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ECOLOGICAL EFFECTS OF CADMIUM POLLUTION IN THE AQUATIC ENVIRONMENT: A REVIEW

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ABSTRACT

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Cadmium is known to be an extremely toxic accumulative poison and has been defined as a stock pollutant. Uncontaminated surface waters may contain an average of less than $1.0~\mu g$ Cd/kg, while concentrations in contaminated waters may reach a value several magnitudes higher. Many aquatic species, especially fish and shellfish, have the ability to concentrate cadmium in their bodies above ambient levels, with long-term effects which are not yet fully determined.

This paper reviews the current literature with reference to the occurrence, uses, properties and effects of cadmium on aquatic biota, with emphasis on fish and shellfish. The limited data available on chemistry and toxicity of cadmium compounds in ecosystems prevent establishment of scientifically valid standards for protection of aquatic life.

Key words: Cadmium, heavy metals, pollution, toxicity, aquatic environment, ecological effects, review.

RESUME

Ray, S. and J. Coffin. 1977. Ecological effects of cadmium pollution in the aquatic environment: a review. Fish. Mar. Serv. Tech. Rep. No. 734: 24 p.

Le cadmium est reconnu comme extrêmement toxique et cumulatif dans les tissus vivants. Sa concentration dans les eaux non contaminées peut être de moins de 1.0 $\mu g/kg$ en moyenne tandis qu'elle peut être beaucoup plus élevée dans les eaux contaminées. Beaucoup d'espèces aquatiques, et plus spécialement les poissons, les mollusques et les crustacés ont la propriété de concentrer le cadmium dans leurs tissus jusqu'à des niveaux bien supérieurs à celui du milieu ambiant. Les consequences à long terme de ce phénomène ne sont pas encore entièrement connues.

Cette publication passe en revue la litérature courante qui traite de la présence, des utilisations, des propriétés et des effets du cadmium sur la biocénose aquatique, particulièrement sur les poissons, les mollusques et les crustacés. La quantité restreinte de données sur la chimie et la toxicité du cadmium dans les écosystèmes prévient l'éstablissement de normes scientifiques valides pour la protection de la vie aquatique.

INTRODUCTION

The role of heavy metals in the biosphere is complex and not well understood. It is certain that various heavy metals such as zinc, copper and cobalt in trace amounts are absolutely necessary to life processes; it is also well known that other trace elements such as arsenic, cadmium, mercury and lead are not only dispensible but toxic to the living organism.

The fate of trace elements in the environment, be it concentration, maintenance at a constant value or discrimination at each of the various trophic levels, is governed by the species of aquatic life present and the metabolic behaviour of the individual element (1). Geochemical properties of the environment may also affect the concentration of trace elements in plant materials and/or aquatic animals. Thus, many influencing factors in the biochemical, environmental and genetic realms interact to determine the concentration of a particular element in the biosphere.

Cadmium is one of the heavy metals of current interest in environmental contamination studies since the appearance in Japan of "itai-itai" disease, or acute human cadmiosis. Due to the common occurrence of this trace metal in industrial effluents, its extreme toxic properties to man and aquatic organisms, and its existence in natural waters as a potential pollutant, cadmium has caused increasing alarm. Concern is again generated by the similiarity of cadmium to zinc, which enables cadmium to replace zinc in its essential roles in the living system (2). Moreover, while there is a rapid turnover of zinc, cadmium is strongly bound to proteins.

Cadmium shows no indication of being an essential trace element in biological processes; on the contrary, it is highly toxic to a wide variety of living organisms, including man (3). It is considered hazardous to health at concentrations above 10 ppb (4) in the drinking water supply. Schweiger (5) determined that lethal concentrations of cadmium in water for several species of fish ranged from 4 to 20 mg/liter, depending on many factors. Effects on growth rate have been observed for concentrations between 5 and 10 ppb (6). Cadmium is especially accumulated by shellfish (7), and has been found in fish tissue of numerous species in natural concentrations from 10 ppb to greater than 100 ppb (8, 9).

OCCURRENCE IN NATURE

Cadmium ranks as the 67th element in order of abundance in the earth's crust (10), which has an average content of about 0.15 ppm (11). Cadmium minerals are scarce. As a result of its chemical similiarity to zinc, cadmium occurs by isomorphous replacement in almost all zinc ores. The most common occurrence of

cadmium is as the sulphide greenockite, which is found associated with zinc ores, especially sphalerite. Zinc ores contain from 0.001% to 0.067% of recoverable cadmium, and zinc concentrates contain as much as 0.7% cadmium (12). Cadmium is also found associated with the zinc minerals ZnS, calamine and ZnCO3. While cadmium is predominantly associated with zinc minerals, it is also found in sulphide deposits of lead and copper.

USES

Potential environmental pollution occurs wherever cadmium metal and/or its alloys and compounds are used. Several sources are responsible for cadmium in the aquatic environment: cadmium electroplating processes—which use approximately 55% of the cadmium produced per year in the U.S., long-life battery industries, and runoff from agricultural areas treated with phosphate (≈1.8 ppm cadmium) and superphosphate (approx. 9 ppm cadmium) fertilizer (13).

Cadmium electroplating gives an attractive, corrosion-resistant finish to ferrous metals; and is widely used in automobile manufacture, household appliances, aircraft, radios, television sets, and electrical equipment. Cadmium coatings have the advantage of needing less film thickness for rust protection than zinc or nickel, and do not chip off when plated objects are subjected to deformation—though they are not very resistant to mechanical abrasion. Other characteristics which make cadmium an appropriate coating are its high ductility, good solderability and good corrosion resistance to alkali and salt water. Cadmium coatings also have a high rate of deposition with an ease of application, and cadmium-coated objects retain the silver-white lustre for extended periods (12).

Other principal uses of cadmium in industry are in paint pigments, and as stabilizing material for polyvinyl plastics, batteries and alloys. Cadmium chemicals, especially pigments and nickel-cadmium alloys, account for 7% of total use. Cadmium alloys are used in bearings, solders, low-melting alloys and silver brazing. Some minor uses of cadmium are in the manufacture of television picture tubes, phosphors, fungicides, control rods and shields in nuclear reactors, curing agents for rubber, ceramics, motor oils, dental amalgams and piano wires, and for protection of ship's gear.

