

## THE PROBLEM OF THE THREAT TO THE MARINE ENVIRONMENT FROM HEAVY METALS

Contents: 1. Introduction, 2. Mercury, 3. Lead, 4. Copper, 5. Zinc, 6. Cadmium; Streszczenie; References

### 1. INTRODUCTION

The threat to the natural environment, including that of the sea, by heavy metals provides one of the most difficult and urgent zoological problems of present day society. The long delay between stimulation of the environment with an atom or ion of a metal and the response in the ecosystem hinders the assessment of cause and effect and to undertake preventive measures to protect the ecosystem.

About one-third of the ninety elements occurring on the Earth have been found to be essential for the growth and development of bacteria, plants and animals. Some of the elements occur and are consumed in large quantities as nutrients, while other, so-called trace elements, can be detected only by highly sensitive analytical procedures. We cannot be quite sure that certain organisms do not require minute amounts of other elements hitherto considered as unnecessary.

Some of the trace elements may become toxic when their levels become higher than the metabolic and physiological requirements. On the other hand, many plants and animals can tolerate excessive physiological levels of trace metals and by their accumulation they provide a serious threat to higher links in the trophic chain.

At present, heavy metals are considered as stimulants or retardants of vital processes, depending on their concentration, oxidation states and forms of occurrence. Apart from a straightforward action, heavy metals may accumulate in different organs of animals in the form of compounds temporarily innocuous, from which they can pass to the

blood stream owing to rapid metabolic transformations, and to poison the whole organism.

The pollution of surface waters and soil with heavy metals is increasing and provides a serious problem, even in our country, particularly in the vicinity of industrial regions.

The paper presents the effect of heavy metals on the marine ecosystem, marine fauna and human health.

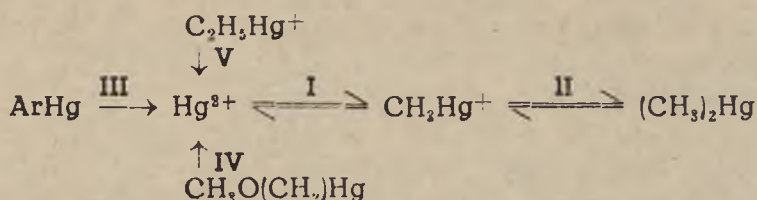
## 2. MERCURY

Mercury occurs in the environment highly dispersed in the upper layers of the earth's crust as HgS, its content amounting on average to 50 ng/g. The upper layer of the soil contains 50 to 510 ng/g and this level may rise locally up to 1000 ng/g due to the use of organomercurials in agricultural operations. Table 1 lists the contents of mercury in the environment after various authors.

Table 1. Mercury levels in the natural environment after various authors  
Tab. 1. Zawartość rtęci w środowisku przyrodniczym według różnych autorów

Environment Środowisko	Concentration Stężenie	Reference Literatura
Volcanic emissions Wyżewy wulkaniczne	up to 20 000 ng/m <sup>3</sup>	34
Earth's crust Skorupa ziemska	50—100ng/g	14, 21, 27
Soil (Europe) Gleba (Europa)	50—510 ng/g	21
Atmosphere — Atmosfera	0,1—20 ng/m <sup>3</sup>	9
Precipitation — Opady	5—120 ng/dm <sup>3</sup>	7, 14
Arctic glacier Lodowiec arktyczny	30—70 ng/dm <sup>3</sup>	33, 34
Spring water Woda źródłana	10—50 ng/dm <sup>3</sup>	14
Inland waters Wody śródlądowe	10—500 ng/dm <sup>3</sup>	8
Sea waters Wody morskie	10—30 ng/dm <sup>3</sup>	15
Ocean	3—30 ng/dm <sup>3</sup>	8, 11, 15
Deep oceanic waters Głębokie wody oceaniczne	3—5 ng/dm <sup>3</sup>	15
Minamata Bay Zatoka Minamata	1600—3600 ng/dm <sup>3</sup>	10, 17

In the aqueous environment both the metallic mercury and its compounds undergo various chemical and biological transformations illustrated by the following scheme:



The methylation reaction (I) occurs in bottom sediments owing to the activity of the *Clostridium* bacteria (particularly *Cl. cochlearium*) and to processes promoted by aerobic methanogenic fungi (e.g. *Neurospora crassa*) and bacteria belonging to the genus *Pseudomonas* [13].

