(1) (Ref. 152). In 1956, many cases of mercury poisoning were observed in the north of Iraq, and more than 100 cases (14 fatal) were admitted to Mosul Hospital. In 1960, many farmers from the central part of Iraq were also affected, and 221 patients were admitted to one hospital in Baghdad alone. There were many deaths; 22 bodies seemed to have been examined in the Medical-Legal Institute in Baghdad, and large amounts of mercury were found in the organs (average level in the liver was 65.8 ± 18.5 ppm). All the poisonings apparently occurred from eating dressed wheat, and patients admitted that they had been warned against eating the wheat. Some of the patients had washed it to rid if of the poison, however, and when they noticed that nothing happened to fowls that consumed it for a few days, they started eating it also, sometimes mixed with larger amounts of untreated wheat or maize. People who had eaten the dressed wheat and remained well for some days or weeks set an unfortunate example to others who did not hesitate to consume the dressed wheat in the form of bread. The product responsible in all cases was Granosan M, a fungicide containing 7.7% ethylmercury-*p*-toluenesulfonanilide. Cases of neural lesions were found in all grades of severity. Symptoms usually appeared within one or two months after eating the treated wheat, sometimes longer, but occasionally in as short a time as a few days, depending on the amount consumed.

(2) (Refs. 154,155). In Minamata, Japan, an acetaldehyde and vinyl chloride plant discharged large quantities of mercuric chloride and methylmercury from 1950 until 1960, when equipment for waste water treatment was installed. The effluent was channeled to the Minamata River and Minamata Bay. From 1953 to 1960, 111 persons living around the Minamata River and Bay areas were reported poisoned after having eaten fish and shellfish caught in the contaminated area, and 41 died. Twenty-five out of forty affected families ate fish and shellfish daily (158). Among the 111 were 19 congenitally defective babies born of mothers who had eaten the contaminated food. Clinical features observed were lack of motor coordination, constriction of visual fields, and difficulty articulating words; pathological findings were regressive changes in the cerebellum and cerebral cortices.

During 1965 in Niigata, Japan, 26 cases of mercury poisoning and 5 deaths were also officially documented under similar circumstances, although in a Japanese report of the Niigata incident, details are given which show that at least 120 persons had one or more of the following symptoms associated with high mercury concentrations in the blood: numbness in the distal parts of the extremities; numbness around the mouth; constriction of the visual fields. Severely affected persons and their families consumed fish from mercury-contaminated water with a frequency of 0.5 to 3 times a day, and the fish seem to have contained 5 to 20 ppm mercury. In both the Minamata and Niigata incidents, the mercury compound involved was methylmercury, since this was the form of mercury found in fish flesh. From data derived from the Niigata incident, it has been calculated that consumption of 1.5 mg mercury as methylmercury per day (representing a daily diet of 250 g fish with 5 to 6 ppm mercury as methylmercury) is probably lethal (100).

iii. Industrial incidents (2). Numerous cases of industrial exposure to mercurials with resultant toxic symptoms have been reported, the most notorious being incidents in the mining and hat-felt industries. However, the most informative cases have been those in which symptoms were related to exposure. In an early study (45,46), symptoms of mercury poisoning in the hat-felt industry were observed only among workers who had been exposed to air mercury levels above 100 $\mu g/m^3$. In a thermometer workshop, one case of mercurialism and eight suspected cases were found among 120 workers exposed to mercury concentrations around 100 $\mu g/m^3$ (43). However, seven cases of mercury poisoning with pronounced tremor were found among 91 workers in a chlor-alkali plant, where mercury levels in the air were generally below 100 $\mu g/m^3$ (42). Evidence from the U.S.S.R. indicates that increased excitability of the central and autonomic nervous systems together with slight anemia and hypothyroidism occurred among workers exposed to as little as 10–30 $\mu g/m^3$ (2). In a careful investigation concerning the chlor-alkali industry (2), time-weighted average exposure levels were calculated for 642 workers, and the maximum average exposure found was 270 $\mu g/m^3$. The majority of workers were exposed to less than 50 $\mu g/m^3$. There was a very strong correlation between weight and appetite loss and the air-mercury levels. There was also a significant positive correlation between the prevalence of certain other neural complications and exposure, with the increase of symptoms being apparent above average concentrations of 100 $\mu g/m^3$. All of the above incidents refer almost exclusively to exposure to metallic mercury vapor.

In one study of 100 workers exposed to phenylmercurial compounds (with some admixture of inorganic mercury salts) in the air, concentrations of mercury in the working environment were found to be almost always greater than 290 $\mu g/m^3$, with one third of the workers exposed to concentrations up to 5,100 $\mu g/m^3$ (132,159). In another investigation, 26 subjects were found to have been exposed up to 6 years to 250–3,200 $\mu g/m^3$ phenylmercuric pyrocatechin (160). No evidence of poisoning was observed in either case, so aerosols and vapors containing phenyl- and inorganic mercurials must be considerably less toxic than vapors of metallic mercury.

Industrial exposure to alkylmercurials is relatively infrequent, with most cases involving laboratory workers or those preparing and handling dressed seed for agricultural use. A survey of reports appearing in the medical literature has been prepared (158), but no information is provided on exposure levels. In these incidents, the typical, generally irreversible symptoms (see section 7Biv) of alkylmercury poisoning were noted, with many cases terminating in death.

iv. Medical observations (2). Acute occupational exposures to high concentrations of inorganic salts of mercury are rare. Clinical manifestations, such as those which occur with accidental or suicidal intake of inorganic salts, are acute gastrointeritis with abdominal pain, nausea, vomiting, bloody diarrhea, and severe kidney injury leading to anuria with uremia. Acute exposure to high concentrations of mercury vapor may give rise to symptoms of pulmonary irritation and sometimes involvement of the central nervous system.

Chronic exposure to inorganic mercury is also uncommon. In the case of chronic exposure to mercury vapor, symptoms and signs involving the central nervous system are most commonly seen, the principal features being tremors and psychological disturbances. Symptoms related to the mouth, such as gingivitis, stomatitis, and excessive salivation, may occur along with a number of nonspecific symptoms such as loss of appetite, weight loss, anemia, and muscular weakness. Intoxication from mercury vapor or from absorption of mercuric salts may be due, in both cases, to the action of the mercuric ion. Metallic mercury is able to diffuse much more extensively into the blood cells and various tissues than inorganic mercury, but once distributed, most of it is oxidized to the mercuric form (139,161).

Acute exposures to phenylmercurials have been rare, and what information exists suggests that primarily kidney damage occurs (162). No conclusive evidence of systemic toxic effects in man after long-term exposure to phenylmercurials has been published, but an irritant effect on the skin has been observed (163). In a few reported cases from exposure to methoxyethylmercury compounds, the symptoms have been loss of appetite, diarrhea, weight loss, fatigue, and kidney damage with albuminuria and occasionally a nephrotic syndrome (149,164). The symptoms are probably due to inorganic mercury, since both phenylmercury and methoxyethylmercury are rapidly converted to inorganic mercury in the body (2).

No cases of damage in children born to mothers exposed to inorganic mercury, phenylmercury, or methoxyethylmercury compounds have been reported. In animal experiments, mercuric mercury was found to concentrate in the placenta, which acted as a barrier for absorption into the fetus (134).

Symptoms of methyl- and ethylmercury poisoning may occur weeks to months after acute exposure to toxic concentrations. The symptomatology of acute and chronic poisoning from both compounds is similar, including numbness and tingling of the lips or hands and feet, ataxia, disturbances of speech, concentric constriction of the visual fields, impairment of hearing, and emotional disturbances (165–167). With severe intoxication the symptoms are irreversible, indicating a threshold level of brain damage has been surpassed (4).

In infants born to mothers with exposure to methylmercury, the symptoms are somewhat different. Most children have mental retardation and cerebral palsy with convulsions. These effects are even found in infants born of asymptomatic mothers (168,169). Thus, alkylmercurials not only can pass the blood brain barrier but also the placental barrier (102,134,170), and fetal intoxication is possible for as long as 3–4 years after the mother is poisoned (170).

C. Biological Studies

Although considerable biological research has been done on the effects of mercury compounds in both intact animals and *in vitro* systems, the toxic effects of mercury remain essentially unexplained, particularly with reference to its mode of action on nerve cells, fibers, or synapses. All mercury compounds are cytotoxic to cells in culture, with organic mercury compounds being an order of magnitude more effective than inorganic mercury compounds (171). Mercuric ions can interact with sulfhydryl groups of proteins, however, thus interfering with enzymes and producing changes in membrane permeability (172). A methylmercury concentration equivalent to 20 ppm interferes with detoxification enzymes in rat liver preparations, and carcinogenic N-oxygenated metabolites accumulate because of a reduced capacity for complete oxidative demethylation (173). Metallic mercury has been shown to be both carcinogenic (174) and to enhance the effect of other carcinogens (175), whereas inorganic mercury has not (176). Any number of mercury compounds will produce chromosomal abnormalities in various cells, along with genetic and teratogenic effects (see ref. 175). As little as 0.25 ppm methylmercury in the food of *Drosophila* can induce the formation of extra chromosomes in offspring (177–179). Both phenyl- and methylmercury compounds have been found to be the most potent known inhibitors of spindle formation during mitosis

(178,180,181), with effective levels being comparable to those found for methylmercury in the blood cells of Swedish control populations (136,182); inorganic mercury is about 200 times less active in this respect, although in all such comparisons this factor may partially represent a difference in cell penetration of the two types of compounds.

D. Determination of an Allowable Daily Intake (ADI) for Mercury

The form of mercury to which the public is most generally exposed is methylmercury present in foods. An attempt has been made to define the maximum ADI of mercury as methylmercury in man (182). Two estimates have been made based on the scanty evidence available:

(1) In a study of persons with a high intake of Swedish fish contaminated to varying degrees with methylmercury, it was calculated that one subject consumed approximately 150 grams of fish per day, with an average mercury concentration of 6.7 ppm fresh weight; this is equivalent to 1.0 mg mercury ingested per day (100). A mercury level of 1.2 ppm in red blood cells was found in this individual, which is about half that found as average for several groups with symptoms of mercury poisoning (see Table 11). Thorough clinical investigation revealed no other signs or symptoms of illness or physical impairment. A daily consumption of 1.0 mg mercury per day thus represents the highest intake ever calculated for an asymptomatic person. Based on this one individual then, an intake of methylmercury equivalent to 1.0 mg mercury per day can be considered a "no effect dose" in man. Calculation of a safety factor of 10, which seems to be customary for "unintentional food additives," would yield an ADI of 0.10 mg mercury.

(2) The metabolism and retention of [^{203}Hg] methylmercury nitrate has recently been studied in three adult male subjects (137,138,183). Each took 3 μCi of the isotope orally, and essentially complete absorption occurred. Measurements made with a whole-body counter over a seven-month period and corrected for isotope decay indicated a physiological half life in the body of 70–74 days, equivalent to a daily excretion of 1% of the body burden (Fig. 11). Similar results are obtained if the ingested [^{203}Hg] methylmercury is covalently bound to food proteins (100,184). Radioactivity was also measured over different parts of the body by scintillation scanning and was found to be mainly localized over the liver area, with 10–20% located in the head (Fig. 12). The amount of mercury in the brain of an adult man which causes toxicity has been calculated to be 12 mg (136,182). If this amount of mercury in the brain is assumed to represent 10–20% of the body burden, the latter would be 60–120 mg. With a daily excretion of 1%, the dose of methylmercury needed to maintain equilibrium would be equivalent to 0.6–1.2 mg mercury per day. If one accepts this as a minimum toxic chronic dose to adult man and applies a safety factor of 10, one obtains an ADI of 0.06–0.12 mg mercury.

The above values should simply be taken as the best present estimates and need considerable reevaluation in the future. It is not clear, for instance, if the relative concentration and turnover of methylmercury in the brain after chronic exposure can be adequately inferred from experiments in which the isotope was "pulsed." Also, *these estimates are only based on neurological dysfunction as an end point and do not take into consideration possible teratogenic or carcinogenic effects, damage to the genetic mechanisms, or very long term effects which would enhance senescence.*

E. Economic and Social Costs of Mercury Pollution

Considerations to this point have involved physical, chemical, and biological aspects of mercury in the environment. As part of any evaluation of mercury pollution and the standards which will serve as guidelines for the release of mercury into the environment, economic and social factors must also be taken into consideration. Such factors are even more difficult to measure and quantify than biological exposures and effects; but they are, in fact, presently being incurred in many areas of the U.S. (see Table 8) and must

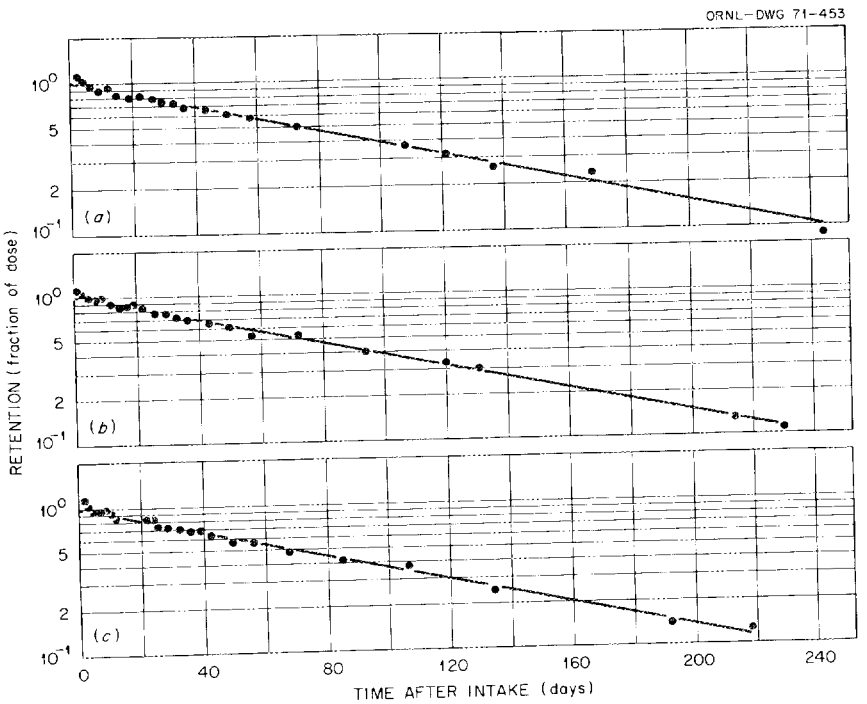

Fig. 11. Whole body retention of ^{203}Hg after a single oral intake of [^{203}Hg]methylmercury nitrate by three (*a*–*c*) human volunteers (semilog plot). From Aberg *et al.* (183).

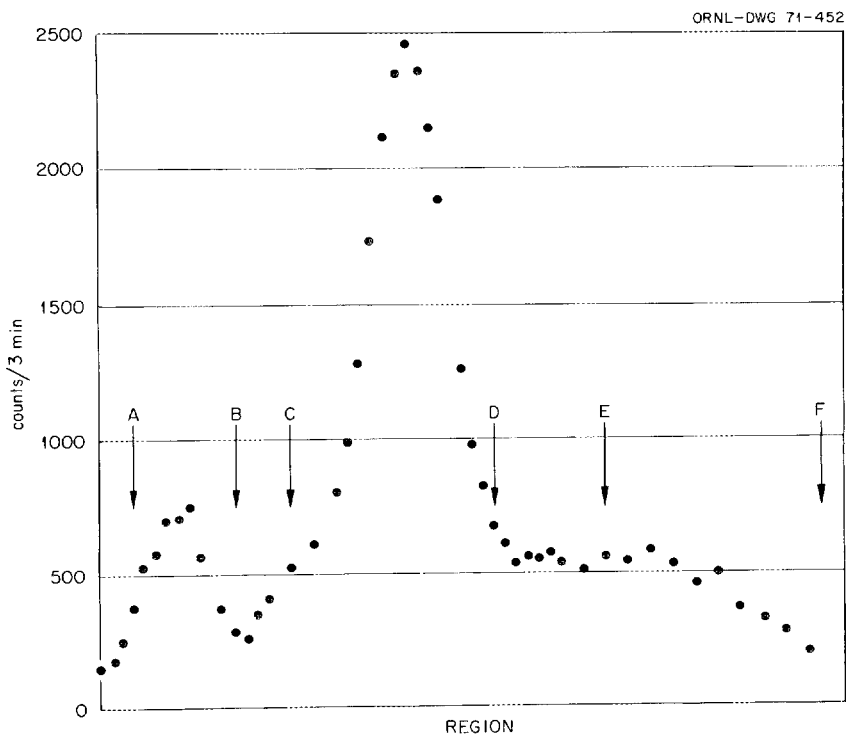

Fig. 12. Step-scanning profile of volunteer (Fig. 11*b*) 43 days after intake of [^{203}Hg]methylmercury nitrate. Borders between anatomical regions are: *A*, skull cap; *B*, plane beneath tragus at one fourth distance between tragus and jugular notch; *C*, jugular notch; *D*, umbilicus; *E*, ischial tuberosity; *F*, knee. From Aberg *et al.* (183).

be balanced against possible biological hazards in order to establish equitable standards. No attempt will be made here to evaluate these costs; rather, they are simply enumerated in order to appreciate the scope of the problem (5):

(1) Cost to manufacturer to meet standards, whether realistic or arbitrary, for their effluents;

(2) Cost of added enforcement, regulation, inspection, and control, and of holding inventories pending decision;

(3) Cost of subsidies (currently under consideration by state governments, for example) to compensate businessmen hurt from either the commercial or sport fish bans;

(4) Cost of health care and loss of labor income attributable to morbidity caused by mercury poisoning;

(5) Loss of income to states resulting from a reduced demand for commercial and sport fishing licenses;

(6) Loss of revenues for commercial fishermen and those whose livelihood is linked to the recreational facilities of impounded or suspect areas (although these businessmen are relatively few in number, the loss to them as individuals is absolute and catastrophic);

(7) Loss to processors, distributors, and retailers of all types of fish due to reduced sales volume and to promotional expenses incurred while dissociating their products from suspect specimens;

(8) Loss to the consuming public in that their range of choice is effectively reduced by fear of a whole class of food products.