Numerous cadmium compounds are also widely employed in industry; viz, compounds with acetate, bromide, carbonate, chloride, flouride, cyanide, nitrate, octoate, sulphate, salicylate, stearate, sulphoselenide, tungstate, benzoate, ferrocyanide, formate, laurate, oxalate, propionate, tartarate, and zirconates. The uses of these compounds are many and varied; viz, chemical testing for sulphides, dyeing, printing textiles, photography, catalysts, external antiseptics, and in x-ray screens, etc. A

comprehensive list of the cadmium compounds used in industry has been compiled (12).

The extensive use of cadmium in industry and the degree of its toxicity indeed pose an environmental problem.

CHEMISTRY

Cadmium is a soft, ductile metal, with an atomic number of 48 and an atomic weight of 112.40. It is a transition metal in Group IIb of the periodic table of elements, and shows a valence of +2. The covalent radius is 1.48Å (10). Cadmium is a bluish-white to silver-white metal, with a density of 8.642, melting point of 321°C, and boiling point of 765°C. The vapor pressure is 1.0 mm at 394°C and 10 mm at 484°C. The standard potential of cadmium is -0.402 (25°C) relative to the normal hydrogen electrode.

Some properties of cadmium include: insolubility in water, non-reactivity with bases, and direct reactivity with halogens and non-metals such as sulphur and phosphorous. Cadmium reacts slowly with organic acids but readily with mineral acids.

The chemical properties of zinc and cadmium are very similar.

	Chen	nical parame	
Ion	"d"	Config.	C.N.
Zn	d10	sp³	4
Cđ	d10	sp³	4

Both are electropositive with a valence of +2 in their compounds, and they are also generally similar in reactivity. Zinc, like cadmium, is a transition metal in Group IIb. Both have completed "d" shells; both react readily with non-oxidizing acids, releasing hydrogen; and both react readily when heated with oxygen, to yield oxides.

It is this similiarity of cadmium and zinc which causes alarm. Zinc is an essential trace element necessary for the function of many common and obligatory enzymes. Cadmium, however, due to its likeness, can replace zinc on the active sites of these enzymes, inhibiting their action. Moreover, while cadmium can replace zinc, the reverse is not possible (2), probably due to the strength of the cadmium-protein bond.

ANALYSIS

Analysis of trace metals is difficult and presents many complications, especially in a biological matrix.

A number of methods have been used to determine trace amounts of cadmium:

- (1) atomic absorption spectroscopy,
- (2) neutron and photon activation analysis,
- (3) anodic stripping voltametry,
- (4) differential pulse polarography,
- (5) spectrophotometric colorimetry,
- (6) isotope dilution,
- (7) spark source mass spectrometry,
- (8) emission spectroscopy,
- (9) specific ion electrode.

Atomic absorption spectroscopy, due to its characteristic speed, similicity and sensitivity, is now the instrument of choice.

ATOMIC ABSORPTION

In atomic absorption spectroscopy, the element of interest is aspirated to a high temperature flame, where it is dissociated from its chemical bonds to an unexcited and unionized ground state level. In this atomized state, it is capable of absorbing radiation at discreet lines of narrow band width. The narrow emission line is provided by a hollow cathode lamp source, which has a cathode made of the same element filled with neon or argon at low pressure. The element concentration is measured as a function of absorbance.

For analysis of trace amounts of cadmium, flame atomic absorption spectroscopy usually employs a pre-concentration step to increase sensitivity and eliminate interferences. However, this technique still poses problems with background effects, low sensitivity and the requirement for a relatively large sample, and often requires sample pre-treatment and concentration steps.

Recent modifications of the atomic absorption technique have offered advantages to the detection of Cd and trace metals in general. These methods—graphite furnace, tantalum ribbon and carbon rod atomizer—fall under the general term "flameless atomic absorption (spectroscopy". When flameless devices are used, smoke from residual organic materials or salts can enter the sample beam, resulting in severe non-specific background absorption, which can, however, be compensated for by use of a background corrector (14).

The graphite furnace has been used extensively as a modification of the conventional flame atomic absorption spectroscopy for the determination of elements at the $\mu g/1$ level. This method allows a marked increase in sensitivity for most elements, offers the advantage of tracemetal determination and avoids problems of pre-concentration. Use of the graphite furnace has increased the detection limit to 0.000003 $\mu g/ml$ from the 0.002 $\mu g/ml$ capability of the usual flame technique (15).

Occasionally, analytical difficulties are encountered with the use of the graphite furnace. These technical problems are due

to the presence of a relatively non-volatile matrix component or to a very volatile analyte. However, certain chemical manipulations (matrix modifications) have been employed to allow analysis of these difficult samples. Matrix modifications either decrease the volatility of the analyte to prevent its volatilization during charring, or increase the volatility of the matrix to promote its removal before atomization, and therefore overcome the unusual matrix difficulties (16).

The tantalum ribbon (17) has also been successfully used as a modification to regular flame atomic absorption spectroscopy. Use of this technique with routine background correction leads to a slight loss in precision for trace-metal detection. However, the increased convenience of use, freedom from absorption and freedom from cross-contamination, were judged to be well worth the small loss in relative precision.

Another modification of regular atomic absorption used for trace-metal analysis is the carbon rod atomizer (18). This method, too, poses experimental complications, for it restricts the maximum size of the sample which can be analyzed. Another problem is that light scattering is experienced due to evaporation from the heated graphite surface into the cool inert gas atmosphere.

The sampling boat system has also been used for determination of cadmium in fish tissue by standard addition method (19).

SAMPLE PREPARATION

There are two main methods of sample preparation for analysis—wet acid digestion and dry ashing. The procedures have been evaluated and found comparable (20).

Both methods offer individual advantages and thus a quandary has arisen concerning the most desirable technique. Wet acid digestion helps to prevent the possible metal loss due to volatilization encountered in dry ashing. The latter also has the disadvantage of possible metal loss due to retention on the crucible. The difficulties encountered in wet ashing are: possible non-homogeneity due to small sample size, incomplete recovery of organically bound cadmium and constant surveillance required during digestion.

Extreme caution must be exercised to avoid contamination of all glassware and reagents. It is also imperative that high purity acids be used for digestion.