Microorganisms, in particular bacteria, participate in the following transformations:

(i) reduction of phenyl-, ethyl- and methylmercury to elemental mercury;

(ii) aerobic conversion of phenylmercury acetate (PMA) to diphenylmercury and elemental mercury;

(iii) reduction of the  $\text{Hg}^{2+}$  ion to mercury (*Pseudomonas*, *Enterobacteriaceae*, *Staphylococcus aureus*).

In an acidic medium, methylmercury remains in water, whereas in a weakly basic medium it is converted to dimethylmercury (II).

Both methylmercury and dimethylmercury are easily assimilable by lower aquatic animals providing the first links of the trophic chain.

Under specific conditions of the marine environment, phenylmercury [III], alkoxymethylmercury [IV] and alkylmercury [V] can be converted to an inorganic form of mercury. Under anaerobic conditions the mercuric ion may react with hydrogen sulphide deposited on the bottom to form sparingly soluble  $\text{HgS}$ .

It is worth noting that transformations of many more complex mercury compounds in the marine environment have not been described so far.

The level of mercury in predatory fish is usually by 3 — 4 orders of magnitude higher than in the surrounding sea water [1]. The relation between the mercury level in fish and in the surrounding water can be calculated from the following empirical equation

$$[11] : \quad y = 0.031x - 0.179$$

where  $x$  is the mercury level in water in  $\text{ng dm}^{-3}$  and  $y$  is the mercury level in fish tissue in ppm of wet weight.

Ingestion in branchial animals occurs mainly directly from the sea water through the body surface and gills. It may also occur through the alimentary tract. Almost 100 per cent of mercury in fish and some other branchial animals occurs as methylmercury, a lower percentage being found in some molluscs.

Methylmercury is accumulated mostly in the muscles of fish and

the half period of excretion amounts on average to about 2 years depending on the species [28].

The cumulation factor for the system fish — fish-eating birds amounts to about 10. In the Baltic Sea area, the critical organisms constitute birds, sea mammals and man who assimilate all the mercury from food. In humans, about 15 per cent of cationic ( $\text{Hg}^{2+}$ ) mercury ingested with food is assimilated and as much as 94 per cent of methylmercury,  $\text{CH}_3\text{Hg}^+$  [22]. The half-period of excretion of  $\text{Hg}^{2+}$  and  $\text{CH}_3\text{Hg}^+$  in man is  $42 \pm 3$  days and  $76 \pm 3$  days respectively.

The potential threat from environmental pollution with mercury compounds can be estimated from the following expression [8]:

$$I = 0.1 \frac{P_g}{c}$$

where  $I$  is the impact index,  $P_g$  is the overall production of mercury and  $c$  is its concentration in the environment.

A more accurate estimate can be obtained by using the index of local impact,  $I_L$ :

$$I_L = I \frac{P_L}{P_g} \cdot \frac{S_g}{S_L}$$

where  $I$  is the impact index from the preceding equation,  $P_L$  and  $P_g$  are the local and overall annual production of mercury respectively,  $S_g$  and  $S_L$  are the total and local area of a particular region respectively.

According to FAO data, the annual production of mercury in the world amounts to approx. 9 000 tons. About 5 000 tons of this reaches the seas and oceans. It is estimated that the waters of the World Ocean have accumulated 136 million tons of mercury and the atmosphere surrounding the Globe, to which it is emitted during the burning of coal, about 22,700 tons [5].

The main sources of pollution of the atmosphere are the paper industry in which mercury compounds are used for preservation of the groundwood and wood pulp, the plastics industry where the compounds are used as catalysts, chlor-alkali production using mercury electrodes and agriculture where mercury fungicides are used.

In Poland, of the 335 tons of mercury consumed annually by the national economy, 140 tons find their way to the environment, i.e. 42 per cent. About 25 tons, i.e. 7 per cent [4], are utilized in the production of pesticides.

