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Metal Bioaccumulation in Fishes and Aquatic Invertebrates A Literature Review

Montana State Univ, Bozeman Fisheries Bioassay Lab

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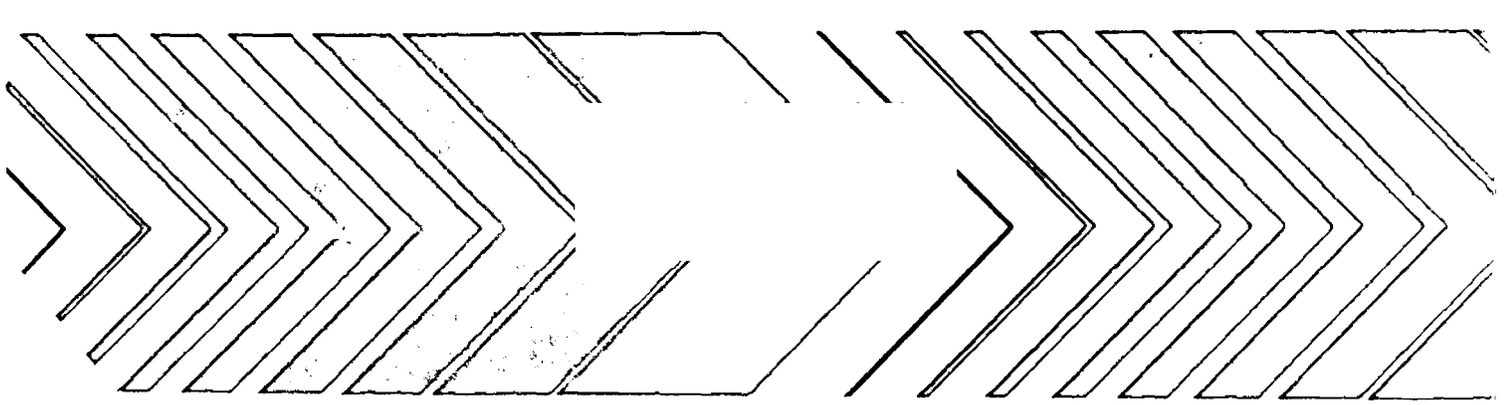
A Literature Review



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Literature concerning the bioaccumulation of metals by freshwater and marine fishes and invertebrates has been reviewed; metal residue levels are also reported for a few mammals and plants. Twenty-one metals are considered in individual sections of the review and a bibliography of over 300 literature citations is included.

The major sources of each metal to the environment are listed as are the causes and symptoms of metal toxicity in humans. Some discussion is included on the health implications of human consumption of metal-contaminated aquatic organisms. Available information is presented on: routes of accumulation, kinetics of accumulation and excretion, distribution within organisms, physiological responses of organisms, residue-toxicity thresholds, chemical speciation relative to biological availability, and microbial and chemical interconversions in aqueous systems. Major areas of insufficient knowledge are identified.

Few metals accumulate in the edible portions of aquatic organisms; moreover, most metals when ingested orally have a relatively low toxicity to humans. However, mercury, arsenic, and radioactive cesium may reach hazardous concentrations in edible tissues of fishes and shellfishes; additionally, in shellfishes, cadmium, lead, and other metal isotopes may exceed levels safe for human consumption.

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by

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FOREWORD

Residues of pollutants in aquatic organisms eaten by man have been the object of concern in recent years. Several metals, notably mercury and cadmium, have been involved in many places.

This report summarizes the literature on the bioaccumulation of heavy metals in aquatic organisms. Since environmental release of metals from energy development activities has been of much concern, this report should be useful as a summary of the current state of knowledte on metal bioaccumulation.

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Director
Environmental Research Laboratory-Duluth

ABSTRACT

Much of the available literature concerning the bioaccumulation of metals by freshwater and marine fishes and invertebrates has been reviewed; this includes literature reports of both laboratory and field investigations. Metal residue levels are also reported for a few mammals and plants. Twenty-one metals are considered in individual sections of this review and a bibliography of over 300 literature citations is included.

The major sources of each metal to the environment are listed as are the causes and symptoms of metal toxicity in humans. Some discussion is included on the health implications of human consumption of metal-contaminated aquatic organisms. If data were available for particular metals, information is presented on: routes of accumulation, kinetics of accumulation and excretion, distribution within organisms, physiological responses of organisms, residue-toxicity thresholds, chemical speciation relative to biological availability, and microbial and chemical interconversions in aqueous systems.

Few metals accumulate in the edible portions of aquatic organisms; moreover, most metals when ingested orally have a relatively low toxicity to humans. However, mercury, arsenic, and radioactive cesium may reach hazardous concentrations in edible tissues of fishes and shellfishes; additionally, in shellfishes, cadmium, lead, and other metal isotopes may exceed levels safe for human consumption.

It is concluded from this review that much remains to be learned about the bioaccumulation of metals in aquatic organisms. Major areas of insufficient knowledge include the interconversions and pathways of metals in natural environments, the relative contributions to the total tissue residue of metals ingested from food compared to metals absorbed via respiratory surfaces, the most bioaccumulative and toxic chemical species, and the relationship between metal toxicity and metal residues in tissues. More research is needed in these areas.

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SECTION I

INTRODUCTION

During recent years considerable attention has focused on the fates of metals and their derivatives in the aquatic environment. Although some metals are essential to aquatic organisms in trace amounts, others offer no known direct benefits. Mercury is an example of a non-beneficial metal which is readily accumulated by aquatic organisms. The consumption of mercury-contaminated fishes and shellfishes has resulted in several incidences of human poisonings which have elicited worldwide concern over the dangers of mercury in the aquatic environment. These incidences, as well as concern for other aspects of environmental health, have prompted researchers to explore the extent to which other metals are concentrated in living tissues, particularly in aquatic organisms. In addition, the widespread development of nuclear energy sources and the continued testing of nuclear weapons has created concern over several metals isotopes.

Metals accumulation studies which focus on the aquatic environment are important for various reasons. The extent to which metals are accumulated by aquatic animals can be related to metals toxicity. The relationship between acute toxicity and the concentration of metals in various tissues is a useful tool for diagnosing the cause of fish kills, and knowledge of relationships between chronic toxicity and metals tissue levels can aid regulatory agencies in adopting and monitoring compliance with water quality standards. Like mercury, other metals concentrated by commercially or recreationally valuable aquatic organisms pose a threat to human consumers and could thereby render these resources less valuable. The United States Food and Drug Administration (FDA) currently lists mercury, lead, cadmium, arsenic, selenium and zinc at the top of its priority list in its program concerning toxic elements in food (Jelinek and Corneliussen 1977). Of these, only mercury has an FDA-specified regulatory limit for fish and shellfish (Anon. 1974); FDA guidelines for other metals in foods have not been established. Survey and monitoring programs aimed at pinpointing metals contamination problems would help regulatory agencies in adopting the necessary restrictions, and an understanding of the processes governing the fates, pathways and distributions of metals in natural waters is necessary for assessing the current status of metals in the environment and for avoiding potential problems due to metals.

Aquatic animals can assimilate metals by ingestion of particulate material suspended in water, ingestion of food, ion exchange, and adsorption on tissue and membrane surfaces. Excretion of metals occurs through the feces, urine and respiratory membranes. In addition, animals with

exoskeletons may lose considerable amounts of accumulated metals during molting. A variety of physical, chemical, biological and seasonal variables existing in natural waters interact to influence the availability of metals to aquatic life. Moreover, certain metals or specific chemical species of a given metal are accumulated and retained by fishes to a much greater degree than others. Thus, the complexity of the factors governing metals uptake and excretion precludes making widely applicable generalizations.

The purpose of this report is to collate the metals bioaccumulation literature, particularly those studies dealing with metals bioaccumulation by aquatic animals. Because the mercury literature has already been thoroughly reviewed elsewhere, primary emphasis in the mercury section is on recent publications. However, a few of the more important earlier findings are summarized. A primary objective of this report is to evaluate the importance of metals other than mercury from the standpoint of their bioaccumulative tendencies; it should be recognized that other metals have received only a fraction of the attention that mercury has. Twenty-one metals covered individually in this review are aluminum, arsenic, beryllium, boron, cadmium, cesium, chromium, cobalt, copper, iron, lead, manganese, mercury, molybdenum, nickel, plutonium, ruthenium, selenium, silver, strontium and zinc. A few additional metals which have received only modest attention in the aquatic environment are covered in a general section. Based on current understanding, recommendations are presented as to which metals represent the greatest hazard to human consumers of aquatic life and which are the least troublesome. Accounts published after 1 July 1977 are not included in this report. Although this report is extensive, it is not exhaustive; the authors accept full responsibility for inadvertent omissions.

A few continually recurring terms should be defined at this point and qualifying statements should be made. "Concentration factor" as used herein refers to the ratio of the concentration (weight/weight) of a substance (in this case a metal) in an organism (or in a particular tissue or organ) to the concentration (weight/volume) of that substance in the water in which the organism had been living. For example, an organism (or tissue or organ) containing 10 μg Cu g^{-1} taken from a lake containing 1 μg Cu ℓ^{-1} has concentrated copper 10,000-fold; thus, by definition, the concentration factor is 10,000. Concentration factors may be derived from either laboratory or natural exposures to substances. "Concentration factors" are most useful when they apply to organisms which had reached or had nearly reached a plateau concentration of a substance under a particular exposure condition. However, concentration factors are often derived and reported for a pre-equilibrium stage in the accumulation of a substance, for purposes of comparing various treatments in a laboratory experiment or for comparing substance uptake rates among various organs or tissues from organisms in a particular treatment. Thus, the exact conditions under which these concentration factors were derived should be known before comparing concentration factors reported by different workers.

"Half-time" (or "half-life"), as used here, is biological half-time and is defined as the amount of time required for an organism to eliminate

half of the total body burden of an accumulated substance. Half-time is a useful index of the relative persistence of metal residues in organisms.

Most workers express concentrations of substances in biological tissues as the mass of substance present per mass of wet tissue; however, some workers report concentrations on a dry weight basis. All metals concentrations for biological substances reported in this review are wet tissue concentrations unless specified otherwise. In general, dry tissue concentrations are enriched about fivefold over wet tissue values because most organisms contain approximately 80 percent water.

The common and scientific nomenclature reported in the text is the same as that used by the original author(s). Both common and scientific names are included in most instances; however, a few of the most common freshwater and marine fishes are referred to by their scientific name the first time they are mentioned in a given section and by their common name for subsequent referrals in the same section. In addition, some of the less commonly known invertebrates are referred to only by scientific name. The Appendix indexes text referrals to the various organisms.

SECTION II

CONCLUSIONS AND RECOMMENDATIONS

Several trends are apparent from the information reported in this review, and research and monitoring priorities are suggested.

1. Unlike mercury, most metals are not accumulated in the edible portions of fishes and do not represent a threat to human consumers of fish unless the fish are eaten in their entirety (Table 1). Metals deserving further attention with respect to their propensity for accumulation in edible fish tissues include mercury, arsenic and radioactive cesium.

2. Shellfishes, particularly oysters, passively accumulate many metals much more readily than fishes (Table 1); this suggests a priority for monitoring in metal-contaminated areas. Potentially dangerous metals in shellfishes include cadmium, arsenic, mercury, lead, silver and the various radioisotopes.

3. Most fishes are capable of accumulating most metals both from their diet via the gastrointestinal system and from water via various membrane surfaces, particularly the gills. With some exceptions, the relative contributions of these two sources of metals to fishes are poorly understood. Considering that food may be an important route of exposure to toxic chemicals of fish, criteria derived from laboratory toxicity experiments during which fish received exposure to chemicals only through the water could be misleading. Further research is needed in this area.

4. Although the distribution of some metals in the tissues of a variety of aquatic organisms has been extensively studied, more information is needed about the actual mechanisms of toxic action, particularly mechanisms of chronic toxicity. Because some metals continue to be accumulated by fishes at the same rate even under conditions which significantly reduce toxicity, and also because some species or individuals are more or less susceptible than others to bioaccumulation of a particular metal, it follows that toxic response is internally determined and that adaptive factors are involved.

5. Sediments are an important sink for most metals in aquatic environments. Further information concerning the biological and physico-chemical factors affecting metals mobilization from and deposition in sediments would be valuable.

TABLE 1. THE RELATIVE HAZARD TO HUMANS PRESENTED BY THE OCCURRENCE OF METALS IN THE EDIBLE PORTIONS OF FISH AND SHELLFISH

Metal	Toxicity to humans from oral ingestion		Bioaccumulative tendency						Human hazard rating		
			Freshwater fish muscle		Marine fish muscle		Marine shellfish or crustaceans				
	low	high	low	high	low	high	low	high	low	med.	high
Aluminum	x			x	x			c	x		
Arsenic		x	x			x		x		x	
Beryllium	x		x			c		c	x		
Boron	x		x		x		x		x		
Cadmium		x	x		x			x		x	
Cesium[a]		x		x	x		x			x	
Chromium	x		x		x		x		x		
Cobalt[a]		x	x		x		x		x		
Copper	x		x		x			x	x		
Iron	x			x		x		x	x		
Lead		x	x		x			x		x	
Manganese	x		x		x		x		x		
Molybdenum	x		x		x			c	x		
Mercury[b]		x		x		x		x			x
Nickel	x		x		x		x		x		
Plutonium[a]		x		c	x		x		x		
Ruthenium[a]		x	x		x		x		x		
Selenium		x	x		x		x		x		
Silver	x		x		x			x		x	
Strontium[a]		x	x		x		x		x		
Zinc	x		x		x			x	x		

[a]Radioisotope is primary form of concern.

[b]Methylmercury is the species of concern.

[c]Insufficient information available.

6. Although some instances have been reported where high levels of metals in natural waters have been attributed to natural sources, the largest share of contamination is due to man. Waters receiving metals inputs resulting from man's activities should receive the highest monitoring priority.

7. The relationships between chronic toxicity thresholds and metal concentrations in tissues have been determined for a few metals with a few fish species. Studies should be undertaken to determine if these relationships are valid in natural environments; if this concept proves useful, then relationships should be established for other metals and with other aquatic species.

8. Some chemical forms of metals, such as methylmercury, are far more toxic and more readily accumulated by aquatic organisms than are others. The most bioaccumulative and toxic forms of other hazardous metals should also be determined.

SECTION III

ALUMINUM

Aluminum is the commonest metallic element in the earth's crust; it is abundant in many rocks and ores but does not occur as pure aluminum in nature. It accumulates in the lungs of humans but is almost nontoxic to man (Berry et al. 1974). Aluminum is an essential metal for human biological function, although its function in tissues has not been clearly established; aluminum levels 5 to 50 times the normal daily intake do not appear to be harmful to humans (Sorenson et al. 1974). Aluminum may enter natural waters from coal strip mining activities (Sorenson et al. 1974), as a byproduct of some oil shale mining processes (Freeman and Everhart 1971), from water treatment facilities using aluminum sulfate (alum) as a coagulant for suspended solids (NAS 1973), and in industrial wastes. Aluminum is amphoteric in water, the solubility of aluminum hydroxide complexes increasing both above and below pH 5.5 (Burrows 1977). However, the conditions existing in most natural waters cause aluminum to be precipitated or absorbed (Kopp and Kroner 1970).

To our knowledge, no laboratory experiments have been completed on the uptake and elimination of aluminum by aquatic organisms; however, several workers have reported aluminum residue concentrations in marine and freshwater fishes. Eviscerated, decapitated lake trout (*Salvelinus namaycush*) from a New York lake averaged from 140 to 300 μg Al g^{-1} depending on their age; however, aluminum content was not age-related. Calico bass (*Paralabrax clathratus*) collected near Catalina Island, California, averaged 8.0 and 25 μg Al g^{-1} of dry tissue in muscle and liver respectively (Stapleton 1968). Comparatively, calico bass collected near the outfall of a Los Angeles steam plant contained 25 μg Al g^{-1} in muscle and 28 in liver on a dry basis. Dover sole (*Microstomus pacificus*) collected near municipal outfalls along the southern California coast contained, on a dry basis, 1.8 to 8.2 μg Al g^{-1} in kidney and 0.6 to 15 in heart (McDermott et al. 1976). Goldberg (1962) determined the elemental composition of ashed tissue samples from yellowfin tuna (*Neothunnus macropterus*). The highest percentages of aluminum compared to other salts were found in heart, pyloric caeca, liver and spleen; however, the concentrations of aluminum in these tissues were not reported.

Consumption by humans of seafoods containing aluminum presents little risk due to the low toxicity of aluminum to humans. Moreover, the insolubility of aluminum under many natural conditions decreases aluminum's importance as a toxicant in water. However, both the toxicity of aluminum to aquatic life under those conditions promoting aluminum's solubility and the aluminum tissue levels resulting in adverse effects on aquatic organisms are poorly understood and warrant further investigation.

SECTION IV

ARSENIC

Arsenic is widely used as an herbicide to defoliate cotton before mechanical picking. Furthermore, the smelting of ores and the burning of fossil fuels contribute significant amounts of arsenic to the atmosphere. Arsenic is known to have a high affinity for sulfhydryl groups in proteins and thus interferes with certain enzymatic reactions (Berry et al. 1974). Acute poisonings in humans are characterized by gastrointestinal problems, irregular heartbeat, coma and possibly death. Chronic exposure symptoms include alimentary canal disturbances, coughing, and graying of the skin. Respiratory and skin diseases have been reported among smelter workers and members of an adjoining community (Birmingham et al. 1965). Some reports suggest that arsenic in drinking water may increase the incidence of skin cancer (NAS 1973). Arsenic may reach the aquatic environment through atmospheric fallout, industrial outfalls and the improper application of arsenical herbicides or pesticides (Sandhu 1977). In water, trivalent arsenic (arsenite) is far more poisonous than the pentavalent form (arsenate); however, under aerobic conditions trivalent arsenic is quickly converted to arsenate (Dabrowski 1976). In addition, arsenic may be bacterially methylated, much like mercury, to form highly toxic methylarsenic or dimethylarsenic (Anon. 1971; Braman and Foreback 1973). Fortunately, these compounds are volatile and are readily oxidized to less toxic forms (Wood 1974).

Gilderhus (1966) observed arsenic uptake by young and adult bluegill (*Lepomis macrochirus*) placed in ponds treated with various concentrations of the herbicide sodium arsenite. By the end of the test arsenic concentrations in water ranged from 0.3 to 9.0 mg As ℓ^{-1}. After sixteen weeks' exposure whole adult bluegills contained arsenic levels very similar to the concentration of arsenic remaining in the pond after that period. Muscle arsenic concentrations in mature fish were about 60 percent that of whole fish. Immature bluegills attained arsenic concentrations nearly twice those present in adults. Immature bluegills displayed reduced survival and growth rates in proportion to the level of arsenic in the pond; adult survival, however, was decreased only at the highest arsenic concentration. Tissue residues of 1.3 and 5.0 µg As g^{-1} were associated with reduced growth rate and increased mortality in immature and adult bluegills respectively. By the end of the experiment, 20 to 80 percent of the arsenic applied to the ponds remained in solution.

Dabrowski (1976) incubated rainbow trout (*Salmo gairdneri*) eggs in water containing various concentrations of either sodium arsenate or arsenic

trioxide. The same pattern of arsenic accumulation was observed for both compounds. Embryos accumulated up to 2.5 µg As g^{-1} after 40 days' exposure to only 0.05 mg ℓ^{-1} arsenic. Accumulation of arsenic became accelerated between 33 and 41 days of exposure, presumably due to increased permeability of the chorion during this stage of development. Concentrations up to 50.0 mg As ℓ^{-1} did not reduce egg survival; in fact, reduced survival was observed at arsenic concentrations less than 5.0 mg ℓ^{-1} because the higher arsenic concentrations reduced fungal growth on eggs.

Sorensen (1976a) exposed green sunfish (*Lepomis cyanellus*) to various concentrations of arsenic (as sodium arsenate) in water and measured accumulation of the metal. There appeared to be a relationship between exposure concentration and arsenic accumulated, but the data were quite scattered. In another study (Sorensen 1976b) green sunfish were exposed to the same chemical under varying temperatures and exposure intervals. Arsenic uptake by liver, gut and muscle increased with arsenic concentration in water, temperature and exposure interval. Dead sunfish did not passively accumulate arsenic, and no useful method was found for confirming arsenic-caused fish kills. Biological half-time for arsenic in gut and liver was about seven days. Because arsenic has been used to control aquatic vegetation, Wiebe et al. (1931) examined the background levels of arsenic in largemouth bass (*Micropterus salmoides*) from several Illinois rivers and then measured accumulation of arsenic by the bass after exposure to arsenic in food and water. Although arsenic was readily accumulated from both sources, elimination was rapid upon termination of exposure. The arsenic levels necessary to control aquatic vegetation did not result in arsenic concentrations in bass considered to be dangerous to human consumers of fish. Similarly, Ullmann et al. (1961) compared the arsenic content of calico bass (*Pomoxis nigromaculatus*) collected from New York lakes before and after treatment with the herbicide sodium arsenite; no differences were detected. Fish contained between 0.10 and 0.47 µg As g^{-1} in muscle fillet.

Pakkala et al. (1972) surveyed arsenic concentrations in decapitated eviscerated fishes from various New York lakes. The maximum level reported was about 0.5 µg g^{-1}, and for a given location fish age did not appear to be correlated with tissue residues. Sandhu (1977) measured arsenic content of fish and water in a pond accidentally sprayed with an arsenical herbicide. Arsenic in the pond reached 2.5 mg As ℓ^{-1}; fish accumulated up to 12.4 µg As g^{-1} in muscle representing a concentration factor of only five. Ellis et al. (1941) surveyed the arsenic content of various freshwater fish species collected from southeastern United States waters; the average arsenic content for all species was 0.75 µg As g^{-1}. Lipids contained more arsenic than other fractions; in particular, liver oil averaged nearly 40.5 mg As ℓ^{-1}. Lake Michigan plankton and benthos were found to contain 6.0 and 6.6 µg·As g^{-1} respectively (Seydel 1972); Lake Superior plankton contained about 30 percent less. The arsenic concentrations present in phytoplankton and zooplankton were similar.

The arsenic levels in some marine organisms have also been measured. LeBlanc and Jackson (1973) analyzed various marine organisms from the western coast of Canada and found that muscle tissue from assorted fishes and clams usually contained between 1 and 5 µg As g^{-1}, but dungeness crab

(*Cancer magister*) muscle averaged nearly 7 μg As g^{-1} with one individual containing 37.8 μg As g^{-1}. Wilber (1969) has reported arsenic levels in marine shellfish exceeding 100 μg g^{-1}. These values are considerably higher than those reported for freshwater organisms. Uthe and Reinke (1975) have demonstrated that post-mortem tissues from fish, shellfish and lobsters were capable of reducing arsenate to the highly toxic arsenite. Evidence suggests that the reduction is chemical. This accounts for the finding that arsenic found in tissues from these organisms collected in nature exists almost entirely as arsenite.

Arsenic is accumulated by fishes both from water and from food but reported concentration factors for arsenic in fishes are generally quite low. Arsenic is lipophilic; thus, fats contain more arsenic than other tissue fractions. Fish muscle tissue also accumulates arsenic; however, the biological half-time of arsenic is only seven days in green sunfish. Shellfishes concentrate arsenic to a much greater degree than fishes, and marine organisms contain more arsenic than freshwater forms. The U.S. Food and Drug Administration (FDA) allows 3.5 μg As g^{-1} in fruits and vegetables, and Canadians recommend a maximum level of 5.0 μg As g^{-1} for food (NAS 1973). These guideline levels have reportedly been exceeded 20-fold in shellfish and more than twofold in fish, but surveys of arsenic in fishes have shown that they usually contain substantially less arsenic than the guideline recommends. Shellfishes, therefore, warrant special attention because of their unusual metals-concentrating ability. Like mercury, arsenic can apparently be methylated by microorganisms in water, but the frequency of this occurrence and the bioaccumulative properties of methylated arsenicals have not been reported. More research is needed regarding methylated arsenicals. Arsenate present in the tissues of consumable seafood organisms is rapidly converted to arsenite following death; thus, arsenic in marketable seafoods is likely present in one of the most poisonous forms.