In all of these cases, the loss to each level and sector of the economy has a "multiplier" impact on many other sectors. It is far too early to anticipate what the net longer-term economic and social consequences of the mercury pollution problem will be.

8. EXISTING STANDARDS AND TOLERANCE LIMITS

Standards are prescriptive; they prescribe pollutant levels that cannot legally be exceeded. They are established in the light not only of applicable criteria but also of such additional factors as technological feasibility, the costs involved and the time required to achieve the goals established by the criteria.

— Birmingham. Federal Government and Air and Water Pollution, 23 Business Lawyer 467.

A. Air

No official standard for mercury presently exists in the U.S. Over the past five years, the American Conference of Governmental and Industrial Hygienists has recommended as threshold limit values a time-weighted average concentration of 100 $\mu g/m^3$ for metallic vapor and inorganic compounds, and 10 $\mu g/m^3$ for organic mercury compounds during working conditions. These values are derived from studies performed in the hat-felt industry on the effects of metallic mercury vapor [(45,46); see section 7Biii] and from an assumption that organic mercurials are more rapidly absorbed through the respiratory tract. Notice has been served, however, that ACGIH intends to change the recommended threshold limit values to 10 $\mu g/m^3$ for alkylmercurials and 50 $\mu g/m^3$ for all other forms of mercury (185).

After a careful consideration of the available evidence, an international symposium convened in Stockholm in 1968 recommended that the "maximum allowable concentration" (MAC) values for mercury should be set at 100 μg mercury/m^3 for inorganic, phenyl-, and methoxyethylmercury salts, 50 $\mu g/m^3$ for mercury vapor, and 10 μg mercury/m^3 for alkylmercury compounds, and further advised that women of child-brearing age should not have *any* exposure to the alkylmercurials (2). MAC values were defined as "that average concentration in the air which causes no signs or symptoms of illness or physical impairment

in all but hypersensitive workers during their working day on a continuing basis, as judged by the most sensitive internationally accepted tests."

In the U.S.S.R. the permissible limits have been stated to be 10 $\mu g/m^3$ for inorganic mercury and 5 $\mu g/m^3$ for alkylmercury compounds (2). These values are considered as *ceiling values* which should never be exceeded even for brief exposure. The recommended safe level of mercury vapor adopted by the U.K. is 100 $\mu g/m^3$ and by Germany is 1 $\mu g/m^3$ (186).

B. Water

Up until 1970, drinking water standards of the U.S. Public Health Service and the World Health Organization did not include limits for mercury (187,188). In the spring of 1970, in response to inquiries received from health authorities, water utilities, and the American Water Works Association, the Bureau of Water Hygiene of the U.S. Public Health Service tentatively proposed a standard of 5 ppb for mercury in drinking water (189). This is the same standard that has been customarily quoted by various officials in this country for a number of years and that was officially set by Russian authorities nearly 20 years ago (190,191). No detailed reasoning was given by the Russian authorities for the adoption of the 5 ppb value, and it probably represents a balance of the recognized toxic effects of mercury and a desire to have "zero" levels on the one hand with the limitations of routine analyses for mercury at the time. The limit of 5 ppb is considered by the Bureau of Water Hygiene "to contain a reasonably safety factor for the protection of human health in consideration of degree of exposure, routes of entry, metabolic rate and excretion rate of the heavy metal" (189).

C. Food

Although the U.S. Food and Drug Administration has seized on occasion mercury-treated seed grains which were diverted by irresponsible persons into food channels, there are presently no established tolerances or standards in the U.S. for mercury residues in any food products (92).

In 1966 the World Health Organization recommended that the ADI for organic mercurials in food be zero and set the "practical residue limit" as 0.02 to 0.05 ppm (192). A "practical residue limit" was defined as that level of residue which is "found in foods from background and natural environmental contamination" and which should not be exceeded.

By late 1966, it had become apparent to Swedish authorities that "background" levels in fish were considerably higher than 0.02 to 0.05 ppm and that a number of fish from Swedish waters contained residues higher than 1.0 ppm. An official toxicological evaluation was therefore made and published in 1967 (193). The argument in this evaluation was that in the Minamata incident in Japan [see section 7Bii(2)], affected persons ate, generally daily, fish and shellfish with an average mercury content of 50 ppm and that according to Japanese pharmacological experience a decrease in mercury intake by a factor of 10, i.e., fish containing 5 ppm, should prevent poisoning. A relatively low safety factor of 5 was adopted, and the proposed limit was thus set at 1 ppm. It has been subsequently pointed out, however, that the toxicological evaluation contained a serious error, in that the Japanese data for mercury content of fish were based on dry weight analyses, whereas determinations in other countries (including Sweden) are based on the wet ("as received") weight of samples (4). Since the normal water content of fish is about 80%, a recalculation of the data yields a final proposed limit of 0.2 ppm, leaving aside any appraisal of the methodology or other assumptions used in the official toxicological evaluation. Sweden has been reluctant to adopt this lower standard, however, since it would involve banning of probably most fishing areas in Sweden (182); rather it has recommended that fish containing between 0.2 and 1.0 ppm mercury not be consumed more than once a week.

The U.S. Food and Drug Administration has recently set an "interim guideline" of 0.5 ppm after taking this error and the average consumption of fish in the U.S. into consideration (92) [the average consumption of fish in Sweden is about four times the U.S. average (194)]. At the time this guideline was proposed, however, analysts for FDA were using an analytical method which was unreliable for fish samples containing less than 0.5 ppm (92), and this consideration was also a factor which entered into the proposed interim guideline [more recent methods used by these scientists can now detect 0.01 ppm in fish (189)]. The Federal Food, Drug, and Cosmetic Act empowers the FDA to take regulatory action against any food product, either imported or in interstate commerce, which is found to contain an amount of adulterant equal to or greater than that specified by the interim guideline.

9. APPENDICES

A. Some Physical and Chemical Properties of Mercury (195)

Several physical properties of metallic mercury are summarized in Table 17 (196). Those properties which have been found particularly useful to industry and science are its conductivity, uniform thermal expansion, high density and surface tension, and liquidity at ordinary temperatures. Another important property of metallic mercury is its ability to dissolve many metals to form amalgams.

With an atomic number of 80, the quantum shells of mercury are filled through 6*s*, which, according to theory, can contain only two electrons. Removal of these to form a cation exposes a shell containing 18 electrons, which is an inert gas configuration. Hence, mercury never shows a valency greater than two, is surprisingly volatile, and, except for the noble gases, is the only element having a vapor which is monatomic at room temperature (197).

The saturation concentration of mercury in air can be calculated from its vapor pressure and is provided as a function of temperature in Fig. 13 (27). It should be noted that at room temperature (20–24°C) the saturation concentration of mercury in air is 130–180 times greater than the presently recommended ACGIH threshold limit value for exposure to man. Hence, it is conceivable that mercury vapor in the environmental air could reach a concentration that would be harmful and even fatal (27).

Metallic mercury is regarded as virtually insoluble in water, yet an anomaly is evidenced when attempts are made to measure its solubility. At room temperature in deaerated water its solubility is 20–30 ppm; at the same temperature but with the water saturated with oxygen, it is around 40,000 ppm. Probably neither value represents solubility, but rather a measure of the inability of mercury to maintain nobility in an increasingly hostile environment. The increase in apparent solubility is caused in part by the oxidation of mercury to mercuric oxide, with subsequent hydrolysis to the relatively soluble mercuric hydroxide.

Metallic mercury is not oxidized by dry air at room temperature, however, and is inert toward most gases except the halogens, hydrogen sulfide, and sulfur vapor, which combine directly with it even at room temperature. In fact, sulfur powder has been recommended as a means of rendering spilled mercury less toxic, since it coats the metal with HgS (196).

Mercury forms two series of salts, traditionally considered as being univalent and bivalent. However, it has been shown that the "univalent" compounds contain the group Hg_2^{2+} (or $^{+}Hg{-}Hg^{+}$), with two mercury atoms covalently bound to each other, so this series is actually bivalent also (198). Univalent (mercurous) salts, except the nitrate, are mostly insoluble, and the bivalent (mercuric) series is mostly soluble, except the iodide and sulfide. The extreme insolubility of the sulfide (solubility product of 4×10^{-53}) is perhaps fortunate, as it has promoted the deposition of millions of tons of mercury in the sulfide form (cinnabar). If the compound were soluble, rain and weathering might have distributed the mercury throughout the planet in sufficient concentrations to be poisonous, and life as we know it might have been impossible (197).

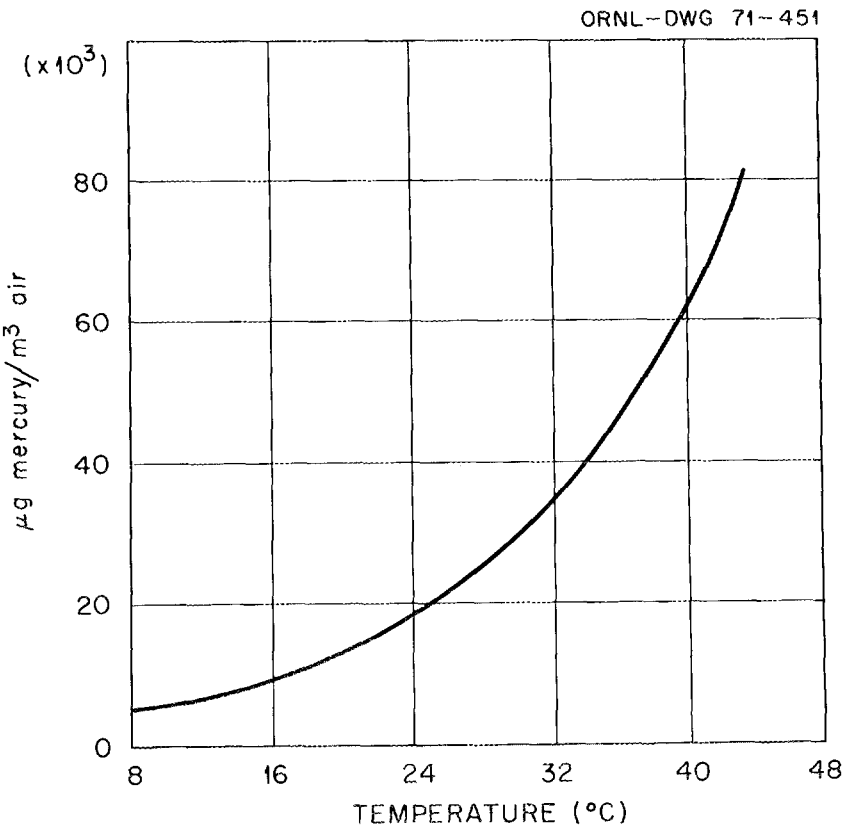

Fig. 13. Saturation concentration of mercury in air *vs* temperature. From Stahl (27).

The standard electrode potentials at 25°C of the inorganic mercury couples are (199,200):

$$2\,Hg^0 \rightleftharpoons Hg_2^{2+} + 2e^- \quad (-0.79 \text{ volts})$$

$$Hg^0 \rightleftharpoons Hg^{2+} + 2e^- \quad (-0.85 \text{ volts})$$

$$Hg_2^{2+} \rightleftharpoons 2\,Hg^{2+} + 2e^- \quad (-0.92 \text{ volts}).$$

Such potentials are a measure of thermodynamic susceptibility to oxidation, and the significant feature of the mercury couples indicated is that all the potentials are so close in magnitude. Thus, practically any oxidizing agent which is able to oxidize mercury (Hg^0) to mercurous ion (Hg_2^{2+}) is also able to oxidize mercury to mercuric ion (Hg^{2+}) or mercurous to mercuric ion. Conversely, almost any agent that can reduce mercuric to mercurous ion can also reduce either mercuric or mercurous ion to mercury.

A very important chemical transformation of mercury involves the electron exchange reaction:

$$Hg_2^{2+} \rightleftharpoons Hg^{2+} + Hg^0 .$$

The degree of disproportionation will depend upon the solubility or amount of dissociation of the mercuric compound formed and the extent to which metallic mercury enters or leaves the system. Divalent mercury also forms a large number of complexes of the general type $M_2[Hg(X)_4]$, where M can be a variety of

cations and X can be represented by halides or cyanide. Complexes with Hg–S bonds are also quite numerous. Most mercuric complexes do not have mercurous analogs, however, since such complexes readily disproportionate into the corresponding mercuric complex and metallic mercury (196).

The above considerations thus partially set the stage for an understanding of the geochemical migration of mercury through the environment as it passes through a welter of complexes, compounds, oxidation states, and phases. The final setting to the stage involves another important property of mercury alluded to before, namely its pronounced tendency to form covalent instead of ionic bonds. Thus it has a remarkable ability among metals to form compounds with organic radicals, normally linking covalently to a carbon atom. The replacement of hydrogen atoms in organic compounds with mercury is almost as easy as bromination or nitration, and the number of compounds which can be formed in this way is exceedingly large.

Organic mercury compounds can be conveniently classified into two types, RHgX and R_2Hg, where R is an organic radical and X an inorganic radical. RHgX compounds in general are crystalline solids whose properties depend upon the nature of X. When X is chlorine, bromine, iodine, cyanide, thiocyanide, or hydroxyl, the compound is a covalent non-polar substance more soluble in organic liquids than in water. When X is a sulfate, nitrate, phosphate, or perchlorate radical, the substance is salt-like, that is, ionic. R_2Hg compounds are non-polar, volatile, toxic liquids or low-melting solids. All are thermally unstable and light-sensitive.

B. Analytical Methodology for Environmental Mercury

The sampling and determination of mercury in the environment present some extremely challenging problems. These relate not only to the variety of sample matrices and to the very low quantities of mercury that are encountered, but also to the recognized volatility of mercury compounds and to their tendency to adsorb on particles and surfaces. These latter characteristics place great importance upon the sampling, sample handling, and chemical treatment portions of the analytical scheme. Obviously, even the most sensitive and sophisticated measurement technique is to no avail if mercury is lost prior to the measurement. One of the purposes of this discussion is to consider these important aspects of the mercury analysis question. A brief evaluation of several analytical measurement techniques along with the methodology for estimating organic-bound mercury is also given, and references are included to further background material.

i. Sampling and sample treatment. Because of its volatility, mercury in trace amounts is likely to be found almost anywhere. Care must be taken to prevent contamination of samples by traces of mercury from the reagents, labware, or even the air of the laboratory. Labware should be well rinsed with nitric acid and water before use. The great tendency of mercury to adsorb on glass is well known. Adsorption on silica is weak, but Pyrex ware is generally thought to be suitable for most work (201). However, this idea is being questioned increasingly because of the extreme sensitivities of present detection methods, and because of the very small mercury concentrations that are encountered. A recent study (202) of mercury losses from acidified 10^{-4} and 10^{-7} *M* $HgCl_2$ solutions on ten different materials suggests that Pyrex, polycarbonate, and Teflon are the best materials for storing and handling mercury, but there are significant losses with time on all these materials, *even at the 10^{-4} M level.* The analytical implications of these findings are twofold. First, suitable materials for sample collection and treatment must be selected. Second, samples must be analyzed quickly after they are collected. It should be emphasized that considerable uncertainty surrounds this question of mercury loss. Filtration and acidification of the samples are frequently recommended in the literature as hedges against mercury loss (203), but such treatments themselves may alter the sample.