After Somer [31], about 4 ton of mercury is discharged annually with municipal sewage to the Baltic Sea and about 20 tons with industrial effluents (Table 2). Rivers supply about 6 tons and the atmosphere about

4 tons annually. Of the total amount of about 34 tons/yr. as much as 29 tons/yr. is deposited in coastal regions and about 3 tons/yr. leaves the Baltic through the straits. As a result, comparatively low levels of mercury (7—9 ng dm<sup>-3</sup>) are observed in the open-sea waters, comparable with those noted in the distant Atlantic waters [26].

Polish studies on mercury levels in fish [2, 20, 24], taken in southern regions of the Baltic Sea and in the Pomeranian and Gdańsk Bays, showed them to range between 0.10 and 0.15 mg kg<sup>-1</sup>.

To protect man — the fish consumer — several governments of Baltic states issued a ban on the sale of fish containing more than 1 mg Hg per kg wet mass. This applies especially to fish taken in Oresund, the Gavlø region, the Gulf of Bothnia and in the coastal regions of the Gulf of Finland [23].

### 3. LEAD

At the beginning of 1970 the global output of lead amounted to approx. 4 million tons, 40 per cent of this amount being consumed in the production of batteries and various oxides, and 25 per cent in the production of cables and pipelines. Of the more important lead compounds, PbCO<sub>3</sub> was used as a component of a white pigment, PbCrO<sub>4</sub> as a yellow pigment, Pb<sub>3</sub>O<sub>4</sub> as the protective pigment, so-called minium, Ca<sub>2</sub>PbO<sub>4</sub> as a corrosion inhibitor for iron and steel, Pb(SiO<sub>3</sub>)<sub>2</sub> for dyeing earthenware, (C<sub>2</sub>H<sub>5</sub>)<sub>4</sub>Pb and (CH<sub>3</sub>)<sub>4</sub>Pb as fuel additives increasing the octane number.

Lead used by man has been dispersed in the environment in various forms over the ages, and is nowadays considered to be the main pollutant. Its high toxicity and continuous cumulation in higher organisms has led to its critical level in the human organism.

The lead content of the atmosphere varies substantially from 0.0002 to 71 µg m<sup>-3</sup>, the latter value being recorded during the peak traffic period in Los Angeles. Atmospheric lead is produced mostly by combustion of fuel containing alkyl lead compounds [13]. The concentration of lead in American gasoline amounts to 2.6 g per US gallon (= 3.785 litre) on average.

Unpolluted Antarctic soil contained small quantities of lead averaging 10 ppm. This value has been assumed to be the natural level of the element in the soil. In urban and industrialized regions the level of lead in the soil is almost ten times higher, particularly along roads and highways.

Natural sources of lead contaminate water reservoirs to a small extent only, owing to the poor solubility of the element and formation of deposits. In clean reservoirs of surface waters the mean level of lead

was  $0.5 \mu\text{g dm}^{-3}$ , whereas in distant ocean waters it was less than  $0.1 \mu\text{g dm}^{-3}$ . During the past 50 years, however, the level of lead in the World Ocean increased rapidly. Ice samples taken in Greenland revealed an increase in the level of lead from 0.0005 to more than  $0.2 \mu\text{g kg}^{-1}$  from 800 B.C. to 1965 [3].

The toxic effect of lead on microorganisms can be explained in terms of displacing essential elements such as Mg, Fe and Mn from the -SH, -NH<sub>2</sub> and =NH groups of enzymes by the Pb<sup>2+</sup> ions. However, no untoward effect of lead has been noted on plants. Many species of plants tolerate high levels of lead in the soil and tend to transport it from the surface to deeper layers of soil by means of the root system. Grass has been reported to grow well over a lead mine area where the level of lead in the tissue ranged from 200 to 9 645 ppm [13]. Particularly resistant to lead were *Plantago lanceolata*, *Agrostis stolonifera* and *Festuca rubra*.

The ability of plants to accumulate lead has been used to retrieve its levels in the past. In particular periods the following levels of lead were found in the veins of a tree growing 50 m off a moderately crowded street:

1865—1879	0.16 ppm Pb
1900—1912	0.12 ppm Pb
1940—1947	0.33 ppm Pb
1956—1959	0.74 ppm Pb
1960	3.90 ppm Pb

All analyses were performed using the same technique simultaneously. If one assumes that lead was adsorbed in the annual growth rings of a tree, an alarming and dramatic factor of environmental pollution within the past 100 years is provided.