SECTION V

BERYLLIUM

Beryllium was at one time used to coat fluorescent lights and is currently used as a hardening agent in metal alloys, an additive in rocket fuels (McKee and Wolf 1963), a catalyst in the electroplating and organic chemical industries (NAS 1973), and a structural material for missiles and spacecraft; it is also used in nuclear reactors as a reflector or moderator and in gyroscopes and computer parts (Weast 1975). Significant amounts of beryllium also occur in the ores of other metals and in coal. Beryllium poisonings in humans (berylliosis) have resulted from industrial exposures to beryllium dusts or fumes in or near beryllium refineries; however, reports indicate that orally ingested beryllium is not toxic to humans (Berry et al. 1974). Symptoms of poisoning include pneumonitus and coughing, fatigue, and weakening of the heart (Knapp 1971). Beryllium metal is insoluble in water, as are its carbonate, oxide and sulfate; however, beryllium nitrate, phosphate and halides are all water-soluble (Weast 1975). Little work has been completed concerning the fates and pathways of beryllium in water. Tarzwell and Henderson (1960) have demonstrated that beryllium is highly toxic to warmwater fishes in soft water.

Slonim (1973) measured ^{7}Be uptake by guppies (*Poecilia reticulata*) in a static freshwater system. Levels were highest in the viscera and intestinal tract followed by kidney and ovary. Uptake was directly related to beryllium concentration in water, inversely related to fish size, and not related to fish age. Water hardness was inversely related to beryllium toxicity but did not influence beryllium uptake by the guppies. Thus, body burden of beryllium is not the controlling factor governing toxicity. The authors suggested that beryllium present in a particular target organ may be related to toxic response.

Although beryllium has a low solubility in water, it is possible that benthos could accumulate beryllium from sediment and thereby transfer the metal to higher organisms via the food chain; however, the danger of beryllium in consumable products to man is minimal because orally ingested beryllium has a low toxicity. Although beryllium toxicity decreases with increasing water hardness, beryllium uptake is unaffected by increasing hardness. Due to the paucity of information concerning the accumulation of beryllium by aquatic animals and because of beryllium's high toxicity under certain conditions, more work is needed concerning the relationship between beryllium accumulation by aquatic animals and beryllium toxicity.

SECTION VI

BORON

Boron is used in a process for bleaching groundwood by the pulp and paper industry (Thompson et al. 1976), as a hardener for other metals, and as a neutron absorber in nuclear installations (NAS 1973); it is also enriched to a considerable degree in fly ash from fossil fuels. Boron exists naturally at high concentrations in seawater as borate (NAS 1973). Boron has a relatively low toxicity to mammals as evidenced by the observations that cattle have consumed nearly 20 g of borax per day and humans 3 g boric acid per day with no adverse symptoms (McKee and Wolf 1963), but only 1 mg B ℓ^{-1} in irrigation water is toxic to some plants (Kopp and Kroner 1970).

Igelsrud et al. (1938) measured boron levels in marine algae and found that terrestrial plants growing in solutions containing boron levels similar to those existing normally in seawater accumulated higher boron concentrations than the marine algae. Calcareous structures such as the shells of marine organisms concentrated boron to a considerable degree. The authors suggested that boron in these structures was likely present as magnesium or calcium borate. The boron contents of various freshwater and neritic and oceanic marine zooplankton and phytoplankton have also been measured (Yamamoto et al. 1973). Generally, zooplankton contained higher boron levels than phytoplankton in the marine environment but phytoplanktons were higher in freshwater. Marine neritic forms contained more boron on the average than oceanic forms. Interestingly, little difference existed in the boron content of the freshwater forms compared to marine species even though the boron content of seawater averages about 460 times that of freshwater.

Thompson et al. (1976) measured boron accumulation and elimination in underyearling sockeye salmon (*Oncorhynchus nerka*) and in juvenile oysters (*Crassostrea gigas*) and found that boron was only modestly accumulated and readily eliminated. In sockeye salmon, bone contained the highest boron concentration followed by gill, liver and kidney. They also surveyed various shellfishes from British Columbia waters and found that the boron content ranged from about 1 to 6 $\mu g\ g^{-1}$.

In view of the fact that boron is a normal constituent of salinity in seawater, has a relatively low toxicity to aquatic animals and to man and is not readily bioconcentrated by aquatic organisms, boron is probably not a serious pollutant in the aquatic environment.

SECTION VII

CADMIUM

Cadmium is rare in nature, but is highly toxic. Cadmium poisonings in humans resulting from oral consumption or inhalation of the metal are well documented (Flick et al. 1971; Fassett 1975) with most exposures occurring in industry. Chronic exposures to cadmium are believed to contribute to cardiovascular diseases, hypertension and cancer. Voors and Shuman (1977) were able to correlate liver cadmium levels with the incidence of heart failure in North Carolina residents. One of the more tragic incidences of cadmium poisoning occurred among rice paddy workers in Japan; exposure resulted from the practice of using mine water to irrigate rice fields (Martin 1971). Workers developed a skeletal disorder resulting in bones so fragile that just coughing caused multiple fractures of limbs and ribs. Major sources of cadmium in natural waters include effluents from electroplating and smelting industries and runoff from agricultural areas where phosphate fertilizers are used (Clubb et al. 1975).

Several authors have measured cadmium uptake by freshwater organisms following short exposures to high levels of cadmium in water. Solbé and Flook (1975) examined cadmium concentrations in vertebrae and muscle from stone loach (*Noemacheilus barbatulus*) exposed to cadmium concentrations bracketing acutely lethal levels and found a direct relationship between concentrations of cadmium in water and in tissue. Clubb et al. (1975) measured cadmium uptake and elimination in two stoneflies, *Pteronarcella badia* and *Pteronarcys californica*, exposed to subacute levels of cadmium followed by return to cadmium-free water. Uptake was curvilinear in both species while elimination was rapid and linear. The latter result suggests that these insects may be capable of recovering from intermittently high exposures to cadmium.

The distribution of cadmium among various tissues and organs from freshwater fishes has also been examined. Rowe and Massaro (1974) looked at the change in body distribution of cadmium over 21 days in white catfish (*Ictalurus catus*) following administration of a single intragastric dose (0.2 mg Cd kg^{-1}). Concentration maximums for gastrointestinal tract organs were reached shortly after injection. Organs continuing to increase in cadmium concentration after 21 days included liver, kidney, spleen, swimbladder, blood and ovaries with kidney and liver containing the highest concentrations. The authors postulated that perhaps metallothionein was being synthesized by liver and kidney thereby facilitating the detoxification process. Marafante (1976) has confirmed that all the cadmium present in the livers and kidneys of goldfish (*Carassius auratus*) was associated

with a specific cadmium-binding protein and Noël-Lambot (1976) has demonstrated the existence of a cadmium-binding protein in the mussel *Mytilus edulis*.

Mount and Stephan (1967) were able to correlate cadmium mortalities in bluegill (*Lepomis macrochirus*) to the amount of cadmium accumulated in gill tissue. Fish killed by cadmium always contained more than 150 μg Cd g^{-1} in gill whereas survivors never exceeded 130 μg Cd g^{-1}. The authors speculated that gill autopsy may be useful for confirming cadmium-caused fish kills. It was further shown that liver accumulated high concentrations of cadmium during chronic exposure but accumulated very little following acute exposure; it was suggested that fish liver concentrations exceeding 300 μg Cd g^{-1} indicate a previous history of chronically damaging cadmium exposure.

Others have studied cadmium uptake in freshwater fishes exposed to sublethal concentrations of cadmium over an extended time period. Rehwoldt (1976) fed adult zebrafish (*Brachydanio rerio*) a diet containing 10 μg g^{-1} cadmium for six months and measured cadmium uptake in whole fish. Male zebrafish accumulated over twice as much cadmium as females. Uptake proceeded linearly for two to three months then leveled off, becoming asymptotic over the last three to four months of exposure. This dietary cadmium level resulted in a marked decrease in reproductive success; however, very little cadmium was present in newly hatched young. Pascoe and Mattey (1977) exposed three-spined stickleback (*Gasterosteus aculeatus*) to various cadmium concentrations in water (0.001-100 mg Cd ℓ^{-1}, hardness near 100 mg ℓ^{-1} as $CaCO_3$) for up to 79 days. Stickleback accumulated cadmium at all concentrations tested; however, concentration factor was inversely and linearly related to exposure concentration. Concentration factors ranged from 511 at the lowest exposure to 0.51 at the highest. All of the concentrations tested were lethal to stickleback.

Kinkade and Erdman (1975) measured ^{115}Cd uptake by various organisms in both hard (150 mg ℓ^{-1} as $CaCO_3$) and soft (0 mg ℓ^{-1} as $CaCO_3$) water. After 21 days' exposure the infusoria snail (*Ampullaria paludosa*), catfish (*Corydoras punctatus*) and guppy (*Lebistes reticulatus*) held in soft water had all accumulated higher cadmium concentrations than individuals held in hard water. Hardness influenced cadmium uptake by the guppies more than the other species; by the end of the experiment, guppies kept in soft water had accumulated over twice as much ^{115}Cd as the hard water group. On the other hand, cadmium uptake by catfish was only slightly reduced by hardness. Interestingly, both snail and catfish accumulated cadmium more rapidly in hard water during the first few days of exposure to the isotope but the soft water groups eventually caught up and surpassed those held in hard water; guppies accumulated cadmium more readily in soft water from the onset. Guppies accumulated cadmium much more readily than catfish, reaching levels almost 12-fold that of catfish by the end of the experiment. These results may partly explain the increased toxicity of cadmium to fish in soft water and the relative resistivity of catfish to cadmium toxicity.

Benoit et al. (1976) measured cadmium levels at various time intervals in several tissues from brook trout (*Salvelinus fontinalis*) exposed to

cadmium in water for up to 38 weeks. Kidney accumulated the highest concentrations of cadmium followed by liver and gill. Muscle did not accumulate significant amounts of cadmium at any of the concentrations tested, including levels at or above the lowest concentration found to be chronically damaging (3.4 μg Cd ℓ^{-1}). Most tissues appeared to reach equilibrium with respect to cadmium after 20 weeks. The authors suggested that equilibrium cadmium levels could prove to be a useful index for determining the fitness of cadmium-exposed fish populations in nature. Fish placed in fresh water after previous exposure to cadmium lost cadmium rapidly from gill tissue but did not lose cadmium from either kidney or liver. Similarly, Kumada et al. (1973) measured the uptake and retention of cadmium by rainbow trout (*Salmo gairdneri*) exposed to cadmium in water for up to 40 weeks; the fish attained maximum cadmium concentrations after 10-20 weeks' exposure with liver and kidney having the highest concentrations. Return to freshwater resulted in rapid loss of cadmium by gills, extended elimination by most organs, and almost no loss of cadmium from kidney. These cadmium clearance patterns suggest that gill is the major site of cadmium accumulation from water and kidney is the route of elimination.

Cearley and Coleman (1974) exposed largemouth bass (*Micropterus salmoides*) and bluegill to various concentrations of cadmium for periods of up to six months and measured cadmium concentrations in gill, gut content and remaining tissue after various time intervals. Gut content contained the highest cadmium levels followed by gill and remaining fish. All tissues reached equilibrium after two months, a shorter time period than that found by Benoit et al. (1976); however, the water temperatures were considerably higher (23.9 C compared to 3-15 C) during this experiment. Behavior of dying fish suggested that the nervous system was affected. Piavaux (1977) tried to determine the influence of cadmium on zinc metabolism by exposing sunfish (*Lepomis gibbosus*) to cadmium in water and measuring changes in activity of the Zn-metalloenzyme alkaline-phosphatase in various organs in addition to measuring changes in zinc and cadmium concentrations in these organs. Exposure to cadmium resulted in cadmium accumulation by all organs analyzed whereas zinc increased in some organs and decreased in others. The activity of the enzyme did not follow any systematic trend. The authors thus concluded that cadmium does not preferentially out-compete zinc for the active metabolic sites involved in this system.

In another chronic study Eaton (1974) exposed bluegill to cadmium in hard water at levels ranging from 31 to 2140 μg Cd ℓ^{-1}. After 11 months' exposure tissue residues were measured in gill, intestine and caecum, liver, and kidney. Although individuals held at all cadmium concentrations contained considerably more cadmium than controls, differences between fish held in the various treatments were not that great. The no-effect level was between 31 and 80 μg Cd ℓ^{-1}.

Cadmium uptake by marine organisms has also been extensively studied. Calabrese et al. (1975) measured the concentrations of cadmium in gill and blood from winter flounder (*Pseudopleuronectes americanus*) exposed to 5 or 10 μg Cd ℓ^{-1} in water but no cadmium was detected. Dethlefsen et al. (1975) subjected eggs and larvae of herring (*Clupea harengus*), garpike (*Belone belone*), and flounder (*Platichthys flesus*) to several cadmium concentrations

in water at various salinities for up to 14 days. Garpike accumulated considerably less cadmium for a given cadmium concentration in water than did flounder or herring. It was postulated that the garpike chorion was more efficient at retaining cadmium, thus newly hatched larvae would begin with less cadmium. Only fish exposed to 0.5 mg ℓ^{-1} cadmium and higher accumulated more cadmium than controls, and salinity did not influence accumulation. Although earlier studies had shown that embryos accumulate cadmium, larvae were shown to accumulate cadmium much more rapidly than embryos.

MacInnes et al. (1977) exposed cunner (*Tautogolabrus adspersus*) to either 0.05 or 0.10 mg Cd ℓ^{-1} in seawater for 30 or 60 days. All tissues analyzed (gill, muscle, liver) contained cadmium concentrations below the detection limits (2.0 µg Cd g^{-1}) for the methods used. However, Benoit et al. (1976), using a much more sensitive analytical method, detected significant uptake for the same tissues at concentrations even lower than those to which fish were reportedly exposed in the above study. Therefore, the analytical methodology employed probably obscured the results.

In another study Greig et al. (1974) measured cadmium uptake and elimination in cunner exposed to concentrations of cadmium in seawater ranging from 3 to 48 mg ℓ^{-1}. Liver attained the highest cadmium level, averaging 8.2 times higher than gill. The relationship between cadmium concentration in water and that in liver was linear, but the relationship appeared curvilinear for gill. Upon transfer to clean water, cunner rapidly cleared cadmium from gills, red blood cells, and serum and retained cadmium in muscle and carcass. Cadmium elimination from liver was highly variable with some fish clearing cadmium rapidly and others retaining high levels of cadmium in liver.

Several cadmium uptake experiments have been performed with mummichog (*Fundulus heteroclitus*) (Eisler 1971, 1974). Pertinent findings included: (1) Survivors accumulated a proportionally smaller percent of the cadmium in their medium with increasing concentration whereas fish that died exhibited the reverse trend. (2) Tissue concentrations exceeding 86 mg Cd kg^{-1} ash were usually lethal. (3) After 21 days of exposure, viscera contained more than 60 percent of the total cadmium, gill had 22 percent, and head and remainder had 8-10 percent. (4) Fish placed in cadmium-free water for 180 days after a 21-day cadmium exposure period lost about 90 percent of their accumulated cadmium; viscera contained most of the retained cadmium. (5) Fish exposed to various cadmium concentrations in water for periods ranging from 6 to 96 hours and then placed in cadmium-free water for 50 days accumulated cadmium levels proportional to their exposure regime and eliminated 54-76 percent of the accumulated cadmium during the post-exposure period. (6) Dead mummichog accumulated cadmium much more rapidly than living individuals; e.g., 89 times as much after 48 hours' exposure and 53 times as much after 24 hours' exposure. (7) Elimination rate of cadmium was somewhat greater for dead fish but at most only four times greater than survivors. The same author and coworkers (Eisler et al. 1972) have shown that American oysters (*Crassostrea virginica*) were able to concentrate cadmium in edible tissues from seawater to a much greater degree than either mummichog, bay scallop (*Aquipecten irradians*) or American

lobsters (*Homarus americanus*). Cadmium concentrations exceeding levels considered dangerous for human consumption (13 mg kg^{-1}) were attained by oysters exposed to cadmium concentrations in water considered safe for drinking (10 µg ℓ^{-1}). Zaroogian and Cheer (1976) reported that American eastern oysters (*C. virginica*) reached the 13 mg kg^{-1} level after exposure to only 5 µg Cd ℓ^{-1} for 40 weeks. American oysters exposed to either 0.1 or 0.2 mg Cd ℓ^{-1} in seawater experienced heavy mortalities after 13-16 weeks preceded by emaciation and discoloration (Shuster and Pringle 1969). Oysters exposed to both cadmium levels accumulated about 100 µg Cd g^{-1} after 13 weeks. Comparatively, American oysters collected from the Atlantic coast of the United States contained 0.08 to 7.78 µg Cd g^{-1} (Pringle et al. 1968).

O'Hara (1973) found that cadmium accumulation in the estuarine fiddler crab (*Uca pugilator*) was directly related to temperature and inversely related to salinity. The salinity effect was believed to result from the necessity for the crab actively to accumulate salts as salinity decreased. Similarly, Wright (1977a) found that increasing salinity decreased cadmium accumulation in the haemolymph and carapace of the shore crab (*Carcinus maenas*); however, salinity didn't influence cadmium uptake by either gill or hepatopancreas. In particular, calcium was found to be responsible for this effect (Wright 1977b). In another study using the shore crab Wright (1977c) found that haemolymphatic cadmium was almost totally bound to the protein fraction. Cadmium levels in the haemolymph tended to rise immediately preceding mortality. Cadmium uptake was believed to be passive, but active uptake was not eliminated as a possibility. Cadmium uptake by marine bivalves (*Mya arenaria, Mytilus edulis, Mulinia lateralis* and *Nucula proxima*) increased with increased temperature and decreased with increasing salinity; however, the magnitude of the response varied with species (Jackim et al. 1977). Moreover, the presence of sediment or zinc ions acted to decrease cadmium accumulation. The sediment effect was attributed in part to a reduced filtration rate observed in bivalves living in sediment. Among these four species, filter feeders accumulated more cadmium than deposit feeders.

Fowler and Benayoun (1974) described the uptake, elimination and retention of ^{109}Cd in both the mussel *Mytilus galloprovincialis* and the benthic shrimp *Lysmata seticaudata*. Test organisms were still accumulating cadmium at the end of two months' exposure to cadmium in water. After this time whole mussels had reached a concentration factor of 130 and whole shrimp 600. Both organisms attained highest cadmium levels in viscera although exposure was only through water. Interestingly, cadmium uptake and elimination rates were directly related to temperature in shrimp but unaffected by temperature in mussel. Biological half-times for test organisms held in the laboratory were 378 days for shrimp and 1254 days for mussel; however, mussels kept in cadmium-free water in the laboratory eliminated cadmium more slowly than animals kept in an estuary. This was believed to result from the laboratory animals being of a poorer nutritional state. Kerfoot and Jacobs (1976) measured cadmium uptake at various levels of a marine food chain consisting of sewage-seawater, plankton and shellfish. Components of the food chain were cultured separately in a sewage treatment system designed to utilize nutrients from the sewage to provide algae indirectly to be used as food for the shellfish population. In this system cadmium accumulated by the

algae was a more important source of cadmium to shellfish than was cadmium in water. However, the authors believed that the importance of these two sources would be reversed in a natural system.

Several surveys of the concentrations of cadmium in various marine and freshwater biota have been completed. Havre et al. (1973) measured cadmium levels in several marine fish species caught off the coast of Norway. Cadmium levels were very low for all fish tested, ranging from 0.002 to 0.033 μg Cd g^{-1}. Lovett et al. (1972) examined cadmium concentrations in eviscerated, decapitated fish from various freshwater lakes and streams in New York state. Maximum concentrations exceeded 0.1 μg Cd g^{-1} but most fish contained less than 0.02 μg Cd g^{-1}. Talbot et al. (1976) measured cadmium levels in oysters and mussels from Port Philip Bay and Corio Bay near Melbourne, Australia. Both shellfish were highly contaminated, with oysters containing more cadmium than mussels. Oysters contained from 35.5 to 174.3 μg Cd g^{-1} and mussels ranged from 2.8 to 17.0 μg Cd g^{-1}. Mussels collected from piers contained less cadmium than mussels living on sediment, implicating sediment as a contributor to uptake. Martin and Broenkow (1975) found that mixed phytoplankton and zooplankton collected off Baja, California, near San Diego averaged 13.2 μg Cd g^{-1} (dry weight basis); samples collected from other coastal areas never exceeded 7.5 μg Cd g^{-1}. Cadmium levels averaged between 184 and 1163 μg Cd g^{-1} (dry basis) in red abalone (*Haliotis rufescens*) from various portions of the California coast (Anderlini 1974). Preston (1973) discussed the sources of cadmium to marine waters off the coast of the United Kingdom. Industrial effluents were the greatest contributors to cadmium in the ocean with atmospheric fallout not considered an important source. Major contamination problems were limited to inshore waters. Shellfishes, particularly oysters and crabs, contained the highest cadmium concentrations.

Cadmium is readily accumulated through both food and water by marine and freshwater organisms, and either source of uptake can result in the development of toxic symptoms by fishes. Fish tissues appear to reach equilibrium with respect to cadmium after 8-20 weeks' exposure, depending on the water temperature. Cadmium uptake increases with increasing water temperature and decreasing salinity. Sex may determine rate of cadmium accumulation in some fish species, perhaps due to sex-related metabolic differences. Fish accumulate highest cadmium concentrations in kidney and liver, probably due to the presence of a detoxifying cadmium-specific binding protein. High concentrations of cadmium in fish liver may indicate a history of chronic cadmium exposure.

An autopsy technique has been developed for freshwater fishes utilizing the finding that a survival threshold concentration exists for gill tissue. However, this technique is probably not useful in the marine environment because dead fish placed in cadmium-spiked saltwater accumulated cadmium at a far greater rate than living individuals. This phenomenon possibly occurs because of the hypertonic nature of the saltwater medium. Chronic cadmium poisoning appears to be correlated with specific tissue cadmium levels, perhaps providing a method for detecting the occurrence of adverse cadmium conditions in natural environments.

Very little cadmium is accumulated in the edible portions of fishes. Cadmium in fishes, therefore, does not appear to represent a hazard to human consumers. However, oysters, abalone and mussels are capable of accumulating extremely high levels of cadmium in edible portions and therefore represent a greater hazard to human consumers than other marine organisms. Because of cadmium's high toxicity, edible shellfishes should be carefully monitored whenever cadmium contamination is suspected.

SECTION VIII

CESIUM

Nuclear testing in the years following World War II has resulted in the release of considerable amounts of cesium into the environment. In addition, radiocesium is a component of discharges from nuclear power plants and fuel reprocessing plants (Hewett and Jefferies 1976). Cesium has a relatively low toxicity to humans; however, radiation sickness may result from exposure to the radioisotope.

Brungs (1967) found that most of the ^{137}Cs introduced into a pond was associated with the sediment within four days following application; however, tadpoles accumulated a considerable amount of the isotope. In another pond study, Pendleton (1959) found that ^{137}Cs concentration factors for different organisms were directly related to trophic level. Cesium-137 levels in sunfish (*Lepomis gibbosus*) from the pond fluctuated widely during the 15-month experiment; lowest concentration factors in sunfish occurred during high and low temperature extremes.

Few studies have dealt with the distribution of cesium in fish, but the Nile catfish (*Clarias lazera*) concentrated ^{134}Cs to the greatest extent in muscle and bone (Ishak et al. 1977). The concentration factor (based on dry weight) at maximum uptake was only 0.37. Gustafson et al. (1966) measured the ^{137}Cs content in the edible portions from selected marine and freshwater fish species purchased from Chicago area fish markets. Freshwater fish contained much higher ^{137}Cs concentrations than marine species, and carnivores generally contained higher concentrations than planktivores. Cesium concentration factors for marine fishes were as low as 20 whereas some freshwater fish concentrated cesium nearly 10,000-fold. An indirect correlation was found between the level of potassium in water and the ability of fish to accumulate radiocesium. Other workers have reported a similar finding (Williams and Pickering 1961; Feldt 1963; Preston et al. 1967). Thus, less cesium is accumulated by fishes living in the potassium-rich marine environment.

Morgan (1964) compared ^{134}Cs uptake among various freshwater and marine organisms and determined the influence of organism size on uptake and elimination of the isotope. The rate of cesium uptake among the various organisms was compared by determining the amount of time required for the organisms to attain a concentration factor of unity. Trends that were noted included: (1) Marine shellfishes, including various crustaceans and molluscs, and marine pelagic fishes reached a concentration factor of unity in four days or less; (2) marine demersal and bottom feeding species,

freshwater mussels and marine eels required 6 to 17 days to attain unity with respect to the isotope; (3) rays and freshwater fishes required between 30 and 120 days to attain unity; (4) freshwater organisms concentrated ^{134}Cs tenfold greater than marine organisms; and (5) rate of uptake and half-time of elimination were inversely related to organism size. Depending on the organism, rates of uptake varied up to eightfold over the range of sizes tested. Half-time of elimination decreased about 3.5-fold with a 100-fold increase in organism weight.