The determination of organic-bound mercury usually requires decomposition of the organic material. The number of organic-bound mercury compounds that can be quantitatively decomposed to yield mercury metal, without preliminary oxidation of the organic material, is limited (204). Combustion is problematical due to the volatility of mercury and its compounds, and hence the usual procedure involves a wet oxidation. Many different oxidizing agents (including nitric acid, perchloric acid, hydrogen peroxide, ammonium persulfate, potassium permanganate, potassium chlorate, and chlorine) have been used under a variety of conditions for this purpose (204). There is no consensus about the relative merits of these agents. The procedures are tedious, time-consuming, and often do not liberate all the mercury in the sample. For analysis of such environmentally based materials as water, sludge, mud, and fish, the permanganate–sulfuric acid digestion technique appears to be used most frequently (205,206). It must be emphasized that recovery of a "spike" of an inorganic mercury compound is not conclusive evidence that organic-bound mercury is quantitatively liberated in a wet oxidation treatment. Neither is recovery of a spike of a type of organomercury compound that differs from that in the sample. Such experiments simply indicate that the procedure may work, not that the procedure does work.

ii. Analytical methods for determining mercury. (*1*) *Spectrophotometric dithizone method.* Several methods have been proposed for the colorimetric determination of trace levels of mercury, and these have been discussed in detail in the reference literature (201,207). Generally, the selectivity of these methods is not great. The diphenylthiocarbazone (dithizone) method is probably the most widely used colorimetric procedure. It involves reaction of Hg_2^{2+} or Hg^{2+} with excess dithizone in a fairly acid (1 *N* HNO_3 or H_2SO_4) solution to form colored complexes that are soluble in carbon tetrachloride or chloroform. The absorbance of the complex itself is measured at 490 nm, or alternatively the decrease in dithizone absorbance at 610 nm is measured. These procedures can be used to determine 0.5 to 50 ppm mercury by suitable choice of sample size. Relative errors are generally 2–4%. Copper, silver, gold, palladium, and platinum(II) interfere in trace amounts. Lead, zinc, nickel, and cobalt interfere if present in large amounts. The use of masking reagents can significantly decrease these interferences (201). Addition of acetic acid to the aqueous mercury solution prior to extraction is often recommended as a precaution against photochemical decomposition of the organic solution of mercury dithizonate.

The dithizone procedure finds greatest utility for routine analysis of dissolved or aqueous samples that contain fairly large (for "trace") concentrations of mercury. It presumes that mercury is present in inorganic form, but some organomercuric compounds are also extracted and detected (203). The procedure therefore cannot be used to distinguish inorganic from organic-bound mercury, nor can it be assumed to give total mercury in organic-based samples without some preliminary treatment that insures decomposition of organic materials.

(*2*) *Atomic absorption spectrophometry.* The determination of mercury by conventional flame emission or absorption spectrometry is not widely used, primarily because there is an unconventional technique that affords superior sensitivity. This so-called flameless or cold-vapor technique (208–211) is being adopted to such an extent that it may soon become the standard method. It involves chemical reduction of mercury in the sample to metallic mercury, volatilization of the mercury into a long-path-length absorption tube, and measurement of the absorption of the 253.7 nm mercury resonance line by the entrained mercury vapor. This procedure is both sensitive (the detection limit is 0.2 ppb) and rapid. There are few metallic interferences, and about ten minutes per determination is required. The procedures are usually designed to avoid strong anionic mercury complexes, which might hinder the reduction and volatilization of mercury. However, the fact is sometimes overlooked that many organic substances absorb in the ultraviolet region and that such absorption can constitute a significant error in the analysis. A rather simple correction for the type of error has been proposed (212), but is not often utilized.

Because of the inherent simplicity and sensitivity of this flameless atomic absorption technique, it is being adopted widely for determining total mercury in many types of samples (203,206,211,213). Again, the prime assumption and limitation is that mercury in the sample is present as or is converted to an inorganic, unbound form and hence can be reduced and volatilized.

(*3*) *Neutron activation analysis.* Mercury, like many other elements, can be determined with great sensitivity by neutron activation analysis. This technique involves exposing a sample to a source of neutrons, during which time a stable isotope of mercury may capture a neutron to produce a radioactive nuclide (214).

The basic equation of activation analysis,

$$A = Nf\sigma[1 - \exp(-0.693t/t_{1/2})] ,$$

states that the induced radioactivity (A) equals the product of the number of target mercury atoms (N), the neutron flux (f), the neutron cross section (σ), a measure of the probability of capture, and an exponential term involving the irradiation time (t) and the half-life ($t_{1/2}$) of the radionuclide product. The latter decays, emitting characteristic gamma rays whose energy can be measured by use of a NaI(Tl) or Ge(Li) detector.

Two radioactive mercury nuclides are produced by activation: ^{197}Hg($t_{1/2}$ = 67 hr) and ^{203}Hg($t_{1/2}$ = 47 d). A short irradiation produces primarily ^{197}Hg, but some ^{203}Hg is also formed; as the irradiation time is increased and/or the decay time of the irradiated sample increases, the ratio of ^{203}Hg to ^{197}Hg becomes greater. The use of ^{197}Hg gives greater sensitivity (215); thus, ideally one would irradiate for a short period and determine mercury through measurement of the ^{197}Hg directly. However, when the total mercury present is at the few-microgram level, the gamma rays from ^{197}Hg may be obscured by gamma rays from more abundant elements present. Then a chemical separation must be made: inactive mercury carrier (a few mg) is added, mercury precipitated or extracted, the isolated mercury fraction is counted, and the total yield through the procedure calculated (216).

Advantages of the neutron activation method include high sensitivity, freedom from chance contamination since there is no treatment of sample prior to irradiation, independence of the chemical form of the element sought, the possibility of nondestructive determination when mercury is above 1 μg or in a non-interfering medium such as water, and high and unequivocal specificity. Furthermore, with a flux of 10^{12} $n/cm^2/sec$, mercury can be determined at levels as low as 1 nanogram. Disadvantages are that neutron activation analysis is not adaptable to field use and that large numbers of samples require special irradiation facilities and data handling. The technique requires minimal working time (but not elapsed time) when radiochemical separations are not required. Neutron activation analysis is particularly useful for special and referee analyses.

(*4*) *X-ray fluorescence.* Recent developments in instrumentation and sources have made x-ray fluorescence analysis attractive for examination of environmental type samples (217). The method requires a source of low-energy photons, which are used to excite the characteristic x-rays of the element sought. Ideally one uses a monochromatic x-ray source with energy just greater than the x-ray energy of the sought element; either L or K x-rays can be excited and used, but because of absorption problems the higher energy of K x-rays makes them more suitable for use. The characteristic x-rays emitted by the elements in the sample are then sorted and measured using a solid-state Ge(Li) or Si detector coupled to a multi-channel analyzer. The exciting source may be an x-ray tube (with slits or other arrangement to single out a particular wavelength), or an isotopic source. The latter has the advantages of portability and relative cheapness (218).

Mercury K x-rays have an energy of about 69 kev; mercury L x-rays are about 10 kev. Where samples are uniform and rather thin, L x-rays can be used; for sludges, soils, residues, etc., it is better to excite and measure K x-rays. Positive identification is made through precise energy measurement (resolution of less than 1 kev is possible). By calibration with known standards and integration of observed x-ray peaks, quantitative values can be obtained. The sensitivity of K x-ray fluorescence for determination of mercury is to some extent a function of the matrix and the other elements present. These interferences can be minimized by narrowing the energy range of the exciting radiation. As the exciting energy approaches the absorption edge of the characteristic x-ray of interest, background as seen by the detector is reduced, and sensitivity and selectivity are improved. By changing this wavelength (either through use of a monochromator in conventional dispersive x-ray production or through change of target in radioisotope excitation) the focus can be changed from one element to another. In analyzing water it has been found advantageous to separate and concentrate the metals in question; one way of doing this is by use of ion exchange paper. By use of this technique and excitation by an x-ray emission spectrograph, for example, 1 μg of mercury has been determined in color additives (219). At ORNL these results have been duplicated using ion exchange paper to concentrate mercury from water solution (220); a radioisotope source was used to produce strontium K x-rays, which excited mercury L x-rays.

For field use a radioisotope-excited source would be desirable because of its portability and non-power requirements. For excitation of mercury L x-rays a source of ^{241}Am (59.6 kev) could be used. To excite K x-rays of mercury, ^{109}Cd (87.7 kev) and ^{57}Co (122 kev) have been used. The sensitivity of the method for mercury is somewhat a linear function of source strength; thus increasing the intensity of exciting x-rays should enable one to detect smaller quantities of mercury. It seems possible that under optimum conditions a lower limit for mercury of 0.1 μg could be reached. If so, the x-ray fluorescence method appears highly suitable for rapid examination of many samples containing 0.1 μg or more of mercury. It is especially adaptable to "yes or no" situations where one is interested in establishing that samples are below a particular level of concentration.

(5) *The determination of organomercury species.* Relatively little work has been done on the determination of organic-bound mercury itself, either *in toto* or as specific compounds, although there is a growing interest in such measurements. Gas-liquid chromatography possesses the necessary resolving power and detector sensitivity to attack these analytical problems, and it is the method that has received greatest attention to date (129,221–224). Thin-layer chromatography has also been suggested for this type of analysis. However, the basic problem here is to separate the organomercurials from their matrix reproducibly (preferably quantitatively), without alteration, and in large enough amounts to permit quantitative detection. Multiple-pass solvent extraction and re-extraction is used invariably as the initial step. Extraction of both the organomercurials and their dithizonates has been studied (221,222). The extracted group of compounds is then separated into individual species by gas-liquid chromatography (using an electron capture detector) or by thin-layer chromatography (using visual indication). Recoveries are of the order of 70–90%. Gas-liquid chromatography is superior to thin-layer chromatography for quantitative analysis.

These proposed procedures have not yet been used widely. They are sufficiently lengthy and complicated that they discourage routine use. The most recent results (224) indicate, however, that extraction of the dithizonates with alkaline cysteine in propan-2-ol, washing with ether, and then re-extracting with ether yields a solution that can be chromatographed with a polyethylene glycol succinate on Chromosorb G column. Methyl-, ethyl-, alkoxyethyl-, tolyl-, and phenylmercury can be resolved and determined. As little as 0.05 nanograms of the alkyl compound can be detected. These results indicate that the chromatographic approach to this problem is a sound one, and that greatly improved procedures are likely to be forthcoming as research in this area increases. Such studies are clearly warranted.

A small amount of polarographic work has been done with the organomercurials (225–227). While satisfactory polarographic procedures for these compounds may be developed, the methods are not likely to have the sensitivity that is needed for handling environmental samples. The polarographic procedures will be useful primarily as supplementary and referee methods.

Recently, an emission spectrophotometric device (228) was described that utilized a radiofrequency helium plasma to determine mercury in water. The device is rather simple, subject to few interferences, and quite sensitive. The most promising characteristic, however, is that it responds to combined and elemental mercury in the same sample. It is likely that this approach could be developed into a rather inexpensive and rapid method for determining organic-inorganic mercury ratio, methylmercury, dimethylmercury, and the singificant arylmercury compounds.

C. The Mercury Cell Chlor-Alkali Process

Approximately 30% of the U.S. supply of chlorine comes from mercury cells, with most of the balance being produced by the so-called diaphragm electrolytic cell, which does not use mercury (13). In recent years, the trend in new construction has greatly favored the mercury cell, presumably because of the higher grade NaOH product, which is highly concentrated and is essentially free of chloride impurities. The total chlorine capacity of mercury cells in the U.S. is about 8000 tons per day from about 35 installations around the country, with plant capacities ranging from 20 to about 700 tons per day. The average cell requires about 1500 lb of mercury per ton of daily chlorine capacity, and in 1969 the average loss of mercury was about 0.5 lb per ton of chlorine produced. However, this loss can apparently be decreased by one to two orders of magnitude by proper plant management. As we shall see below, some of this management involves internal recycling of streams which were formerly considered wastes.

Chlor-alkali plants are largely captive, i.e., either the chlorine or the NaOH is sold within the company. About 50% of the chlorine goes to the plastics industry. Other major uses include bleaching (pulp and paper industry, textile industry), sewage treatment, and as a component of many chemicals including pesticides. Alkali users with captive plants include the aluminum industry, which is the biggest user, as well as the glass, paper, petroleum, and detergent industries. With the exception of the photographic industry, which requires mercury-free alkali, the direct output of the mercury cell process is satisfactory for most users. On the other hand, the direct output of the diaphragm cells (only 11–12% NaOH) may require both concentration and chloride removal for some purposes. Still, the diaphragm cell often competes favorably with the mercury cell.

The power consumption of the mercury cells is about 4300 kwhr per ton of chlorine, compared to an average of 3200 kwhr per ton for the diaphragm cells. The total power consumption for the chlorine industry is 3.9×10^6 kw, or 1.5% of the power consumption of the country.

Before going into the mercury effluent problem, a brief description of the process is in order. A schematic representation of a mercury cell chlor-alkali plant is shown in Fig. 14. It should be pointed out that there are several different varieties of commercial mercury cells, including De Nora (50% of total), Uhde (20%), Sauve (10%), Mathieson (10%), and others (10%). The schematic only represents a general idea of what any specific operation would be like. A circulating sodium chloride solution (in some cases potassium chloride is used) is electrolyzed in the cell in which mercury serves as the cathode; the anode is usually graphite. Sodium ions react with the mercury to form an amalgam, and chlorine gas is produced at the anode. The mercury flows through the cell, picking up sodium on the way, and is pumped to a regeneration cell, where the amalgam is mixed with water and forms one half of a self-shorted electrolytic cell, with iron serving as the other electrode. The regeneration cell removes sodium from the amalgam and

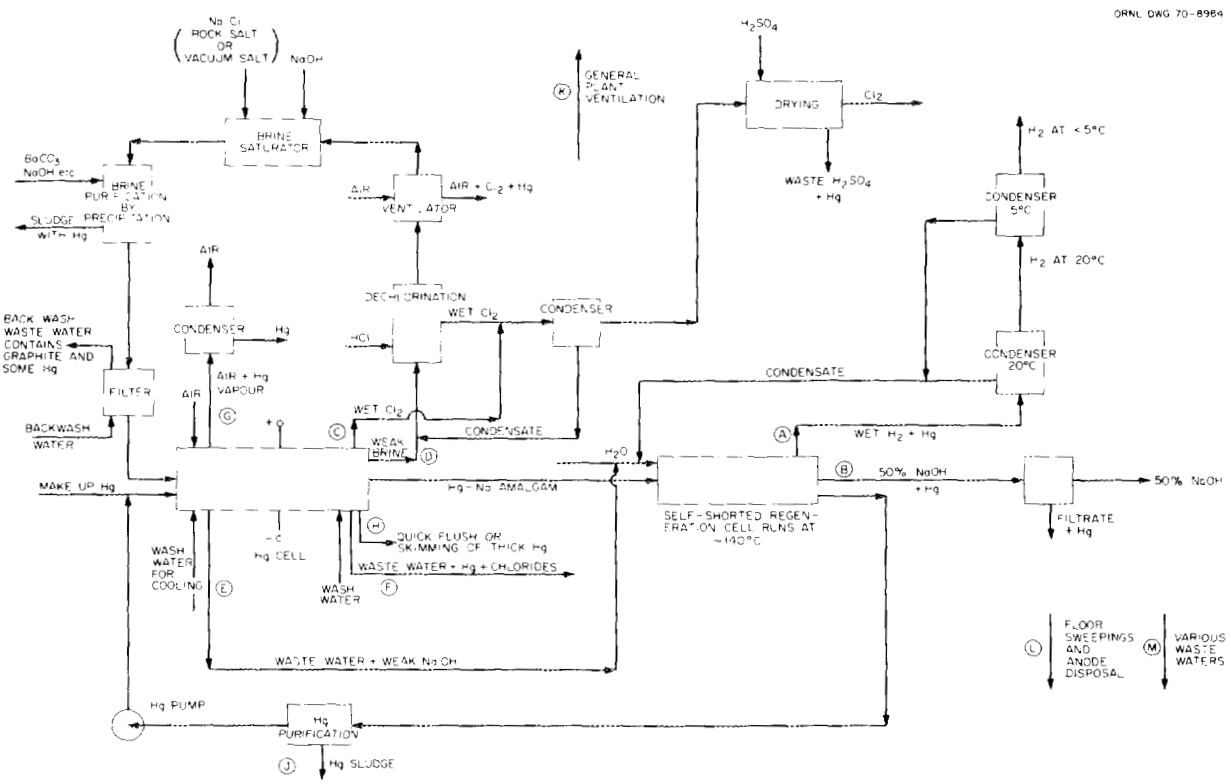

Fig. 14. Flow diagram for the chlor-alkali mercury cell process showing potential sources of mercury effluents.

produces NaOH (~50% solution) and hydrogen gas. The regenerated mercury is then returned to the mercury cell, and the process is repeated. The brine solution leaves the mercury cell and is vacuum de-chlorinated and then purged with air to remove the last of the chlorine. The solution is then saturated with salt. This is followed by a purification step, in which various impurities from the newly added salt are precipitated. The brine is then passed through a filter and back to the mercury cell.