DISTRIBUTION IN ORGANISMS

MAN

Scientific evidence indicates that cadmium is toxic to humans (21, 22, 23, 24, 25). The average daily intake of cadmium by humans is approximately 4-60 µg, depending on the foods chosen (24). The body burden of cadmium in an adult is

estimated to be about 30 mg; in the newborn it is 1 μ g, indicating that there is an increase in accumulation with age. Of the total body burden, 50%-75% is in the liver and kidney, with 1/3 in the kidney (25). This accumulation of cadmium in human tissues, especially the liver and kidney, was linked to "itai-itai" disease in Japan and has also been linked to hypertension (26).

FISH

Cadmium is primarily ingested in the aquatic environment due to activities of man and is a cumulative poison. The U.S. Environmental Protection Agency recommends safe levels for crustaceans or the eggs and larvae of salmon should not exceed "0.003 mg/l in hard water or 0.0004 mg/l in soft water at any time or place" (27). An adult organism may accumulate a quantity of metal that does not cause death but may be deleterious to some other stage of the life cycle. Studies have shown that freshwater fish can concentrate cadmium to levels 10-1,000 times higher than levels in ambient water (25).

In all studies of the cadmium level in freshwater fish, the greatest accumulation occurred in the kidney; however, it is not clear whether accumulation level is directly proportional to exposure level. Mount and Stephan (28) found significant accumulation of cadmium in the kidney, liver, gill and gut; but not in bone or muscle of surviving exposed bluegill, Lepomis macrochirus. Eaton (29) observed similar results in bluegill; but an increase in exposure level showed increased residue level only in liver, intestine, and caecum, but not kidney. Cearley and Coleman (30) observed greatest accumulation in the internal organs, and the metal level increased with the exposure concen-Smith et al. (31) found that in tration. the catfish, Ictalurus punctatus, an increase in cadmium exposure led to an increased level in the kidney. The greatest biological magnification (41x) occurred at the lowest (50 ppb) cadmium exposure level, but a magnification of only 5x occurred at the highest (800 ppb) cadmium level.

The liver also accumulates cadmium to a high degree; however, the accumulation is normally less than in the kidney (32). With the liver, the direct correlation between cadmium exposure level and accumulation has been confirmed (31), with maximum biological magnification at the lowest exposure level.

Lovett et al. (7)—in a survey of 406 fish of 29 species from New York State—found 68.5% of the fish contained 20 ppb of cadmium or less; and, with few exceptions, the rest contained up to 100 ppb. Cadmium accumulation only occasionally appeared species-dependent. No relation was obvious between total residues of the metal and size, sex or age of the fish. All species from only one stream

contained cadmium at consistently high levels, reflecting the high background level in the water.

Studies of cadmium concentrations in whole fish and fish liver samples from the Great Lakes were performed by Lucas et al. (9). In 19 whole fish from 3 species, the cadmium level was 94 ppb; while in 40 liver samples from 10 species, cadmium levels ranged from 0.6 to 1.4 ppm, again indicating accumulation of metal in the liver. The concentration of cadmium in whole fish samples varied according to species and lake sampled. The values also varied among individual fish of the same species. Chemical pollution or variations in habitat have been speculated as causes of this variability. Concentrations were lowest in Lake Michigan smelt, whitefish and lake trout, and were highest in Lake Erie goldfish. Cadmium concentrations were higher in lake whitefish and possibly in bloaters and lake trout from Lake Superior than in samples of those species from Lake Michigan. Uthe and Bligh (33) found cadmium concentrations in selected Canadian freshwater fish (whitefish, pike, rainbow smelt and yellow perch) all below 0.05 ppm (wet weight).

A study by Cearley and Coleman (30) indicated that bass and bluegill accumulated cadmium in concentrations greater than those of the exposure water. Metal accumulation increased with the exposure concentration. Maximum total body accumulations by the bass were from 8-fold (0.008 mg Cd/l exposure) to 15-fold (0.08 mg Cd/l exposure) greater than by the controls; maximum accumulations by the bluegill were 6-, 20- and 210-fold greater (at exposures of 0.008, 0.08 and 0.85 mg Cd/1, respectively) than by the controls. Portmann (34) studied heavy-metal levels in fish from coastal waters around England and Wales. The mean concentration of cadmium in deep-water fish was less than 0.05 ppm, although concentrations up to 0.15 ppm were found in individual fish. The mean concentrations of cadmium in fish from the North and Irish seas were 0.12 and 0.07ppm, respectively. Cod contained slightly more cadmium than plaice.

Fish species from several waters of the Northwest Territories were analyzed for cadmium content (35). Metal concentrations in liver samples of northern pike and whitefish were 0.70 and 0.21 ppm, respectively, in one location, and 0.83 and 0.10 ppm in another. Cadmium concentrations of trace amount to 0.10 ppm were found in the muscle tissue of whitefish, northern pike, walleye, sucker, lake trout and cisco from the same area.

Cadmium concentrations were determined in five organs of Atlantic salmon, Salmo salar, grilse and parr from the Miramichi River in New Brunswick, and were compared to hatchery-parr samples (36) (Table 1). Generally higher levels of cadmium were found in the parr-sample organs, irrespective of the source. Kidney and liver contained most, with kidney levels about 4-5 times greater than those of the liver, excepting in wild parr. Cadmium concentration in liver samples from wild parr was 4 times greater than in samples from hatchery parr. Cadmium concentration in grilse muscle was extremely low.

Lake Erie yellow perch, Perca flavescens; white bass, Morone chrysops; and smallmouth bass, Micropterus dolomieui, had generally low cadmium content, in the range of 0.039-0.063, 0.007-0.034 and 0.043-0.047 µg/g, respectively (37). Tong (38) found very low concentrations of cadmium (2.1-6.3 ppb fresh weight) in whole tissue of lake trout, Salvelinus namaycush, and did not observe any relation to the age of the fish. Striped bass, Morone saxatilis, from Annapolis River, Nova Scotia, (39) had mean cadmium concentrations in muscle and liver of 0.08 ppm (range 0.04-0.10) and 1.4 ppm (range 0.6-2.7 ppm), respectively, on a dry-weight basis.