In a saltwater laboratory study Baptist and Price (1962) looked at ^{137}Cs accumulation and elimination by summer flounder (*Paralichthys dentatus*), croaker (*Micropogon undulatus*), bluefish (*Pomatomus saltatrix*) and little tuna (*Euthynnus alleteratus*). Cesium was readily accumulated both from food and from water. Concentration factors for all species ranged from 10 to 20. Cesium elimination rate functions varied among tissues. The half-time for edible muscle tissue approached 100 days.

Häsänen and Miettinen (1963) noted that fishes collected from Finnish lakes contained ^{137}Cs in proportion to their position in the aquatic food chain, suggesting that food was an important route of uptake. Similarly, Jefferies and Hewett (1971) described the importance of various sources of cesium accumulation in plaice (*Pleuronectes platessa* L.) and thornback ray (*Raja clavata*) and determined biological half-times. Ray were estimated to derive over 80 percent of their cesium from food whereas plaice derived approximately 50 percent via this route. A significant route of cesium uptake from water for plaice was through the gut. The biological half-time for cesium in ray was calculated to be 180-190 days and in plaice 120-140 days. Excretion of cesium was primarily extrarenal. In another experiment Hewett and Jefferies (1976) exposed brown trout (*Salmo trutta*) to ^{137}Cs in freshwater and found a concentration factor near 10 for muscle and a biological half-time of about 100 days. This low concentration factor is similar to those reported for marine fish but probably is a result of the high potassium content of the test water (4.0 mg ℓ^{-1}). Earlier work (Preston et al. 1967) had shown that the half-time of ^{137}Cs in brown trout from Lake Trawsynydd, North Wales, (a potassium-deficient lake receiving nuclear power effluents) was 500 days and the concentration factor was near 40,000.

Cesium can be accumulated by fishes both from food and through water. Cesium is chemically similar to potassium; thus waters with high potassium levels such as the ocean inhibit cesium uptake, accounting for the wide variety of concentration factors reported by various workers. Organism size is inversely related to rate of cesium uptake or elimination probably due to size-related metabolic differences. Although cesium has a relatively low toxicity to humans, the isotope is very hazardous. Because a large percentage of the cesium accumulated by fishes lodges in edible muscle tissue, sport and commercial fisheries suspected to be contaminated by radiocesium should be carefully monitored.

SECTION IX

CHROMIUM

Chromium is used in electroplating, steelmaking, photography, and some chemical syntheses. Although not as toxic as many metals, chromium has been known to cause ulcers, skin lesions and cancer in humans (Berry et al. 1974). In water trivalent chromium exists as a complex, colloid, or precipitate, depending on pH; hexavalent chromium is usually present only as an ion (Knoll and Fromm 1960). Potential sources of chromium in water include industrial wastes and nuclear effluents.

Chromium content was shown to increase with age in lake trout (*Salvelinus namaycush*) collected from a New York state lake (Tong et al. 1974). Knoll and Fromm (1960) exposed rainbow trout (*Salmo gairdneri*) to 2.5 mg ℓ^{-1} hexavalent chromium in water for up to 24 days and measured chromium uptake in various tissues. Some fish were also exposed for only 12 days then placed in chromium-free water for 24 days to measure elimination. Pyloric caeca attained the highest concentrations of chromium followed by gut, kidney and liver. Except for spleen and kidney, all tissues lost chromium rapidly after exposure to freshwater conditions. Results indicate that chromium is transported by the body to the gut where it can be eliminated through the feces. Chromium administered to the gut through a stomach tube was over 50 percent eliminated after one day and none was distributed to other organs. This result implicates the gill as the major route of chromium accumulation. Edible tissues did not accumulate significant amounts of chromium.

In another study Kuhnert and Kuhnert (1976) measured chromium content of rainbow trout gill, kidney, liver and intestine following 48 hours' *in vivo* exposure to 2.5 mg Cr ℓ^{-1} as chromate. Kidney and gill accumulated about four times as much chromium as either intestine or liver. Fromm and Stokes (1962) found that rainbow trout took 10 days to reach whole body equilibrium chromium concentration upon exposure to hexavalent chromium (as chromate) levels below 0.01 mg Cr ℓ^{-1}. However, fish exposed to chromium concentrations of 0.05 mg ℓ^{-1} and higher continued to accumulate chromium linearly in time until the test was terminated after 30 days. In a laboratory study Buhler et al. (1977) analyzed two groups of rainbow trout raised in two natural waters differing in chromium content. Trout contained chromium levels in proportion to the chromium in their environment. Trout exposed to 2.5 mg ℓ^{-1} hexavalent chromium accumulated chromium rapidly during the first day of exposure but did not accumulate appreciable chromium during further exposures for up to 22 days. Apparently an equilibrium condition was rapidly reached. Tissues having the highest chromium levels were

spleen, kidney, gastrointestinal tract, gall bladder and opercular bone. In goldfish (*Carassius auratus*) ^{51}Cr was accumulated to the largest extent in air bladder, kidney and head kidney (Hibiya and Oguri 1961). The marine polychaete *Nereis virens* also accumulated chromium in proportion to exposure level, demonstrating accumulation through both gut and epithelium (Raymont and Shields 1963).

Shuster and Pringle (1969) exposed American eastern oysters (*Crassostrea virginica*) to either 0.05 or 0.1 mg Cr ℓ^{-1} for 20 weeks. At the termination of the experiment oysters exposed to the two levels averaged about 6.0 and 11.0 µg Cr g^{-1} respectively. Comparatively, east coast oysters collected from Maine through North Carolina ranged from <0.12 to 3.40 µg Cr g^{-1} (Pringle et al. 1968). Chromium concentration factors for marine organisms have been reported to approach 2000 for various planktonic forms, 500 in shellfishes and 100 in crustaceans and fishes (Lowman et al. 1971).

Chipman (1967) measured chromium (as ^{51}Cr) uptake by the marine polychaete worm, *Hermione hystrix*. Orally ingested trivalent chromium was not assimilated by the worms; however worms accumulated hexavalent chromium from water. A concentration factor of 12 was attained after 19 days' exposure, and worms were still accumulating chromium at the end of the test; uptake was proportional to exposure concentration indicating that accumulation was a passive process. Chromium elimination was from two compartments each accounting for about half of the initial burden. Biological half-times for the two elimination phases were 123 and 4 to 8 days. It was suggested that longer exposures to the metal would result in a higher percentage of the accumulated chromium being present in the slowly eliminated compartment.

Some fishes are capable of attaining chromium levels nearly 100-fold the concentrations of chromium in water; however, reports of the time required for these fish to reach equilibrium tissue levels of chromium vary from one day to over 30 days. Apparently exposure level and some unknown factors influence this relationship. What is clear is that fish rapidly eliminate chromium upon return to freshwater following exposure. Thus, fish exposed intermittently to high chromium levels would not experience cumulative chromium uptake.

In fishes chromium is apparently accumulated from water through the gills followed by transport via the blood to the various organs and tissues. Eventually chromium reaches the gut where it is eliminated through the feces. Because fishes accumulate relatively little chromium in edible tissues and because chromium is low in toxicity to humans, consumption of chromium-contaminated fish by humans should not result in toxicosis.

SECTION X

COBALT

Cobalt is widely used as an alloy, a pigment for glassware and as a binder for manufacturing tools (McKee and Wolf 1963). Cobalt is also used in nuclear generated electrical power plants and may enter water in the effluents from this industry. Cobalt dusts from refinery and alloy plants have resulted in human poisonings characterized by dermatitis, gastrointestinal pain, vomiting and low blood pressure (Berry et al. 1974). However, trace amounts of dietary cobalt are essential to human health.

Brungs (1967) released ^{60}Co along with several other radionuclides into a pond containing a typical warmwater fauna. Following introduction cobalt rapidly became associated with bottom sediments and suspended solids. Most of the cobalt accumulated by pond organisms was present in soft tissues. In a laboratory study, Nile catfish (*Clarias lazera*) attained the highest levels of ^{60}Co in muscle, bone and gill (Ishak et al. 1977) but the whole body concentration factor (dry weight basis) was only 0.36. Gill attained higher levels when ^{60}Co was present in water rather than food.

Eggs of pike (*Esox lucius*), perch (*Perca fluviatilis*), and whitefish (*Coregonus lavaretus*) were exposed to ^{60}Co for up to 120 days (Kulikov and Ozhegov 1975). Equilibrium concentration factors differed considerably (~1 to ~36) depending on fish species, life stage and incubation water temperature. Uptake of cobalt by eggs was believed to be based on sorption of cobalt on the egg surface and not on physiological and biochemical processes inside the egg. Evidence for this was the fact that pike larvae hatched from cobalt-exposed eggs were practically cobalt-free; also, the length of time required to establish an equilibrium distribution of ^{60}Co between eggs and water was the same in all cases regardless of the concentration factor. Eggs exposed to ^{60}Co for two days then placed in cobalt-free water until hatching eliminated cobalt rapidly. By the end of incubation, perch eliminated 80 percent, whitefish 57 percent and pike 40 percent; the rate of elimination was faster at higher temperatures.

Amiard-Triquet and Amiard (1975) measured the transfer of ^{60}Co through a food chain comprised of diatom (*Navicula* sp.), bivalve (*Scrobicularia plana*), shore crab (*Carcinus maenas*) and rat (*Rattus rattus*). Diatoms accumulated substantial ^{60}Co but the accumulation of ^{60}Co by other organisms was inversely related to trophic level. Rat accumulated highest ^{60}Co concentrations in liver, and crabs and bivalves had highest levels in hepatopancreas. Similarly, Mathis and Cummings (1973) noted an inverse relationship between trophic position and cobalt tissue levels among various

biota from the Illinois River; in this study fishes averaged 0.1 µg Co g^{-1}. In comparison, whole fish from the Danube River, Austria, averaged 0.3 µg Co g^{-1} dry weight (Rehwoldt et al. 1975) while great lakes fishes averaged 0.022 to 0.042 µg Co g^{-1} (Lucas et al. 1970) and eviscerated decapitated New York lake trout (*Salvelinus namaycush*) averaged 0.043 to 0.081 µg Co g^{-1}.

Cobalt concentrations have also been reported for some marine organisms. Calico bass (*Paralabrax clathratus*) collected off southern California contained 0.012 to 0.052 µg Co g^{-1} dry tissue (Stapleton 1968); clams (*Ensis* sp.), mussel (*Mytilus* sp.) and common shrimp (*Crangon crangon*) from several locations along the Belgian coast averaged (in µg Co g^{-1} dry tissue) 0.028-0.105, 0.014-0.11 and 0.18-1.04 respectively (Bertine and Goldberg 1972). Pacific oysters (*Crassostrea gigas*) contained 0.10 to 0.20 µg Co g^{-1} and American eastern oysters (*Crassostrea virginica*) ranged from 0.06 to 0.20 µg Co g^{-1} (Pringle et al. 1968); and zooplankton collected near Puerto Rico averaged 40 µg Co g^{-1} dry weight (Martin 1970).

Van Weers (1975) measured ^{60}Co uptake and retention by the common shrimp under a variety of exposure conditions. Shrimp readily accumulated the isotope from both food and water. Decreasing the water temperature from 15 to 5 C resulted in a slight decrease in cobalt uptake by the shrimp. A considerable portion of accumulated cobalt was lost during molting; the elimination process was comprised of two phases. With a 10 C increase in water temperature, the biological half-times of the slow and fast phases were reduced from 12.8 to 6.9 days and from 2.0 to 1.2 days, respectively.

Although planktonic organisms accumulate considerable cobalt, higher animals including fishes appear to accumulate very little. Upon entering water, cobalt apparently tends to associate quickly with particulate matter and sediments thereby becoming unavailable for accumulation by most organisms. Because cobalt is an essential element, its toxicity is relatively low; however, radiocobalt is more hazardous. Marine shellfishes, because of their metals concentrating ability and because they live on or in the sediment, should be monitored in areas where high radiocobalt levels are suspected.

SECTION XI

COPPER

From the standpoint of human health, copper is relatively low in toxicity compared to metals like mercury and cadmium, although prolonged consumption of large doses has been known to cause emesis and liver damage in humans (NAS 1973). The non-corrosive properties and low price of copper make it highly desirable for use in electrical wire and water pipes. In water, copper has been used as an algicide and is a common constituent of acid mine drainage. Divalent copper ion (Cu^{2+}) and its hydroxy complexes are believed to be the toxic chemical species to fishes; and alkalinity and pH are believed to be the major factors controlling copper speciation (Chakoumakos 1977).

Goettl et al. (1972) determined baseline concentrations for copper in various tissues from rainbow trout (*Salmo gairdneri*) collected from a research hatchery with a pristine water supply. On a dry weight basis values were (in µg Cu g^{-1}): opercular bone 7.3, eye 4.3, gill 4.9, intestine 9.6, kidney 12.9 and muscle 1.7. McKim and Benoit (1974) measured copper levels in gill, kidney, liver and muscle from brook trout (*Salvelinus fontinalis*) previously exposed to copper in water from the egg stage through spawning. Even the highest copper concentration employed (9.4 µg ℓ^{-1}) resulted in no detectable copper accumulation in any of the tissues analyzed; however, the copper levels used were considered quite low, since none of the exposure levels adversely affected the trout. Using considerably higher copper levels Benoit (1975) found that bluegill (*Lepomis macrochirus*) exposed for up to 22 months accumulated copper at all concentrations 40 µg ℓ^{-1} and above; this same level was the lowest concentration having an adverse effect (decreased larval survival) on bluegill. This result suggests that fishes may be adversely affected by copper if they are attaining copper tissue levels exceeding natural background levels.

Brungs et al. (1973) measured copper uptake by brown bullhead (*Ictalurus nebulosus*) exposed to various copper concentrations in water in hopes of establishing an autopsy technique useful for confirming copper-caused fish kills. No useful relationship was found; moreover, lethal exposure preceded by subacute exposure resulted in higher tissue copper levels than in fish having experienced only the lethal conditions. Bullhead accumulated copper at all water concentrations equaling or exceeding 27 µg Cu ℓ^{-1}. Copper concentrations in liver and gill tissues most accurately reflected the copper exposure conditions. Equilibrium concentrations were reached in these tissues after 30 days' exposure; however, Goettl et al. (1974) found that rainbow trout continued to accumulate copper in liver for up to 107 weeks; moreover, trout accumulated copper in liver after

exposure to only 3 µg Cu ℓ^{-1} in water. Copper elimination by stone loach (*Noemacheilus barbatulus*) was studied following short exposures to relatively high copper concentrations in water (Solbé and Cooper 1976). Gill, eye, vertebrae and muscle all lost copper rapidly, but liver tended to retain copper. In the marine mummichog (*Fundulus heteroclitus*) the presence of copper was shown to enhance cadmium accumulation (Eisler and Gardner 1973); in addition, dead mummichog accumulated copper more readily than living individuals.

Nehring (1976) suggested that it may be possible to detect instances of intermittently acute copper pollution in streams by monitoring copper levels in aquatic insects because some stream insects including the mayfly *Ephemerella grandis* and the stonefly *Pteronarcys californica* were more resistant to copper than fishes and because copper residue accumulation reflected the insects' copper exposure history. In the isopod *Asellus meridianus* copper was accumulated from both food and water, particularly in the hepatopancreas (Brown 1977). The hepatopancreas was believed to be an important storage site for copper, possibly helping avoid the accumulation of copper at more sensitive sites.

In a food chain consisting of copper-enriched sediment, bacteria and tubificid worms (*Tubifex* sp.), copper level increased with increasing trophic level (Patrick and Loutit 1976). However, Windom et al. (1973) found that for several North Atlantic fish species, copper level was inversely related to trophic position. Similarly, Cross et al. (1973) observed no increase in copper content with age among bluefish (*Pomatomus saltatrix*) and morids (*Antimora rostrata*) collected off the North Carolina coast. Marks (1938) analyzed California coastal organisms for copper including various snails, mussels and an octopus, *Polypus bimaculatus*. Copper concentrations increased with size (age) in the common terrestrial snail *Helix aspersa*, decreased with size in the sea mussel *Mytilus californianus*, and remained the same regardless of size in the octopus. These species as well as the majority of assorted other species analyzed contained from 1 to 10 µg Cu g^{-1}. Martin and Flegal (1975) measured the concentrations of copper and various other metals in livers from squid (*Loligo opalescens, Ommastrephes bartrami, Symplectoteuthis oualaniensis*) collected off southern California. Squid were able to concentrate copper in their livers to incredibly high levels with some individuals attaining 15,000 µg Cu g^{-1} on a dry basis, representing a concentration factor of 2.1 million. Increased copper levels were highly correlated with increasing levels of silver, cadmium and zinc. It was hypothesized that squid actively accumulated copper for synthesis of their major respiratory pigment, hemocyanin, and that other similar metals were accumulated in the process.

Scott and Major (1972) measured copper uptake by the mussel (*Mytilus edulis*) exposed to 0.3 mg Cu ℓ^{-1} for four days in a static seawater system. Mussels accumulated copper for the first 22 hours but then began eliminating copper with tissue returning to background levels after 48 hours. This loss of copper resulted from the mussels' excreting large quantities of mucus, thus rendering the excreted copper unavailable for reaccumulation. The loss of copper by mussels during these experiments would probably not occur in a

flow-through system where copper was continually being renewed in the solution. The marine polychaete worm (*Nereis virens*) accumulated copper in laboratory experiments through both gut and epithelium, and uptake was proportional to exposure level (Raymont and Shields 1963). Oysters (*Crassostrea virginica*) exposed to 0.025 or 0.05 mg Cu ℓ^{-1} in seawater for 20 weeks averaged about 700 or 1050 µg Cu g^{-1} of soft tissue respectively (Shuster and Pringle 1969). In comparison, the same species collected from various portions of the U.S. Atlantic Coast ranged from 6.83 to 517.4 µg Cu g^{-1} (Pringle et al. 1968). High copper tissue levels caused the oysters to take on a greenish tint. Martin et al. (1977) measured copper levels in red abalone (*Haliotis rufescens*) suspected to have been exposed to excessive copper from the discharges of a California nuclear power plant. Copper was believed to have leached from copper tubing in the condensing system during a non-operational period. Only living red abalone were analyzed, although over 1000 mortalities had been observed at an earlier date. Abalone contained on the average 65 µg Cu g^{-1} in gill with values ranging from 48 to 78 µg Cu g^{-1}. In addition, copper accumulation experiments were performed on red and black abalone (*Haliotis cracherodii*) in the laboratory. The gills of red abalone that died due to copper contained 62 to 185 µg Cu g^{-1}, whereas survivors contained 5 to 98 µg Cu g^{-1} in gill. Dead black abalone ranged from 92 to 291 µg Cu g^{-1} in gill; survivors accumulated gill copper levels of 12 to 116 µg Cu g^{-1}. For a given copper concentration in water black abalone averaged higher copper levels in gill than red abalone, but black abalone were also more resistant to copper. Copper levels in water near 56 µg Cu ℓ^{-1} resulted in both species attaining gill copper concentrations that were acutely lethal to some individuals; the experimental evidence suggests that copper could have been responsible for an abalone kill observed in the field.

Copper is accumulated by freshwater and marine fishes and shellfishes and by aquatic insects. There appears to be a good correlation between the onset of copper accumulation above background levels and the development of chronic symptoms in fishes. This relationship may prove useful for detecting conditions capable of causing chronic copper poisoning in natural waters. Although the reports of various workers vary considerably as to what is the lowest copper concentration in water resulting in copper accumulation by fish, these differences can probably be explained on the basis of how the chemical characteristics of the different test waters influenced copper speciation. Liver and gill tissues from fish most accurately reflect copper exposure conditions with liver retaining copper the longest after cessation of exposure. The copper content of stream insects may be a useful index for detecting acute copper exposures after it is too late to measure the copper content of the water. It is doubtful that even the highest concentration of copper concentrated by fish could harm human consumers because copper is low in toxicity to humans and does not tend to accumulate in the edible tissues of fishes. Oysters and squid, however, could represent a problem in areas of copper contamination due to their high enriching tendency.

SECTION XII

IRON

Iron is the second most abundant metal in the earth's crust. It enters natural waters from corrosion, steel pickling, mineral processing and acid mine drainage (NAS 1973). Although iron is an essential element and has a relatively low toxicity to humans, large doses have caused internal hemorrhaging, necrosis of the stomach, intestine and liver, and pulmonary congestion (Berry et al. 1974). Iron(II) is readily oxidized to iron(III) in most natural surface waters and a substantial fraction of this iron is present in suspended form (Stumm and Morgan 1970).

Iron concentrations in freshwater fishes from various areas have been reported. Lake trout, *Salvelinus namaycush* (eviscerated, decapitated homogenate) of known ages from Lake Cayuga, New York, averaged 0.14 to 0.34 μg Fe g^{-1} (Tong et al. 1974). On a dry basis whole bluegill (*Lepomis macrochirus*), blueback herring (*Alosa aestivalis*), brook silverside (*Labidesthes sicculus*) and chain pickerel (*Esox niger*) from a South Carolina reservoir averaged 148.7, 130.9, 149.3 and 39.3 μg Fe g^{-1} respectively (Giesy and Wiener 1977). Rehwoldt et al. (1975) found 7.6 to 42.1 μg Fe g^{-1} in whole carp (*Cyprinus carpio*) or whitefish (*Alburnus*) from the Danube River, Austria. The distribution of iron in Danube carp was 9 percent in kidney, 70 in liver, 20 in flesh and 1 in bone (Rehwoldt et al. 1976); these percentages were not adjusted for iron in gill. Mean concentrations (μg Fe g^{-1} dry tissue) for the various tissues were: gill 14,597, kidney 2.49, liver 19.39, flesh 5.54 and bone 0.277. In comparison, calico bass (*Paralabrax clathratus*), a marine fish collected near Catalina Island, California, averaged 44 μg Fe g^{-1} in dorsal muscle and 160 μg Fe g^{-1} in liver on a dry weight basis (Stapleton 1968); and American eastern oysters (*Crassostrea virginica*) collected off the United States eastern seaboard contained 30 to 238 μg Fe g^{-1}, and clams contained 50-1710 (Pringle et al. 1968). None of these workers reported age-related increases in iron content. Iron concentration factors have been determined for several marine and freshwater organisms. Values (dry basis) include: 40 for soft tissues of shrimp (*Ensis ensis*) collected off the coast of Belgium (Bertine and Goldberg 1972), 14,400 for mixed zooplankton collected near Puerto Rico (Martin 1970), 1,840 for Lake Michigan benthos (Thomas 1975) and 10,000 and 1,000 for the soft parts of marine invertebrates and vertebrates respectively (Krumholz et al. 1957).

Iron is concentrated to a considerable degree by some marine organisms, and fish accumulate high levels of iron in gill; the iron concentrations in fish do not appear to increase with age. Because of the low toxicity of iron to humans, iron in seafoods and freshwater fishes does not constitute a hazard to human consumers.

SECTION XIII

LEAD

Lead has long been known to cause poisonings in humans. Some historians have attributed the decline of the Roman Empire to the Roman practice of storing wine in lead-glazed pots. Early symptoms of lead poisoning include anemia and behavioral problems, and advanced plumbism is characterized by cramps, vomiting, kidney damage and neurological disturbances (Berry et al. 1974). Important sources of lead in the environment include automobile exhaust, smelting smoke and lead base paints. Lead mine runoff, outboard motor exhaust (Aronson 1971), highway runoff (Laxen and Harrison 1977), snowmobile exhaust (Adams 1975) and atmospheric fallout (Shukla and Leland 1973) have all been shown to contribute significantly to the lead content of some natural waters. However, most lead is probably precipitated in natural waters due to the presence of carbonates and hydroxides.

Chow et al. (1974) found that epidermis from various tuna fishes including yellowfin tuna (*Neothunnus macropterus*), skipjack (*Katsuwanus pelamis*), and albacore (*Thunnus alalunga*) contained nearly 10,000 times as much lead as muscle tissue. Most of the lead present in epidermis was associated with mucus. Dermis contained nearly 100-fold less lead than epidermis. Thus, muscle tissue being analyzed for lead can easily be contaminated by mucus. Also revealed was the fact that much canned tuna is contaminated over 1000-fold with industrial lead from the factory. Prepared tuna samples of known lead content were sent to various laboratories for independent lead analyses; analytical results confirmed that most analysts could not accurately measure the lead content of fish. Results also suggest that current analytical techniques are not sensitive enough to measure natural levels of lead in marine and freshwaters. These findings should be taken into account when interpreting the following reports.