The principal sources of loss as estimated for Swedish and U.S. plants are given in Table 18 (229,230). The range of values provided for the Swedish plants (229) is indicative of process variations. For example, the amount of mercury lost in the sludge from the brine purification process depends significantly on the particular impurities in the makeup salt. Rock salt tends to lead to large losses, whereas the losses are relatively small if salt from vacuum evaporation is used. Thus, a significant saving in mercury can be accomplished by using the proper salt. Unfortunately, the losses estimated in Table 18 for a number of U.S. plants (230) are not in particularly good agreement with the Swedish data. However, a recent Hungarian study (231) using isotopic tracers would tend to corroborate the Swedish data in that most of the mercury was found to escape to the hydrogen gas condensors.

Thus, the most significant recycle step which can be made in the process is to return the condensate from the hydrogen stream condensers to the regeneration cell makeup water. Another improvement would be to use the mercury cell wash and cooling water as part of the regeneration cell makeup water. Floor sweepings and anode deposits can also be recovered. The mercury in the sludges from the brine purification step, as well as in the filtrates of the brine and NaOH filters, could potentially be recovered, but at the

present time a good percentage of this mercury is probably being deposited in settling ponds or in landfills. How difficult it is to recover the mercury from such deposits is an open question.

D. Some Recommendations for Further Work

"Would you tell me, please, which way I ought to walk from here?"
"That depends a good deal on where you want to get to," said the Cat.
"I don't much care where – " said Alice.
"Then it doesn't much matter which way you walk," said the Cat.
" – so long as I get *somewhere*," Alice added as an explanation.
"Oh, you're sure to do that," said the Cat, "if you only walk long enough."

– Carroll. Alice in Wonderland.

i. Economic and social factors related to mercury usage. We do not have a *detailed* inventory of mercury flow through U.S. society. This is particularly needed with respect to recycle flow, since for the present we only know that some of it comes from batteries, electrical apparatus, scrapped mechanical and control devices, unused dental amalgam, and sludges from electrolytic processes in which mercury is used as a catalyst (3). Specific data do not appear to be available either through the Department of the Interior (Bureau of Mines) or elsewhere in the Federal Government, so we need to obtain this information by contacting the processors, scrap dealers, and representative industries. *Perhaps what is ultimately required is an accountability system for mercury and other persistent, toxic substances similar to the one used by the Atomic Energy Commission for fissionable materials.*

In our brief discussion of the societal flow of mercury (section 2), we mentioned several important externalities which impinge upon materials operations. Such factors also need further consideration in much greater detail. A beginning has been made by an economic analysis of the mercury industry prepared for the U.S. General Services Administration (232). However, the primary emphasis of this study was on price determinants and the development of an econometric model; several of its conclusions were that exploration is not an important determinant of mercury supply and that there is some responsiveness of demand to price after a one-year lag, but mercury demand on the whole is not very elastic. *Future studies are needed which place particular emphasis both on the social costs of present use practices and on those factors (prospecting subsidies, tariff regulations, market needs, etc.) which would apply to recycle potential and to alternate use and materials technology.*

ii. Research in analytical measurements. There is a major necessity for research into the techniques for sampling, sample storage, and sample preparation. The object of this research would be to establish which of the many methods in use are valid, and to devise new and/or improved techniques where they are needed. Reliability and validity in analytical measurements are fundamental; yet sophisticated detection schemes and equipment are worthless if the sample integrity itself is not maintained. We need the answers to such questions as: How to collect the sample? What type container? How long can it be stored and under what conditions? How can inhomogeneities be minimized? How should results be based (dry, wet, as received)? Which oxidation procedures really work? Is freeze-drying an acceptable practice? Are there alternatives to wet oxidation?

Research is also needed into ways and means for rapid, preferably portable, and non-destructive analysis of total mercury. X-ray fluorescence, with both conventional and radioisotope-excited sources, should be explored. Neutron activation analysis with a portable neutron source (^{252}Cf) also offers promise, particularly as a "mapping" technique.

Analytical techniques for determining organomercurials, individually and as a group, need improvement (particularly for methylmercury, because of its toxicity and prevalence). Studies into separation procedures

for these compounds *in toto* should be accelerated; advances in the separations schemes are needed much more than for the quantitation portions of the procedures. There is also a need for simple and rapid instrumental methods for determining organic-to-inorganic mercury ratio, or total-mercury-to-methylmercury, or perhaps total mercury and percent methylmercury, so that these determinations can be made widely and routinely. Pyrolysis techniquis should be explored; the helium plasma emission spectrometry approach appears to be especially promising, and its development should be expedited.

Closely related to the above work is a need for the development of standards of representative types that could be widely circulated. Some work is underway already on this problem at several laboratories, including those of the International Atomic Energy Agency and the Water Quality Office on the U.S. Environmental Protection Agency. This work should be accelerated, and a program of correlative analyses should be initiated. Standards of fish, sediments, particulates, etc., should be developed, and they should contain both organic and inorganic mercury compounds in meaningful quantities. These could do much to minimize discrepancies among various procedures and laboratories.

iii. Restitution of mercury-polluted lakes and streams. Mercury introduced by industrial effluents into water systems is primarily incorporated into bottom sediments, and these sediments may subsequently exchange their mercury load with the overlying water for a period of 10–100 years (112). At the recent conference on mercury pollution held at Ann Arbor (33), it was revealed that mercury contamination remains biologically active today in a Swedish lake into which mercury dumpage was stopped 25 years ago. Similarly, in the U.S., mercury-contaminated sediments of a lake in Wisconsin are still active 12–15 years after mercury emissions from a pulp and paper mill were halted. On the other hand, mercury-contaminated sediments in another Swedish lake have been naturally deactivated by being covered with several decimeters of deposits during the ten years following the curtailment of pollution. The point is that some lakes may be closed indefinitely to commercial and sport fishing unless artificial means can be found to decontaminate them. Some of the methods which have been suggested for this purpose are listed in Table 19, along with the advantages and disadvantages. All such processes, together with their economic costs and biological effects, need further investigation. It will also be important to screen a given deactivation method through a number of simulated sets of possible aquatic conditions. Thus, one must consider aerobic and anaerobic environments as well as pH and the type of food chain involved.

iv. Evaluation and elimination of atmospheric mercury pollution. Considerable mercury vapor emanation can occur during the smelting of mercury as well as other ores and concentrates. Mercury vapor is also apparently emitted during the burning of fossil fuels, and a preliminary estimate indicates that as much as 1800 tons of mercury may be released annually in the U.S. from coal burning alone (section 3A). However, more detailed information is needed on mercury emissions from all such processes in order to evaluate adequately the seriousness of the problem. Mercury vapor released into the atmosphere is effectively brought back to earth by rain, yet it would subsequently contaminate the aquatic environment to a more serious extent than the terrestrial. The assumption made here is that it would not be directly incorporated into the food chain of terrestrial animals, as are alkylmercury seed dressings, but rather would eventually migrate by hydrogeochemical and other processes to river and lake bottoms and municipal sewage-treatment plants. The validity of this assumption also needs corroboration.

However, the ultimate need, pending confirmation of a serious problem, is for methods which would remove dissolved mercury from petroleum and its products and mercury vapor from stack gases generated by smelters and coal-burning plants. Natural gases containing mercury vapor appear to be effectively decontaminated simply by mixing with "sour" gases containing hydrogen sulfide. A system operating on a similar principle and incorporating a filter to remove precipitated mercuric sulfide could perhaps be developed for stack gases.

v. Mercury levels in water and fish. Many pristine lakes have recently been found to contain fish with elevated levels of mercury (1). Large pelagic oceanic fish such as tuna and swordfish have also been shown to contain high mercury concentrations (94). Neither of these findings is completely understood, and we need to know whether they have occurred by natural processes or the intervention of man. In this regard, it would be well worth performing analyses on feathers of avian fish-eaters taken from museum specimens collected over the past 100 years, similar to the study which was done in Sweden (80). The kingfisher (*Megaceryle alcyon*) is probably a prime candidate for such a survey. At the same time, more information needs to be collected on the migratory behavior and feeding habits of the oceanic species along with a detailed analysis of their food chains.

A preliminary study has also indicated that there is a definite but unknown threshold level of mercury in aquatic ecosystems above which fish cannot eliminate mercury from their axial musculature faster than it is incorporated (80) (Fig. 10). This needs documentation in much greater detail, with particular attention to the relationship between water mercury levels and the accumulation of mercury in fish. Water mercury levels in this case should be defined not only in terms of the ambient water content of various mercurials, but also in terms of the potential for generation and turnover of mercury compounds by the bottom sediments of a given body of water. Such studies would help officials set realistic standards for industrial effluents.

vi. Genetic and long-term effects of mercury exposure. Cytological investigations on plant and animal cells have shown that mercury compounds give rise to chromosome breakage and act as inhibitors of the mitotic spindle mechanism, with the result that polyploidy or abnormal distribution of single chromosomes occurs. As spindle inhibitors, methyl- and phenylmercury compounds are more potent than any other substances known, including colchicine (2). Such preliminary observations would imply that both an increased genetic load of mutations within living populations and long-term effects such as carcinogenesis may be influenced by mercury in the environment. Much more basic information is thus needed to sharpen our understanding of these matters. Sources of this information would range from binding studies of mercury with DNA (233) to cytological studies on sufficiently large groups of subjects exposed particularly to alkylmercury compounds (234). Further investigations should also be undertaken on the various mercury compounds used in pesticides and pharmaceuticals, with immediate attention given to compounds of the latter group which are now used as constituents of many contraceptives. Test systems are available for both *in vitro* and *in vivo* analyses, and in order to select the most appropriate procedure to use in these evaluations, the Environmental Mutagen Society (235) should be consulted. Because of the relative lack of information, a carcinogenesis testing program for mercurials also needs to be initiated.

vii. Site and mode of action of methylmercury. More knowledge is required concerning the distribution of mercury in the body, and in particular within the central nervous system. Its mode of action on nerve cells, fibers, or synapses is essentially unknown. Methylmercury levels in several organs, particularly the liver, would appear to be higher after exposure than in the brain (183) (Fig. 12), and yet the brain is the first organ which is affected by increasing levels of methylmercury. With severe intoxication the symptoms are irreversible, indicating a threshold level of brain damage has been surpassed. These observations would imply that the brain is susceptible because methylmercury destroys cells, and neural tissue (unlike liver) is non-regenerative. A thorough study is thus needed on the mechanism by which methylmercury enters cells and can initiate cell death. It is generally assumed that mercury and methylmercury act by blocking sulfhydryl groups of enzymes (236), yet such a mechanism is an inadequate explanation for the effect of methylmercury on neural tissue (237). Furthermore, mercury is capable of combining with phosphoryl groups in cell membranes (238) and with other organic ligands of enzyme systems such as amino and carboxyl groups (236,239). It is also known from bacterial studies that once mercury enters the cell,

very small amounts will promote the breakdown of ribonucleic acid (240). Thus, primary emphasis should be placed on dynamic mercury distribution studies by cell fractionation (241) and electron microscopic techniques which aim to define the intracellular action site. Finally, since it is obvious that the apparent health of a person exposed to methylmercury does not exclude the possibility that his residual brain capacity has been lowered, histopathological and psychological studies of possible brain cell damage in suitable animals exposed to sublethal doses of methylmercury are also needed (4).

viii. A protective agent for methylmercury poisoning. A variety of substances, probably acting in different ways, have been found to reduce the toxicity or promote the excretion of inorganic mercury. These have included maleate (242,243), penicillamine (243,244), 2,3-dimercaptopropanol or BAL (242), and most recently spironolactone (245). Treatment with BAL has been the most widely used therapeutic procedure for mercury poisoning. Unfortunately, however, none of these substances have been shown to be effective against methylmercury poisoning, and there is evidence that BAL may even promote the passage of phenyl- and methylmercury into the brain (246). One of the most urgent needs, therefore, is an antidote to block the action of methylmercury, and support should be given to any efforts along this line. Most likely, however, an effective protective agent will not be found until more is known about the site and mode of action of methylmercury.

10. REFERENCES

A. Specific

1. W. Prouty. Statement to the Environmental Subcommittee to the U.S. Senate Committee on Commerce. Sen. Congr. Rec. S12279 (July 29, 1970).
2. M. H. Berlin *et al.* Arch. Env. Health **19**: 891 (1969).
3. G. N. Greenspoon. *In* Mineral Facts and Problems. U.S. Bureau of Mines (1970).
4. G. Löfroth. Ecological Res. Com. of the Swedish Nat. Sci. Res. Council, Bull. No. 4 (1969).
5. H. L. Seagran. Limnos 3(2): 3 (1970).
6. N. A. Smart. Residue Reviews **23**: 1 (1968).
7. Mercury in the Environment. U.S. Geol. Survey Prof. Paper 713 (1970).
8. E. S. Muskie. Bioscience **20**: 267 (1970).
9. Metal Statistics, 56th ed. Metallgesellschaft, Frankfurt am Main (1969).
10. Bureau of Mines Minerals Yearbook (1968).
11. Trends in Usage of Mercury. National Materials Advisory Board Report NMAB-258 (1969).
12. J. R. Manion, Jr., Business and Defense Services Administration, Miscellaneous Metals and Minerals Div., Dept. of Commerce. Private communication (Aug. 13, 1970).
13. North American Chlor-Alkali Industry Plants and Production Data Book (Chlorine Institute Pamphlet No. 10). The Chlorine Institute, Inc., 342 Madison Avenue, New York, N.Y. 10017 (1970).
14. J. M. West. *In* Bureau of Mines Minerals Yearbook (1968).
15. Anonymous. Chem. Engineering 75(2): 18 (1968).
16. D. E. H. Frears. Pesticide Handbook, 18th ed. State College, Pa., Entomological Society of America, College Science Publishers (1966).
17. M. N. Gleason, R. E. Gosselin, and H. C. Hodge. Clinical Toxicology of Commercial Products. Williams and Wilkins Co., Baltimore, Md. (1957).
18. Geological Survey Research 1968. U.S. Geological Survey Professional Paper 600 (1968).
19. J. E. Shelton. *In* Mineral Facts and Problems. U.S. Dept. of the Interior, Bureau of Mines Bull. No. 630 (1965).
20. Anonymous. Chem. and Eng. News **48**(26): 36 (June 22, 1970).
21. Dept. of Interior News Release, Sept. 16, 1970. Covered in SFI Bull. **220**: 6 (1970).
22. C. M. Tarzwell. House Congr. Rec. H8576 (Sept. 10, 1970).
23. A. W. Hoover and L. J. Goldwater. Arch. Env. Health **12**: 506 (1966).
24. E. M. Farkas, Office of Public Affairs, Environmental Protection Agency. Private communication (Jan. 19, 1971).
25. M. Dall'Aglio. *In* Origin and Distribution of the Elements – A Symposium (L. H. Ahrens, ed.), p. 1065. Pergamon Press, New York, N.Y. (1968).
26. I. R. Jonasson. Geological Survey of Canada, Paper 70-57 (1970).
27. Q. R. Stahl. Air Pollution Aspects of Mercury and Its Compounds. Litton Systems, Inc., Bethesda, Md. (1969).
28. E. A. Sergeev. Internat. Geology Rev. **3**: 93 (1961).
29. Mercury Potential of the United States. Bureau of Mines Information Circular 8252 (1965).
30. S. W. Maher, Chief Geologist, East Tenn. Branch, Tenn. Div. of Geology. Private communication (Jan. 6, 1971).
31. A. Stock. Sv. Kem. Tidskr. **50**: 242 (1938).
32. A. J. Johnels, T. Westermark, W. Berg, P. I. Persson, and B. Sjöstrand. Oikos **18**: 323 (1967).
33. International Conference on Environmental Mercury Contamination, held at Univ. of Michigan, Ann. Arbor, Mich., Sept. 30–Oct. 2, 1970. Proceedings summarized in Env. Science Technol. **4**: 890 (1970).
34. J. H. McCarthy, Jr., W. W. Vaughn, R. E. Learned, and J. L. Meuschke. U.S. Geological Survey Circular 609 (1969).
35. O. Weiss. Abst. 3rd International Geochem. Symp. (Toronto), p. 63 (1970).
36. S. H. Williston. J. Geophys. Res. **73**: 7051 (1968).