Benoit et al. (40) determined the cadmium distribution in different organs of three generations of brook trout, Salvelinus fontinalis, exposed to several concentration levels. Cadmium residues were greatest in kidney, liver and gill tissues; and were directly related to exposure concentrations in the test water. In the first generation, the residue level attained equilibrium after 20 weeks exposure at 3.4 μ g/1; reached values of about 50 and 10 μ g/g in kidney and liver, respectively; and did not undergo any change for the duration of the experiment (38 weeks). Levels in muscle were extremely low and no increase was noted in the red blood cells. Mean residue levels in the second generation after 70 weeks of

TABLE 1. Cadmium concentration in Atlantic salmon (ng/g dry wt).

	Mirami	chi grilse	Mirami	chi parr	Hatchery parr			
Organ	Average	Range	Average	Range	Average	Range		
Muscle	0.79	0.28-1.36	423	30-1,031	74	54-112		
Gill	122	74-194	546	143-1,743	562	223-956		
Spine	18	3-33	96	56-152	83	58-106		
Kidney	2,037	1,309-3,407	1,380	699-2,075	1,261	601-2,509		
Liver	386	135-707	1,392	512-3,683	304	105-497		

exposure at the same concentration were kidney, 64.8; liver, 9.0; gill, 5.1; and muscle, 0.05 $\mu g/g$; and compared closely with the first generation fish after 38 weeks.

While the majority of current literature focuses on cadmium content of freshwater organisms, a large number of marine species has been analyzed as well. Analysis of 91 individuals representing 35 species of North Atlantic Chondrichthys and Osteichthys (41) indicated that cadmium concentrations were similar among Osteichthys tissues and organs, with the exception of smaller planktivorous fishes such as anchovies and myctophids. These fish, occupying a lower trophic level, have much higher cadmium concentration than the other fish studied. This suggested a depletion of cadmium up the food chain. In the Chondrichthys studied, most tissues had similar cadmium concentrations excepting the liver, which had a higher concentration.

Brooks and Rumsey (42) analyzed eight species of commercial seafish from New Zealand and found the edible parts to have 0.002-0.02 ppm; and, of the eight other organs analyzed, the liver showed the highest concentration, up to 12.15 ppm. In the northern Adriatic anchovey, Engraulis encrasicholus; and sardine, Sardina pilchardus, concentrations of cadmium in various tissues ranged from <0.1 to 1.4 ppm wet weight. No significant difference was observed between the two species (43). Hardisty et al. (44) noted distinct correlation between the cadmium concentration of tissues and the proportion of crustaceans in the diet of estuarine fish. Zook et al. (45) analyzed several species of ocean fish and found a mean cadmium content of 0.068 ppm. Eisler et al. (46) observed a measurable accumulation of cadmium in Fundulus heteroclitus when exposed to salt water for three weeks. Arctic cod, Boreogadus saida, has <0.5 and 0.68 ppm in fillet and liver, respectively (47).

AQUATIC INVERTEBRATES

Shellfish display the capacity of selectively concentrating trace metals up to many hundreds of times those levels found in their environment. Accumulation occurs through several pathways, chiefly by ingestion of particulate materials and by absorption. Aquatic organisms may also have the capacity to control uptake and/or excretion of cadmium in the body independent of concentration in water. In a comparative study, Eisler et al. (46) found that exposure to 10 µg Cd/l for 21 days led to an increase from 0.33 to 1.49 ppm cadmium in oyster, Crassostrea virginica, and from 0.33 to 0.48 ppm in the fish Fundulus heteroclitus. Seasonal variation of trace-metal levels has been observed (48). Cadmium concentrations have been related to the trace metals entering estuarine or coastal areas from industrial areas and effluents. Ratkowsky et al.

(49) found a close correlation between proximity to heavily urbanized areas in Tasmania and the concentration of metals in the oysters Ostrea angasi, Crassostrea gigas and C. commercialis—those growing near urban areas having the highest concentrations.

Many data have been compiled on cadmium levels in shellfish. The American eastern oyster appears to have a great propensity for trace-metal uptake (50, 51), particularly cadmium, along with copper and zinc (Table 2).

TABLE 2. Cd content in molluscs (48).

Species	ppm	(wet	weight	basis)
Pacific oyster		0.8	8-1.4	
Eastern U.S. oyster		0.3	1-7.8	
Northern quahaug		0.3	1-0.7	
Soft shell clam		0.3	1-0.9	
1				

On exposure to 0.1 ppm cadmium, they may accumulate up to 122 ppm in a 20-week period, and the maximum level attained by the oysters before their death may be several hundred ppm.

Oysters in Mobile Bay, Alabama, were shown to contain less cadmium than the average concentrations reported for Atlantic coast oysters (52). Huggett et al. (51) found concentrations of cadmium in oysters, Crassostrea virginica, in the Chesapeake Bay to be a function of proximity to the source of pollution and of the animal's position in the estuary. A recent survey of heavy metals in Pacific oysters, Crassostrea gigas, showed that cadmium may be present at concentrations as high as 63 ppm (wet weight) in contaminated areas (53) and was correlated with metal concentration of the sediment.

Other molluscs have also been studied in relation to cadmium content. Boyden (54), in a study of trace-metal relationship to mollusc body size, has documented that cadmium concentration is directly related to the body weight of the mussel Mytilus edulis and is related to the square of the body weight of the limpet Patella vulgata. Further values (55) found in New Zealand bivalve soft tissues are: scallop, Pectenovae zelandiae, 249; oyster, Ostrea sinuata, 35; and mussel, Mytilus edulis acteanus, <10 ppm dry weight—with enrichment factors of 2,260,000, 318,000 and 100,000, respectively.

Figures cited (56) for levels of cadmium in Gastropoda (snails and slugs) are widely different, 0.69-178 ppm dry weight.

Parsons et al. (57) found that molluscs and the crab Cancer magister, analyzed from the Sturgeon Bank of the Fraser River mudflats, contained larger amounts of cadmium than did the same species from similar environments on the coast of British Columbia. Various other crustaceans have also been studied and values of cadmium content recorded. Shrimp and lobster on a dry-weight basis have respective cadmium levels of 0.10 and 0.12 ppm (25).

Eisler et al. (46) showed that all species analyzed (oyster, Crassostrea virginica; scallop, Aequipecten irradians; lobster, Homarus americanus) accumulated measurable amounts of cadmium after three weeks in sea water containing 10 ppb of cadmium. Cadmium content in the experimental group was consistently higher than in the controls (Table 3).