Varanasi and Markey (1977) studied lead accumulation in rainbow trout (*Salmo gairdneri*) skin and examined the influence of calcium concentration on lead accumulation from water. Most of the lead in skin was associated with scales. After six weeks in lead-free water fish retained over 70 percent of the lead they had accumulated. Calcium markedly decreased lead accumulation on skin suggesting that lead might in turn interfere with the ability of fish to accumulate calcium for bony structures.

Merlini and Pozzi (1977a) measured lead uptake in pumpkinseed sunfish (*Lepomis gibbosus*) exposed to ^{203}Pb at pH 6.0 and 7.5. Fish at the lower pH accumulated three times as much lead as fish kept at pH 7.5. Gill,

liver and fin accumulated the most lead and muscle the least. The authors attributed the increased lead uptake at low pH to the increasing concentration of divalent lead with decreasing pH. In another experiment Merlini and Pozzi (1977b) found a direct correlation between lead accumulation by pumpkinseed sunfish and the concentration of ionic lead in water at various concentrations of total lead. Results suggest that the conditions existing in the majority of natural waters render most lead unavailable for accumulation by aquatic animals.

Holcombe et al. (1976) exposed brook trout (*Salvelinus fontinalis*) to various lead concentrations over three generations. Kidney and gill accumulated the highest concentrations of lead followed by liver; muscle accumulated very little lead. Based on the results of these experiments the Maximum Acceptable Toxicant Concentration (MATC) was concluded to be between 58 and 119 µg Pb ℓ^{-1} total lead; the incidence of spinal scoliosis was the determining factor. Trout exposed to 119 µg Pb ℓ^{-1} accumulated over 100 µg Pb g^{-1} (on a dry basis) in gill, liver and kidney after 38 weeks. The authors postulated that the equilibrium tissue levels (~50 µg Pb g^{-1} for liver and ~180 for kidney) for fish exposed to 119 µg Pb ℓ^{-1} could be used as an index to detect sublethal chronic lead damage in nature. Gill, liver and kidney lost about 75 percent of their accumulated lead after fish spent 12 weeks in a lead-free environment.

Adams (1975) exposed groups of caged brook trout to water in a pond having a history of heavy snowmobile use the previous winter. Snowmobile use on the pond was controlled and trout were held in the pond for three weeks following ice-out. Lead content of the water increased from 4.1 µg ℓ^{-1} in the falls of 1972 and 1973 to 88 and 135 µg ℓ^{-1} in the respective springs following ice-out. During these same years control fish averaged 0.37 and 0.64 µg Pb g^{-1} while caged fish contained 5.82 and 5.66 µg Pb g^{-1}. Trout kept in melted snow containing snowmobile exhaust accumulated lead in proportion to the concentration of exhaust. Of the various tissues analyzed, gut had the highest lead content.

Pagenkopf and Neuman (1974) measured the lead content of various cold-water fish species taken from a stretch of the West Gallatin River, Montana, paralleling a highway with a moderate traffic flow. Lead values in these fishes were not significantly different from the lead content of fishes from areas with no known source of lead pollution. In another survey, Pakkala et al. (1972) measured the lead content of decapitated, eviscerated fish taken from various regions of New York state. Most fish contained between 0.3 and 1.5 µg g^{-1} of lead. No trend was apparent between age and lead content in lake trout (*Salvelinus namaycush*) taken from Lake Cayuga. The lack of any apparent trend could be due to the fact that tissues known to concentrate lead were discarded from the samples.

In laboratory experiments, isopods (*Asellus meridianus*) accumulated lead from both food and water with hepatopancreas attaining the highest concentration (Brown 1977). Three different natural isopod populations were used during the experiments and it was found that individuals from the most lead-tolerant population also accumulated the most lead. Suggested mechanisms for this tolerance included improved ability to store lead

and a better lead detoxification system. Interestingly, sulfur was detected at much higher levels in the tolerant group suggesting perhaps that toxicity was avoided due to sulfhydryl binding of the metal.

Lead accumulation by marine organisms has also been studied. Gajewska et al. (1976) surveyed saltwater fishes collected from the South Baltic and North Atlantic, and freshwater fishes from Poland, for their lead content. Saltwater fishes ranged from 0.020 to 1.330 mg Pb kg^{-1} and freshwater fishes ranged from 0.020 to 2.640 mg Pb kg^{-1}. Kauranen and Järvenpää (1972) measured the biological half-time of lead and polonium in various marine organisms including *Mytilus edulis, Mesidotae entonom, Gammarus zaddachi* and *Harmothoë sarsi* in an attempt to explain the fact that the Po/Pb ratio is greater than unity in many marine organisms. All organisms except *Harmothoë* displayed an initial fast elimination phase for both metals followed by an extended period of slow elimination. Lead had a considerably longer half-time than polonium in all organisms tested, particularly the mussel *Mytilus* sp. The polychaete *Harmothoë* had the longest half-time for polonium (180 days). Because lead has a longer half-time than polonium, the high polonium-to-lead ratio in marine organisms must be due to a preferential uptake of polonium.

Stewart and Schulz-Baldes (1976) fed abalone (*Haliotis rufescens*) a diet of lead-treated brown algae (*Egregia laevigata*) for three to six months. Lead accumulation by abalone was directly related to the concentration of lead in their diet. Kidney and digestive gland accumulated the highest levels of lead but edible muscle (foot) accumulated very little lead. Tissue burdens of up to 21 µg Pb g^{-1} had no adverse effects on the abalone.

Chow et al. (1976) were able to correlate the lead content in two species of marine mussels (*Mytilus californianus* and *M. edulis*) with the degree of human activity in the area from which they were collected. The highest concentration found was 4.2 µg Pb g^{-1}, and gill tissue contained the highest lead level among organs. Exposure of American eastern oysters (*Crassostrea virginica*) in seawater to 0.1 or 0.2 mg Pb ℓ^{-1} for 10 weeks resulted in the development of an emaciated condition (Shuster and Pringle 1969). Ten-week exposures to 0.025, 0.05, 0.10 or 0.20 mg Pb ℓ^{-1} resulted in oysters averaging 35.1, 57.6, 102.9 and 276.8 µg Pb g^{-1} respectively, representing concentration factors ranging from 1030-1400. Oysters collected from the eastern coast of the United States contained <0.12 to 2.29 µg Pb g^{-1} (Pringle et al. 1968). The Canadian Food and Drug Directorate has established 2 µg g^{-1} as the maximum concentration of lead allowable in fish food (Adams 1975). This level could therefore be attained by oysters after exposure to less than 2 µg Pb ℓ^{-1}. This is not an unlikely possibility since the average lead content of major U.S. rivers has been reported to be 23 µg ℓ^{-1} (Kopp and Kroner 1970).

Lead may enter natural waters through a variety of sources. Upon entering water most lead is precipitated as carbonates or hydroxides; however, decreasing pH increases the availability of divalent lead, the principal form accumulated by aquatic animals. Many values reported for lead in fish tissue could be in error due to contamination from mucus.

Fishes accumulate very little lead in edible tissues; however, oysters and mussels are capable of attaining unacceptably high lead concentrations in edible portions after exposure to very low levels of lead in water. Based on this information, fishes are probably not a major source of lead in the human diet but shellfishes should be monitored closely. Calcium decreases lead accumulation by fishes and lead may inhibit calcium accumulation and deposition. Lead levels in fish livers exceeding 50 μg Pb g^{-1} and fish kidney above 180 μg Pb g^{-1} may indicate a history of unacceptable lead exposure.

SECTION XIV

MANGANESE

Manganese ores are common in nature but the pure element does not exist (McKee and Wolf 1963). Metallic manganese is used as an alloy in steel, and manganese salts are extensively used in inks, dyes, ceramics, matches, fireworks, batteries and paints (NAS 1973). Manganese has a low toxicity to humans but poisonings have occurred from excessive exposures to the oxides of manganese in manganese plants (Berry et al. 1974); symptoms include headache, weakness of muscles, tremors, reduced mental capacity and increased susceptibility to pneumonia and other respiratory diseases. Manganese may enter water from industrial outfalls and as a component of acid mine drainage. Although manganese chlorides, nitrates and sulfates are quite soluble in water, its carbonates, oxides and hydroxides are relatively insoluble (Kopp and Kroner 1970). Manganese has been reported to be one of the least acutely toxic metals to fishes (Doudoroff and Katz 1953).

Orally administered ^{54}Mn was accumulated almost exclusively in the bone of plaice (*Pleuronectes platessa*) (Pentreath 1973). Plaice concentrated manganese from water 1650-fold. The elimination of manganese accumulated from water was divided into two phases having biological half-times of 3.9 and 329.2 days. The slower phase accounted for 80 percent of the initial body burden. Intraperitoneally injected manganese was more readily eliminated; mean half-times for the two phases of elimination were 5.6 and 166.3 days.

Various workers have surveyed freshwater and marine fishes for their manganese content. Headless, dressed, homogenized samples of lake whitefish (*Coregonus clupeaformis*) and northern pike (*Esox lucius*) from Canadian lakes contained from 0.66 to 3.16 µg Mn g^{-1} (Uthe and Bligh 1971). Similarly prepared samples of lake trout (*Salvelinus namaycush*) from Lake Cayuga, New York, contained 0.013 to 0.052 µg Mn g^{-1} (Tong et al. 1974). No correlation existed between lake trout age and manganese tissue concentration. Abdullah et al. (1976) found that brown trout (*Salmo trutta*) and Atlantic salmon smolts (*Salmo salar*) collected from British waters contained high manganese concentrations in their scales, with most individuals containing from 30 to 100 µg Mn g^{-1} in scale. Several estuarine fishes collected near Beaufort, North Carolina, averaged 19 to 35 µg Mn g^{-1} dry weight (Cross and Brooks 1973). Bluefish (*Pomatomus saltatrix*) and morids (*Antimora rostrata*) collected off the North Carolina coast averaged about 0.2 µg Mn g^{-1} in muscle (Cross et al. 1973); and calico bass (*Paralabrax clathratus*) from the California coast averaged 0.5 µg Mn g^{-1} of dry dorsal muscle tissue. Similar to reports for freshwater fishes, manganese concentration was not related to fish size. American eastern oysters (*Crassostrea virginica*)

collected at various locations from Maine to North Carolina contained 0.14 to 15.00 μg Mn g^{-1} (Pringle et al. 1968). Marine gastropods (*Thais lapillus* and *Littorina littorea*) collected off the coast of Wales were found to contain over 80 percent of their body burden of manganese in gonad and digestive gland (Ireland and Wootton 1977), and Bryan (1971) has noted high manganese levels in the hepatopancreas of marine crustacea. These results suggest that food may be a major source of manganese to these organisms.

Patrick and Loutit (1976) have shown that tubificid worms (*Tubifex* sp.) can accumulate manganese from their diet, and Thomas (1975) has reported that lake benthos can concentrate manganese 3700-fold. Thus invertebrate food organisms may be important sources of manganese to higher organisms.

Manganese has been detected in marine and freshwater fishes and has been shown to be accumulated via the food chain by marine and freshwater invertebrates. However, manganese appears to be a relatively non-hazardous element in most waters due to the low toxicity of manganese to humans and to aquatic life and the insolubility of manganese under most natural conditions. Further investigations are, however, needed to relate manganese tissue residues in aquatic organisms to manganese toxicity under those conditions, such as acid mine drainage, which increase the solubility of manganese.

SECTION XV

MERCURY

The mercury literature has been extensively reviewed by Löfroth (1970), Ackefors (1971), Study Group on Mercury Hazards (1971), Wojtalik (1971), D'Itri (1972), Skerfving (1972), Saha (1972), Gavis and Ferguson (1972), Kojima and Fujita (1973), Löfroth (1973), Peterson et al. (1973), Neville and Berlin (1974), Jernelöv et al. (1975), Stopford and Goldwater (1975), and Doi and Ui (1975). Because of the breadth of the mercury literature and because mercury has been thoroughly reviewed by many workers since 1970, only those papers published subsequent to the most recent review will be considered in detail here. However, a few of the most important earlier findings are briefly summarized.

The worldwide concern over mercury in the aquatic environment is reflected by the fact that the Minamata Bay and Agano River, Japan, mercury poisonings resulting from human consumption of contaminated fishes and shellfishes are now almost common household knowledge. The Study Group on Mercury Hazards (1971) has described the human health aspects of methylmercury poisoning including clinical symptoms and the major sources of mercury to the environment. Victims suffer from paresthesia, ataxia, deafness, blindness and deterioration of the central nervous system followed by death. Most mercury in the environment results from the chlor-alkali industry, the pulp and paper industry, seed fungicide treatment, burning of fossil fuels, mercurial catalysts used in industry and natural weathering processes. In the United States and Canada the most severe cases of contamination to aquatic environments have been attributed to the chlor-alkali and pulp and paper industries.

Most of the mercury found in fish tissue, particularly edible portions, has been shown to be methylmercury with few exceptions (Uthe et al. 1973; Westöö 1973; Laarman et al. 1976; Hildebrand et al. 1976). Matsumura et al. (1975) have shown that liver preparations from a variety of freshwater and marine fishes are capable of converting mercuric ion to methylmercury in vitro. However, in view of the fact that exposure of fishes to mercuric ion in water results in mostly inorganic mercury in their tissue (Hannerz 1968; Cox et al. 1975) it is likely that the high levels of methylmercury found in fishes in nature result from exposure to methylmercury in their environment.

Bacteria common to most natural waters have been proven capable of converting many mercury compounds to methylmercury (Jensen and Jernelöv 1969; Wood et al. 1968; Bisogni and Lawrence 1975). Therefore, virtually any

mercury compound entering water may become a bioaccumulation hazard if the environmental conditions are favorable for methylation. Other microbial conversions of mercury have also been reported. Iverson et al. (1975) have shown that certain bacteria, primarily *Pseudomonas* sp., are capable of transforming mercuric ion and phenylmercuric acetate to volatile elemental mercury, and Spangler et al. (1973) have described a process whereby methylmercury is demethylated. Thus under certain conditions the most noxious forms of mercury can be converted to less toxic forms.

Bisogni and Lawrence (1975) described the influences of inorganic mercury concentration, availability of inorganic mercury, pH, microbial activity and redox potential on mercury methylation rates. In general, more methylmercury is produced when more inorganic mercury is present. Chemical agents which precipitate mercury, such as sulfide, reduce the availability of mercury for methylation, but only when present in large quantities. At neutral pH the primary product of mercury methylation is monomethylmercury. Methylation can occur under both aerobic and anaerobic conditions, but more mercury is produced when more bacteria are present. Hence, highly organic sediments which favor bacterial growth have a higher methylation potential than inorganic sediments. The authors suggested several methods of decontaminating mercury-laden aquatic environments including (1) addition of strong complexing agents, (2) elimination of nutrient inputs, and (3) reducing the amount of inorganic mercury available in the sediment by dredging, covering, or application of a removable mesh having a high mercury affinity. All of these methods would be extremely costly. In another study Shin and Krenkel (1976) quantified the influences of temperature, BOD, pH, chloride ion concentration and mercury ion concentration on the methylation process and subsequent methylmercury accumulation by mosquitofish (*Gambusia affinis*) or guppy (*Poecilia reticulata*). Fish accumulated more mercury as temperature and mercury content of sediment increased. A chloride ion concentration of 200 mg ℓ^{-1} and a pH near neutrality were ideal for methylation with variation in either direction resulting in reduced methylation. BOD's in the range 8-800 mg ℓ^{-1} did not influence mercury accumulation. This last result is inconsistent with earlier findings showing that conditions favoring bacterial growth enhanced methylation. Demethylation of mercury was observed to occur when methylmercury levels became excessive.

Ramamoorthy et al. (1977) measured the uptake of mercury from water by both bacteria and sediment. The bacterium *Pseudomonas fluorescens* and the sediment (Ottawa River sediment, mostly kaolinite and illite) were suspended in a mercury-spiked solution of Ottawa River water. Bacteria accumulated mercury much more rapidly than sediment, taking up nearly 20-fold as much mercury as sediment after 72 hours. Mercury loss from the system during the experiment was attributed to the bacteria converting divalent mercury to the volatile Hg°. This loss did not occur in water systems containing no bacteria.

Kudo (1976) exposed guppies to water over a bed of ^{203}Hg-enriched sediment (1.0 μg Hg g^{-1}) and measured the guppies' mercury uptake. Mercury uptake from water by the guppies was compensated for by increased mobilization of mercury from sediment into water. The half-life of mercury in the

sediment under these conditions was estimated to be 12-20 years. Over half of the mercury present in the fish was organic, suggesting that conditions were such that mercury was being methylated. In another study with inorganic mercury Tsai et al. (1975) exposed fathead minnows (*Pimephales promelas*) and emerald shiners (*Notropis atherinoides*) to various concentrations of mercuric chloride in water and measured the influence of pH on accumulation. Mercury uptake increased as pH decreased, increasing sharply at pH values below 7.0. Nearly 50 percent of all the mercury in the fish was associated with external mucus. The decreased accumulation of mercury at high pH values was believed to result from the increased formation of less reactive mercury hydroxide complexes. The presence of HPO_4^{2-} was also believed to inhibit mercury accumulation, whereas the presence of sulfide increased mercury uptake. Similarly, the presence of iodide and bromide increased mercury accumulation by factors of 18 and 6 respectively. The authors suggested that more readily accumulated mercury-halide or mercury-sulfide complexes were formed.

Communities of animals including snails, tadpoles, several species of insects and mosquitofish were exposed to mercuric ion in artificial streams (Cox et al. 1975). Mercury content of the organisms was related to habitat and trophic level with carnivores and bottom dwellers having higher mercury levels than herbivores and species living in the water column. Over 80 percent of the mercury present in the organisms including the mosquitofish was inorganic mercury.

Similarly, Kramer and Neidhart (1975) measured mercury uptake and elimination from water in guppy using inorganic mercury and methylmercury and found that methylmercury was more readily accumulated and retained than inorganic mercury and that uptake rate increased with exposure level. Half-time for methylmercury was 70 days, a much lower value than that reported by other workers. These findings support the hypothesis that inorganic mercury is not the major source of mercury to fish in most natural environments.

Ruohtula and Miettinen (1975) measured uptake and elimination of ^{203}Hg-labeled methylmercury in rainbow trout (*Salmo gairdneri*) following a single dose injected into the stomach or exposure through the water. The biological half-time ranged from about 200-500 days. Elimination time was inversely related to water temperature. Miettinen (1975) discussed the biological half-times of various mercury compounds in the mussel (*Pseudanodonta complanata*) and in rainbow trout. In the mussel half-times for inorganic, phenyl- and methylmercury were 23, 43 and 100-400 days respectively. In rainbow trout half-times for methyl-, ethyl- and propylmercury were 346, 119 and 233 days respectively. Half-times decreased with increasing tissue burden. Orally administered methylmercury was retained longer than methylmercury accumulated from water. Smith et al. (1975) compared the uptake patterns of several mercury compounds including methylmercury, inorganic mercury and phenylmercuric acetate by the freshwater clam *Anodonta grandis*. All of the mercury compounds were readily accumulated but only methylmercury was significantly retained by clams following their transfer to mercury-free water. After exposure was terminated, methylmercury redistributed within the organism so that foot increased in concentration, gill rapidly decreased, and liver remained about the same. Surprisingly, temperature did not significantly influence rate of uptake.

Laarman et al. (1976) transferred yellow perch (*Perca flavescens*) and rock bass (*Ambloplites rupestris*) from mercury-contaminated Lake St. Clair (midwestern U.S.) to essentially mercury-free earthen ponds. After two years in the mercury-free environment, all of the reduction in the mercury content of these fish could be accounted for by growth dilution.

Heisinger and Green (1975) exposed the eggs of the Japanese medaka (*Oryzias latipes*) to various mercuric ion concentrations in water. Concentrations exceeding 15 μg Hg ℓ^{-1} resulted in significantly reduced survival. The observed toxicity resulted from hemolysis of the red blood cells. Calabrese et al. (1975) examined the gill and blood uptake of mercury in winter flounder (*Pseudopleuronectes americanus*) exposed to 5 or 10 μg Hg ℓ^{-1} in water for 60 days. Mercury averaged 20.6 μg g^{-1} in gill and 2.9 in blood for the fish exposed to 5 μg ℓ^{-1} and 42.8 μg g^{-1} in gill and 3.8 in blood for the fish exposed to 10 μg Hg ℓ^{-1}.

Giblin and Massaro (1975) studied the role of blood in the accumulation and transport of methylmercury in rainbow trout; they found that hemoglobin was the major site for methylmercury binding in blood, containing almost 95 percent of the methylmercury present. The rainbow trout hemoglobin molecule was found to have four reactive -SH groups per hemoglobin molecule (compared to two for humans), accounting for methylmercury's high affinity for hemoglobin. Experiments showed that the binding of methylmercury to hemoglobin was reversible and that even methylmercury injected into the trout as methylmercury-S-cysteine eventually became bound to hemoglobin. These results suggest that methylmercury bound to red blood cells can be transferred to other tissues and organs and that red blood cells can also receive methylmercury bound to other proteins. The rate of methylmercury transport in and out of red blood cells was dependent on the concentration of -SH groups on both sides of the cell membrane.

Fromm (1977) studied several physiological aspects of mercury accumulation by rainbow trout using both $^{203}HgCl_2$ and $CH_3{}^{203}HgCl$. Gill was found to be the major site of mercury accumulation from water as opposed to gastrointestinal tract (swallowed water) or skin. Methylmercury was accumulated far more readily than inorganic mercury, but inorganic mercury bound to gill mucus over 14 times more readily than organic mercury. This difference was believed to be due to the greater lipid solubility of methylmercury thus allowing its entry across a cell membrane comprised primarily of lipid. Exposure for up to 12 weeks to 10 μg Hg ℓ^{-1} as methylmercury did not significantly alter the concentration of plasma electrolytes including Na^+, K^+, Cl^-, Ca^{2+} and Mg^{2+}; however, an unexplainable increase in hematocrit was noted. Oxygen consumption was unaffected by exposure.

Olson et al. (1975) exposed fathead minnows to concentrations of methylmercury in water ranging from 0.018 to 0.247 μg Hg ℓ^{-1} and measured whole body levels after 48 weeks' exposure. Uptake was not proportional to exposure concentration but increased with concentration. Fish exposed to the lowest methylmercury level attained concentrations nearly three times the FDA action level (0.5 μg Hg g^{-1}), and minnows receiving the highest exposure concentration exceeded the FDA level nearly 22-fold. In another chronic toxicity study McKim et al. (1976) exposed brook trout (*Salvelinus fontinalis*)

to various methylmercury concentrations in water over three generations and found that fish attained mercury concentrations in edible tissues exceeding the FDA guideline at exposure levels (as low as 0.03 μg Hg ℓ^{-1}) having no adverse effects on growth, reproduction or survival. These findings suggest that water quality standards should be based on a concentration of methylmercury in water which will provide protection for human consumers of fish.

Hartung (1976) pooled the data of previous workers to construct a model of methylmercury accumulation from water. Uptake was found to conform to zero-order kinetics during the initial uptake phase. The relationship between water temperature and methylmercury accumulation rate for a given methylmercury exposure was direct and also nearly linear. Experiments performed at different laboratories with similar test species and at similar temperatures agreed quite well.

Methylmercury is also readily accumulated by both fish and mammals through their diets. Wobeser (1975) fed groups of rainbow trout fingerlings diets containing from 4 to 24 μg Hg g^{-1} as methylmercury for 105 days. No mortality was attributed to methylmercury even though some fish accumulated as much as 30 μg Hg g^{-1}. Fish at the highest exposure level suffered from minor gill hyperplasia. Scherer et al. (1975) fed walleye (*Stizostedion vitreum vitreum*) a diet of shredded northern pike (*Esox lucius*) collected from mercury-contaminated Clay Lake, Ontario, and measured mercury accumulation in various tissues as well as the locomotor response of exposed fish. Lens accumulated extremely high concentrations of mercury (over 200 μg g^{-1}) and exposed fish suffered from increased mortality and decreases in growth, locomotor activity, coordination, and response to light. Considerably less fat was deposited in livers of mercury-fed fish as compared to controls.