37. A. Andersson and L. Wiklander. Grundförbättring **18**: 171 (1965).
38. D. E. White and C. E. Roberson. *In* Petrologic Studies – A Volume to Honor A. F. Buddington, p. 397. Geol. Soc. America (1962).
39. J. Cholak. Proc. 2nd Nat'l. Air Pollution Symp., Pasadena, Calif. (1952).
40. S. S. Brar, D. M. Nelson, E. L. Kanabrocki, C. E. Moore, C. D. Gurnham, and D. M. Hatahori. Nat'l. Bur. Standards Spec. Pub. 312, Vol. 1, p. 43 (1969).
41. L. J. Goldwater. Cited by Stahl (27).
42. L. Friberg. Nord. Hyg. Tidskr. **32**: 240 (1951).
43. P. Seiffert and H. Neudert. Zbl. Arbeitsmed. **4**: 129 (1954).
44. P. L. Birdstrup, J. A. Bonnell, D. G. Harvey, and S. Locket. Lancet **261**: 856 (1951).
45. P. A. Neal *et al.* U.S. Public Health Service Bull. No. 234 (1937).
46. P. A. Neal *et al.* U.S. Public Health Service Bull. No. 263 (1941).
47. G. A. Neville. Canadian Chem. Education **3**: 4 (1967).
48. A. Stock and F. Cucuel. Naturwiss. **22**: 390 (1934).
49. F. Heide and G. Böhm. Chemie Erde **19**: 198 (1957).
50. H. Hamaguchi, R. Kuroda, and K. Hosohara. Nippon Kagaku Zasshi **82**: 347 (1961).
51. K. Hosohara. Nippon Kagaku Zasshi **82**: 1107 (1961).
52. N. K. Aidin'yan. Akad. Nauk SSSR, Trudy Inst. Geol. Rudn. Mestorozhd., Petrog., Mineral., Geokhim. **70**: 9 (1962).
53. N. K. Aidin'yan, N. A. Ozerova, and S. K. Gipp. Akad. Nauk SSSR, Trudy Inst. Geol. Rudn. Mestorozhd., Petrog., Mineral, Geokhim. **99**: 5 (1963).
54. K. Hosohara, H. Kozuma, K. Kawasaki, and T. Tsuruta. Nippon Kagaku Zasshi **82**: 1479 (1961).
55. L. Wiklander. Grundförbättring **21**: 151 (1968).
56. N. V. Kvashnevskaya and Y. I. Shablovskaya. Akad. Nauk SSSR, Doklady **151**: 426 (1963).
57. E. H. Bailey, P. D. Snavely, Jr., and D. E. White. U.S. Geol. Survey Prof. Paper 424D, p. D306 (1961).
58. B. J. Skinner, D. E. White, H. J. Rose, Jr., and R. E. May. Econ. Geol. **62**: 316 (1967).
59. A. P. Vinogradov. The Geochemistry of Rare and Dispersed Chemical Elements in Soils. Consultants Bureau, Inc., New York, N.Y. (1959).
60. A. Andersson. Grundförbättring **20**: 95 (1967).
61. S. Furutani and Y. Osajima. Kyushu Daigaku Nogakubu Gakugei Zasshi **22**: 45 (1965).
62. J. A. Pickard and J. Grainger. Ann. Rep. Agr. Hort. Res. Sta., Long Ashton, Bristol, p. 65 (1962).
63. T. Westermark. Press communication, cited by Lofroth (4).
64. J. E. Larsson. Environmental Mercury Research in Sweden. Swedish Environment Protection Board, Research Secretariat, Stockholm (1970).
65. D. H. Klein and E. D. Goldberg. Env. Science Technol. **4**: 765 (1970).
66. J. N. Spencer and A. F. Voigt. J. Phys. Chem. **72**: 464 (1968).
67. C. K. Kim and J. Silverman. Anal. Chem. **37**: 1616 (1965).
68. W. A. Haller *et al.* *In* Modern Trends in Activation Analysis, p. 177. National Bureau of Standards Publication No. 312 (1969).
69. R. E. Jervis, D. Debrun, W. LePage, and B. Tiefenbach. Summary of Progress, Canadian National Health Grant No. 605-7-510 (Trace Mercury in Environmental Materials) for the period Sept. 1969–May 1970. Partially summarized in Chem. Eng. News **48**(42): 8 (Oct. 5, 1970).
70. N. A. Smart and A. R. C. Hill. J. Sci. Fd. Agric. **19**: 119 (1968).
71. T. Goto and A. Sato. J. Jap. Soc. Fd. Nutr. **19**: 245 (1966).
72. C. Tomizawa, A. Kobayashi, M. Shibuya, Y. Koshimizu, and Y. Oota. Shokuhin Eiseigaku Zasshi **7**: 33 (1966).
73. M. G. Raeder and E. Snekvik. Kongl. Videnk. Selsk. Forhandl. **13**: 169 (1941).
74. N. A. Smart. Nature **199**: 1206 (1963).
75. B. S. Legatowa, B. S. Hordynska, and I. Bernstein. Roczniki Pzh. **14**: 221 (1963).
76. E. G. Pappas and L. A. Rosenberg. J. Assoc. Off. Agr. Chemists **49**: 792 (1966).
77. G. Westöö. *In* Chemical Fallout (M. W. Miller and G. G. Berg, eds.), p. 75. C. C. Thomas, Springfield, Ill. (1969).

78. G. Westöö. Vår Föda **19**: 1 (1967).
79. R. Christell, L. G. Erwell, K. Ljunggren, B. Sjöstrand, and T. Westermark. Proc. Internat. Conf. in Modern Trends in Activation Analysis, Dallas, Tex. (1965).
80. A. G. Johnels and T. Westermark. *In* Chemical Fallout (M. W. Miller and G. G. Berg, eds.), p. 221. C. C. Thomas, Springfield, Ill. (1969).
81. J. Boëtius. Med. Danm. Fiskeri-og Harunderögelser **3**(4): 93 (1960).
82. K. Borg. Proc. VIII Nord. Veterinärmötet, Helsingfors, p. 394 (1958).
83. K. Borg, H. Wanntorp, K. Erne, and E. Hanko. J. Appl. Ecol. **3**(suppl.): 171 (1966).
84. P. J. Conder. H.M.S.O. Sixth Rep. from the Estimates Committee, M.A.F.A., p. 298 (1961).
85. J. H. Koeman and H. Van Genderen. Proc. 17th Internat. Symp. Phytopharmacy, Gent. p. 1879 (1965).
86. J. F. Eades. Nature **210**: 650 (1966).
87. D. Lihnell and A. Stenmark. Statens Vaxstskyddsanstalt Meddenlanden **13**: 110 (1967).
88. S. Dalgaard-Mikkelsen. Nord. Hyg. Tidskr. **50**: 34 (1969).
89. V. Sjöblom and E. Häsänen. Nord. Hyg. Tidskr. **50**: 37 (1969).
90. B. Underdal. Nord. Hyg. Tidskr. **50**: 60 (1969).
91. W. Berg, A. G. Johnels, B. Sjöstrand, and T. Westermark. Oikos **17**: 71 (1966).
92. A. C. Kolbye. Testimony before the Subcommittee on Energy, Natural Resources, and the Environment of the Senate Committee on Commerce (May 8, 1970).
93. A. C. Celeste and C. G. Shane. FDA Papers, p. 27 (November 1970).
94. Assoc. Press. Chattanooga Times (Dec. 25, 1970).
95. Anonymous. New York Times (Oct. 30, 1970).
96. L. Altman. New York Times (Nov. 7, 1970).
97. K. Norén and G. Westöö. Vår Föda **19**: 13 (1967).
98. M. M. Joselow, L. J. Goldwater, and S. B. Weinberg. Arch. Environ. Health **15**: 64 (1967).
99. G. C. Griffith, E. M. Butt, and J. Walker. Ann. Intern. Med. **41**: 501 (1954).
100. G. Birke, A. G. Johnels, L. O. Plantin, B. Sjöstrand, and T. Westermark. Svenska Läkardidn. **64**: 3628 (1968).
101. A. K. Perkons and R. E. Jervis. J. Forensic Sci. **11**: 50 (1966).
102. S. Tejning. Report No. 68 02 20, Dept. Occup. Med., Univ. Hosp., Lund, Sweden (1968).
103. A. Jernelöv. *In* Chemical Fallout (M. W. Miller and G. G. Berg, eds.), p. 68. C. C. Thomas, Springfield, Ill. (1969).
104. S. Jensen and A. Jernelöv. Nature **223**: 753 (1969).
105. C. G. Rosén. Nord. Hyg. Tidskr. **50**: 180 (1969).
106. J. M. Wood, F. S. Kennedy, and C. G. Rosén. Nature **220**: 173 (1968).
107. R. Romer and W. A. Noyes, Jr. J. Am. Chem. Soc. **71**: 3390 (1949).
108. T. Suzuki, K. Furukawa, and K. Tonomura. J. Ferment. Technol. **46**: 1048 (1968).
109. K. Furukawa, T. Suzuki, and K. Tonomura. Agr. Biol. Chem. **33**: 128 (1969).
110. K. Rissanen, J. Erkama, and J. K. Miettinen. Paper submitted to F.A.O. Technical Conference on Marine Pollution and Its Effects on Living Resources and Fishing. Rome (Dec., 1970).
111. T. Fagerström and A. Jernelöv. In the Press (1970).
112. A. Jernelöv and S. Freyhuss. Communication from the Swedish Water and Air Pollution Research Laboratory, Stockholm (Feb. 19, 1968). Cited by Löfroth (4).
113. C. A. Campacci, P. Pigati, and A. M. S. Marcondes. Biologica **35**: 213 (1969).
114. M. Lagervall and G. Westöö. Vår Föda **21**: 9 (1969).
115. J. A. Pickard and J. T. Martin. Ann. Rep. Agr. Hort. Res. Sta., Long Ashton, Bristol, p. 93 (1959).
116. R. G. Ross and D. K. R. Stewart. Can. J. Plant Sci. **40**: 117 (1960).
117. R. G. Ross and D. K. R. Stewart. Can. J. Plant Sci. **42**: 280 (1962).
118. W. A. Glooschenko. J. Phycol. **5**: 224 (1969).
119. T. B. Hasselrot. Inst. Freshw. Res. Drottningholm Rep. **48**: 102 (1968).
120. L. Hannerz. Inst. Freshw. Res. Drottningholm Rep. **48**: 120 (1968).
121. R. R. Rucker and D. F. Amend. Prog. Fish Culturist **31**: 197 (1969).

122. A. Jernelöv. Discussion *In* Chemical Fallout (M. W. Miller and G. G. Berg, eds.), pp. 93 and 240. C. C. Thomas, Springfield, Ill. (1969).
123. M. Berlin and S. Ullberg. Arch. Env. Health **6**: 589 (1963).
124. T. Suzuki, T. Miyama, and H. Katsunuma. Industr. Health **4**: 69 (1966).
125. V. L. Miller, R. A. Klavano, and E. Csonka. Toxic. Appl. Pharmacol. **2**: 344 (1960).
126. J. C. Gage and A. A. B. Swan. Biochem. Pharmacol. **8**: 77 (1961).
127. J. C. Gage. Brit. J. Industr. Med. **21**: 197 (1964).
128. G. Westöö. Vår Föda **19**: 121 (1967).
129. G. Westöö. Acta Chem. Scand. **22**: 2277 (1968).
130. V. L. Miller, P. A. Klavano, A. C. Jerstad, and E. Csonka. Toxic. Appl. Pharmacol. **3**: 459 (1961).
131. K. D. Lundgren, A. Swensson, and V. Ulfvarsson. Scand. J. Clin. Lab. Invest. **20**: 164 (1967).
132. L. J. Goldwater, A. C. Ladd, and M. B. Jacobs. Arch. Env. Health **9**: 735 (1964).
133. M. Berlin. Arch. Environ. Health **6**: 626 (1963).
134. M. Berlin and S. Ullberg. Arch. Environ. Health **6**: 616 (1963).
135. M. Berlin, C. A. Grant, and G. Nordberg. Cited by (2, 136) as "to be published" in Nord. Hyg. Tidskr.
136. F. Berglund and M. Berlin. *In* Chemical Fallout (M. W. Miller and G. G. Berg, eds.), p. 258. C. C. Thomas. Springfield, Ill. (1969).
137. L. Eckman, U. Greitz, G. Persson, and B. Åberg. Nord. Med. **79**: 450 (1968).
138. L. Eckman, U. Greitz, J- O. Snihs, and B. Åberg. Nord. Med. **79**: 456 (1968).
139. L. Magos. Brit. J. Industr. Med. **25**: 315 (1968).
140. M. Berlin, L. G. Jerksell, and H. von Ubisch. Arch. Environ. Health **12**: 33 (1966).
141. M. Berlin, J. Fazackerly, and G. Nordberg. Arch. Environ. Health **18**: 719 (1969).
142. G. Nordberg and F. Serenius. Cited by (2) as "to be published" in Acta Pharmacol.
143. L. Friberg. Arch. Industr. Health **20**: 42 (1959).
144. R. Ukeles. Appl. Microbiol. **10**: 532 (1962).
145. R. C. Harriss, D. B. White, and R. B. Macfarlane. Science **170**: 736 (1970).
146. P. A. Weir and C. H. Hine. Arch. Env. Health **20**: 45 (1970).
147. E. D. S. Corner and B. W. Sparrow. Marine Biol. Assoc. United Kingdom J. **35**: 531 (1956).
148. L. Popper. Abstracted in Can. Med. Assoc. J. **96**: 1225 (1967).
149. L. Dérobert and O. Marcus. Bull. Off. Soc. Med. Lég. Crim., France, **36**: 294 (1956).
150. M. Waldron. New York Times (Aug. 10, 1970).
151. B. Storrs *et al.* Morbidity and Mortality **19**(3): 25 (1970).
152. M. A. Jalili and A. H. Abbasi. Brit. J. Industr. Med. **18**: 303 (1961).
153. S. A. Raza Ali. Communication to W.H.O., summarized in Interagency Coordination in Environmental Hazards (Pesticides), Senate Hearings before the Subcommittee on Reorganization and International Organizations of the Committee on Government Operations, Append. IV to Part 1, p. 905 (March 1, 1965).
154. K. Irukayama. Third Intern. Conf. Water Pollution Res., Water Pollution Control Federation, Washington, D.C., p. 153 (1966).
155. Minamata Disease. Study group of Minamata disease, Kumamoto Univ., Japan (1968).
156. T. B. Eyl. J. Am. Med. Assoc. **215**: 287 (1971).
157. H. Heimann. Arch. Environ. Health **14**: 488 (1967).
158. L. T. Kurland, S. N. Faro, and H. Siedler. World Neurol. **1**: 370 (1960).
159. A. C. Ladd, L. J. Goldwater, and M. B. Jacobs. Arch. Environ. Health **9**: 43 (1964).
160. W. Massman. Zbl. Arbeitsmed. 7: 9 (1957).
161. T. W. Clarkson, J. Gatzy, and C. Dalton. Univ. Rochester A.E.C. Rep. No. 582 (1961).
162. L. J. Goldwater, A. C. Ladd, P. G. Berkhout, and M. B. Jacobs. J. Occup. Med. **6**: 227 (1964).
163. J. Lecomte. Compt. Rend. Soc. Biol. **143**: 1296 (1949).
164. H. Wilkening and S. Litzner. Deutsch. Med. Wschr. 77: 432 (1952).
165. D. Hunter, R. R. Bomford, and D. S. Russel. Quart. J. Med. **33**: 193 (1940).
166. J. Hosokawa. Kumamoto Med. Soc. **30**: 1252 (1956).
167. H. Tokuomi *et al.* Kumamoto Med. J. **14**: 47 (1964).