TABLE 3. Uptake of cadmium by marine organisms.

mg Cd/kg wet wt					
Experimental	Control				
1.49	0.33				
2.46	1.15				
0.72	0.51				
	1.49 2.46				

Several zooplankton from Arctic cod habitat in Strathacona Sound (47) were found to have from 1.2 ppm (chaetognaths) to 7.0 ppm (amphipods) cadmium.

A preliminary survey (45) of American seafood showed Long Island oysters had a cadmium concentration of 2.06 ppm, whereas Maryland and Virginia oysters had only 0.61 ppm. Calieo scallops had 2.34 ppm. Edible parts of six other species (clamsboth hard, Mercenaria mercenaria and soft shell, Mya arenaria; lobster, Panulirus borealis; blue crab, Callinectes sapidus; king crab, Paralithodes camschatica; and sea scallops, Placopecten magellanicus) had average values of more than 0.10 ppm cadmium. Zaroogian and Cheer (58) reported that adult American oysters reared in seawater containing 5 ppb cadmium accumulated up to 10.75 ppm in 40 weeks, and the rate of accumulation in summer was more than double that of winter and spring. Naturally high levels of cadmium (up to 9 mg/g, wet wt) were observed in dredge oysters, Ostrea lutaria, from New Zealand (59).

Hutcheson (60) found rates of uptake of cadmium by the blue crab, Callinectes sapidus, to be greatest at low salinities and high temperatures. Benayoun et al. (61) showed that cadmium can be accumulated either directly from water or through the food chain, and flux of cadmium through the tissue of the euphausiid Meganychtiphanes norvegica was a relatively slow process—suggesting that ingestion of cadmium was

probably the more important route of accumulation. Cadmium incorporated in the limpet Patella vulgata was shown to be associated with proteins of low molecular weight (62). Casterline and Yip (63) reported that most cadmium in the oyster Ostrea edulis was bound to proteins of 9,200-13,800 molecular weight. In a study by Bender (64) of beach dipterans and amphipods collected from four locations in California, it was noted that dipterans (beach flies) showed consistently higher levels (up to 149.7 ppm) of cadmium than did amphipods.

AQUATIC PLANTS

In two floating aquatic plants, Lemna valdiviana and Salvinia natans, concentration factors of up to 9,500 (0.01-0.1 ppm cadmium) were observed (65).

Cadmium accumulation (\approx 1,000-fold) in pondweed, Najas quadulepensis, was shown to be a direct function of exposure level (66). Exposure for 21 days at a concentration of 7.0 ppb resulted in accumulation of 60.4 μ g/g ash; and, at 90 and 830 ppb, the concentration levels obtained were 4,357 and 5,429 μ g/g ash, respectively.

Water quality appeared to be an influential factor in determining cadmium behavior in freshwater ecosystems. The initial rates of cadmium uptake by the alga Nitella flexilis and the rooted plant Elodea canadensis were faster in hard water than in soft water. However, the total cadmium concentrations of the organisms were greater in soft water than in hard water, and hardness influenced both rate of uptake and total residues of cadmium (67). Witkamp et al. (68), working with 115Cd, observed a very rapid absorption (5-15 min), with a concentration factor of approximately 1,000 times by algae (species unknown). Bowen (69) has suggested that the predominant factor governing concentration level within the algae is the prevailing dissolved concentration of the element. Fucus vesiculosus and Ascophyllum nodosum have been shown (70, 71) to reflect the ambient cadmium concentration. Again, seasonal variations have been observed in F. vesiculosus (72).

Ray and White (73) have shown that the green algae Ulothrix and two vascular plants, Potamogeton richardsonii and Equisetum fluviatile, concentrate cadmium from the stream to a great extent. Subsequent work showed that Equisetum arvense, Sparganium chlorocarpum and Sparganium americanum also display some affinity for cadmium (74). The algae Ascophyllum nodosum (75) from Norwegian fjords showed abnormally high cadmium concentration up to 90 km from the source. The water hyacinth Eichornia crassipes is also known to remove cadmium from contaminated lakes (76). The alga Cladaphora glomerata showed a maximum cadmium concentration of 3.9 ppm with a concentration factor of 49,000 (77).

TOXICITY

FISH

A number of toxicity studies, both acute and chronic, have been done on fish. The acute lethal action of cadmium is unusually slow. Schweiger (5) had earlier suggested 4 mg Cd/l as being lethal to rainbow trout in seven days and 3 mg/l as being safe; however, in a later study by Ball (78), the 7-day TLm was found to be only 0.01 mg Cd/l. Pickering and Henderson (79) made acute toxicity studies of several species of warm-water fish. The chronically safe level for bluegill, Lepomis macrochirus, after 11 months exposure in hard water is between 30 and 80 µg/l (80). Egg and larvae survival rates were affected even at the lowest concentration tested.

Pickering and Gast (81) found the maximum toxicant concentration which had no adverse effects on fathead minnow, Pimephales promelas, in flow-through bioassay to be between 37 and 57 µg Cd/1. However, in static bioassay, the 96-hour TLm was found to be 0.47-0.84 and 52.7-105 mg/l for soft and hard water, respectively (79). In a study of the chronic toxicity of a copper, cadmium and zinc mixture to the the fathead minnow, a lethal threshold was attained when each metal was present at a concentration of 0.4 or less of its individual lethal threshold (82).

Eisler (83) determined the 96-hour TLm of cadmium to mummichogs, Fundulus heteroclitus, and striped killifish, Fundulus majalis, at 20°C and 20°/00 salinity. The values obtained there were 21.0 and 55 mg Cd/l, respectively. In a further study (84), it was observed that the addition of 10 mg/l of cadmium to Zn/ Cu mixtures tested produced a statistically significant increase in mortality. Cearley and Coleman (30), on exposing large-mouth bass, Micropterus salmoides, and bluegill to 0.85 mg Cd/l, found 50% mortality within 56 and 138 days, respectively. At 0.08 mg Cd/l, the bass had 50% mortality in 82 days, whereas the bluegill survived six months exposure. Negilski (85) found 168-hour LC 50 for the marine fishes yellow-eye mullet, Aldrichetta forsteri, and small-mouthed hardyhead, Atherinasoma microstoma, to be 16 and 21 mg/l, respectively, in static tests. The median survival periods for stone leach, Noemacheilus barbatulus, (86) at 3.4 and 5.2 mg Cd/1 were 34.4 and 3.5 days, respectively, in hard water (240 ppm).