Mink (*Mustela visen*) fed a diet of fish for 145 days containing mercury concentrations approaching the FDA action level accumulated mercury levels as high as 7.8 μg g^{-1} in liver, 6.5 in kidney and 8.3 in brain with no adverse effects (Wobeser et al. 1976).

Suzuki et al. (1976) examined the socio-economic factors governing the fish-eating habits and subsequent mercury accumulation by residents of several small Japanese islands. The mercury content of both hair and blood from islanders was correlated with the frequency of fish consumption and the mercury content of the fish consumed. Factors governing the availability of fish influenced the frequency of fish consumption. In another study concerning mercury in the diets of humans Smith and Armstrong (1975) analyzed the mercury levels in various dietary components of Northwest Territory natives (Inuits) as well as in Arctic canines. Both bearded seals (*Erignathus barbatus*) and ringed seals (*Phoca hispida hispida*) were found to contain exceedingly high levels of mercury in their livers. In addition, liver contained high selenium levels. Most of the mercury present in seal livers, unlike the case for fish, was inorganic suggesting a demethylation process by this organ. Arctic char (*Salvelinus alpinus*), caribou (*Rangifer tarandus*), Arctic fox (*Alopex lagopus*) and wolf (*Canis lupus*) contained only modest mercury levels. However, sledge dogs (*Canis familiaris*), which feed primarily on seal, contained extremely high mercury levels. Since the Inuits' diet consists primarily of caribou and Arctic char for most of the

year and seal for only a short time, there appears to be no immediate hazard; a problem could develop should seal become a more important dietary constituent.

Conflicting reports exist in the literature regarding the relative importance of food and water as sources of mercury to fishes. The Swedish workers Fagerström and Asell (1976) insist that mercury uptake from water via the gills accounts for most of the mercury present in fishes, but most other reports show food to be a more important source. Norstrom et al. (1976) modeled methylmercury uptake by Ottawa River yellow perch deriving coefficients for the various factors influencing methylmercury accumulation from the literature and from their own laboratory experiments. Earlier work had shown that 80 percent of the methylmercury present in food and 12 percent of that passing over the gills was accumulated by the fish. Elimination was described as a function of body weight and methylmercury body burden. If the coefficients in the model are correct, then about 40 percent of the methylmercury present in Ottawa River yellow perch was derived from the water and 60 percent from their food. Experiments conducted by Terhaar et al. (1977) demonstrated that fathead minnows accumulated more mercury when their food source (*Daphnia magna*) was raised in the test water. Suzuki and Hatanaka (1974) estimated the percentage of methylmercury in young yellowtail (*Seriora quinqueradiata*) attributable to their food, based on the relationship between food consumption rate and growth rate and the efficiency of methylmercury extraction from food. Their estimates suggested that food accounted for almost all of the mercury present in young yellowtail; however, the efficiency of methylmercury extraction from food was based on laboratory experiments during which yellowtail were fed anchovies (*Engraulis japonica*) which had been exposed previously to methylmercury-dosed seawater for a short duration. Conceivably, longer exposures to lower mercury concentrations, such as exist in nature, would result in a body distribution of mercury in the food organism that would decrease mercury's availability to a predator fish.

Many field investigations have been conducted during which various workers have collected numerous freshwater and marine organisms and analyzed them for their mercury content. In the United States Kelly et al. (1975) measured the mercury concentrations found in walleye from several Michigan lakes and compared these to values obtained from museum specimens collected from the same lakes 40 years earlier. In comparing fish from seven collection sites, walleyes from three lakes increased in mercury, but specimens from the other four sites decreased or remained the same. Anderson and Smith (1977) measured the mercury levels in fish from an Illinois lake located downwind from a coal-fired electrical power plant emitting large amounts of mercury into the atmosphere. Fish were found to contain unusually low concentrations of mercury compared to other Illinois lakes with no known source of mercury, suggesting that conditions existing in the lake were acting to suppress mercury accumulation by the fish; no hypotheses were suggested. In a similar study, Aronson et al. (1976) compared the muscle mercury levels in fish from three Ohio lakes including industrialized Lake Erie and two less industrialized lakes. Of the species analyzed, carp (*Cyprinus carpio*) contained almost twice as much mercury as other species from the non-industrialized lakes, but Lake Erie carp had the same average

mercury content as coinhabitors. This discrepancy was believed to result from a difference in the nutrient status of the two systems with Lake Erie being more oligotrophic than the other lakes.

Hildebrand et al. (1976) measured total mercury and methylmercury levels in fishes and benthic fauna collected from the Holston River, Virginia, at various distances downstream from the waste disposal ponds from an abandoned chlor-alkali plant. Mercury concentrations were highest in fish collected immediately below the pond (within two miles), but fish collected as far as 80 miles downstream exceeded the FDA guideline. Nearly 90 percent of the mercury in fish and 50 percent of that in benthic invertebrates was methylmercury.

The mercury content of largemouth bass (*Micropterus salmoides*) from three South Carolina reservoirs was correlated with trophic stage of the reservoir (Abernathy and Cumbie 1977). Newer reservoirs were oligotrophic in character and contained bass with higher mercury levels than bass from older, more eutrophic reservoirs. The lower pH, higher dissolved oxygen content, and lower alkalinity of the oligotrophic reservoir were believed to enhance microbial methylation of mercury and subsequently promote mercury accumulation by fish. Freshwater fish from various other South Carolina water bodies have also been surveyed for their mercury content (Koli et al. 1977). Some northern pike and mudfish (*Amia calva*) exceeded the FDA guideline level but most fish contained permissible amounts. Kidney, liver and muscle contained higher mercury levels than other tissues. In another southern United States study Crockett et al. (1975) measured the mercury concentrations in commercially grown channel catfish (*Ictalurus punctatus*) collected from various catfish farms in Arkansas and Mississippi. Mercury levels were very low, averaging 0.05 μg Hg g^{-1} with none of the fish analyzed exceeding the FDA guideline. Cumbie (1975) found that mercury concentrations in muscle tissue from fishes collected from the Suwanee River in Georgia exceeded the FDA guideline. Hair samples from fish-eating mammals (otter, *Lutra canadensis*, and mink) collected from the same area were found to contain up to 68 μg Hg g^{-1} on a dry basis. These high mercury levels in mammals were believed to have been accumulated via the food.

In the western United States Benson et al. (1976) looked at the mercury content of channel catfish and smallmouth bass (*Micropterus dolomieui*) from the Snake River, Idaho. Most bass three years and older and catfish seven years and older exceeded the FDA guideline, with bass attaining higher mercury levels than catfish. Similarly, Richins and Risser (1975) examined the mercury levels found in fish and crayfish from various areas of the Carson River watershed in Nevada. Intensive gold and silver mining in the drainage during the 1800's implicated this area for mercury contamination. The survey revealed that edible tissues from some fishes exceeded the FDA guideline; most notably, white bass (*Roccus chrysops*) from Lehontan Reservoir averaged 1.3 μg Hg g^{-1}. Potter et al. (1975) reported on the mercury content of various tissues from fishes and invertebrates collected from Lake Powell near Page, Arizona. Interestingly, muscle contained the highest mercury level of any tissue in most species: walleye, largemouth bass, carp, black crappie (*Pomoxis nigromaculatus*), and flannelmouth sucker (*Catostomus latipinnis*); however, liver, kidney, heart, spleen, stomach,

brain and gill all exceeded muscle mercury levels in rainbow and brown trout (*Salmo trutta*). Apparently, modes of uptake, retention and elimination vary among species. Factors which were believed to influence the observed levels of mercury in plants and animals at different trophic levels included age, surface area, metabolism, habitat and activity.

Various workers have also examined the mercury concentrations found in organisms from foreign countries. Annett et al. (1975) measured the mercury content of muscle tissues from fish and waterfowl collected from the Ball Lake area of the Wabigoon-English River system of Ontario, Canada. Ball Lake is downriver from Dryden, Ontario, a town supporting a large paper mill and chlor-alkali manufacturing facility. Both fishes and waterfowl were found to be highly contaminated. The average mercury content in fishes ranged from 0.51 to 13.54 μg Hg g^{-1} depending on species and sampling location; waterfowl breast tissue ranged from 0.62 to 8.36 μg Hg g^{-1}. All of the waterfowl and 95 percent of the fish exceeded the FDA guideline, most by several orders of magnitude; mercury levels in fish were near those reported for Minamata Bay.

Renzoni and Bacci (1976) collected freshwater mussel (*Unio cfr. elongatulus*) from a river system located downstream from a large cinnibar mine and refinery in Italy. The concentration of mercury in adductor muscle was linearly related to mussel size, with digestive gland and gill tissues containing the highest concentrations of mercury. Half-time varied from nearly 60 days in digestive gland to only 15 days in gonad.

Since the 1966 ban on alkylmercury seed dressing in Sweden the mercury content of feathers from selected raptorial and seed-eating birds has decreased. Feathers from museum-preserved birds collected prior to the use of mercurial seed dressings also contained low mercury concentrations (Westermark et al. 1975). Olsson (1976) performed a similar before-and-after study below a Swedish paper mill. Four years after mercury discharges were discontinued, northern pike contained significantly less mercury; males contained higher mercury levels than females and size was more closely correlated with mercury level than age. For fish of the same length, individuals with a low condition factor contained more mercury than those with high condition factors, indicating that mercury concentration increases in the fish during periods of starvation. Norwegian workers (Steinnes et al. 1976) found that fishes collected below the outfalls of a pulp and paper mill five years after a ban on mercury contained considerably less mercury than fishes collected prior to the ban; however, the levels were still unacceptable for human consumption. Livers from these fish contained unusually high mercury levels with some livers exceeding 200 μg Hg g^{-1}. The ratios of mercury in liver to that in muscle for these fish were much higher than ratios reported by earlier workers (Jernelöv and Lann 1971). Caines and Holden (1976) examined a case of mercury pollution in a Scottish river. Mercury entered the river from an industry prophylactically treating seed potatoes with methoxyethylmercuric chloride. Some brown trout and grayling (*Thymallus thymallus*) attained mercury levels in muscle approaching 20 and 12 μg g^{-1} respectively. However, mercury levels in fish returned nearly to background within one year after the discharge was stopped. This unusually fast recovery was

attributable to the finding that most of the mercury in these fish was present in forms other than methylmercury.

Looking at dam lakes in Bohemia Hejtmánek et al. (1975) found one lake where 40 percent of the fish analyzed contained mercury levels surpassing 0.5 μg Hg g^{-1}. Northern pike generally contained higher mercury levels than other species from a given lake. In a Bavarian study Knöppler and Dorn (1976) surveyed the mercury content of fishes from various waters including fish culture ponds and the rivers Danube, Naab and Altmühl. Pond fishes had very low mercury levels (0.01 to 0.22 μg g^{-1}) and most fish from the Naab and Altmühl contained less than 0.5 μg g^{-1}. Danube River fishes were much higher in mercury with some fish containing nearly 2.5 μg Hg g^{-1}. However, Gergely et al. (1977) found that fish from the Hungarian portion of the Danube averaged only 0.59 μg Hg g^{-1} and fish from various other Hungarian lakes averaged less than 0.5.

Jeyachandran and Raj (1975) analyzed several fish from a Tamil Nadu, India, reservoir for mercury and found none exceeding 0.50 μg Hg g^{-1}. In Japan Matsunaga (1975) demonstrated that crucian carp (*Carassius carassius*) concentrated mercury nearly 25,000 times the concentration present in river water. Yamanaka and Ueda (1975) reported the unusual finding that high levels of ethylmercury were present in fishes and sediments in the Jinzu River, Japan, below the outfall of a pharmaceutical company synthesizing an antiseptic containing an ethylmercury derivative. Upon news of this occurrence, the Japanese government dredged the river to eliminate the contamination. Fish were monitored for mercury content following dredging and it was found that four years were required for fish to return to normal mercury concentrations. The half-time of ethylmercury in these fishes was greater than one year.

The distribution and concentrations of mercury in marine organisms is important because marine fishes and shellfishes are frequent human dietary constituents. Arima and Umemoto (1976) found an uneven distribution of mercury in muscle tissue from bigeye tuna (*Thunnus obesus*), bluefin tuna (*T. thynnus*) and swordfish (*Xiphias gladius*). This finding was attributed to the fact that mercury has a higher affinity for myofibrillar protein and sarcoplasmic protein than for non-protein nitrogenous compounds or insoluble muscle residue. Mercury had the highest affinity for myofibrin.

German workers (Krüger et al. 1975) collected fishes from north Atlantic waters utilized by the German commercial fleet and analyzed edible portions for mercury. Fish from most areas including Iceland, Newfoundland, and the Faeroes did not exceed the 1.0 μg Hg g^{-1} permitted by the Federal Republic of Germany. However, fish from the Elbe estuary contained extremely high mercury levels. In another survey of German fishing waters, older ling (*Molva molva*) and red-fish (*Sebastes marinus*) were found to exceed 1 μg Hg g^{-1} (Jacobs 1977). Bluefin tuna averaged 0.40 μg Hg g^{-1}, ray (*Hypotremata* sp.) averaged 0.77 and various sharks averaged 1.83. Between 70 and 98 percent of the mercury was present as methylmercury. The mercury levels in market fishes from the port of Genoa, Italy, were surveyed by Cugurra and Maura (1976). Most fish species contained mercury levels below the FDA guideline level but tuna (*Thunnus thynnus*, *Oblata melamura* and *Umbrina*

cirrhosa) exceeded the guideline. New Zealand snapper (*Chrysophrys auratus*) were analyzed for mercury due to their commercial importance in the area (Robertson et al. 1975). Although fish from some bays averaged up to 0.72 μg Hg g^{-1} and extremely large individuals (50+ cm) averaged 1.00 μg Hg g^{-1}, the overall average marketable fish contained only 0.25 μg Hg g^{-1}.

Nuorteva and Häsänen (1975) looked at the relationship between mercury accumulation and size in fourhorn sculpin (*Myoxocephalus quadricornis* L.) collected from two areas of the Baltic and compared the mercury levels found in this species to the mercury concentrations present in other species from the same area. The relationship between mercury content and weight was linear for fish from both areas; however, sculpins contained more mercury than another bottom fish, the flounder (*Platichthys flesus*). The higher mercury content of sculpins was attributed to their having a more mercury-rich diet than flounder.

Reimold and Shealy (1976) analyzed young-of-the-year finfish from bays along the Georgia and South Carolina coasts for mercury during various times of the year. Most individuals contained mercury concentrations well below the FDA guideline. However, the spring 1973 samples of silver perch (*Bairdiella chrysura*) from the South Santee River and Port Royal Sound, and Atlantic croaker (*Micropogon undulatus*) from Winyah Bay contained mercury levels in excess of 0.5 μg Hg g^{-1}. The same species contained much lower mercury levels the preceding and following falls; no explanation was advanced. Similarly, Greig et al. (1977) measured mercury concentrations in organs and muscles from three fish species collected off the northeast coast of the United States. Cusk (*Brosme brosme*) had higher concentrations of mercury in muscle and liver than in gill or kidney but a blackbellied redfish (*Helicolenus dactylopterus*) had similar mercury levels in all tissues. Muscle tissue from spiny dogfish (*Squalus acanthias*) had higher mercury levels than organs, averaging 0.35 μg Hg g^{-1}.

In the same species collected off the Oregon coast Childs and Gaffke (1973) found an average level of 0.60 μg Hg g^{-1}. Hall et al. (1977) measured mercury levels in spiny dogfish collected from inland marine waters of the state of Washington. The mean mercury content of fish from all sampling stations exceeded 0.9 μg Hg g^{-1}; mercury tissue level was directly related to fish weight, and males contained more mercury than females for a given weight. Because females of this species are known to grow faster than males, this latter finding was attributed to growth dilution. This evidence suggests that more mercury is present in water off the Oregon-Washington coast than in the North Atlantic and that inland waters are more severely contaminated. Hall et al. (1976a) collected halibut (*Hippoglossus stenolepis*) from various locations along the Pacific coast of North America and measured the mercury content of muscle tissue. Mercury was evenly distributed in the entire edible portion. The average concentrations for fish of similar size steadily increased moving south from the Bering Sea down to the Oregon-Washington coast. The authors speculated that this trend may reflect the degrees of mercury contamination for the various latitudes of the Pacific Ocean. The same north to south trend was noted in a similar study of Pacific sablefish, *Anoplopoma fimbria* (Hall et al. 1976b).

Eganhouse and Young (1976) measured the mercury content of mussel (*Mytilus californianus*) from various locations along the California coast between San Diego and just north of Santa Barbara. Specimens collected near harbors and municipal and industrial outfalls generally contained more mercury. Flegal (1977) reported a similar finding for seston from San Francisco Bay; phytoplankton and organic detritus contained more mercury than zooplankton. The same trend was noted for most fish and shellfish collected from the Georges River--Botany Bay estuary, an area known to receive large amounts of industrial and domestic effluents from Sydney, Australia (Williams et al. 1976). Sidney rock oysters (*Crassostrea commercialis*) from Botany Bay were significantly higher in mercury than oysters from areas receiving less pollution; but other organisms analyzed, including blacklip abalone (*Haliotis ruber*), blackfish (*Girella tricuspidata*) and bream (*Acanthopagrus* sp.) had about the same mercury content regardless of collection location.

Shultz et al. (1976) reported on the mercury content of blue marlin (*Makaira nigricans*) caught off the coast of Hawaii. Unlike most fish, marlin contained more inorganic mercury than methylmercury. Muscle averaged 2.42 μg Hg g^{-1} of which only 0.36 μg g^{-1} was organic. The relationship was particularly striking in liver where the ratio of inorganic mercury to methylmercury was 35 to 1. Perhaps marlin are unusually efficient at demethylating mercury. The authors suggested that the frequent volcanic activity in the islands area may contribute mercury to surrounding waters. Hawaiian inshore organisms were found to contain relatively low mercury levels (Klemmer et al. 1976); all individuals including a variety of sessile and mobile invertebrates and benthic and pelagic fishes averaged less than 0.33 μg Hg g^{-1} and most species averaged less than 0.15.

In summary, methylmercury is the form of mercury present in most fish tissue and is the most readily accumulated and retained form of mercury in biological systems. Most mercury occurring in water, particularly problem amounts, can be traced to man-caused sources; however, fish mercury levels exceeding the FDA guideline have in some instances been attributed to natural sources. Upon entering water, virtually any mercurial compound may be microbially converted to methylmercury. Conditions reported to enhance the methylation process include large amounts of available mercury, large numbers of bacteria, absence of strong complexing agents such as sulfide, neutral pH, high temperature, and a moderately aerobic environment. Demethylation processes also occur but apparently only when methylmercury levels become excessive. Bacteria not only act as methylators of mercury but also preferentially accumulate large amounts of mercury; however, sediment and water are probably the two most important mercury sinks. Conditions reducing the mercury content of overlying waters, such as the accumulation of mercury by aquatic organisms, result in the mobilization of mercury from sediment.

Methylmercury is readily accumulated by fishes both from their food and through the water. Although conflicting evidence exists as to the relative importance of these two sources of mercury to fishes, most reports suggest that both sources can be significant. Upon entering a fish, methylmercury is very difficult to eliminate; most studies imply that the

biological half-time of methylmercury in fishes is between one and three years. Hemoglobin has been shown to be the major transporting agent for methylmercury in the body, not only transporting methylmercury to the various tissues and organs but also receiving methylmercury from proteins during elimination. In muscle protein mercury is not uniformly distributed because methylmercury has a higher affinity for myofibrin and sarcoplasmic protein than for other muscle fractions.

Fishes are able to tolerate very high tissue burdens of mercury. Fathead minnows exposed to 0.078 μg Hg ℓ^{-1} as methylmercury and brook trout exposed to 0.03 μg Hg ℓ^{-1} as methylmercury attained mercury levels in edible portions exceeding the FDA action level (0.5 μg Hg g^{-1}) without suffering adverse effects. In fact, rainbow trout have accumulated up to 30 μg Hg g^{-1} without noticeable effects. Thus, regulations regarding mercury in water must primarily be concerned with protecting human consumers of fish. Differences in the mercury content reported between and among species of fish from a given environment are reportedly due to variations in age, surface area, metabolic rate, habitat, and activity of the fish.

Methods have been suggested for decontaminating mercury-laden environments, but all are time-consuming and costly. Although in most instances economic factors preclude effective removal of mercury from most lakes and streams, several studies have shown marked improvements in aquatic environments once mercury discharges were ceased.

SECTION XVI

MOLYBDENUM

Molybdenum is used as a lubricant, an alloy in steel and as a catalyst in petroleum processing. In addition, molybdic acid is used by the ceramic industry (NAS 1973). Although molybdenum is a trace nutrient for plants, livestock consuming plants from areas containing high levels of molybdenum in soil have developed toxic symptoms (Chappell 1975). Toxicity results from a copper deficiency resulting from molybdenum's replacing copper. Molybdenum may enter the aquatic environment through leaching processes near molybdenum mines, burning of fossil fuels, or natural weathering processes. Molybdenum is known to be an important micronutrient for algae; however, very little information is available on the accumulation of molybdenum by fishes.

Ward (1973) measured the molybdenum content of tissues from rainbow trout (*Salmo gairdneri*) and kokanee salmon (*Oncorhynchus nerka*) collected from waters containing 0, 6, and 300 µg Mo ℓ^{-1}. Although some tissues such as bone, kidney, and brain increased in molybdenum content as the molybdenum content of the water increased, other tissues including skin and muscle contained similar molybdenum concentrations regardless of the molybdenum concentration in water. Rainbow trout contained consistently more molybdenum than kokanee salmon collected from the same water; however, the relative ages of these two species were not known. In lake trout (*Salvelinus namaycush*) from Lake Cayuga, New York, molybdenum content actually decreased with fish age (Tong et al. 1974). Goettl and Davies (1977) exposed rainbow trout to various molybdenum concentrations in water for periods of up to 492 days and measured molybdenum accumulations by liver. Trout exposed to the highest molybdenum level (18.7 mg Mo ℓ^{-1}) accumulated significantly more molybdenum than controls, but lower exposures resulted in insignificant molybdenum uptake.

Molybdenum does not tend to accumulate in the edible portions of fishes and has a relatively low toxicity to humans. In addition, trace amounts of molybdenum are important for the growth of phytoplankton. Molybdenum in aquatic environments is, therefore, of little danger to humans. Because molybdenum replaces copper, it might be instructive to explore molybdenum's influence on the toxicity of copper to fishes.

SECTION XVII

NICKEL

Nickel is a common component of some metal plating industry wastes, is a constituent of metal alloys (Pickering 1974), and is present in the emissions from coal combustion. Some organic nickel derivatives, particularly nickel carbonyl, are highly toxic to humans. However, orally ingested nickel has a very low toxicity to man. Although metallic nickel is insoluble in water, some nickel salts are quite soluble (Kopp and Kroner 1970). Because of nickel's low toxicity to humans, almost no information is available on the accumulation of nickel by aquatic animals.

Friedrich and Filice (1976) studied the accumulation of nickel by mussel (*Mytilus edulis*) kept in artificially prepared seawater under static conditions. No significant accumulation was noted after four weeks' exposure to 0.03 mg Ni ℓ^{-1}, but significant uptake was noted at all concentrations exceeding 0.056 mg Ni ℓ^{-1}; rates of nickel elimination were not measured. In a study of the accumulation of iron, zinc, lead, copper and nickel by algae collected near a zinc smelting plant it was found that nickel exhibited the lowest concentration factor for all the metals tested (Trollope and Evans 1976). Panel on Nickel (1975) have summarized available information on the nickel concentrations found in various marine and freshwater fishes or shellfishes. Most foods including clams, scallops, shrimp, lobsters, crabs, marine fishes and freshwater fishes contained nickel levels below 0.75 μg Ni g^{-1}; however, fresh oysters and Pacific salmon contained higher nickel levels, averaging 1.50 and 1.70 μg Ni g^{-1} respectively. Pringle et al. (1968) found from 0.12 to 1.74 μg Ni g^{-1} in oysters (*Crassostrea virginica*) collected along the eastern coast of the United States. Wright (1976) observed nickel concentrations exceeding 7.0 μg Ni g^{-1} in muscle from marine fishes collected from the northeast coast of England, and Romeril and Davis (1976) reported that European eels (*Anguilla anguilla*) maintained in Trent River water averaged (dry basis) 21 μg Ni g^{-1} in muscle and 16 in liver.

Apparently, elemental nickel is not a human health concern in the aquatic environment because nickel is not accumulated in significant amounts by aquatic animals, and since orally ingested nickel has a very low toxicity to humans. Concern over nickel in water should focus on its effects on aquatic life.