168. J. Engleson and A. B. Herner. Acta Paediat. Scand. **41**: 289 (1952).
169. H. G. Matsumoto, T. Koya, and T. Takeuchi. J. Neuropath. Exp. Neurol. **24**: 563 (1965).
170. A. V. Bakulina. Sov. Med. **31**(6): 60 (1968).
171. M. Umeda, K. Saito, K. Hirose, and M. Saito. Jap. J. Exp. Med. **39**: 47 (1969).
172. R. Weed, J. Eber, and A. Rothstein. J. Gen. Physiol. **45**: 395 (1962).
173. E. Arrhenius. Oikos **9**(suppl): 32 (1967).
174. H. Druckery, H. Hamperl, and D. Schmähl. Z. Krebsforsch. **61**: 511 (1957).
175. H. F. Malling, J. S. Wassom, and S. S. Epstein. Newsletter Env. Mutagen Soc. **3**: 7 (1970).
176. A. Granati, D. Scabo, and M. Andreoli. Folia Med. **40**: 496 (1957).
177. C. Ramel. Hereditas **57**: 445 (1967).
178. C. Ramel. Hereditas **61**: 208 (1969).
179. C. Ramel and J. Magnusson. Hereditas **61**: 231 (1969).
180. G. Fiskesjö. Hereditas **62**: 314 (1969).
181. G. Fiskesjö. Hereditas **64**: 142 (1970).
182. F. Berglund and M. Berlin. *In* Chemical Fallout (M. W. Miller and G. G. Berg, eds.), p. 423. C. C. Thomas, Springfield, Ill. (1969).
183. B. Aberg, L. Ekman, R. Falk, U. Greitz, G. Persson, and J-O Snihs. Arch. Env. Health **19**: 478 (1969).
184. J. K. Miettinen, T. Rahola, and T. Hattula. Fifth Symp. on Radioactivity Investigations in Scandinavia, Helsinki (1969).
185. Threshold Limit Values of Airborne Contaminants and Intended Changes Adopted by the American Conference of Governmental and Industrial Hygienists for 1970. A.C.G.I.H., 1014 Broadway, Cincinnati, Ohio.
186. Anonymous. Food Cosmet. Toxicol. **6**: 397 (1968).
187. Water Quality Criteria. Federal Water Pollution Control Administration, Washington, D.C. (1968).
188. V. Lambou. Testimony before the Subcommittee on Energy, Natural Resources, and the Environment of the Senate Committee on Commerce (May 8, 1970).
189. Anonymous. Am. Water Works Assoc. J. **62**: 285 (1970).
190. P. Kirkor. Sewage Industr. Wastes **23**: 938 (1951).
191. S. N. Czerkinski. Lit. Ber. Wass. Abwass. Luft u. Boden **4**: 98 (1955).
192. Pesticide Residues in Food. World Health Organization Technical Report No. 370 (1967).
193. F. Berglund and A. Wretling. Vår Föda **19**: 9 (1967).
194. Anonymous. SFI Bull. **217**: 6 (Aug. 1970).
195. A. J. Smith (Tenn. Valley Auth.) has provided several helpful comments for this section. Private communication (Jan. 13, 1971).
196. J. Roesmer. Radiochemistry of Mercury. Nat. Tech. Information Service, U.S. Dept. of Commerce, Springfield, Va. (1970).
197. Anonymous. Chemistry **40**(10): 18 (1970).
198. A. G. Sykes. Kinetics of Inorganic Reactions. Pergamon Press, New York, N.Y. (1966).
199. W. M. Latimer. The Oxidation States of the Elements and Their Potentials in Aqueous Solution. Prentice Hall, New York, N.Y. (1938).
200. W. Forsling, S. Hietanen, and L. G. Sillen. Acta Chem. Scand. **6**: 901 (1952).
201. E. B. Sandell. Colorimetric Determination of Traces of Metals, 3rd ed., p. 621. Interscience Publishers, Inc., New York, N.Y. (1959).
202. M. R. Greenwood and T. W. Clarkson. Amer. Ind. Hyg. Assoc. J. **31**: 250 (1970).
203. Y. Chau and H. Saitoh. Environ. Science Technol. **4**: 839 (1970).
204. J. F. Coetzee. *In* Treatise on Analytical Chemistry (I. M. Kolthoff and P. J. Elving, eds.), Part II, Vol. 3, p. 231. Interscience Publishers, New York, N.Y. (1961).
205. H. S. Booth, N. E. Schreiber, and K. G. Zwick. J. Am. Chem. Soc. **48**: 1815 (1926).
206. Anonymous. Determination of Mercury by Atomic Absorption Spectrophotometer Method. The Dow Chemical Company, Midland, Mich. 48640.
207. F. D. Snell and C. T. Snell. Colorimetric Methods of Analysis, 3rd ed., Vol. II, p. 63. Van Nostrand Co., New York, N.Y. (1949).

208. M. S. Dill. Report No. Y-1572, Union Carbide Corp., Oak Ridge, Tenn. (1967).
209. C. Ling. Anal. Chem. **40**: 1877 (1965).
210. W. R. Hatch and W. L. Ott. Anal. Chem. **40**: 2085 (1968).
211. A. O. Rathje. Amer. Ind. Hyg. Assoc. J. **30**: 126 (1969).
212. A. E. Ballard, D. W. Stewart, W. O. Kamm, and C. W. Zuehlke. Anal. Chem. **26**: 921 (1954).
213. B. B. Mesman and B. S. Smith. Atomic Absorption Newsletter **9**: 81 (1970).
214. Guide to Activation Analysis (W. S. Lyon, ed.). Van Nostrand Co., Princeton, N.J. (1964).
215. L. Kosta and H. R. Byrne. Talanta **16**: 1297 (1969).
216. C. K. Kim and J. Silverman. Anal. Chem. **37**: 1616 (1965).
217. Anonymous. Analytical Methods for Energy Dispersive X-ray Spectrometry (1969). Kevex Corp., 898 Mahler Rd., Burlingame, Calif. 94010.
218. J. R. Rhodes. Analyst **91**: 683, 1088 (1966).
219. W. B. Link, K. S. Heine, J. H. Jones, and P. Wattlington. J. Assoc. Offic. Agr. Chemists **47**: 391 (1964).
220. T. H. Handley, Oak Ridge National Laboratory. Private communication (December 1970).
221. G. Westöö. Acta Chem. Scand. **20**: 2131 (1966).
222. G. Westöö. Acta Chem. Scand. **21**: 1790 (1967).
223. K. Teramoto, M. Kitabatake, M. Tanabe, and Y. Noguchi. J. Chem. Soc. Japan, Ind. Chem. Sect. **70**(9): 1601 (1967).
224. J. O'G. Tatton and P. J. Wagstaffe. J. Chromatogr. **44**: 284 (1969).
225. T. M. Hopes. J. Assoc. Off. Agr. Chemists **48**: 585 (1965).
226. E. B. Bayer. J. Assoc. Off. Agr. Chemists **52**: 844 (1969).
227. R. Bensch and R. C. Bensch. J. Am. Chem. Soc. **73**: 3391 (1951).
228. R. W. April and D. N. Hume. Science **170**: 849 (1970).
229. H. O. Bouveng and P. Ullman. Reduction of Mercury in Waste Waters from Chlorine Plants. Report of the Swedish Water and Air Pollution Research Laboratory, reprinted by the Chlorine Institute, 342 Madison Ave., New York, N.Y. Also, Ind. Water Eng., 24 (June 1969).
230. J. Nichols and L. C. Fuhrmeister, Monsanto Chem. Co. Private communication (July 30, 1970).
231. B. Balla and L. Kalman. Kem. Kozlem. **32**(2): 173 (1969).
232. Economic Analysis of the Mercury Industry. Charles River Associates, Inc., 16 Garden St., Cambridge, Mass. (July 1969).
233. R. O. Rahn, M. D. C. Battista, and L. C. Landry. Proc. Nat. Acad. Sci. **67**: 1390 (1970).
234. S. Skerfving, K. Hansson, and J. Lindsten. Arch. Environ. Health **21**: 133 (1970).
235. A. Hollaender, Pres., Biology Division, Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830.
236. M. C. Battigelli. J. Occupational Med. **2**: 337 (1960).
237. Y. Yoshino, T. Mozai, and K. Nakao. J. Neurochem. **13**: 1223 (1966).
238. H. Passow, A. Rothstein, and T. W. Clarkson. Pharmacol. Rev. **13**: 185 (1961).
239. E. S. Cook and G. Parisutti. J. Biol. Chem. **167**: 827 (1947).
240. T. Beppu and K. Arima. J. Bacteriol. **98**: 888 (1969).
241. Y. Yoshino, T. Mozai, and K. Nakao. J. Neurochem. **13**: 397 (1966).
242. T. W. Clarkson and L. Magos. Brit. J. Pharmacol. **31**: 560 (1967).
243. T. Stoytcher, L. Magos, and T. W. Clarkson. Eur. J. Pharmacol. **8**: 253 (1969).
244. J. R. Brown, F. R. Jose, and M. V. Kulkarni. Med. Serv. J. Canada **23**: 1089 (1967).
245. H. Selye. Science **169**: 775 (1970).
246. M. Berlin and S. Ullberg. Nature **197**: 84 (1963).
247. G. H. Friedrick and H. E. Hawkes. Mineralium Deposita **1**: 77 (1966).
248. H. V. Warren, R. E. Delavault, and J. Barasko. Econ. Geology **61**: 1018 (1966).
249. J. T. Martin. Analyst **88**: 413 (1963).
250. H. V. Warren and R. E. Delavault. Oikos **20**: 537 (1969).
251. N. K. Aidin'yan, A. I. Troitskii, and G. A. Balavskaya. Geochem. Internat. **4**: 670 (1964).
252. A. G. Dvornikov. Akad. Nauk SSSR, Doklady **150**: 894 (1963).
253. A. G. Dvornikov and V. Y. Petrov. Geochem. Internat. **1**: 1021 (1961).

254. V. I. Morozov. Akad. Nauk SSSR, Doklady **163**: 209 (1965).
255. M. A. Karasik and V. I. Morozov. Geochem. Internat. **3**: 497 (1966).
256. Anonymous. Environment **11** (June): 53 (1970).
257. O. I. Joensuu. To be published in Science. Private communication (Jan. 22, 1971).
258. M. Brandenstein, I. Janda, and E. Schroll. Tschermaks Minerolog. Petrog. Mitt. **7**: 260 (1960).
259. M. A. Karasik, A. E. Vasilev'skaya, V. Y. Petrov, and E. A. Rutekhin. Akad. Nauk Ukrayin. RSR, Geol. Zhurn. 22(2): 53 (1962).
260. N. A. Ozerova. Akad. Nauk SSSR, Trudy Inst. Geol. Rudn. Mestorozhd., Petrog., Mineral., Geokhim. **72**: 1 (1962).
261. A. G. Dvornikov. Akad. Nauk SSSR, Doklady **150**: 894 (1963).
262. A. P. Bol'shakov. Geochem. Internat. **3**: 459 (1964).
263. A. G. Dvornikov. Akad. Nauk SSSR, Doklady **172**: 199 (1967).
264. A. G. Dvornikov. Akad. Nauk RSR, Dopovidi, Ser. B, **29**: 293 (1967).
265. A. G. Dvornikov. Akad. Nauk RSR, Dopovidi, Ser. B, **30**: 732 (1968).
266. Anonymous. Environment **12**(2): S-1 (March 1970).
267. Dept. of Interior News Release (July 2, 1970).
268. Dept. of Interior News Release (Aug. 20, 1970).
269. Anonymous. TVA Today **1**(4): 1 (1970).
270. Assoc. Press. Chattanooga Times (July 7, 1970).
271. R. Reed (New York Times News Svce.). Newport News Daily Press (July 16, 1970).
272. A. Stock. Biochem. Z. **304**: 73 (1940).
273. R. B. Forney and R. N. Harger. Fed. Proc. **8**: 292 (1949).
274. K. Samsahl, D. Brune, and P. O. Webster. AE-124, Aktiebolaget Atomenergi, Stockholm (1963).
275. L. A. Dal Cortivo *et al.* J. Forensic Sci. **9**: 501 (1964).
276. S. Tejning. Report No. 67 02 06, Dept. Occup. Med., Univ. Hosp., Lund, Sweden (1967).
277. P. Sumari *et al.* Cited by Löfroth (4) as "to be published" in Nord. Hyg. Tidskr.
278. S. Tejning. Report No. 67 08 31, Dept. Occup. Med., Univ. Hosp., Lund, Sweden (1967).
279. S. Tejning. Report No. 68 05 29, Dept. Occup. Med., Univ. Hosp., Lund, Sweden (1968).
280. O. Hoshino *et al.* Eisei Kagaku **12**: 94 (1966).

B. General

A recent review (3) of the technological aspects of mercury sources and uses is provided in the 1970 edition of "Mineral Facts and Problems," and much of the geological information has been put together and evaluated by the U.S. Geological Survey (7) and the Geological Survey of Canada (26). The Swedish experience is reviewed in the monograph on "Methylmercury" by Löforth (4) and various chapters (77, 80, 103, 136, 182) of the book "Chemical Fallout," whereas details of the Japanese experience are covered by review articles (154, 158) and a book (155) put out by a Kumamoto University study group. Much of the agricultural literature is discussed by Smart (6), while Stahl (27) has written a review specifically directed at the air pollution aspects of mercury and its compounds. The toxicological information has been evaluated by an international committee of experts (2), and references to many basic biological studies are provided in Malling *et al.* (175). A comprehensive 940-page, three-volume index/bibliography ("Environmental Aspects of Mercury Usage") with over 15,000 entries and listing more than 2,000 articles has been recently prepared by the Dow Chemical Company (Dow Ecology Council, c/o B. J. McNaught, 2020 Dow Center, Midland, Michigan 48640).

Table 1. Known mercury dischargers in the United States (as of September 1970)

Name and address	Receiving waters	Remarks
Allied Chemical Co., Buffalo, N.Y.	Buffalo River to Lake Erie	Discharged 0.66 lb/day on July 27.
Allied Chemical Co., Moundsville, W.Va.	Ohio River	Discharged 3.7 lb/day on July 15. Reduced load to 0.5–1.0 lb/day on Aug. 28. (Company value.)
Allied Chemical Co., Solvay, New York	Onondaga Lake	Discharged 4.4 lb/day on July 14.
Aluminum Co. of America, Point Comfort, Texas	Lavaca Bay	Early analyses not available. Sediment analyses indicate previously higher discharges. On July 29 to Aug. 2, 1.45–1.96 lb/day were discharged.
Buckeye Cellulose, Memphis, Tenn.	Wolf River to Mississippi River	Discharged 0.10 lb/day on Aug. 13.
Buckman Labs., Memphis, Tenn.	Lateral sewer to Wolf River interceptor to Mississippi River	Discharged 0.06 lb/day on Aug. 11.
Chapman Chem. Co., Memphis, Tenn.	Non Connah Creek to Mississippi River	Discharged 0.09 lb/day on Aug. 14.
Chesbrough-Ponds, Inc., Faichney Inst., Watertown, N.Y.	Black River to Lake Ontario	On Aug. 5, 1.50 lb/day of mercury were being discharged. (Subject to further investigation.)
Detrex Chem. Ind., Ashtabula, Ohio†	Ditch to Lake Erie	Discharged 2–60 lb/day in Mar.–Apr. and 3.0 lb/day on June 26. Reduced load to 1.48 lb/day on July 14.
Diamond Shamrock Chem. Co., Deer Park, Texas	Houston Ship Channel	Discharged 13.2 lb/day on May 15. Reduced load to 1.72–6.0 lb/day on July 18–27. On Sept. 1, discharge reduced to 1–2 lb/day and expects to reach less than 0.5 lb/day by Oct. 15. (Company value.)
Diamond Shamrock, Delaware City, Dela.*	Delaware River	Discharged 29.1 lb/day on July 14. Reduced load to 3.03 lb/day on Aug. 21.
Diamond Shamrock, Muscle Shoals, Ala.*	Pond Creek to Tennessee River	Discharged 8.6 lb/day on May 7. Reduced load to 3.25 lb/day on July 15.
Dow Chem. Co., Plaquemine, La.	Mississippi River	Company estimated earlier losses to be 40–50 lb/day. Discharged 3.2 lb/day on May 18. On July 15, mercury was not detected.
Garrett-Callahan Co., Millbrae, Calif.	Millbrae STP to San Francisco Bay	On Aug. 5, found to be discharging a small amount of concentrated waste.
General Aniline & Film Corp., Linden, New Jersey‡	Arthur Kill	Discharged 29.2 lb/day on July 17. Reduced load to 6.7 lb/day on Aug. 7.
General Electric Chem. Prod. Plant, Cleveland, Ohio	Lake Erie	Discharged 0.003 lb/day on Aug. 20.
General Electric Co., Edmore, Michigan	Cedar Lake to Pine River	Discharged 0.002 lb/day on Aug. 3–4.
General Mercury Corp., Tempe, Arizona	Ground water (via leach field)	On Aug. 14, found to be discharging a small amount of concentrated waste (equiv. to 0.001 lb/day).
Georgia Pacific, Bellingham, Wash.	Puget Sound	Discharged 10.5 lb/day on July 14. Reduced load to 0.17 lb/day on Aug. 10–12.
Goodrich Chem. Co., Calvert City, Ky.	Tennessee River	Company claims reductions prior to July 14. On July 14, 0.05 lb/day of mercury was discharged.
Hill Air Force Base, Ogden, Utah	North Davis Co. STP	Discharged 0.005 lb/day on Aug. 6.
Hooker Electrochem., Niagara Falls, N.Y.	Niagara Falls sewer system and Niagara River	Discharged 1.34 lb/day on July 23. Company will report improvement (Sept. 18 reporting date).