INVERTEBRATES AND PLANTS

Very few data are available on toxicity levels of cadmium to aquatic invertebrates and plants. However, research is imperative in this field, since plants and insects serve as important fish food organisms, and obviously the protection of fishery resources depends upon the continued propagation of the aquatic plants and insects. Contamination at the lower

trophic level will also eventually lead to contamination of fish, and ultimately mammalian species. Eisler (83) determined the 96-hour TL_m of cadmium for a number of marine organisms at 20°C and 20% salinity (Table 4) by static bioassay.

TABLE 4. Lethal concentration of cadmium to marine invertebrates.

Species	mg/liter Cd
Sand shrimp,	· ·
Crangon septemspinosa	0.32
Hermit crab,	
Pagurus longicarpus	0.32
Glass shrimp,	
Palaemonetes vulgaris	0.42
Common starfish,	
Asterias forbesi	0.82
Common soft shell clam,	
Mya arenaria	2.20
Green crab,	
Carcinides maenus	4.10
Atlantic oyster drill,	
Urosalpinx cinerea	6.60
Eastern mud snail,	
Nassarius obsoletus	10.50
Sandworm,	
Nereis virens	11.00
Blue mussel,	
Mytilus edulis	25.00

Information on cadmium toxicity levels of various aquatic insect species is very limited. Clubb et al. (87) studied cadmium toxicity to nine species of aquatic insects selected for their wide distribution and importance in the aquatic food chain. Ninety-six-hour TLm values in continuous-flow bioassays at 10°±2°C for stonefly, Pteronarcella badia, and mayfly Ephemerella grandis grandis, were 18.0 and 28.0 mg Cd/1, respectively. Seven other organisms studied-true flies, Atherix variegata; Hexatoma sp.; Holorusia sp.; the stoneflies, Acroneuria pacifica, Arcynopteryx signata, Pteronarcys californica; and the caddis fly, Brachycentrus americanus—were found to be relatively insensitive to cadmium.

Median tolerance limits (50% - 24-96) hr) of various benthic organisms to cadmium is static bioassays at 17°C have been tabulated (Table 5) (88).

Cadmium was found lethal to various freshwater zooplankton species obtained from a subalpine lake in Italy in static bioassay at 10°C. Cadmium concentrations of 3.8, 0.55 and 0.055 mg/l were found lethal (LC₅₀ - 48 hr) to Cyclops abyssorum

prealpinus, Eudiaptomus padanus padanus and Daphnia hyalina, respectively (89).

TABLE 5. Toxicity of cadmium to freshwater invertebrates.

TL _m (ppm) 24-96 hr				
4.6-1.7				
0.14-0.07				
5.1-3.4				
11.0-8.1				
5.1-1.2				
10.1-8.4				

In a static bioassay with three species of freshwater insects—a stonefly, Acroneuria lycorias; a mayfly, Ephemerella subvaria; and a caddis fly, Hydropsyche betteni-Warnick and Bell (90) observed that the species Ephemerella was most sensitive to cadmium (96-hr ${\rm TL}_{\rm m}$ - 2.0 mg/l), but the other test organisms lived beyond 96 hours even at concentrations up to 64.0 mg/l. In a static bioassay of seven sublittoral and intertidal Australian invertebrate species (91), the LC50 values ranged from 0.2 to 14 mg/l. Continuous-flow bioassay gave very similar results. LC50 of cadmium for some freshwater invertebrates were determined: 0.04 mg/l for the amphipod Austrochiltonia subtenuis, 0.06 mg/l for the shrimp Paratya tasmaniensis, 0.84 mg/l for the ephemeropteran nymph Atalophlebia australis, 250 mg/l for the zygopteran nymph Ischnura heterosticta, and well over 2,000 mg/l for a trichopteran larva of the Leptoceridae (92).

Algicidal concentration for Selanastrum capricornutum has been found to be 0.65 mg Cd/l (93). Chlorella showed a growth inhibition at 0.05 ppm Cd, and Scenedesmus at less than 0.1 ppm (94).

FACTORS AFFECTING TOXICITY

Many factors act to influence the effects of cadmium on the living organism. The form of the metal in the water is particularly important, since a complex or chelated form may be less toxic than the ionic form, depending upon its stability and how easily it can dissociate. It has been suggested that toxicity of cadmium is relatively independent of hardness (6). Again, there may be antagonistic or synergistic influences of one metal on another, and toxic effects of cadmium are known to be partially or

totally prevented by some metals, such as zinc and selenium, and thiol compounds (21, 94, 95).

There appears to be a synergistic effect between dissolved oxygen and cadmium toxicity to aquatic insects, and results indicate that the toxicity of this trace metal increases as the dissolved oxygen concentration increases (96). Conflicting results have been reported as to the nature of the relationship between cadmium and zinc regarding toxicity. In the aquatic plants Lemna valdiviana and Salvinia natans, cadmium and zinc acted synergistically to inhibit growth (65). Hutchinson (94) observed an antagonistic effect of selenium on cadmium toxicity to green alga, Haematococcus, and Chlorella. Eisler and Gardner (84) examined the acute toxicities of mixtures of cadmium, copper and zinc to mummichogs, Fundulus heteroclitus, in synthetic seawater. It was observed that levels of cadmium not ordinarily lethal when tested alone show adverse effect on survival in combination with zinc and copper. O'Hara (97) observed that the toxicity of cadmium to the fiddler crab, Uca pugilator, is greatest at high temperatures and low salinities. In freshwater shrimp, Paratya tasmaniensis, zinc and cadmium appeared to interact less than additively at concentrations below one toxic unit but, above it, it was strictly additive. Again there was seasonal difference in its sensitivity to cadmium (92).

BIOLOGICAL EFFECTS

Cadmium has a particular affinity for organic ligands involving sulphur, nitrogen and other eletronegative functional groups. The ability of the metals to interfere with enzyme-mediated pathways is attributed to complex formation between the metals and ligands on the enzyme, substrate, activators and cofactors containing any of the above groups that serve as electron donors. This effect of inhibition by cadmium is seen in α-ketoglutarate dehydrogenase, a-dehydrolipoic acid dehydrogenase and transhydrogenase-all of which are common enzymes in intermediary metabolism. Cadmium has also been found to be strongly bound to mitochondrial proteins. Cadmiumbinding proteins have been isolated from marine vertebrates (98).