SECTION XVIII

PLUTONIUM

Nuclear testing programs carried out among the major world powers have resulted in the release of a considerable amount of radioplutonium to the environment (Schell and Watters 1975). Plutonium from this source enters water primarily through atmospheric fallout. In addition, plutonium may enter water in the wastes from nuclear powered electrical facilities and from nuclear fuel reprocessing plants. Over the pH range of most natural waters, plutonium is present either in the trivalent or hexavalent form.

Studying plutonium in organisms collected off the coast of France in an area receiving effluents from a nuclear fuel reprocessing plant, Guary et al. (1976) found that crab (*Cancer pagurus*) accumulated highest plutonium levels in gill and exoskeleton, whereas plaice (*Pleuronectes platessa*) attained highest concentrations in gut. This result suggests that food is an important source of plutonium to plaice. However, in a more comprehensive study of marine littoral organisms from the same area, Guary and Fraizier (1977a) observed a decrease in plutonium concentration factor with increasing trophic level. Plutonium was preferentially accumulated by animals with calcareous exostructures such as mussels and lobsters; calcareous plants also attained high plutonium levels. Although food was definitely a source of plutonium to animals at higher trophic levels, the increment of plutonium provided by food was not great enough to cause a positive correlation between trophic level and tissue plutonium accumulation. Miettinen et al. (1975) noted a similar lack of correlation between trophic level and plutonium content in marine organisms collected from the Baltic Sea near Finland.

A spatial study of plutonium in various molluscs collected within the plume of a nuclear fuel reprocessing plant revealed that only animals within 50 km or less of the reprocessing plant contained plutonium concentrations greater than could be attributable to fallout (Guary and Fraizier 1977b). Unlike molluscs from most areas, molluscs collected near the reprocessing plant contained higher plutonium concentrations in soft tissues than in shell. This observation was attributed to possible differences in the isotopic composition of plutonium near the plant.

Noshkin (1972) reviewed various surveys of the plutonium levels found in marine organisms and pointed out that bottom feeding organisms always accumulated more plutonium than other organisms from the same environment. Edible muscle from marine fishes and shellfishes accumulated very little

plutonium, usually having a concentration factor of less than ten; but gut and bony structures accumulated much higher levels. Similarly, Schell and Watters (1975) reported low plutonium concentrations in fishes collected near former nuclear testing sites. Ward (1966) reported that lobster (*Homarus vulgaris*) exposed to ^{239}Pu in seawater for over 200 days also preferentially accumulated plutonium in hard parts. Almost 90 percent of the ^{239}Pu was in the shell; flesh contained only 1.2 percent of the ^{239}Pu in the body. The concentration factor for flesh was only three (dry basis). Fowler et al. (1975) also noted plutonium's high affinity for bony structures, namely the shells of common Mediterranean mussel (*Mytilus galloprovincialis*) and the exoskeletons of benthic shrimp (*Lysmata seticaudata*). The biological half-time of plutonium in the mussels was near two years but in shrimp the half-time was only 1.5 months due to the loss of plutonium during molting.

Chelation of metals with certain ligands is known to increase the solubility of some metals in water but little is known about the biological availability of these ligand-metal complexes. Eyman and Trabalka (1977) examined the intragastric availability of two plutonium complexes (Pu-fulvate and Pu-citrate) to channel catfish (*Ictalurus punctatus*) and compared the retention of these complexes to that of plutonium hydroxide. Each of several catfish was exposed via a single injection to one of the isotopically labeled plutonium compounds. Pu-citrate was found to be retained much more readily than Pu-hydroxide presumably due to its net negative charge but Pu-fulvate was only modestly retained. This latter finding was believed to result from the stability of this complex in the digestive tract and its high molecular weight.

Plutonium is accumulated by aquatic organisms from both food and water, and calcareous structures such as bone or shell attain the highest levels. Chelation of the metal influences its biological availability but the extent and direction of this influence depends on the particular ligand-metal complex. Edible muscle tissue from fish accumulates very little of the isotope, but some reports indicate that the form of isotopic plutonium present in nuclear fuel reprocessing plant effluents is readily accumulated in the soft tissues of some marine molluscs. Because the isotope is extremely hazardous, the consumption of marine molluscs and aquatic species which are usually eaten in their entirety (e.g., sardines and herring) should be restricted if contamination is suspected.

SECTION XIX

RUTHENIUM

Ruthenium is rare in nature, occurring as the metal and in arsenide, sulfide, and other ores. Ruthenium is also a fission product of uranium and is used as a platinum and palladium alloy, as a catalyst, and as a dye in ceramics (Weast 1975; McKee and Wolf 1963). Ruthenium-106 is common in the effluents from nuclear fuel reprocessing plants, therefore its presence in the aquatic environment has received some attention.

Ishikawa et al. (1976) measured ^{106}Ru uptake and elimination by the marine clam (*Meretrix meretrix lusoria*) as an initial step in determining the influences of ruthenium from a Japanese nuclear fuel reprocessing plant on the marine environment. Clams attained highest ruthenium levels in mid gut gland followed in decreasing order by gill, visceral mass, mantle, shell and foot. The concentration factor for gill was 10. Prepared ruthenium was present either as purified $^{106}Ru \cdot Cl_x$ or $^{106}RuNO \cdot Cl_x$, and purchased ruthenium was present as crude $^{106}Ru \cdot Cl_x$ or $^{106}RuNO \cdot (NO_3)_x$. Prepared ruthenium was accumulated slightly more readily than purchased forms, and $RuNO \cdot Cl_x$ was eliminated faster than $Ru \cdot Cl_x$. Ruthenium elimination was characterized by two phases, the first having a biological half-time of 39.3 to 48.7 days and the slower second phase having a half-time of 121.2 to 166.7 days. The fast phase represented elimination from soft parts, whereas the slow phase was due to elimination from shell.

Jones (1960) studied ruthenium uptake by marine organisms and sediments and determined the influences of the source of the ruthenium and the presence of iron on uptake. Marine algae absorbed ^{106}Ru in amounts proportional to their surface areas, but the extra-cellular composition of the algae was also a determining factor. Mussel (*Mytilus edulis*) accumulated highest amounts of ^{106}Ru in shell whereas plaice (*Pleuronectes platessa*) contained highest ^{106}Ru levels in gill, gut and skin with very little in muscle. Nitrosyl ^{106}Ru was complexed by iron, making it less available to biota. Organisms accumulated commercial ruthenium more readily than ruthenium present in nuclear reactor effluents.

Berg and Ginsberg (1976) collected and analyzed ruthenium-contaminated crayfish (*Orconectes obscurus, O. rusticus rusticus, Cambarus robustus* and *C. bartoni bartoni*) from a New York creek and performed ruthenium uptake studies with two species of crayfish (*C. robustus* and *C. rusticus*) in the laboratory. The laboratory experiments showed that crayfish species and sex, and whether the tracer was present as a chloride or nitrate derivative, had little influence on uptake, but physical form of the additive, route of

uptake, and mode of administration all influenced accumulation. Suspended particulates decreased uptake, ruthenium present in food increased the visceral content of ruthenium, and ruthenium was concentrated to a greater degree by conditioned individuals than by new crayfish added to the same medium. Concentration factor ranged from three to nine depending on the medium. Most ruthenium in the crayfish was associated with exoskeleton. whereas gill and muscle contained very little ruthenium. Viscera contained substantial amounts of ruthenium when ruthenium was present in the diet, but accumulated negligible amounts when water was the only source of the isotope. Crayfish collected from Buttermilk Creek, New York, had highest ruthenium levels in digestive gland followed by gill, carcass, abdominal muscle and body fluids. Fish from the same creek contained considerably less ruthenium than crayfish. Crayfish were suggested as indicator organisms for detecting ruthenium contamination. Harrison (1973) also found that fishes accumulated very little ruthenium (as ^{103}Ru); however, crayfish (*Astacus* sp.) in freshwater and crabs (*Cancer productus*) in saltwater exhibited concentration factors ranging from three to seven in visceral organs.

The chemical form of the ruthenium and the chemical characteristics of the water influence the accumulative properties of the isotope. Although the concentration factors reported for aquatic organisms are quite low, the biological half-time of ruthenium is high. Shellfishes appear to concentrate ruthenium more readily than fishes, and ruthenium in fishes does not lodge in edible muscle tissue. Instead, the internal organs, gills and skin of fishes preferentially accumulate ruthenium. In crayfish, the mode of exposure influences the distribution in the body. Although ruthenium is not readily accumulated by aquatic organisms, the occurrence of the radioisotope in seafoods may present a potential hazard to humans.

SECTION XX

SELENIUM

Selenium is commonly used in industry and agriculture and occurs in relatively high concentrations in fossil fuels and some natural sediments. Selenium, therefore, reaches natural waters via both fallout and runoff. Plants growing in geographic regions possessing highly seleniferous soils usually contain high concentrations of selenium. The consumption of these plants by grazing animals has resulted in alkali disease or selenium poisoning (Berry et al. 1974). Symptoms include lesions of internal organs, characterized by congestion and hemorrhages. Industrial accidents have resulted in selenium poisonings in humans, but poisonings are more common in other animals. Beal (1974) described the symptoms of human selenium poisoning in detail; poisoning is characterized by anemia, nervousness, hypertension, depression, gastrointestinal disturbances and garlic odor of the breath and perspiration.

The toxic action of selenium reportedly results from its inhibition of sulfur enzymes (Berry et al. 1974). However, selenium in the diet is known to exert a protective influence against mercury poisoning. Japanese quail and rats fed a tuna diet containing selenium and methylmercury were less susceptible to methylmercury poisoning than individuals fed a corn soya diet containing only methylmercury (Ganther and Sunde 1974). Sell and Horani (1976) reported a similar finding for chicks and Japanese quail; in their experiments dietary selenium reduced methylmercury accumulations by 50 percent. Koeman et al. (1973) have shown that mercury and selenium are present in the livers of marine mammals in a 1:1 molar ratio whereas marine fishes usually contain upwards of 40 times more selenium than mercury. This result suggests that selenium and mercury may occur together in marine mammals, perhaps resulting in some degree of protection against mercury.

Kim et al. (1977) found that creek chubs (*Semotilus atromaculatus*) immersed in water containing 3.0 mg Se ℓ^{-1} for 48 hours were less susceptible to mercuric chloride in water than untreated individuals. Interestingly, at mercury concentrations below 0.07 mg Hg ℓ^{-1} selenium treatment increased mercury accumulation; but at mercury levels above 0.10 mg Hg ℓ^{-1} selenium inhibited mercury accumulation. No speculations were offered as to the mechanism of this action.

Sandholm et al. (1973) studied selenium uptake in a laboratory food chain consisting of water, phytoplankton, zooplankton and fish; both selenite and selenomethionine were used. In these experiments food was

determined to be the most important source of selenium to fish because very little selenium was accumulated via the water route.

In a saltwater study, Fowler and Benayoun (1976a) measured selenium uptake in a marine shrimp (*Lysmata seticaudata*) exposed to ^{75}Se in food and water and in a mussel (*Mytilus galloprovincialis*) exposed to ^{75}Se in water. Shrimp exoskeleton contained 60 to 90 percent of the ^{75}Se accumulated when exposure was through the water, but viscera had the highest activity when selenium was accumulated via food. Exoskeleton contained 20 to 45 percent of the selenium accumulated from food. Only 10 percent of the selenium present in exoskeleton was lost during molting. The selenium distribution in shrimp accumulating selenium from food was similar to the distribution of selenium found in shrimp in nature. Mussels accumulated the highest concentrations of selenium in viscera. Selenium concentrations continued to increase in all tissues analyzed (gill, muscle, shell, viscera, mantle and whole body) after 63 days' exposure. The greatest percentage of total selenium in the animal (40 to 60 percent) occurred in the shell. Fowler and Benayoun (1976b) reached a similar conclusion with euphausiids (*Meganyctiphanes norvegica*). Dietary selenium was retained at an efficiency of 66 percent. Viscera attained the highest concentration of selenium and molted exoskeletons contained from 3 to 8 percent of the selenium which had been accumulated. The biological half-time of selenium was 37 days and whole body concentration factors were estimated at 1500 to 7500 for this species.

Beal (1974) measured the selenium content of fishes from various freshwaters in central Canada and found an average whole body selenium content of 0.33 $\mu g\ g^{-1}$ with values ranging from 0.04 to 2.00. In a similar survey of fishes from New York State waters Pakkala et al. (1972) observed comparable values. Barnhart (1958) found high levels of selenium in fish from an artificial Colorado lake located in a highly seleniferous region. The inability of stocked fish to survive for extended periods of time in this impoundment was attributed to fish accumulating excessive selenium through their food chain. Analyses of aquatic organisms from various fresh and marine waters near Finland revealed that marine fishes usually contained 1 to 2 μg Se g^{-1} whereas freshwater fishes ranged from 2 to 3 (Sandholm et al. 1973). Marine plankton contained selenium levels similar to those in fish; however, aquatic flowering plants had extremely low concentrations of selenium.

What little information is available suggests that dietary selenium is the most important source of selenium to many marine and freshwater organisms; however, this has not been confirmed. Fishes do not appear to concentrate selenium at levels which would be dangerous to human health; in fact, in some instances the accumulation of selenium by fishes may be beneficial to both fishes and to human consumers of fishes due to the protective action selenium provides against mercury. However, because of selenium's high toxicity, the relationship between selenium toxicity to aquatic organisms and selenium accumulation requires further attention.

SECTION XXI

SILVER

Silver is one of the most toxic metals to aquatic life, ranking ahead of mercury on a relative acute toxicity basis (Doudoroff and Katz 1953). Silver is used in photography, silverware, metal alloys, electroplating, ink, food and beverage, and porcelain (Weast 1975, McKee and Wolf 1963). In addition, silver iodide is an effective nucleating agent in weather modification (Cooper and Jolly 1970). Although natural weathering processes contribute some silver to natural waters, most silver salts are insoluble in water (McKee and Wolf 1963). Effluents from the photoprocessing industry contribute silver to natural waterways (Terhaar et al. 1977). Over-exposure to silver by humans results in graying of the skin, eyes, and mucous membranes (argyrosis). However, because of the low solubility of most silver compounds, very little is accumulated by mammals.

Cearley (1971) found that juvenile largemouth bass (*Micropterus salmoides*) and bluegill (*Lepomis macrochirus*) exposed to 0.01 or 0.001 mg Ag ℓ^{-1} for six months continued to accumulate silver for two months then leveled off. Internal organs contained more silver than muscle tissue. Similarly, Coleman and Cearley (1974) exposed largemouth bass and bluegill to several silver concentrations in water (from 0.3 to 70 µg Ag ℓ^{-1}) for up to six months and measured silver accumulation in various tissues from bass and in whole bluegill. Silver uptake by both species was rapid during the first two months but then slowed considerably. Gill and internal organs from bass (including liver, kidney, spleen and digestive system) reached much higher silver levels than did the remainder of the fish, demonstrating that muscle tissue accumulates little silver. Concentration factor for the gills of bass was near 200, and whole bluegill concentrated silver in gill up to 120-fold. These findings are in agreement with those of Goettl et al. (1974) who measured silver concentrations in tissues of cutthroat trout (*Salmo clarki*) collected from an alpine lake located in a region underlying an atmospheric zone undergoing extensive cloud seeding with silver iodide. Silver concentrations (in µg Ag g^{-1} dry tissue) ranged from: 1.92-4.40, bone; 0.09-0.99, muscle; 1.08-2.21, liver; 0.32-0.92, gonads; 0.00-0.79, skin; 0.28-0.50, gut; and 0.18-0.50, kidney. Hibiya and Ogura (1961) measured the distribution of ^{110}Ag, five to seven days after a single dose of the isotope was injected into the air bladders of goldfish (*Carassius auratus*). During this experiment liver and air bladder attained higher silver levels than other organs.

Terhaar et al. (1977) have shown that an alga (*Scenedesmus* sp.), Daphnia (*Daphnia magna*), freshwater mussels (*Ligumia* sp. and *Margaritifera*

sp.) and fathead minnows (*Pimephales promelas*) are all capable of accumulating silver from water; but the food chain was not an important route of silver accumulation for animals at the higher trophic levels. Thurberg et al. (1974) measured silver levels in marine bivalves following 96 hours' exposure to either 0.5 or 1.0 mg Ag ℓ^{-1}. Gills contained higher silver levels than the rest of the body and individuals exposed to the higher concentration accumulated slightly more silver than those exposed to 0.5 mg Ag ℓ^{-1}, but the difference was not proportional to exposure level. Elimination rates were not measured.

In surveys of the metals content of fish from both marine (Greig et al. 1976; McDermott et al. 1976) and freshwater (Lucas et al. 1970; Tong et al. 1974) environments, silver was always present at low concentrations, usually less than 0.1 µg Ag g^{-1}. Red abalone (*Haliotis rufescens*) from the California coast contained 13 to 129 µg Ag g^{-1} in gill, mantle or digestive gland (dry basis) but only 1.1 to 44 µg Ag g^{-1} in foot (Anderlini 1974). The silver concentration in foot averaged only one-tenth that found in other organs. An apparent inverse relationship was noted between the silver and copper content of abalone.

Silver is not present in aquatic animals at very high concentrations because most of its compounds are virtually insoluble in water and silver has a very short biological half-time; moreover, ingested silver has a very low toxicity to humans and does not accumulate significantly in the edible portions of fish. This combination of characteristics decreases the hazards associated with consuming silver-exposed aquatic organisms; but because of silver's high toxicity to aquatic life, threshold levels of silver in key organs should be determined.

SECTION XXII

STRONTIUM

Radioactive strontium may enter the environment as a result of nuclear detonation or in effluents containing nuclear wastes. Strontium fallout easily enters ground and surface waters since strontium is not appreciably absorbed by soils (McKee and Wolf 1963). Strontium is chemically similar to calcium and is therefore deposited in bony tissues. Because of this bone-seeking tendency, radiostrontium is extremely dangerous; however, non-radioactive strontium is almost nontoxic to man.

Brungs (1967) observed that animals with exoskeletons (crayfish) or shells (clams, snails) were capable of accumulating much higher ^{85}Sr levels than animals containing higher percentages of soft tissues. However, crayfish (*Cambarus longulus longerostris*) have been shown to lose most of their accumulated ^{85}Sr during molting (Schurr and Stamper 1962). Although strontium was readily accumulated by crayfish, the biological half-time was only about two days. In a lake receiving radioactive wastes strontium concentration factors in the hard parts of fish reportedly exceeded 30,000 (Krumholz 1956). Increasing calcium concentrations in water decreased strontium uptake by aquatic organisms due to the similarity of the two elements (Williams and Pickering 1961; Preston et al. 1967). Feldt (1963) demonstrated this trend by showing that strontium levels in fishes from various seas and lakes were inversely related to salinity in saltwater and to hardness in freshwater.

Schiffman (1961a) measured strontium flux in perfused gills from rainbow trout (*Salmo gairdneri*). Trout were able to excrete strontium even against a concentration gradient, suggesting that the ability of trout to concentrate strontium must result from a binding mechanism reducing the diffusibility of ionic strontium. Dialysis studies with strontium in blood confirmed that nearly 50 percent of the strontium was present in a nondialyzable form. In another study Schiffman (1961b) injected a single dose of ^{85}Sr into the dorsal aorta of a urinary bladder cannulated rainbow trout. After 24 hours 6 to 7 percent of the ^{85}Sr was in the urine, 3 to 4 percent in the gut and 50 to 75 percent remained in the fish. These results suggest that 15 to 40 percent was excreted via the gills or skin.

Nakatani and Foster (1963) fed varying levels of ^{90}Sr-^{90}Y to rainbow trout for up to 25 weeks. About 25 percent of the oral dose administered was retained by the trout, and bony tissues contained the highest concentrations. Ophel and Judd (1967) examined some of the factors governing radiostrontium accumulation from food in the goldfish (*Carassius auratus*). Fish

were force-fed the radioactive diet followed by a voluntary feeding of a control diet. Calcium and magnesium in the diet reduced the retention of ^{90}Sr from the diet. The concentration factor for dietary ^{90}Sr was 250-500. Increasing the time interval between the isotope force-feeding and the voluntary control feeding increased the efficiency of dietary ^{90}Sr uptake. Tripling the specific activity of the food resulted in only a slight decrease in retention efficiency of the isotope. Similarly, Schiffman (1959) measured the retention of ingested ^{90}Sr-^{90}Y by rainbow trout when the isotope was present in a natural diet or in a gelatin capsule. Twenty-one percent of the encapsulated strontium was retained by the trout but only seven percent of the isotope present in the natural diet was retained. Strontium accumulated from food and water were additive. Based on the retention of strontium from the natural diet, water was calculated to be ten times more important than food as a source of strontium to fish in nature.

Shealy and Carlson (1973) exposed various life stages of largemouth bass, (*Micropterus salmoides*) including embryo, prolarval, postlarval and juvenile, to ^{85}Sr in water and measured accumulation and retention of the isotope. Strontium was retained longer as the bass became older, presumably due to an age-related increase in percentage of bony structures. Rosenthal (1963) measured strontium uptake and turnover in the guppy (*Lebistes*). Uptake was linear over the 15-day experiment. Viscera lost strontium rapidly with a biological half-time of only eight days but the half-time for the rest of the body including muscle tissue was over two years. Guppies showed only a slight preference for calcium over strontium. Martin and Goldberg (1962) followed the uptake and elimination of ^{90}Sr in various organs and tissues from Pacific mackerel (*Pneumatophorus diego*) following a single oral inoculation of the isotope. Ninety-five percent of the dose was excreted during the first 24 hours following inoculation, but the remaining five percent remained in the fish for the duration of the 235-day experiment. The retained ^{90}Sr (80 percent) was located in bony tissue, where the concentration remained constant throughout the experiment. Strontium increased rapidly in gill followed by a steady decrease, suggesting that this organ is important in strontium excretion. Edible muscle tissue retained very little of the isotope.

Strontium is readily accumulated and retained by fish from either their food or water. Calcium competes with strontium in the uptake process, thus organisms accumulate less strontium in calcium-rich waters. Upon entering fish, strontium is eliminated primarily through the gills. Strontium is chemically similar to calcium and is therefore bone-seeking; thus aquatic organisms with high percentages of bone tend to accumulate high levels of the metal. Bone-bound strontium is retained for long periods of time by non-molting animals; however, non-radioactive strontium has a very low toxicity both to aquatic animals and to man. Fishes such as sardines which are consumed in their entirety represent the greatest risk to humans, and soft waters contaminated by the radioisotope offer the optimum conditions for isotopic bioaccumulation.

SECTION XXIII

ZINC

Zinc has relatively low abundance in nature, but occurs widely in a number of minerals. Zinc is extensively used in alloys, for galvanizing, and for die castings; it is also used in the manufacture of paints, cosmetics, pigments, electrical equipment, and other products (Weast 1975). Zinc is an essential element for human and animal growth. Zinc is also an important cofactor for certain enzymes (Lehninger 1970) and has a relatively low toxicity to man. However, over-exposure by humans to zinc oxides or chlorides has resulted in flu-like symptoms and pneumonia (Berry et al. 1974). Zinc enters water in numerous industrial effluents and through acid mine drainage. In addition radioactive zinc is released during nuclear explosions. Zinc sulfate and halides are soluble in water but the carbonate, oxide and sulfides are insoluble (Weast 1975). Zinc toxicity to fishes decreases with increasing water hardness (Mount 1966; Pickering and Henderson 1966; Sinley et al. 1974). In static bioassays zinc toxicity was reported to decrease with increasing pH (Sprague 1964; Cairns et al. 1972). However, Mount (1966), using flow-through tests, observed that zinc was more toxic with increasing pH; a possible explanation for this was that in static bioassays insoluble zinc settles out of solution, whereas in flow-through systems it remains in suspension.

Zinc has been extensively studied in the freshwater environment. Zinc-65 was accumulated much more readily than ^{60}Co, ^{137}Cs or ^{85}Sr by soft tissues of carp, snails, tadpoles and clams during radioisotope experiments conducted in ponds (Brungs 1967). During laboratory experiments brown bullhead (*Ictalurus nebulosus*) accumulated ^{65}Zn rapidly for the first seven hours' exposure followed by a reduced accumulation rate (Joyner 1961). Gill and viscera reached the highest zinc concentrations of the tissues analyzed. The esophagus was plugged on some fish to determine the fraction of zinc accumulation attributable to swallowed water; this route of uptake was found to be negligible. Zinc-exposed fish transferred to fresh water lost half of their accumulated zinc after six days followed by a period of reduced zinc elimination. Willis and Jones (1977) determined that zinc elimination in juvenile mosquitofish (*Gambusia affinis*) was derived from three separate zinc reservoirs having biological half-times of 2, 14 and 235 days; relative compartment sizes (as percent of total zinc present in the fish) were reported to be 9, 4, and 91, respectively. Thus, most of the zinc accumulated is slowly eliminated.