Table 1. (continued)

Name and address	Receiving waters	Remarks
International Mining & Chem. Co., Orrington, Maine*	Penobscot River	Discharged 2.65 lb/day on July 14. Reduced load to 0.22 on Aug. 19. (Company value.)
Mallinckrodt Chem., Erie, Pa.	City of Erie STP to Lake Erie	Calculated to be discharging 0.051 lb/day on July 28 (load determined by subtracting NOSCO Plastics load from combined NOSCO-Mallinckrodt load).
Monochem. Inc., Geismar, La.	Mississippi River	Discharged 0.91 lb/day on July 18. On Sept. 2, load less than 0.25 lb/day. (Company report.)
Monsanto Chem. Co., Texas City, Texas	Galveston Bay	Discharged 0.45 lb/day on Aug. 1.
NASA, Lewis Research Center, Cleveland, Ohio	Rocky River	Early analyses not available. Sediment analyses indicate previously higher discharges. On July 21, 0.02 lb/day was discharged.
NOSCO Plastics, Erie, Pa.	City of Erie STP to Lake Erie	Discharged 0.002 lb/day on July 28.
Olin Mathieson Chem. Corp., Augusta, Ga.*	Savannah River	Discharged 12.9 lb/day on May 20. Reduced load to 0.51 lb/day on July 14.
Olin Mathieson Chem., Charleston, Tenn.	Hiwassee River	Discharged 2.2 lb/day on July 16. Reduced load to 1.0 lb/day on Aug. 26. (Company value.)
Olin Mathieson Chem., McIntosh, Alabama	Tombigbee River	State issued statement that discharges were substantially reduced July 13. On July 14, 0.12 lb/day of mercury was discharged.
Olin Mathieson Chem., Niagara Falls, N.Y.*	Niagara River	Discharged 26.6 lb/day on July 14. Reduced load to 0.38–0.85 lb/day on Aug. 12–16. (Company values.)
Olin Mathieson Chem., Saltsville, Virginia	North Fork, Holston River	Early analyses not available. Sediment analyses indicate previously higher discharges. On Aug. 12, 0.58 lb/day was discharged.
Oxford Paper Co., Rumford, Maine*	Androscoggin River	Discharged 26.2 lb/day on July 14. Plant closed Aug. 15.
Pennwalt Chem. Co., Calvert City, Ky.*	Tennessee River	Discharged 1.54 lb/day on July 14.
Pioneer Paint & Varnish Co., Tucson, Arizona	Santa Cruz River	Discharged 0.006 lb/day on Aug. 21–22.
PPG Industries, Lake Charles, La.	Bayou d'Inde	Discharged 26.5 lb/day on July 20. On Sept. 1, no mercury being discharged; using a temporary lagoon. Permanent facilities are to reduce loading to less than 0.05 lb/day by Sept. 30. (Company report.)
PPG Industries, Natrium, W.Va.	Ohio River	Discharged 4.0 lb/day on July 15. Reduced load to 0.5–1.0 lb/day on Aug. 28. (Company value.)
Quicksilver Prod., San Francisco, Calif.	City of San Francisco STP to San Francisco Bay	On Aug. 5, found to be discharging a small amount of concentrated waste (equiv. to 0.004 lb/day).
Reactive Metals, Inc., Ashtabula, Ohio	West Branch, Fields Brook, to Fields Brook to Ashtabula, River	Discharged 0.199 lb/day on Aug. 3–4.
Riegel Paper Co., Riegelwood, N.C.	Cape Fear River	Discharged 6.32 lb/day on July 17. Reduced load to 0.59 lb/day on Aug. 10. (Company value.)
Stauffer Chem. Co., Axis, Alabama	Mobile River	State issued statement that discharges were substantially reduced July 13. On July 14, 0.07 lb/day of mercury was discharged.
Tenneco Chem. Co., Pasadena, Texas	Houston Ship Channel	Discharged approx. 1.0 lb/day on May 19. Reduced load to 0.02–0.15 lb/day on July 17–21.
Westinghouse, Fairmont, W.Va.	Monongahela River	Early analyses not available. Sediment analyses indicate previously higher discharges. On July 15, 0.19 lb/day was discharged.

Table 1. (continued)

Name and address	Receiving waters	Remarks
Weyerhaeuser Co., Longview, Wash.*	Columbia River	Discharged 15.1 lb/day on July 14. Reduction to approx. 1.0 lb/day by Aug. 17. (Company value.)
Williams Gold Refining Co., Buffalo, N.Y.	City of Buffalo STP to Niagara River	Discharged 0.001 lb/day on July 30.
Woodbridge Chem., Woodbridge, N.J.	Berrys Creek to Hackensack River	Sample of a discharge on Aug. 12 indicated 2.08 lb/day mercury. (Subject to further investigation)
Wyandotte Chem., Geismar, La.	Mississippi River	Discharged 1.70 lb/day on May 19. Reduced load to 0.91 lb/day on July 18.
Wyandotte Chem., Port Edwards, Wis.	Wisconsin River	Early analyses not available. Sediment analyses indicate previously higher discharges. On July 27, 0.08 lb/day was discharged.
Wyandotte Chem., Wyandotte, Mich.†	Detroit River	Discharged 11–74 lb/day from Mar. 27 to Apr. 10. Reduced load to 0.35–0.50 lb/day July 22–24.

*Firms subject to Federal suit under 1899 Refuse Act.
†Firms subject to State action.
‡Firms subject to 180-day notice abatement action under Federal Water Pollution Control Act.

Table 2. Maximum mercury concentration in air measured at scattered mineralized and nonmineralized areas of the western United States

Sample location	Maximum Hg concentration ($\mu g/m^3$)[1]	
	Ground surface	400 feet above the ground[2]
Mercury mines		
Ord mine, Mazatzal Mtns., Ariz.	20.000 (50)	0.108 (4)
Silver Cloud mine, Battle Mtn., Nev.	2.000 (50)	0.024 (8)
Dome Rock Mtns., Ariz.	0.128 (6)	0.057 (20)
Base and precious metal mines		
Cerro Colorado Mtns., Ariz.	1.500 (5)	0.024 (2)
Cortez gold mine, Crescent Valley, Nev.	0.180 (60)	0.055 (4)
Coeur d'Alene mining district, Wallace, Idaho	0.068 (40)	
San Xavier, Ariz.		0.025 (3)
Porphyry copper mines		
Silver Bell mine, Ariz.		0.053 (3)
Esperanza mine, Ariz.		0.032 (3)
Vekol Mtns., Ariz.		0.032 (4)
Ajo mine, Ariz.		0.030 (3)
Mission mine, Ariz.		0.024 (3)
Twin Buttes mine, Ariz.	0.020	0.022 (3)
Pima mine, Ariz.		0.013 (3)
Safford, Ariz.		0.007 (2)
Unmineralized areas		
Blythe, Calif.		0.009 (20)
Gila Bend, Calif.		0.004 (2)
Salton Sea, Calif.		0.004 (2)
Arivaca, Ariz.		0.003 (2)

[1] Number of measurements shown in parentheses.
[2] Samples taken from single-engine aircraft.

Table 3. Mercury in selected rivers of the United States, 1970

(detection limit 0.1 ppb)

Source and Location	Time sample collected		Mercury (in ppb)
	Mo.-day	Hour	
Gold Creek at Juneau, Alaska	6-10	1350	<0.1
Colorado River near Yuma, Ariz.	6-18		<0.1
Welton Mohawk Drain near Yuma, Ariz	6-19		<0.1
Quachita River downstream from Camden, Ark.	6-18	0900	<0.1
St. Francis River at Marked Tree, Ark.	6-19	1000	0.1
Santa Ana River below Prada Dam near Riverside, Calif.	6-29		<0.1
South Platte River at Henderson, Colo.	5-19	1410	0.3
Blue River upstream of Dillon Reservoir, Colo.	6-22		<0.1
French Creek near Breckenridge, Colo.	6-22		<0.1
Animas River at Silverton, Colo.	6-22		0.1
Cement Creek at Silverton, Colo.	6-22		<0.1
Red Mountain Creek near Ouray, Colo.	6-22		17.0
Red Mountain Creek at Ironton, Colo.	6-22		<0.1
Nuuanu Stream near Honolulu, Hawaii	6-8	0930	.6
Honolii Stream near Papaikou, Hawaii	6-8	1405	<0.1
North Fork Kaukonahua near Wahiawa, Hawaii	6-11	1800	0.4
Ohio River near Grand Chain, Ill.	6-26	1040	0.1
Floyd River at Sioux City, Iowa	6-9	1645	0.2
Kansas River downstream from Topeka, Kans.	5-19	1130	3.5
Mississippi River near Hickman, Ky.	6-25	1030	<0.1
Merrimack River above Lowell, Mass.	6-8	1100	1.2
Wolf Creek near Cedar Lake, Mich.	6-7	1100	<0.1
Unnamed tributary to Wolf Creek near Edmore, Mich.	6-7	1000	0.1
Rainy River at International Falls, Minn.	5-14	1245	<0.1
St. Louis River at Scanlon, Minn.	6-8	1015	<0.1
Pearl River at Byram, Miss.	6-17	1445	0.1
Pascagoula River at Merrill, Miss.	6-9	1500	3.0
Yellowstone River near Billings, Mont.	5-14	1500	<0.1
Missouri River near Great Falls, Mont.	5-18	1730	<0.1
Missouri River near St. Louis, Missouri	6-23	1430	2.8
Missouri River at Hermann, Missouri	6-24	1030	0.2
Salt Creek near Lincoln, Neb.	6-24	0915	0.5
Las Vegas Wash at Henderson, Nev.	5-14		<0.1
Pemigewasset River at Woodstock, N.H.	6-8	1700	3.1
Canadian River near Glenrio, N. Mex.	6-10	1100	<0.1
Hudson River downstream from Poughkeepsie, N.Y.	4-7		0.1
Hoosic River near North Pownal, Vt. in Rennsselaer County, N.Y.	4-7		0.1
Wappinger Creek near Wappingers, Falls, N.Y.	4-23	1045	<0.1
Delaware River at Port Jervis, N.Y.	4-23	1420	<0.1
Beaver Kill at Cooks Falls, N.Y.	4-24	1320	0.1
Deer River near Helena, N.Y.	5-5	0735	<0.1
Raquette River at Raymondville, N.Y.	5-5	0945	0.2
Oswegatchie River at Gouverneur, N.Y.	5-6	0800	0.7
Oswegatchie River at Gouverneur, N.Y.	6-16	1200	1.2
Black River at Watertown, N.Y.	5-6	1015	<0.1
Black River near Watertown, N.Y.	5-6	1155	<0.1

Table 3. (continued)

Source and Location	Time sample collected		Mercury (in ppb)
	Mo.-day	Hour	
Lake Champlain near Whitehall, N.Y.			<0.1
Lake Champlain near Ticonderoga, N.Y.			<0.1
Lake Champlain near Crown Point, N.Y.			0.1
Raquette River at Massena, N.Y.	6-16	0840	<0.1
Raquette River at Raymondville, N.Y.	6-16	0910	<0.1
Raquette River at Potsdam, N.Y.	6-16	0950	0.1
Oswegatchie River below Natural Dam, St. Lawrence County, N.Y.	6-16	1130	<0.1
Oswegatchie River at Hailsboro, N.Y.	6-16	1230	0.2
Chemung River near Wellsburg, N.Y.	7-6	1015	0.2
Susquehanna River at Johnson City, N.Y.	7-6	1330	0.1
Maumee River at Antwerp, Ohio	6-10	1215	6.0
Scioto River near Chillicothe, Ohio	6-25	1115	<0.1
Great Miami River near Miamisburg, Ohio	6-11	1815	0.9
North Canadian River near Harrah, Okla.	6-30	1000	1.1
North Canadian River near Oklahoma City, Okla.	6-30	1345	0.1
Whitewood Creek near Vale, S. Dak.	5-22	1100	<0.1
Paper Mill Creek near Herty, Tex.	6-9	1015	0.1
San Antonio River near Elmendorf, Tex.	6-11	1100	<0.1
Blackwater River at Franklin, Va.	6-15	0930	1.1
Jackson River near Covington, Va.	6-16	0820	<0.1
Bailey Creek near Hopewell, Va.	6-18	0945	0.4
Snohomish River near Monroe, Wash.	7-1	1050	<0.1
North Branch Potomac River near Barnum, W.V.	6-3	1600	1.2
Wisconsin River at Wisconsin Rapids, Wis.	6-10	1300	0.9
Wisconsin River near Nekoosa, Wis.	6-10	1230	2.4
North Platte River near Casper, Wyo.	6-23	1215	0.1
Bighorn River at Kane, Wyo.	6-30	1600	<0.1

Table 4. Analysis of soils for mercury in ppm

Area	Number of samples analyzed	Range		Average	Ref.
		Min.	Max.		
Nevada, Texas, California		0.02	0.04		36
Unmineralized areas, California		0.04	0.06		247
Franciscan Formation, California		0.10	0.20		36
Unmineralized areas, British Columbia		0.01	0.05		248
Near mineralization, British Columbia		0.05	2.50		248
Very near mineralization, British Columbia		0.25	2.50		248
England		0.01	0.06		249
England (topsoils)		0.25	15.00		250
Germany		0.03	0.29		48
Sweden (topsoils)	273	0.02	0.92	0.07	60
Africa (topsoils)	14			0.02	60
European U.S.S.R.	130	0.04	5.80		251
Donets Basin	248	<0.05	10.00	0.30	252
Donets Basin		0.10	2.40	1.30	253
Kerch Peninsula	264	<0.10	3.00		254
Kerch-Taman area		0.24	1.90		255
Viet Nam		0.02	1.00	0.30	251

Table 5. Mercury levels in water sediments

Body of water	Area	Level (ppm)	Reference
St. Clair River–Lake St. Clair	U.S. Side	Trace	20
Upper Detroit River	Upstream from Rouge River (U.S. side)	Up to 1.4	20
	Downstream from Rouge River (U.S. side)	Up to 2.0	20
Lower Detroit River	Within mile downstream of Wyandotte Chemical	5.4–86.0	20
	Further downstream to Lake Erie	Trace–26.0	20
Detroit River (main channel)	Between Grassy Island and Grosse Island	4.4	20
	East side of Fighting Island	1.2	20
	Near river's mouth	0.6	20
Western Lake Erie	Near mouth of Detroit River	1.0–2.1	20
	Along Michigan shore	0	20
	Laplaisance Bay	Up to 0.8	20
	Near West Sister Island	1.6–2.1	20
	0–15 miles east of Detroit River (5 miles from Ontario shore)	1.3–2.7	20
	Pelee Island vicinity	0	20
Eastern Lake Erie	Offshore from Cleveland harbor	2.4	20
	Mouth of Grand River	2.0	20
	Offshore from Easterly STP	4.0	20
	Presque Isle Bay	1.1	20
Black River		>1.0	20
Ashtabula River		>1.0	20
Buffalo River		>1.0	20
Missouri River		Up to 32	7
Wisconsin River		Up to 560	256