Animals take in lead with water and food (and accumulate approx. 10 per cent of that amount), also breathing it in with air (and accumulate approx. 50 per cent of that amount). Inorganic forms of lead (Pb<sup>2+</sup>) provide the main poison of metabolic processes and accumulate in erythrocytes, the liver and kidneys. It inhibits the enzymatic activity of dehydrogenase of  $\delta$ -aminolevulinic acid (ALA) which is essential for producing hems — dyes forming haemoglobin with proteins in the bone marrow. Children are more sensitive to lead poisoning than adults, toxic blood levels of lead amounting to  $0.25$  and  $0.8 \mu\text{g dm}^{-3}$ , respectively.

An estimate of the balance of lead in the Baltic Sea waters is shown in Table 2. The following conclusions can be drawn from these data: (i) the atmosphere provides the main source of lead in water; (ii) the removal of lead by sedimentation provides a very effective process both in the coastal zone and in the open sea; (iii) the total content of lead in water is low.

Table 2. Concentration and material balance of heavy metals in the Baltic Sea (after ICES 1977)  
 Tab. 2. Koncentracja i bilans metali ciężkich w Morzu Bałtyckim (wg ICES 1977)

	Hg	Pb	Cu	Zn	
Municipal pollution Zanieczyszczenia komunalne	0.2 g/person/yr g/osoba/rok	10	10	60	Discharges - Zrzuty
Rivers — Rzeki	4 15 x 10 <sup>-3</sup> ton/km <sup>3</sup>	200 ? 10 <sup>3</sup>	200 10 4000	1000 10 4000	
Przemysł — Industry	6 20? ton/yr — t/rok	10 <sup>3</sup> ?	4000 ?	4000 ?	
Atmosphere — Atmosfera	20 x 10 <sup>-3</sup> ton/yr — t/rok	?	3	20	
Sedimentation — Osadzanie się	4 ton/yr — t/rok	10 <sup>3</sup> —10 <sup>4</sup>	600	4000	
Inflow — Napływ	ton/km <sup>3</sup>	?	?	?	
	ton/yr — t/rok	?	?	?	
Outflow — Odplyw	7 x 10 <sup>-3</sup> ton/km <sup>3</sup>	?	5	10	Removal - Usunanie
Sedimentation in the open sea	3 (netto) ton/yr — t/rok	10 — 100	2000	4000	
Sedymentacja w morzu otwartym	0.5 conc. µg/g	?	40	120	
(0.1 mm/yr of 40 per cent dry matter, density 2.5 g/cm <sup>3</sup> )	ton/yr — ton/rok	10 <sup>3</sup> —10 <sup>4</sup>	2000	5000	
0.1 mm/rok 40% suchej masy, gęstość 2,5 g/cm <sup>3</sup>	29 ton/yr — t/rok	10 <sup>3</sup> + industry przemysł	?	?	
Sedimentation in the coastal zone	small ton/yr — t/rok	?	?	?	
Sedymentacja w strefie przybrzeżnej	7 x 10 <sup>-3</sup> conc. in µg/l = ton/km <sup>3</sup>	?	5	10	
Fish catches — Połowry ryb	stężenie w µg/l = t/km <sup>3</sup>	10 <sup>3</sup>	10 <sup>5</sup>	2 x 10 <sup>5</sup>	Load
Sea water — woda morską,	Total tons — Ton razem	140	10 <sup>5</sup>	2 x 10 <sup>5</sup>	

It is likely that lead does not accumulate from food to any noticeable extent. While its level in *Mytilus edulis*, which feeds on suspended matter is low, *Macoma baltica*, which feeds on deposited matter contains higher levels of lead. In seals the lead content is low [3].

#### 4. COPPER

The main sources of copper penetration to the environment are the mining industry and metallurgy. As a matter of fact, a deficiency of copper in the environment may provide a limiting factor in the production of biomass, however an increased level inhibits primary production.

An important feature of copper is its capacity to form very stable complex compounds with complexing and chelating natural compounds present in sea water. The copper complexes are many times more stable than those formed by other metals [6].