Hodson (1975) studied the influence of temperature on zinc accumulation by the gills of Atlantic salmon (*Salmo salar*) and related zinc uptake to

lethal response. Temperatures of 3, 11 and 19 C were tested, and fish were exposed to about 14 mg Zn ℓ^{-1}. Zinc uptake increased as temperature increased, presumably due to a temperature-related increase in metabolic rate. Salmon killed by zinc at 19 C contained significantly more zinc than individuals killed by zinc at lower temperatures. This finding corresponds with the observations that the lethal threshold for zinc increases with temperature and demonstrates that higher tissue residues are necessary to kill salmon as temperature increases, suggesting that the site of toxic action is more resistant at higher temperatures.

Goettl et al. (1972) measured zinc uptake in various tissues from rainbow trout (*Salmo gairdneri*) exposed to zinc in water for up to 92 weeks. Eye accumulated the highest concentration of zinc followed by gill, bone, intestine, liver, kidney and skin. Baseline zinc levels (dry weight basis) for selected tissues from unexposed fish were (in $\mu g\ g^{-1}$): eye 400, skin 90, gill 200, opercular bone 195, stomach 1, liver 150, muscle 20, kidney 125 and intestine 190. In another study Goettl et al. (1974) found that rainbow trout accumulated zinc in eye, gill arch and opercular bone in proportion to zinc concentration in water after exposure to zinc levels ranging from 71 to 260 μg Zn ℓ^{-1} for up to 77 weeks. However, for fish exposed to any given exposure concentration, zinc in opercular bone decreased in time. Slater (1961) found that fingerling brook trout (*Salvelinus fontinalis*) and cutthroat trout (*Salmo clarki*) accumulated ^{65}Zn more readily than fingerling rainbow trout; gill filaments accumulated the highest levels of ^{65}Zn in all three species, and rainbow trout gill tissue accumulated less zinc than gill from the other two species. Joyner and Eisler (1961) immersed chinook salmon (*Oncorhynchus tshawytscha*) fingerlings in a freshwater solution containing 0.2 mg ^{65}Zn ℓ^{-1} for 24 hours followed by exposure to zinc-free water for 63 days. Fish were then sampled at various time intervals. Fish accumulated about 2 percent of the zinc present in the initial test solution and retained almost all of their accumulated zinc throughout the 63-day period in zinc-free water. However, zinc redistributed within the fish during the zinc-free portion of the test, increasing in vertebral column, head and viscera, and decreasing in muscle, skin, scales and fins. After a seven- to nine-day incubation period, isotopic zinc injected into the air bladders of goldfish (*Carassius auratus*) was accumulated to the largest extent in intestine (Hibiya and Oguri 1961). This result implies that fish excrete zinc through the intestine.

Three-spined stickleback (*Gasterosteus aculeatus*) exposed to ^{65}Zn in freshwater accumulated zinc initially but were then able to reduce their internal zinc concentration to a level approaching that of control fish (Matthiessen and Brafield 1977). This unusual ability was attributed to the euryhaline nature of stickleback. These workers also found that zinc uptake (whole fish measurements) was higher in water of high hardness even though zinc toxicity was inversely related to hardness. The authors hypothesized that the low uptake of zinc in calcium-free water could be due to zinc precipitation of mucus on body surface and gills, with release into the water. They suggested that zinc toxicity is lower in hard water because calcium interferes with the internal mechanism of zinc toxicity, perhaps by occupying potential zinc-binding sites on proteins.

Marafante (1976) found that intraperitoneally injected zinc, mercury and cadmium were incorporated into the cadmium-binding proteins present in the livers and kidneys of goldfish (*Carassius auratus*). All of the cadmium in liver and kidney was associated with this protein. However, only 40 percent of the zinc and 17 percent of the mercury in liver and 1.9 percent of the zinc and 12 percent of the mercury in kidney were associated with the cadmium-binding protein. The rest was attached to higher molecular weight proteins. The very low percentage of zinc associated with cadmium-binding proteins in kidney compared to liver suggests that a specific zinc-binding protein may exist in this organ.

Wedemeyer (1968) studied the physiology of ^{65}Zn accumulation in coho salmon (*Oncorhynchus kisutch*) eggs. Of the ^{65}Zn accumulated, 70 percent was in the chorion, 26 percent in the perivitelline fluid, 2 percent in the yolk and 1 percent in the embryo. Ten minutes was required for the yolk to reach maximum zinc concentration and the level attained was a direct function of exposure magnitude. Changing pH greatly influenced zinc accumulation by the chorion and perivitelline fluid, with eggs accumulating the most zinc over the pH range 4 to 9. Exposure to iodoacetate increased zinc uptake by the perivitelline fluid but did not influence accumulation by yolk or embryos thus suggesting that the vitelline membrane was not altered. The mechanism of this increased uptake was believed to be an increased diffusion of zinc across the chorion, resulting from reduced zinc binding to the chorion due to a sulfhydryl blockage. Azo dye and malachite green also increased zinc permeability, but with these chemicals yolk accumulated substantial amounts of zinc suggesting that the vitelline membrane had been altered. The author speculated that the pH dependence of chorion zinc uptake and the influence of iodoacetate on accumulation suggests that negative charge groups participate at zinc-binding sites on the chorion. The fact that an amino blocking agent (2,4-dinitrofluorobenzene) had no influence on uptake supports this supposition. When copper was present at concentrations below 2 mg Cu ℓ^{-1}, zinc uptake was inhibited, but above this concentration zinc uptake was stimulated. Wedemeyer concluded that the uptake process involves first a physicochemical sorption onto the chorion along with a passive diffusion of zinc into the perivitelline fluid, yolk and embryo. Chemicals acting to decrease zinc binding at the chorion increase diffusion by causing a greater concentration gradient and therefore should increase toxicity.

Spehar (1976) exposed flagfish (*Jordanella floridae*) to various zinc concentrations in water and observed significant uptake at concentrations exceeding 47 $\mu g\ \ell^{-1}$. Fish reached a plateau level of zinc in less than 30 days. The lowest zinc concentration having an adverse effect on the fish (reduced growth in females) was 51 $\mu g\ \ell^{-1}$, indicating that adverse symptoms are first realized near the metal concentration where accumulation begins to occur.

Mount (1964) developed an autopsy technique for fish killed from acute exposure to zinc. The technique utilized the principle that zinc accumulates in opercular bone very slowly regardless of the magnitude of exposure, whereas gill tissue accumulates zinc at a modest rate during chronic exposures but rapidly during acutely lethal exposures. Thus, the opercular

bone-zinc to gill-zinc ratio proved to be a useful tool for detecting acute fish mortality due to zinc. Of more than 20 fish species examined from zinc-pollution-free natural waters, only carp (*Cyprinus carpio*) had gill to opercular bone zinc ratios that precluded the use of this technique. Previous exposure to sublethal concentrations of zinc did not influence the usefulness of the method; also, dead fish placed in zinc-contaminated water gave negative results when analyzed. The validity of this technique was further confirmed by Cairns et al. (1971) who analyzed tissues from adult bluegill (*Lepomis macrochirus*) of various sizes that had undergone acute zinc exposure under a variety of experimental conditions. These workers detected zinc above the designated threshold level in some zinc-exposed survivors; however, this finding was attributed to the fact that these bluegills were very near death.

Radiozinc uptake by pumpkinseed sunfish (*Lepomis gibbosus*) from a natural and a synthetic diet were compared by Merlini et al. (1976). In addition, zinc uptake from food was compared to that from water. Zinc-65 was accumulated much more readily from an artificially prepared diet than from a natural diet (the snail *Viviparus ater*). Both diets contained similar levels of zinc. After 25 days of feeding, the fish fed the artificial diet contained nearly six times more zinc than the snail-fed fish. Moreover, the kind of diet influenced zinc accumulation from water. Fish fed uncontaminated synthetic diet accumulated three times as much zinc from water as fish fed snails. This difference in uptake of zinc from water was believed to be a function of the degree to which the zinc was organically bound. In fish fed the artificial diet, zinc was available in the ionic form; in natural food the zinc was more apt to be organically bound and this was believed to be dependent upon whether the food organism was in the process of accumulating or eliminating zinc at the time it was eaten. This hypothesis is consistent with zinc elimination studies which show different half-times of elimination from different zinc pools within an organism. Renfro et al. (1975) designed experiments to determine the relative importance of food and water as sources of ^{65}Zn to fish (*Gobius*), crabs (*Carcinus maenas*) and benthic shrimp (*Lysmata seticaudata*). Test organisms were exposed to the isotope only through water or via both routes. Shrimp and crabs received a lower percentage increment of ^{65}Zn from food than did fish. At the end of these experiments the final proportions of ^{65}Zn activity accumulated from food and water and from water alone were approximately 54:45 in shrimp, 71:31 in crabs and 4:1 in fish. However, it should be noted that the relative ratio of the concentrations of zinc in water and in food was arbitrarily chosen; the results would obviously change with any alteration of this ratio. On the average, crabs lost 61 percent of their zinc activity during molting and shrimp lost 45 percent. Interestingly, the mode of ^{65}Zn accumulation by the food organism (*Artemia salina*) of shrimp did not affect ^{65}Zn uptake by the shrimp. Whole-body concentration factors after 90 days for the organisms exposed to ^{65}Zn via both food and water were 380 for shrimp, 210 for crabs and 25 for fish.

Bryan (1967) studied zinc regulation in the freshwater crayfish (*Austropotamobius pallipes pallipes*) to determine if the trend towards higher zinc blood levels observed in decapod crustaceans in moving from marine to estuarine types continued into freshwater. However, freshwater

crayfish were found to accumulate lower zinc blood concentrations than either marine or estuarine forms. Interestingly, this crayfish accumulated much higher zinc levels in muscle than did its estuarine or marine relative. The latter finding suggests that crayfish regulated zinc in muscle. High zinc levels in stomach fluids imply that food is a major route of zinc uptake. The hepatopancreas was believed to be the principal zinc-regulating organ. In the marine lobster *Homarus vulgaris*, Bryan (1964) found that excretory organs, hepatopancreas and gill increased in zinc concentration following exposure to zinc through either water or food, but muscle and gonad remained the same. Zinc was removed from circulation by both hepatopancreatic absorption and urinary excretion. Zinc concentration factor was inversely related to its concentration in water. Zinc was not accumulated as readily as copper.

Other workers have also reported on zinc's accumulation in and influence on marine organisms. Scott (1977) compared the mercury and zinc concentrations in cleithrum bones from recently caught Atlantic cod (*Gadus morhua*) to the content of these metals in cleithra from cod recovered from an 1865 shipwreck. The historical fish had mercury concentrations similar to those in recently collected fish, but the zinc concentration was higher in the contemporary fish suggesting that the zinc content of the oceans may be increasing. Wright (1976) found unusually high levels of zinc in fishes collected off the northeast coast of England although no known sources of zinc existed in the area. Some individuals were found to contain more than 100 μg Zn g^{-1} in axial muscle tissue.

Seymour (1966) measured ^{65}Zn uptake in Pacific oysters (*Crassostrea gigas*) transferred from radionuclide-free water in Puget Sound to Willapa Bay, Washington, located just north of the confluence of the Columbia River with the Pacific Ocean. Cooling water from the Hanford nuclear reactor containing trace amounts of ^{65}Zn flows down the Columbia River reaching this portion of the Washington coast. Uptake became asymptotic after 500 days with a final concentration factor of 1.5×10^4. Zinc-65 elimination rate was measured by transferring oysters in the opposite direction. Upon transfer to uncontaminated water, elimination proceeded linearly with the biological half-time calculated to be 255 days. Similarly, Osterberg (1962) was able to detect elevated ^{65}Zn concentrations in euphausiids (*Euphausia pacifica*) and tunicates (*Salpa* spp.) collected within the Columbia River plume off the Oregon coast. Wolfe (1970) measured zinc distribution in South Carolina coastal oysters (*Crassostrea virginica*). Although zinc was present throughout the oyster, those tissues with exposed surfaces such as gills and mantles had the highest concentrations. In eastern oysters (*C. virginica*), hard-shell clams (*Venus mercenaria*), and bay scallops (*Pecten irradians*) collected near Beaufort, North Carolina, labial palps and gill accumulated the highest levels of zinc whereas adductor muscle was very low (Chipman et al. 1958). Oysters collected from uncontaminated areas contained 0.3 to 1.0 μg Zn g^{-1}, supposedly representing background levels; clams and scallops contained less zinc than oysters. Studying the distribution of ^{65}Zn administered to artificial estuarine ponds, Duke et al. (1966) and Duke (1967) noted that eastern oysters, scallops (*Aquipecten irradians*) and clams (*Mercenaria mercenaria*) accumulated significantly higher levels of the isotope than other organisms

including fish; oysters and clams had the highest concentrations in edible portions. Sediment acted as a sink for ^{65}Zn in the ponds; over 99 percent of the ^{65}Zn was in the sediment 100 days after application. Shuster and Pringle (1969) subjected American eastern oysters to either 0.1 or 0.2 mg Zn ℓ^{-1} for up to 20 weeks and compared the zinc levels attained to zinc levels found in oysters collected along the Atlantic coast of the United States (Pringle et al. 1968). After 20 weeks the experimental oysters exposed to the two zinc levels averaged about 2650 and 3500 µg Zn g^{-1} respectively. Comparatively, collected oysters ranged from 204.4 to 4120 µg Zn g^{-1}. These values are much higher than those reported earlier by Chipman and coworkers (1958).

A large percentage of the ^{65}Zn accumulated by common shrimp (*Crangon crangon*) was present in exoskeleton and was, therefore, lost during molting (van Weers 1975). Increasing the water temperature increased the frequency of molting, thereby increasing the rate of zinc elimination. Elimination appeared to be from two compartments. Changing the water temperature from 10 to 15 C resulted in the biological half-time for the slow compartment decreasing from 33.7 to 16.5 days. In experiments with the salt marsh snail (*Littorina irrorata*), ^{65}Zn elimination rate was shown to be directly related to water temperature and inversely related to snail body size (Mishima and Odum 1963). Elimination was characterized by a short rapid phase followed by an extended slower phase; metabolic rate was believed to be the most important factor regulating zinc elimination rate.

Kameda et al. (1968) measured ^{65}Zn uptake by various organs and tissues from two marine fishes including marine goby (*Chasmichthys gulosus*) and filefish (*Rudarius ercodes*), a mussel (*Mytilus edulis*), short-necked clam (*Tapes japonica*) and a sea urchin (*Strongylocentrotus pulcherrimus*). Fish reached highest zinc concentration in digestive tract, gill and viscera; muscle and vertebrae accumulated very little zinc. Clams accumulated highest levels in gill and mantle, but mussels had highest zinc concentrations in adductor muscle, viscera and shell. Mussels accumulated considerably more ^{65}Zn than clams. The digestive tract of the sea urchin contained much higher zinc levels than its other organs.

In the plaice (*Pleuronectes platessa*) orally administered ^{65}Zn was retained at relatively high concentrations by most organs tested including heart, spleen, liver, kidney, gonad, gut, gill and skin (Pentreath 1973). Gonads from males were particularly high in zinc, containing approximately three times as much zinc as other organs. The biological half-time for zinc accumulated from water was 313.1 days while intraperitoneally injected zinc had a half-time of only 210.7 days. Zinc-65 was assimilated almost twice as efficiently from gelatin or starch pellets compared to ^{65}Zn fed in live *Nereis* which had previously been exposed to the isotope. Croaker (*Micropogon undulatus*) fed zinc accumulated high zinc levels in gill, kidney, liver and spleen (Chipman et al. 1958). Upon return to zinc-free water, most zinc was quickly eliminated.

Eisler (1967) exposed adult mummichog (*Fundulus heteroclitus*) under static conditions in saltwater to various zinc concentrations ranging from 0.78 to 180 mg ℓ^{-1} (these levels bracketed acutely lethal concentrations)

and measured zinc concentrations in gill arch and whole fish after various exposure intervals. After 192 hours all fish exposed to 42 mg Zn ℓ^{-1} or less survived and contained no more detectable zinc than controls, whereas all fish exposed to 157 and 180 mg Zn ℓ^{-1} died within 48 hours and contained on the average seven times as much zinc as control individuals for whole fish and eight times as much in gill arch. In another study Eisler and Gardner (1973) exposed mummichog to various combinations of cadmium, copper and zinc in seawater with several interesting findings. Low levels of cadmium inhibited zinc accumulation, whereas copper and zinc increased cadmium accumulation. Dead fish immersed in solutions of zinc or copper accumulated more metal than living individuals. Similarly, Eisler (1971) found that dead fish accumulated more cadmium than survivors. These findings should be carefully considered by those interested in using autopsy indices for determining the cause of fish kills in saltwater.

Hoss (1964) exposed three species of flounder of the genus *Paralichthys* to ^{65}Zn added to both food and water. Major conclusions were that zinc uptake from water was proportionally related to the zinc concentration in water and zinc uptake from food and water were additive. Nearly 20 percent of the dietary zinc and 0.2 percent of the aqueous zinc were accumulated; however, fish were exposed to a static test solution containing a fixed volume. The percentage of zinc accumulated from water might well vary with a change in either of these conditions. Accumulation from water would be more meaningfully related to respiratory volumes. Moreover, food consumption rates were not measured, making comparisons of zinc accumulated from food and water difficult.

Zinc is readily accumulated by both marine and freshwater fishes from both food and water, but internal organs and bones accumulate much higher zinc levels than edible muscle tissue. The time required for fish to reach threshold levels of zinc appears to be dependent upon species and the chemical nature of the environment. Upon entering fish some zinc associates with cadmium-binding proteins and evidence suggests that a zinc-binding protein may exist. The level at which zinc becomes chronically toxic is very near the concentration at which zinc begins to accumulate. However, the acute toxicity of zinc to sticklebacks has been shown to decrease with increasing calcium concentration even though calcium stimulates zinc uptake. In marine fishes cadmium reportedly decreases zinc accumulation. Zinc accumulation in salmon eggs has been shown to be a diffusion rate process which can be altered by chemical factors that influence the diffusion gradient at the egg membrane surfaces. Because gill tissue accumulates zinc much more rapidly than bony tissue, a method for detecting zinc-caused fish mortalities has been developed utilizing the ratio of zinc in gill to zinc in opercular bone. This technique has proved valid for a variety of fish species.

The zinc content of the oceans appears to be increasing in time. Although marine fishes readily accumulate zinc, evidence suggests that upon return to zinc-free water marine fishes eliminate zinc much more rapidly than freshwater fishes. This occurrence may be due to the different osmoregulatory problems encountered by fishes in these two environments. Oysters are particularly adept at accumulating zinc, showing concentration

factors as high as 26,500. The half-life of zinc in oysters is reportedly 255 days. Although orally ingested zinc has a low toxicity to humans, oysters should probably be monitored in areas where zinc contamination is severe.

SECTION XXIV

GENERAL

Many papers deal largely with the concentrations of groups of metals found in fishes or other aquatic animals from specific geographic regions, or with the role played by specific groups of organisms in transporting or mobilizing metals in general. Many of the conclusions drawn from these studies were broad in nature and, therefore, were not appropriately applied to a particular metal. However, some of the conclusions from some of these papers did apply to a specific metal and when this occurred, the results were also included in the individual metal section.

Tong et al. (1974) measured the concentrations of 36 metals in tissue homogenates from various aged lake trout (*Salvelinus namaycush*) from Lake Cayuga, New York. Of all the metals analyzed, chromium concentration increased with fish age while molybdenum decreased. No age-related trends were apparent for the other 34 metals. Lucas et al. (1970) measured concentrations of 15 metals in liver or muscle of spottail shiner (*Notropis hudsonius*), alewife (*Alosa pseudoharengus*) and trout-perch (*Percopsis omiscomaycus*) from Lakes Michigan, Superior and Erie. In addition, various other Great Lakes fishes were analyzed for selected metals. Small sample sizes and the absence of information permitting correlation of concentration with fish size (age) make the data difficult to interpret. However, a general understanding of the then current levels of some metals in Great Lakes fishes was derived. Kelso and Frank (1974) also surveyed Lake Erie fishes including yellow perch (*Perca flavescens*), white bass (*Morone chrysops*) and smallmouth bass (*Micropterus dolomieui*) for their cadmium, copper and mercury contents. Metals occurred at low levels in all species except for two specimens of white bass which exceeded 0.5 μg Hg g^{-1}. Results were similar to those reported by Lucas et al. (1970) for these three metals.

The Wisconsin Department of Natural Resources (1974) collected fish from various Wisconsin waters to determine the arsenic, cadmium, chromium, lead and zinc contents of edible portions. Cadmium was not detectable in any of the samples but zinc was present at concentrations ranging from 3.0 to 18.3 μg Zn g^{-1}. Arsenic, chromium and lead were for the most part present at concentrations less than 1.0 μg g^{-1}. The authors concluded that consumption of these fish should not result in any adverse effects on humans. In Iowa, Morris et al. (1972) surveyed fishes from various waters for their metals contents. Fishes from the Iowa River contained mercury levels exceeding the FDA guideline, but fishes from all other rivers surveyed contained permissible mercury levels. Other metals, including barium,

cadmium, chromium, copper, lead, nickel and zinc were present at relatively low concentrations in all samples analyzed.

Uthe and Bligh (1971) compared the concentrations of 13 metals in dressed fish collected from several industrialized lakes with those in fish from pristine lakes in Canada and found that, except for mercury, differences were slight. However, Brown and Chow (1977) compared cadmium, copper, lead, mercury and zinc levels in fish from a polluted area of Lake Huron with those in fish from a relatively unpolluted area and found that all metals were present at higher concentrations in fish from the polluted area. The ratio between metal concentration in kidney or liver and metal concentration in muscle was higher in fish from the polluted area for all metals analyzed. Likewise, Atchison (1975) analyzed warmwater fishes from an industrialized Indiana lake for cadmium and chromium and reported levels exceeding those reported for more pristine areas. During a similar study in Wales (Trollope and Evans 1976) the metals content of algae was shown to increase moving nearer to a zinc smelter emitting large amounts of metals wastes. For the metals analyzed the order of concentration factor was highest for iron followed in decreasing order by zinc, lead, copper and nickel. Likewise, fish collected near industrialized areas of England's Medway estuary contained slightly higher levels of lead and cadmium than organisms from undeveloped areas (Wharfe and Van Den Broek 1977). Stapleton (1968) analyzed various tissues from calico bass (*Paralabrax clathratus*) collected near Los Angeles. Some of the bass were collected from the pollution-free Catalina Island area and others were collected near the effluent pipe from a local steam plant. Aluminum, cadmium and nickel were present at higher levels in fish collected near the steam plant and the livers from these fish were enlarged. However, none of the metals present, including cadmium, copper, lead, mercury and zinc, was found at levels considered to be hazardous.

Mathis and Cummings (1973) measured cadmium, chromium, cobalt, copper, lead, lithium, nickel and zinc levels in various biotic and abiotic components of the Illinois River system. For all metals analyzed, sediment and animals living in or on the sediment such as clams and tubificid worms contained higher metals concentrations than either omnivorous or carnivorous fish. Water contained the lowest concentrations of all metals except lithium of any of the components analyzed. The partitioning of cadmium, lead, mercury and thallium in a eutrophic Illinois lake was also studied (Mathis and Kevern 1975). Thallium was present only in sediment, and mercury was present in only fish and sediment. The other two metals were found in all components analyzed including water, sediments, plants, plankton and fish. The feces of migratory waterfowl were also analyzed and were found to contain high levels of cadmium and lead; the authors suggested that waterfowl contribute significant amounts of lead and cadmium to this lake. In another partitioning study Enk and Mathis (1977) looked at the distribution of cadmium and lead in a small Illinois stream. Both metals increased in the order: water, fish, sediment, invertebrates. Aquatic insects contained the highest cadmium levels, and snails contained the most lead. The association of insects and snails with the sediment was believed to contribute to their body burden of these metals.