At concentrations of 0.18 ppm, the oxidative phosphorylation of rat liver mitochondria was uncoupled to about 50%; and at 0.77 ppm, a complete inhibition was observed (99). This concentration is much lower than the average kidney and liver cadmium content of a normal adult man.

The first concern over cadmium poisoning was sparked by the appearance of the "itai-itai" disease which occurred in Japan. The disease was characterized by osteomalocia, osteoporosis and renal damage (25). It was postulated that while cadmium was a causative factor, there must

have been other profound disturbances in the calcium metabolism, and also calcium and vitamins A and D deficiencies. However, it has since been noted that dietary cadmium affects the components of the bone collagen crosslinks and therefore adversely affects bone formation (100).

Other manifestations of cadmium poisoning are acute pulmonary edema, caused by exposure to fumes or dusts of cadmium and/ or cadmium compounds. Chronic exposure to cadmium results in chronic emphysema (22) and renal disturbances (23). Morphologic alterations of the kidney were confined to proximal tubule cell alterations. Even lowdose levels of cadmium produced degeneration of proximal tubule cell mitochondria and increased numbers of dense, granular lysosome-like bodies (101). Cadmium Cadmium also has a depressive effect in DNA snythesis, which is not energy dependent and may be due to disruption of the replicative process (102).

Cases of acute gastroenteritis have been connected to ingestion of acid-type foods that have been stored in cadmiumlined containers. Mammalian testicular damage has been noted in some instances of high cadmium content.

The initial induction of metallothionein in the liver by administration of cadmium previously appeared as a protective mechanism against initial small doses of cadmium (25) in the liver. Zinc has been shown to protect against cadmium toxicity and to ensure metallothionein synthesis. It appears that the amount of metallothionein is dependent on cadmium level or on the cadmium/zinc ratio. Cadmium-thionein is synthesized in the liver and transported to the kidney. Recent evidence indicates greater toxicity (103) of cadmium-thionein in comparison to ionic cadmium, due to rapid liberation of the cation at specific locations once it reaches the kidney. The strength of the chemical bond of cadmium to metalloprotein facilitates transport of the cation to the kidney.

FISH

The effect of cadmium on fish is well known. It has been reported (104) that the death of fish occurs "chiefly due to the coagulation or precipitation of mucus secreted by the gills or damage to gill tissues". There are physiological manifestations of exposure to chronic levels of cadmium. Behavioral changes have also been observed (29, 30); the breathing rate and respiration rate increase, rate of oxygen consumption falls gradually until death, and the fishes' production of CO₂ declines.

Damage to gill tissues was noted (5) in carp, Cyrinus carpio; brook trout; and rain-bow trout exposed to cadmium. They also experienced respiratory difficulties and displayed symptoms of slow suffocation. However, Eaton (80) observed that most bluegill appeared physically normal till they

died. The adult bluegill spawned at 239 µg/ l and 2,140 µg/l, but most larvae were severely deformed six days after hatching at these concentrations. Larval survival was 10% at 90 µg/l after 30 days; whereas it was 60% and 78% at 33 µg/l and in control water, respectively, after 60 days. In a study (30) of largemouth bass and bluegill, it was noted that those fish which died "exhibited erratic, uncoordinated swimming movements, muscle spasms and convulsions, followed by loss of equilibrium, with periods of quiescence and paralysis". behavior suggested that the nervous system was the site of damage and the inhibition of actyl-cholinesterase was the cause of The rate of weight gain of bluegill death. tended to be lower as the cadmium concentration was increased. Pickering and Gast (81) observed a small but significant reduction of 16.5% in hatchery success in minnow eggs exposed to 57 µg/l, as compared to the control group in one $\mu g/1$. However, in a trimetal study (82) of cadmium, copper and zinc, the hatchability of eggs exposed to 59 µg/1 was actually better than of the control group (93.7% vs 79.4%).

Rehwoldt and Karimian-Teherani (105) noted two distinctly different levels of cadmium uptake for male and female zebrafish, with maximum concentration in females about 2.5 times more than the male, and a marked decrease in the number of offspring when fed a diet containing 10 ppm cadmium. It has been observed by Bengtsson et al. (106) that on chronic exposure of minnows to cadmium, vertebral damage occurred at concentrations as low as 7.5 µg Cd/l and the percentage of surviving minnows with spinal deformity increased with increases of cadmium concentration. regeneration in killifish, Fundulus heteroclitus, was retarded by cadmium concentrations of 0.01 mg/l (107). Gardner and Yevich (108a, b) observed changes in the intestinal tract, in the kidney and in the blood, as indicated by a rise in the percentage of eosinophils. They also observed histopathological changes in the gill tissue. Also observed were pronounced histophthological changes and inhibition by over 50% of lacate oxidation by gills of rainbow trout, Salmo gairdneri, on exposure to 1.12 mg Cd/1 for 24 hr (109). Lower metal concentration induced mortality without any detectable effect on oxidative activity.

Cadmium has been found to alter energy production as indicated by oxygen and phosphate metabolism of bluegill liver mitochondria (110). A concentration of 3.3 x 10⁻³ µmoles/ml severely blocked oxygen uptake, indicating the mode of toxic action.

Christensen (111) noted several biochemical responses of brook trout embryos and alevins. Tests with cadmium showed decreased weight, increased protein content, and increased GOT, ALP and ACH activity. Sangalang and O'Halloran (112) presented evidence that cadmium can directly affect testicular steriodogenesis in brook trout, in vitro, and can inhibit biosynthesis of 11-ketotestosterone. Testes of mature brook

trout exposed to 25 μg Cd/l for 24 hours were injured, as evidenced by histological examination, although no detectable damage was done to primordial germ cells.