Table 6. Analyses of fossil fuels for mercury in ppm

Sample and source	Number of samples analyzed	Range		Average	Ref.
		Min.	Max.		
Coal, Ohio				0.5	33
Coal, U.S.A.	36	0.07	33.0	3.3	257
Coal (pit), Sweden		0.06	0.40		64
Coal, Germany (bituminous)	11	0.001	0.025	0.012	48
Coal, Germany (anthracite)	119	<1.0	2.7		258
Coal, Donets Basin U.S.S.R.		4.5	70.0	11.1	259
Coal, Donets Basin U.S.S.R.		0.14	300.0	46.0	260
Coal, Donets Basin U.S.S.R.	206	0.05	10.00	1.10	261
Coal, Donets Basin U.S.S.R.		2.5	6.5	3.7	262
Coal, Donets Basin U.S.S.R.	756	0.02	20.00		263
Coal, Donets Basin U.S.S.R.		0.1	7.0		264
Coal, Donets Basin U.S.S.R.	13	0.1	300.0	46.0	265
Fuel oil, Sweden				0.003	64
Crude oil, California		1.9	21.0		33,57
Light petroleum, California				100	7
Tarry petroleum, California				500	7
Tar, California				1.0	7

Table 7. Mercury levels in animals from various areas in the United States

Area	Animal*	Level (ppm)	Ref.
California	Pheasant	Up to 4.7	266
Minnesota	Eagle (kidney)	Up to 117	267
Wisconsin	Eagle (kidney)	Up to 7.9	267
North Dakota	Shoveler	0.17–2.26	268
	Shoveler (liver)	0.53–7.5	268
	Pintail	0.04–0.9	268
	Pintail (liver)	0.23–2.9	268
Michigan	Mallard	0.10–0.80	268
	Mallard (liver)	0.23–1.4	268
	Teal	0.10–0.18	268
St. Clair River and Lake St. Clair	Walleye pike	1.4–3.6	20
	Northern pike	0.6	20
	White bass	0.5–0.8	20
	Yellow perch	0.3–1.7	20
	Coho salmon	0.2–1.0	20
	Steelhead	<0.15	20
	Sheepshead	0.2	20
	Gizzard shad	0.2	20
	Channel catfish	0.3–1.8	20
	Carp	0.1–0.3	20
	Sucker	0.9	20
Lake Erie (western basin)	Walleye pike	0.5–2.0	5
	Yellow perch	0.39	5
	Smelt	0.16	5
Lake Erie (central basin)	Yellow perch	0.22	5
	Smelt	0.17	5
Lake Erie (eastern basin)	Yellow perch	0.17	5
	Smelt	0.11	5
Lake Champlain	Fish	Up to 1.4	1
Pickwick Lake	Fish	Up to 2.1	269
Mississquoi River (Vt.)	Fish	Up to 2.0	1
N. Fork of Holston River (Va.)	Fish	0.94–4.4	269
Mobile River (Ala.)	Fish	>0.5	270
Tombigbee River (Ala.)	Fish	>0.5	270
Lake Calcasieu	Crab	1.4	271

*Analyses performed on muscle tissue unless otherwise indicated.

Table 8. State fishing restrictions because of mercury (as of September 1970)

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

Table 9. Analysis of Canadian birds, fish, and animal products for mercury in ppm

Source	Animal*	No. of samples analyzed	Range		Average
			Min.	Max.	
Ontario	Partridge	5	0.020	0.041	0.031
	Pheasant	8	0.008	0.042	0.019
Alberta	Partridge	6	0.014	0.075	0.034
	Pheasant	18	0.004	0.460	0.046
	Pheasant (liver)	2	0.02	0.22	0.12
British Columbia	Pheasant	5	0.004	0.020	0.008
	Pheasant (liver)	3	0.005	0.015	0.011
Lake Nipissing	Pickerel	1			0.42
	Perch	1			0.20
Lake Nipissing (Callender)	Walleye pike	3	0.13	0.20	0.17
Lake Nipissing (Sturgeon River)	Walleye pike	3	0.08	0.17	0.11
Lake Simcoe	Walleye pike	3	0.08	0.38	0.18
	Whitefish	1			0.05
Lake Simcoe (Talbot River)	Walleye pike	3	0.28	0.94	0.62
Lake Superior (Thunder Bay)	Whitefish	6	0.03	0.40	0.16
Lake Ontario	Northern pike	1			0.21
	Whitefish	1			0.08
	Herring	1			0.09
	Perch	1			0.25
	Bass	1			0.07
	Sheepshead	1			0.12
	Sucker	3	0.05	0.05	0.05
Lake St. Clair	Walleye pike	6	0.22	1.54	0.65
	Northern pike	3	0.32	0.63	0.44
	Pickerel	3	0.58	0.66	0.63
Lake Erie	Perch	4	0.22	0.26	0.24
Commercial	Chicken	12	0.025	0.061	0.038
	Chicken (liver)	4	0.022	0.059	0.046
	Chicken (egg)	1			0.009
	Turkey	3	0.012	0.033	0.022
Commercial	Halibut	2	0.14	0.31	0.22
	Haddock	1			0.06
	Mackerel	1			0.17
	Cod	4	0.03	0.08	0.05
	Sole	1			0.04
	Flounder	2	0.06	0.17	0.12
	Crabmeat	1			0.16
	Scallops	2	0.009	0.012	0.010
	Oysters	1			0.068

*Analyses performed on muscle tissue unless otherwise indicated.

Table 10. Analysis of human tissues for mercury in ppm

History of subjects	Ref.	Tissue	No. of samples analyzed	Range		Average
				Min.	Max.	
Normal	272	Liver	6	0.006	0.118	0.042
		Kidney	6	0.030	0.102	0.063
		Thyroid	6	0.005	0.094	0.023
		Pituitary	4	0.040	0.133	0.079
		Olfactory lobe	4	0.006	0.087	0.036
		22 other	6	0	0.021	0.007
Exposed[1]	272	Liver	5	0.061	0.460	0.161
		Kidney	5	0.141	5.130	1.307
		Thyroid	5	0.029	0.354	0.107
		Pituitary	4	0.040	1.580	0.496
		Olfactory lobe	4	0.132	0.480	0.257
		22 other	5	0	1.180	0.074
Normal	273	Liver	92	<0.1	17.2	
		Kidney	92	<0.1	127	
Exposed[2]	273	Liver	25	1.1	25.0	
		Kidney	25	9.4	275	
Normal	99	Liver	15			3.7[3]
		Kidney	15			20.5[3]
		Spleen	15			1.2[3]
Exposed[4]	99	Liver	14			3.4[3]
		Kidney	14			29.6[3]
		Spleen	14			0.4[3]
Exposed[5]	99	Liver	16			13.8[3]
		Kidney	16			152.9[3]
		Spleen	16			3.2[3]
Normal	274	Liver	5	0.015	0.033	
Normal	275	Liver	4	0.05	0.30	
		Kidney	11	<0.05	>0.50	
		3 other	4			<0.05
Normal	98	Liver	29	<0.05	0.9	0.30
		Kidney	39	<0.05	26.3	2.75
		Brain	27	<0.05	0.6	0.10
		Heart	25	<0.05	0.7	0.15
		Spleen	22	<0.05	0.7	0.05
		Lung	23	<0.05	1.0	0.10
		Pancreas	20	<0.05	0.7	0.05
		Intestine (sm)	14	<0.05	0.4	0.05
		Intestine (lg)	13	<0.05	0.9	0.10
		Muscle	22	<0.05	1.0	0.15

[1] Amalgam workers, etc.
[2] History of exposure to mercury diuretics.
[3] Values are based on dry weight; all other values are based on fresh (wet) weight.
[4] Received 400 mg mercury or less before death.
[5] Received 400 mg mercury or more (average 4,692 mg) from 3 weeks to 40 months before death.

Table 11. Daily intake of mercury and mercury concentrations in blood and hair of man

Case	Daily intake (μgHg)	Mercury in blood (ppm)		Mercury in hair (ppm)	Ref.
		Corpuscles	Plasma		
4 normal persons (Sweden)		0.006	0.003	1.35	100
83 normal persons (Sweden)	10	0.010	0.002		276
3 normal persons (Finland)		0.051	0.010	2.30	277
51 fish eaters (Sweden)	44	0.058	0.008	7.9	278
21 fish eaters (Finland)		0.060	0.007		279
20 fish eaters (Finland)		0.196	0.029	17.3	277
8 afflicted persons (Sweden)	204	2.59	0.039	36.1	100
Minamata - diseased Japanese		2.40		500	280

Table 12. Total mercury and methylmercury contents of Swedish foods in 1966

Foods	Total mercury (ppm)	Methylmercury	
		(ppm as Hg)	% of total Hg
Meat (ox)	0.074	0.068	92
Meat (poultry)	0.023	0.017	74
Liver (pig)	0.130	0.095	73
Liver (pig)	0.096	0.075	78
Egg yolk	0.010	0.005	50
Egg yolk	0.010	0.009	90
Egg white	0.012	0.011	92
Egg white	0.025	0.024	96

Table 13. Total mercury and methylmercury content in the axial musculature of fish caught upstream and downstream from industrial outfalls

Type of factory and mercury discharged	Fish	Weight (kg)	Locality where caught	Total mercury (ppm)	Percent methylmercury
Paper and pulp mill (phenylmercuric acetate)	Perch	0.064	Upstream	0.18	100
		0.071	Upstream	0.20	91
		0.10	Upstream	0.70	93
		0.14	Upstream	0.42	100
		0.12	Downstream	1.91	100
		0.12	Downstream	2.18	99
		0.19	Downstream	3.02	86
		0.41	Downstream	2.81	91
	Pike	0.40	Upstream	0.55	98
		0.59	Downstream	3.13	95
		0.91	Downstream	3.48	92
Chlor-alkali plant (inorganic and metallic mercury)	Eelpout	0.24	Upstream	0.35	90
		0.32	Upstream	0.50	93
		0.32	Upstream	0.70	95
		0.36	Upstream	0.37	100
		0.42	Upstream	0.53	79
	Perch	0.015	Downstream	0.83	94
		0.017	Downstream	1.20	92
		0.21	Downstream	2.48	86
	Pike	0.40	Downstream	1.81	95
	Pike-perch	0.37	Downstream	2.39	94
		0.41	Downstream	2.05	95
	Whitefish	0.035	Downstream	1.40	100
		0.067	Downstream	1.06	100

Table 14. Effect of a paper mill on the mercury content in aquatic organisms

Organism	Locality	Mercury (ppm)
Fountain moss (*Fontinalis*)	15 km above mill	0.08
	Below mill	3.70
Water lily	1 km above mill	0.02
	1 km below mill	0.52
Leech (*Helobdella*)	Above mill	0.02
	Below mill	3.10
	Below mill	2.60
	Below mill	2.35
	Below mill	4.40
Isopod (*Asellus*)	15 km above mill	0.06
	1 km above mill	0.06
	20 km below mill	1.90
Caddis-fly larva (Trichoptera)	15 km above mill	0.05
	14 km above mill	0.05
	1 km below mill	17.00
	5 km below mill	5.60
	6 km below mill	10.70
Stone-fly larva (Plecoptera)	15 km above mill	0.07
	17 km below mill	2.40
Alder-fly larva (*Sialis*)	20 km above mill	0.05
	1 km above mill	0.05
	1 km below mill	5.50
	6 km below mill	4.80

Table 15. Content of mercury in organs of pike specimen in ppm

Organ	Content
Heart muscle	1.00
Axial muscle	0.85
Liver	0.78
Kidney	0.64
Intestine	0.61
Ovary	0.56
Epidermal finrays	0.39
Gill	0.30
Brain	0.29
Spleen	0.28
Scales	0.10

Table 16. Lethal concentrations of mercury compounds for various aquatic organisms

Organism	Lethal concentration (ppb)	Mercury compound
Bacteria:		
Escherichia coli	200	Mercuric chloride
Escherichia coli	200	Mercuric cyanide
Escherichia coli	300	Ethylmercuric bromide
Escherichia coli	300	Phenylmercuric chloride
Escherichia coli	300	Ethylmercuric oxalate
Phytoplankton:		
Marine mixture	60	Ethylmercury phosphate
Scenedesmus	30	Mercuric chloride
Scenedesmus	150	Mercuric cyanide
Protozoa:		
Microregma	150	Mercuric chloride
Microregma	160	Mercuric cyanide
Zooplankton:		
Daphnia pulex	5	Phenylmercuric acetate
Daphnia magna	20	Mercuric cyanide
Daphnia magna	6	Mercuric chloride
Amphipod:		
Marinogammarus marinus	100	Mercuric chloride
Isopod:		
Mesospheroma oregonensis	15	Mercuric nitrate
Flatworm:		
Polycelis nigra	270	Mercuric chloride
Polychaete:		
Mercierella enigmatica	1000	Mercuric nitrate
Mollusca:		
Bivalve larvae	27	Mercuric chloride
Australorbis glabratus	1000	Mercuric chloride
Fish:		
Stickleback	20	Mercuric nitrate
Stickleback	20	Mercuric chloride
Guppy	20	Mercuric nitrate
Guppy	20	Mercuric chloride
Shiner	800	Ethylmercury phosphate
Eel	27	Mercuric chloride
Channel catfish	580	Phenylmercuric acetate
Channel catfish	1300	Ethylmercury phosphate
Rainbow trout	2000	Pyridylmercuric acetate
Rainbow trout	9200	Mercuric chloride
Salmon	20	Phenylmercuric acetate
Salmon	50	Mercuric acetate

Table 17. Some physical properties of metallic mercury

Atomic weight	200.6
Melting point, °C	-38.9
Boiling point, °C	356.6
Density, g/cc (at 20°C)	13.546
Surface tension, dynes/cm (at 20°C)	476.5
Vapor pressure, mm Hg (at 20°C)	1.20×10^{-3}
Electrical resistivity, ohms/cm (at 20°C)	9.58×10^{-5}
Coefficient of linear thermal expansion, per °C (at 20°C)	3×10^{-5}

Table 18. Sources of mercury losses from the chlor-alkali process

Source of loss	Sweden		U.S., % of total mercury lost
	Grams mercury lost per ton chlorine produced	% of total mercury lost	
H_2 gas	2–10	4–5	1
H_2 condensate	25–75	40–45	
Atmosphere	15–25	13–28	3
Brine sludge	2–50	4–26	47
Wash water	10	5–18	24
NaOH	0.4–20	1–11	1
Sweepings and anode disposal			4
Unknown			20

Table 19. Schemes for decontamination of mercury-polluted lakes

Method	Advantages	Disadvantages	Unknowns
Dredging	Removes mercury entirely; possibility of mercury recovery	Disposal of dredged material; the process itself can perhaps release a large pulse of mercury; requires elaborate equipment	Ancillary ecological effects; cost
Covering with inactive clay or mercury-binding particulates (e.g., freshly ground quartz, feldspar, etc.)	Simplicity	May not be effective (lake turnover, erosion, reversible binding, etc.); mercury pulse	Cost; effectiveness
Covering with iron pyrite and clay overburden	Simplicity; works to put mercury in a more chemically inactive state; physically covers mercury sediments	Same as for clay and particulates; introduction of iron into lake	Ancillary ecological effects; cost
Reacting with H_2S to convert mercury to HgS	Works to put mercury in chemically inactive state	Difficulty to control action of this powerful reagent; difficult to direct the H_2S directly to the right physical spot; the resulting HgS is not physically covered	Ancillary ecological effects; cost; reaction time
Raise pH of water	Might use lime which would tend to cover mercury deposits	Might cause air pollution; could spread mercury over wider area by releasing mercury from sediments	Ancillary ecological effects; cost
Plastic coatings	Simplicity; avoids lake turnover or sediment migration problems	Permanence?	Ancillary ecological effects; cost
Amalgamation with aluminum or other active metal	Simplicity	Puts another metal in the lake	Ancillary ecological effects; cost; effectiveness
Biological mining (e.g., with clams)	Minimum effect on ecology	Could increase the speed of methylmercury conversion; might not be possible for some streams	Cost; rate of mining; effectiveness