Patrick and Loutit (1976) investigated the transport of various metals including chromium, copper, iron, lead, manganese and zinc through a food chain consisting of metal-enriched sediment, bacteria and tubificid worms. The system studied was a New Zealand river receiving large amounts of domestic and industrial waste. Results confirmed the hypothesis that these elements are concentrated as they move through this type of food chain. Berner et al. (1962) measured the gamma activity in various marine zooplankton collected from the Pacific Ocean. No attempt was made to distinguish among isotopes. A general trend was noted between level of radioactivity (i.e., uptake of fission products) and the animals' feeding method. Ciliary and mucous filter feeders contained the most radioactivity, followed in decreasing order by setal filterers, rapacious forms and tentacular feeders. Goettl et al. (1971) collected aquatic insects from Colorado streams located near mining and milling sites and analyzed them for copper, lead and zinc content; orders of insects analyzed included Diptera, Ephemeroptera, Plecoptera and Trichoptera. On a dry weight basis, insects were found to contain up to 6440, 6000, and 10,250 $\mu g\ g^{-1}$ of copper, lead, and zinc, respectively. The authors suggested that these high metals levels might be harmful to fish ingesting these insects.

Giesy and Wiener (1977) studied the frequency distributions of cadmium, chromium, copper, iron and zinc in whole body homogenates from several freshwater fish species collected from a South Carolina pond. Essential elements including copper, iron and zinc were distributed normally, but non-essential elements such as cadmium and chromium showed a positively skewed lognormal distribution. These results suggest that the statistical procedures used to describe normally distributed populations should not be applied during certain studies of metals accumulation.

Lee and Wilson (1974) looked at the relationship between the water levels of calcium, magnesium and strontium in various Wisconsin lakes and the composition of clam shells (*Lampsilis siliquoidea rosacea*) from these lakes. No correlation was found, suggesting that factors governing the immediate environment of the clams influence the relationship. Similarly in the marine environment Pilkey and Goodell (1963) examined the relationship between salinity, temperature and the metals composition (including barium, iron, magnesium, manganese and strontium) of various marine mollusc shells. Although some relationships were found, correlations were highly variable among species. The environmental conditions determining mollusc shell composition were probably more complex than this study was designed to elucidate.

Abdullah et al. (1976) analyzed scales from Atlantic salmon (*Salmo salar*) smolts and brown trout (*S. trutta*) collected from various locations in the River Dovoy and Lake Bala in North Wales, for manganese and zinc. Scale manganese and zinc concentrations were compared with concentrations of these metals in water; it was found that there was a direct linear relationship, suggesting that the metals levels in scales reflect environmental exposure. Also in Wales, Ireland and Wootton (1977) measured copper, lead, manganese and zinc concentrations in two marine gastropods (*Littorina littorea* and *Thais lapillus*) collected from various coastal locations. Whole body levels of copper, lead and zinc were higher in

Littorina but manganese was present at higher concentrations in *Thais*. Digestive gland contained the highest concentrations of copper, manganese and zinc for both species but shell contained the highest level of lead. Shell also contained high manganese levels in *Littorina*. Differences in metals accumulation patterns between the two species were believed to result from different diets or preferential accumulation of a particular metal by one species. Papadopoulou et al. (1976) measured metals levels in echinoderms collected near Greece and calculated concentration factors. Metals whose radioisotopes will become more common as nuclear power installations become more widespread, including antimony, cesium, chromium, cobalt, iron, rubidium, scandium, selenium and silver, were intentionally chosen. Interestingly, specific species of echinoderms showed preferential propensities for specific metals. The authors suggested that these "metals specific species" would be good indicator organisms for the particular metal they favored. Bowness and Morton (1952) measured copper and zinc concentrations in various portions of the eyes from frogs (*Rana esculenta* and *R. temporaria*) and fish (*Perca fluviatilis* and *Salmo trutta*). Both metals were present at very high concentrations in eye tissue from these animals with the pigment-protein portion of the eye having the highest concentrations.

Aquatic animals from the Danube River and Danube Canal in Vienna, Austria, were found to contain only background levels of antimony, chromium, cobalt, iron, selenium and zinc (Rehwoldt et al. 1975). In carp (*Cyprinus carpio*) from this same river Rehwoldt et al. (1976) examined the tissue distributions of chromium, cobalt, iron, lanthanum, scandium and zinc. The levels of metals in suspended solids in the water were also measured. Cobalt, lanthanum and scandium were present at highest levels in bone whereas the other metals preferentially accumulated in gill, liver and kidney. The gill metals levels were very similar to the levels in suspended solids, suggesting that the metals were on particles imbedded on the gill surfaces. Justýn and Lusk (1976) examined the accumulation of uranium and ^{226}Ra in stream fishes living above or below the outfall from a uranium mine and mill located in northern Bohemia. Both elements were present at higher concentrations in fish below the mill. Uranium content of brown trout was correlated with age, with uptake remaining nearly linear for the four year classes of fish sampled. Musculature contained much lower levels of both elements than did bone or entrails, thus reducing the risk to human consumers.

The changes in iron, manganese and zinc content of spot (*Leiostomus xanthurus*), Atlantic croaker (*Micropogon undulatus*), pinfish (*Lagodon rhomboides*), bay anchovy (*Anchoa mitchilli*) and Atlantic menhaden (*Brevoortia tyrannus*) collected off the coast of North Carolina were measured in relation to time (Cross and Brooks 1973). Excepting manganese in menhaden, the metals content of all these species decreased with age. This trend was attributed in part to the proportionate decrease in size with age of tissues which tend to accumulate the highest levels of these metals (e.g., gastrointestinal tract) and partly to the increased offshore migration tendencies of older fish.

During another study Cross et al. (1973) looked at the relationship between age and concentrations of copper, iron, manganese, mercury and zinc in bluefish (*Pomatomus saltatrix*), a pelagic fish, and morids (*Antimora rostrata*), a bathyl-demersal fish, collected off the North Carolina coast. For both species, only mercury concentration increased with age, suggesting that the other metals may be at equilibrium between the fish and their environment. Childs and Gaffke (1974) measured cadmium and lead levels in fillets from groundfish caught off the northern and southern Oregon coasts. In general fish contained less than 0.10 μg Cd g^{-1} and less than 0.20 μg Pb g^{-1}. Fish from the northern portion of the coast contained on the average slightly higher concentrations of both metals. Windom et al. (1973) compared arsenic, cadmium, copper, mercury and zinc levels in several inshore and offshore species of fish from the North Atlantic. No significant differences were found between fish from the two areas; however, cadmium, copper and zinc levels were inversely related to position in the aquatic food chain.

Patterson and Settle (1977) measured the distribution of several metals in albacore, *Thunnus alalunga*. Cesium, potassium and rubidium were uniformly distributed throughout the various organs and tissues but barium, calcium, lead and strontium were preferentially accumulated by bone. Comparing the distribution of these metals in tuna with their distribution in a terrestrial mammal, the martin (*Martes americana*), showed that the metals were distributed identically in both animals. Similarly, Goldberg (1962) examined the distribution of several metals in various Pacific tuna. General trends noted were that the transition elements (copper, manganese, nickel, zinc) concentrated in internal organs whereas alkaline earths (calcium, strontium) were concentrated in bony tissues. These trends are similar to those reported by other workers. Havre et al. (1972) measured concentrations of six metals in fish from a Norway fjord suspected to be contaminated from a zinc factory. Levels were found to be higher than in uncontaminated areas but not as high as was expected. It was suggested that zinc may inhibit the uptake of other metals. Greig et al. (1976) found that fish and invertebrates collected near a deep-water disposal site contained lower concentrations of cadmium, copper, nickel and zinc than inshore fish. Various marine fishes and algae from the Bay of Haifa off the Mediterranean coast of Israel contained only background levels of cadmium, chromium, copper, lead, nickel and zinc (Roth and Hornung 1977). Leatherland et al. (1973) found low levels of antimony, arsenic, cadmium, mercury and zinc in various fishes and invertebrates collected off the northwest coast of Africa and in the Azores; metals were generally present at higher concentrations in invertebrates than in fish. Rossi et al. (1976) analyzed canned tuna in Italy for metal content and then calculated the contribution of metals from tuna to the total metals consumed by the Italian citizenry. Based on the food habits of the average Italian and the metals content of other dietary constituents, it was determined that antimony, cesium, chromium, cobalt, iron, mercury, nickel, selenium and zinc in tuna did not contribute significantly to the total intake of these metals from other sources.

Watling and Watling (1976a) measured the concentrations of various metals including cadmium, copper, iron, manganese, nickel, silver and zinc

in three species of oysters (*Crassostrea gigas, C. margaritacea* and *Ostrea edulis*) collected from the Knysna estuary, South Africa. Comparisons of the metals concentrations found in these oysters with those observed in the same species from other estuaries led the authors to conclude that this estuary was relatively pristine. All three species of oysters had similar metals concentrations. The mean levels for each metal on a dry tissue basis were (in $\mu g\ g^{-1}$): Ag 1.9 to 6.4, Cd 2.5 to 3.7, Cu 17-38, Fe 57-167, Mn 2-16, Ni 1.6-1.7, Zn 396-886. The same authors (1976b) collected mussels (*Choromytilus meridionalis*) from Saldanha Bay, South Africa, and measured their metals content in relation to weight and sex. The absolute amount of metal increased as mussel weight increased. Interestingly, copper, iron, manganese and zinc were significantly higher in females than in males whereas bismuth and lead were present at higher concentrations in males. Darracott and Watling (1975) have suggested that a variety of molluscs could serve as metals pollution monitoring organisms for the various bays and estuaries of South Africa. These organisms were chosen on the basis of their relative abundance, longevity, size, sedentary nature and metals concentrating ability.

Thomas (1975) summarized available information on the role of benthos in transporting, mobilizing and accumulating metals. Conclusions were that (1) benthic fauna accumulate more metals from water than from sediment, (2) benthos metabolize and thereby change the chemical form of some metal compounds, and (3) benthic organisms themselves are not important sources of metals to fish but benthos may increase the metals content of overlying waters by disturbing metals concentrated in sediment. Sediment matrix is also important in determining the availability of metals to aquatic organisms. Luoma and Jenne (1976) studied the availability of silver, cobalt and zinc to the deposit-feeding marine clam (*Macoma balthica*) using various sediments including organic detritus, biogenic carbonates (crushed clam shells), synthetic calcium carbonates and iron or manganese oxides. Hydrous oxide-bound zinc and cobalt were not available to the clam but silver was readily accumulated in the presence of iron oxides. Conversely, organic complexes of zinc and cobalt increased the availability of these metals to the clam whereas silver accumulation was inhibited when the clams were placed in an organic sediment matrix. All three metals were readily accumulated from both synthetic and biogenic calcium carbonate type sediments. Rates of metals accumulation by the clam were directly related to the characteristic rate of metals desorption from a particular sediment type.

Martin (1970) collected copepods off the coast of Puerto Rico both near the surface and at 100 m depth. The concentrations of copper, iron, manganese, nickel, lead, strontium and zinc were higher in the deep samples than in the surface samples. Martin postulated that this was because the larger species collected in deep water were older and molted less often than the surface specimens because of the inverse relationship between food availability and depth. The sinking of molted copepod exoskeletons was considered to be an important factor in the biogeochemical cycling of metals in the world's oceans.

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APPENDIX

Index to the scientific and common names of organisms referred to in the text. Nomenclature is that used by the original author(s).

FISHES

Scientific Names

Acanthopagrus sp. 46

Alburnus sp. 29

Alosa aestivalis 29

Alosa pseudoharengus 68

Amia calva 42

Ambloplites rupestris 39

Anchoa mitchilli 71

Anguilla anguilla 49

Anoplopoma fimbria 45

Antimora rostrata 27, 34, 72

Bairdiella chrysura 45

Belone belone 15, 16

Brachydanio rerio 14

Brevoortia tyrannus 71

Brosme brosme 45

Carassius auratus 13, 23, 56, 58, 61, 62

Carassius carassius 44

Catostomus latipinnis 42

FISHES (continued)

Chasmichthys gulosus 65

Chrysophrys auratus 45

Clarias lazera 20, 24

Clupea harengus 15, 16

Coregonus clupeaformis 34

Coregonus lavaretus 24

Corydoras punctatus 14

Cyprinus carpio 29, 41, 42, 63, 71

Engraulis japonica 41

Esox lucius 24, 34, 40, 42, 43, 44

Esox niger 29

Euthynnus alletteratus 21

Fundulus heteroclitus 16, 27, 65, 66

Gadus morhua 64

Gambusia affinis 37, 38, 60

Gasterosteus aculeatus 14, 61

Girella tricuspidata 46

Gobius 63

Helicolenus dactylopterus 45

Hippoglossus stenolepis 45

Hypotremata sp. 44

Ictalurus catus 13

Ictalurus nebulosus 26, 60

Ictalurus punctatus 42, 51

Jordanella floridae 62

FISHES (continued)

Katsuwonus pelamis 30

Labidesthes sicculus 29

Lagodon rhomboides 71

Lebistes 59

Lebistes reticulatus 14

Leiostomus xanthurus 71

Lepomis cyanellus 9, 10

Lepomis gibbosus 15, 20, 30, 31, 63

Lepomis macrochirus 8, 14, 15, 26, 29, 56, 63

Makaira nigricans 46

Micropogon undulatus 21, 45, 65, 71

Micropterus dolomieui 42, 68

Micropterus salmoides 9, 15, 42, 56, 59

Microstomus pacificus 7

Molva molva 44

Morone chrysops 68

Myoxocephalus quadricornis 45

Neothunnus macropterus 7, 30

Noemacheilus barbatulus 13, 27

Notropis atherinoides 38

Notropis hudsonius 68

Oblata melamura 44

Oncorhynchus kisutch 62

Oncorhynchus nerka 12, 48

Oncorhynchus tshawytscha 61

FISHES (continued)

Oryzias latipes 39

Paralabrax clathratus 7, 25, 29, 34, 69

Paralichthys dentatus 21

Perca flavescens 39, 41, 68

Perca fluviatilis 24, 71

Percopsis omiscomaycus 68

Pimephales promelas 38, 39, 41, 47, 57

Platichthys flesus 15, 16, 45

Pleuronectes platessa 21, 34, 50, 52, 65

Pneumatophorus diego 59

Poecilia reticulata 11, 37, 38

Pomatomus saltatrix 21, 27, 34, 72

Pomoxis nigromaculatus 9, 42

Pseudopleuronectes americanus 15, 39

Raja clavata 21

Roccus chrysops 42

Rudarius ercodes 65

Salmo clarki 56, 61

Salmo gairdneri 8, 15, 22, 26, 30, 38, 39, 40, 43, 47, 48, 58, 59, 61

Salmo salar 34, 60, 70

Salmo trutta 21, 34, 43, 70, 71

Salvelinus alpinus 40

Salvelinus fontinalis 14, 26, 31, 39, 47, 61

Salvelinus namaycush 7, 22, 25, 29, 31, 34, 48, 68

Sebastes marinus 44

FISHES (continued)

Semotilus atromaculatus 54

Seriora quinqueradiata 41

Squalus acanthias 45

Stizostedion vitreum vitreum 40, 41, 42

Tautogolabrus adspersus 16

Thunnus alalunga 30, 72

Thunnus obesus 44

Thunnus thynnus 44

Thymallus thymallus 43

Umbrina cirrhosa 44, 45

Xiphias gladius 44

Common Names

albacore--see *Thunnus alalunga*

alewife--see *Alosa pseudoharengus*

anchovy--see *Engraulis japonica*

arctic char--see *Salvelinus alpinus*

Atlantic cod--see *Gadus morhua*

Atlantic croaker--see *Micropogon undulatus*

Atlantic menhaden--see *Brevoortia tyrannus*

Atlantic salmon--see *Salmo salar*

bay anchovy--see *Anchoa mitchilli*

bigeye tuna--see *Thunnus obesus*

blackbellied redfish--see *Helicolenus dactylopterus*

black crappie--see *Pomoxis nigromaculatus*

FISHES (continued)

blackfish--see *Girella tricuspidata*

blueback herring--see *Alosa aestivalis*

bluefin tuna--see *Thunnus thynnus*

bluefish--see *Pomatomus saltatrix*

bluegill--see *Lepomis macrochirus*

blue marlin--see *Makaira nigricans*

bream--see *Acanthopagrus* sp.

brook silverside--see *Labidesthes sicculus*

brook trout--see *Salvelinus fontinalis*

brown bullhead--see *Ictalurus nebulosus*

brown trout--see *Salmo trutta*

calico bass--see *Paralabrax clathratus* and *Pomoxis nigromaculatus*

carp--see *Cyprinus carpio*

catfish--see *Corydoras punctatus*

chain pickerel--see *Esox niger*

channel catfish--see *Ictalurus punctatus*

chinook salmon--see *Oncorhynchus tshawytscha*

coho salmon--see *Oncorhynchus kisutch*

creek chub--see *Semotilus atromaculatus*

crucian carp--see *Carassius carassius*

cunner--see *Tautogolabrus adspersus*

cusk--see *Brosme brosme*

cutthroat trout--see *Salmo clarki*

dover sole--see *Microstomus pacificus*

emerald shiner--see *Notropis atherinoides*

FISHES (continued)

European eel--see *Anguilla anguilla*

fathead minnow--see *Pimephales promelas*

filefish--see *Rudarius ercodes*

flagfish--see *Jordanella floridae*

flannelmouth sucker--see *Catostomus latipinnis*

fourhorn sculpin--see *Myoxocephalus quadricornis*

garpike--see *Belone belone*

goldfish--see *Carassius auratus*

grayling--see *Thymallus thymallus*

green sunfish--see *Lepomis cyanellus*

guppy--see *Poecilia reticulata, Lebistes,* and *Lebistes reticulatus*

halibut--see *Hippoglossus stenolepis*

herring--see *Clupea harengus*

Japanese medaka--see *Oryzias latipes*

lake trout--see *Salvelinus namaycush*

lake whitefish--see *Coregonus clupeaformis*

largemouth bass--see *Micropterus salmoides*

ling--see *Molva molva*

little tuna--see *Euthynnus alletteratus*

marine goby--see *Chasmichthys gulosus*

morid--see *Antimora rostrata*

mosquitofish--see *Gambusia affinis*

mudfish--see *Amia calva*

mummichog--see *Fundulus heteroclitus*

Nile catfish--see *Clarias lazera*

FISHES (continued)

northern pike--see *Esox lucius*

Pacific mackerel--see *Pneumatophorus diego*

Pacific sablefish--see *Anoplopoma fimbria*

perch--see *Perca fluviatilis*

pike--see *Esox lucius*

pinfish--see *Lagodon rhomboides*

plaice--see *Pleuronectes platessa*

pumpkinseed sunfish--see *Lepomis gibbosus*

rainbow trout--see *Salmo gairdneri*

red-fish--see *Sebastes marinus*

rock bass--see *Ambloplites rupestris*

silver perch--see *Bairdiella chrysura*

skipjack--see *Katsuwonus pelamis*

smallmouth bass--see *Micropterus dolomieui*

snapper--see *Chrysophrys auratus*

sockeye salmon--see *Oncorhynchus nerka*

spiny dogfish--see *Squalus acanthias*

spot--see *Leiostomus xanthurus*

spottail shiner--see *Notropis hudsonius*

stone loach--see *Noemacheilus barbatulus*

three-spined stickleback--see *Gasterosteus aculeatus*

summer flounder--see *Paralichthys dentatus*

thornback ray--see *Raja clavata*

trout-perch--see *Percopsis omiscomaycus*

walleye--see *Stizostedion vitreum vitreum*

FISHES (continued)

white bass--see *Morone chrysops*

white catfish--see *Ictalurus catus*

whitefish--see *Coregonus lavaretus*

winter flounder--see *Pseudopleuronectes americanus*

yellow perch--see *Perca flavescens*

yellowfin tuna--see *Neothunnus macropterus*

yellowtail--see *Seriora quinqueradiata*

zebrafish--see *Brachydanio rerio*

INVERTEBRATES

Scientific Names

Ampullaria paludosa 14

Anodonta grandis 38

Artemia salina 63

Aquipecten irradians 16, 64

Asellus meridianus 27, 31

Astacus sp. 53

Austropotamobius pallipes pallipes 63

Cambarus bartoni bartoni 52

Cambarus longulus longerostris 58

Cambarus robustus 52

Cambarus rusticus 52

Cancer magister 9, 10

Cancer pagurus 50

Cancer productus 53

INVERTEBRATES (continued)

Carcinus maenas 17, 24, 63.

Choromytilus meridionalis 73

Crangon crangon 25, 65

Crassostrea commercialis 46

Crassostrea gigas 12, 25, 64, 73

Crassostrea margaritacea 73

Crassostrea virginica 16, 17, 23, 25, 28, 29, 32, 34, 49, 64, 65

Daphnia magna 41, 56

Ensis ensis 29

Ensis sp. 25

Ephemerella grandis 27

Euphausia pacifica 64

Gammarus zaddachi 32

Haliotis cracherodii 28

Haliotis ruber 46

Haliotis rufescens 18, 28, 32, 57

Harmothoë sarsi 32

Helix aspersa 27

Hermione hystrix 23

Homarus americanus 16, 17

Homarus vulgaris 51, 64

Lampsilis siliquoidea rosacea 70

Ligumia sp. 56

Littorina irrorata 65

Littorina littorea 35, 70, 71

INVERTEBRATES (continued)

Loligo opalescens 27

Lysmata seticaudata 17, 51, 55, 63

Macoma balthica 73

Margaritifera sp. 56, 57

Meganyctiphanes norvegica 55

Mercenaria mercenaria 64

Meretrix meretrix lusoria 52

Mesidotae entonom 32

Mulinia lateralis 17

Mya arenaria 17

Mytilus californianus 27, 32, 46

Mytilus edulis 14, 17, 27, 32, 49, 52, 65

Mytilus galloprovincialis 17, 51, 55

Mytilus sp. 25, 32

Nereis 65

Nereis virens 23, 28

Nucula proxima 17

Ommastrephes bartrami 27

Orconectes obscurus 52

Orconectes rusticus rusticus 52

Ostrea edulis 73

Pecten irradians 64

Polypus bimaculatus 27

Pseudanodonta complanata 38

Pteronarcella badia 13

INVERTEBRATES (continued)

Pteronarcys californica 13, 27

Salpa sp. 64

Scrobicularia plana 24

Strongylocentrotus pulcherrimus 65

Symplectoteuthis oualaniensis 27

Tapes japonica 65

Thais lapillus 35, 70, 71

Tubifex sp. 27, 35, 69, 70

Uca pugilator 17

Unio cfr. elongatulus 43

Venus mercenaria 64

Viviparus ater 63

Common Names

American lobster--see *Homarus americanus*

American eastern oyster--see *Crassostrea virginica*

American oyster--see *Crassostrea virginica*

bay scallop--see *Pecten irradians* and *Aquipecten irradians*

benthic shrimp--see *Lysmata seticaudata*

black abalone--see *Haliotis cracherodii*

blacklip abalone--see *Haliotis ruber*

common Mediterranean mussel--see *Mytilus galloprovincialis*

common shrimp--see *Crangon crangon*

common terrestrial snail--see *Helix aspersa*

dungeness crab--see *Cancer magister*

INVERTEBRATES (continued)

fiddler crab--see *Uca pugilator*

hard-shell clam--see *Venus mercenaria*

infusoria snail--see *Ampullaria paludosa*

Pacific oyster--see *Crassostrea gigas*

red abalone--see *Haliotis rufescens*

sea mussel--see *Mytilus californianus*

shore crab--see *Carcinus maenas*

short-necked clam--see *Tapes japonica*

Sydney rock oyster--see *Crassostrea commercialis*

tubificid worms--see *Tubifex* sp.

MAMMALS

Scientific Names

Alopex lagopus 40

Canis familiaris 40

Canis lupus 40

Erignathus barbatus 40

Lutra canadensis 42

Martes americana 72

Mustela vison 40, 42

Phoca hispida hispida 40

Rangifer tarandus 40

Rattus rattus 24

MAMMALS (continued)

Common Names

arctic fox--see *Alopex lagopus*

bearded seal--see *Erignathus barbatus*

caribou--see *Rangifer tarandus*

martin--see *Martes americana*

mink--see *Mustela vison*

otter--see *Lutra canadensis*

rat--see *Rattus rattus*

ringed seal--see *Phoca hispida hispida*

sledge dog--see *Canis familiaris*

wolf--see *Canis lupus*

AMPHIBIANS

Rana esculenta 71

Rana temporaria 71

ALGAE

Egregia laevigata 32

Navicula sp. 24

Scenedesmus sp. 56

BACTERIA

Pseudomonas fluorescens 37

Pseudomonas sp. 37