Much concern has been raised by the finding that cadmium affects the reproductive organs (113) of brook trout held in water containing as low as 1 ppb after 41 days. It is not certain whether this is a dose-related effect or not. In all experiments, the testes which were damaged were either nearly mature or fully mature; and, therefore, it appears there is a relationship between testis injury and age. An increase in testosterone and 11ketotestosterone in the plasma of the cadmium-treated fish was noted, and suggested an adverse effect of cadmium on the clearance and utilization of these hormones. The level of these steroids remained high during regression of the testes, suggesting a "possible negative feedback response in the cadmium-treated fish that may involve the pituitaryinterrenal-gonadal axis".

Benoit et al. (40) studied three generations of brook trout, Salvelinus fontinalis, exposed to cadmium. The fish were able to spawn successfully at 3.4 µg/1 and below. The trout exposed to 3.4 µg/1 did not show any hyperactive behavior before spawning, but suddenly became extremely hyperactive when females began their probing activity, "lapsed in paralysis and died within a few minutes". Females were less susceptible than males. Significant reduction in numbers of young produced per female and weight loss of young were observed. In the third generation, there was a 73% decrease in weight from that of the control fish. The toxic mode of action of cadmium for the brook trout males was similar to that observed for flagfish, Jordanella floridae, (114). However, this difference in sensitivity between sexes was not observed by Cearley and Coleman (30) in the case of largemouth bass and bluegill.

Spehar observed that in flagfish, spawning and embryo production were adversely affected at 8.1 µg Cd/1 (114). Largemouth bass, Micropterus salmoidaes, when exposed to 0.1 mg Cd/1, experienced respiratory disfunction and an increase in opercular activity continued at an abnormally high rate till death (115). Rosenthal and Alderdice (116) correlated survival of Pacific herring eggs with cadmium concentration and pre-exposure time. The metal affected water intake and formation of perivitelline space after fertiziliation. It was concluded that short-term exposure of herring eggs can result in changes in the external egg membrane. Goldfish injected with cadmium showed destruction of male germinal epithelium and reduced production of sperm (117). Production of ova was also retarded.

Mounib et al. (118) found decreased activity in four carbon dioxide fixing enzymes (NAD, NADP - malic enzyme, propionyl COA carboxylase and phosphoenol-

pyruvate carboxykinase) in Pacific herring. Since these enzymes are involved in biosynthetic processes, it was speculated that the depression of their activity may explain why larvae were small and inactive at hatching after exposure to cadmium. Cadmium at very low concentrations has an activating effect on glutamine synthetase from Chinook salmon, Oncorhynchus tshawytscha, and inhibits at concentrations >30x10⁻⁶M (119).

Jackim et al. (120) have studied the effect of cadmium on fish liver enzymes. In vitro studies with alkaline phosphatase indicated strong inhibition of activity by cadmium; however, in vivo studies did not indicate any significant change. Both acid phosphatase and xanthine oxidase were marginally inhibited, and moderate inhibition had been noted for RNAase and catalase. Gould and Karolus (121) have found decreased liver enzyme activity in cadmium-exposed cunner, Tantogolabrus adspersus.

INVERTEBRATES AND PLANTS

Scant data are available on biological effects of cadmium on aquatic invertebrates and plants.

Thurberg et al. (122) found green crabs, Carcinus maenas, and rock crabs, Cancer irroratus, exposed to cadmium chloride had depressed rates of gill-tissue oxygen consumption—an indication of metabolic distress. Rock crabs showed significantly increased aspartate aminotransferase activity and elevated level in serum magnesium when exposed to cadmium as the chloride, but not as the nitrate salt (123). Shuster and Pringle (50) observed in cadmium-exposed oysters little shell growth, pigmentation loss in the mantle edge and coloration of digestive diverticulae, and high mortality. Shore et al. (124) observed a relationship between the cadmium concentration in tissue and depression of glycolysis in limpets, Patella vulgata.

Scallops, Argopecten irradians, have been shown to exhibit significantly higher oxygen consumption rates when exposed to 0.94 ppm cadmium for 96 hours (125). MacInnes and Thurberg (126) observed slightly increased oxygen consumption in the gastropod Nassarius obsoletus over an exposure range from 0.5 to 4.0 ppm.

In Daphnia magna, reproductive impairment to the extent of 50% at 0.7 µg/l and 16% at 0.17 µg/l of cadmium was noted. It also caused reduced weight gain and changes in total protein and glutamic oxalacetic transaminase (GOT) activity (127).

It is known that cadmium inhibits the biosynthesis of chlorophyll (25). Klass et al. (128) found that development and growth in laboratory cultures of freshwater green alga, Scenedesmus quadracauda, were adversely affected by a cadmium concentration of about 6 ppb. In Selanastrum

capricornutum (93), the growth inhibition starts at 50 µg/l, with complete inhibition at 80 µg/l. Cadmium-exposed Najas quadulepensis showed reductions of chlorophyll, turgor and inhibited development, even at 7 ppb exposure level (66).

STIMMARY

Cadmium has been defined as a stock pollutant (129). It is extremely harmful in various concentration levels in water, it is toxic to most aquatic organisms studied, and its toxicity is accumulative. Cadmium accumulation and its toxicity—acute and chronic—to aquatic life generates concern, for all levels of the food chain are affected. A food-chain model (130) has been proposed.

Cadmium operates slowly and it appears that a 96-hour bioassay test may not be of sufficient duration to evaluate its toxicity to fish. Again, tremendous differences have been observed between static and continuous bioassay results. The flow-through bioassay, which better simulates natural conditions, should be adopted as a standard procedure if any ecologically meaningful results are to be achieved. The static test can be used only for preliminary screening. Shortterm exposure tests on embryonic or larval stages to estimate chronic toxicity may be advantageous, since these are the most sensitive stages and would be indicative of chronic toxicity.

The impact of cadmium on aquatic organisms themselves should be thoroughly studied, as very little has been accomplished in this respect. Major research effort should also be directed to the chronic toxicity effects of cadmium on the primary producer level, since they may present a problem at higher trophic level by reducing food availability for fish and shellfish, and so result in starvation and larval death. While cadmium coatings in household articles such as ice traps and food containers have been prohibited by law, the production of cadmium is increasing at an alarming rate. Measures must be taken to prevent its entry into the ecosystem, or at least to keep it at the barest minimum, if another disaster on the scale of Minamata is to be prevented. Until further knowledge is gained on its adverse effects on aquatic organisms, caution must be exercised in recommending the permissible levels of cadmium in natural waters containing fish and shellfish.

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