A high accumulation factor of copper has been found in phytoplankton and in macrophytes. Particularly high copper levels were found in the crustaceans *Mesidotea entomon* caught in the Gulf of Bothnia and in the Gulf of Finland [16], where they provide an important component in food for seals. No reports have as yet been submitted on the copper levels in seal meat.

The copper level in fish is fairly low, that in the liver being about 30 times as high as in the muscular tissue. The copper level in the liver of eel caught in copper-polluted areas was about 40 mg kg<sup>-1</sup> wet mass, whereas in unpolluted areas the level was half of this [35].

In aquarium experiments with fresh water, small quantities of copper (30—60 µg dm<sup>-3</sup>) were associated with the initiation of a lethal attack of a virus disease evoked by *Vibrio anguillarum* in eel.

Despite of the fact that the balance of copper in the Baltic Sea is far from complete (Table 2), the level of the metal in this basin can be estimated as being high.

#### 5. ZINC

The main industrial sources of environmental pollution with zinc are the nonferrous mining industry, metal refining works and electroplating works. Zinc is much more volatile than copper, hence its higher concentration in the atmosphere (Table 2). In municipal sewage the major part of zinc is released from zinc-plated pipelines and objects made from zinc-plated metal sheets.

Zinc has been found to be markedly accumulated by phytoplankton. Fish from coastal regions have higher levels of zinc than those from the



open sea (contrary to copper) [16]. It is estimated that half the total zinc in the Baltic Sea (Table 2) is from the atmosphere, the rest being from municipal and industrial sewage.

## 6. CADMIUM

Cadmium has a high lethal toxicity. The highest admissible level (NDS according to the Government Regulations and Legal Gazette of the Polish People's Republic, No. 13, 1976) for an 8-hour working post is 0.1 mg per dm<sup>3</sup> of air. Lethal intoxication in man at a cadmium concentration in air of 10 mg per dm<sup>3</sup> occur after 5 hours [25, 29].

In 1946, Dr Hagino of Japan discovered and described an unknown disease and called it „itai-itai”. It was first noted in 1920 in Fuchu, Toyama region, but it spread particularly after World War II causing hundreds of deaths. Autopsy revealed high cadmium levels in the corpses. The direct cause of the poisoning was the Kamioka mine which discharged considerable amounts of Cu, Pb, Zn and Cd to the Jinzu river. The water from the river was used for drinking by the population and for irrigation of the rice fields. As late as 1968 over 100 people in this region were still ill.

Numerous fatal cases of cadmium poisoning were recorded in the U.S.A. and in Europe (in France 300 persons died in 1948 after drinking wine stored in cadmium vessels), which resulted in the WHO undertaking of complex studies on the metabolism of cadmium and on its content in the biosphere, in 1973—1977. Cadmium was found to be absent in children, but in 45-year-old adults the total amount was 30—40 mg, i.e. the mean annual accumulation amounts to 0.67—1.3 mg (=1.8—3.6 µg per 24 hrs.) [18].

Cadmium and its compounds are used in electroplating, the production of low-melting alloys, stable enamel pigments, alkaline accumulators, plastics, as well as in nuclear reactor, radar and television technology.

The global production of cadmium in 1970 amounted to 15000 tons and increased at a rate of 14 per cent annually. Since cadmium-rich minerals (grenokite, CdS, containing 77.6 per cent Cd and otavite CdCO<sub>3</sub>, containing 61.5 per cent Cd) are meagrely distributed, the main source of cadmium is provided by zinc ores containing 0.01—0.5 per cent Cd. Zinc smelting works in Poland, together with those in the U.S.A. are the biggest suppliers of cadmium and, at the same time, they are the biggest producers of cadmium-containing effluents [12].

The concentration of cadmium in the air over the Atlantic Ocean is very low, ranging from 0.03 to 0.62 ng m<sup>-3</sup>. Over uncultivated regions the concentration ranges from 0.001 to 0.005 µg m<sup>-3</sup>, and over Polish towns it

ranged from  $0.002 \mu\text{g m}^{-3}$  in Gdańsk, to  $0.051 \mu\text{g m}^{-3}$  in Katowice, in 1967 [19]. In many countries concentrations as high as  $0.6 \mu\text{g m}^{-3}$  were found in the air around industrial plants.

In Sweden the degree of contamination of atmospheric air with cadmium was determined indirectly by assaying the Cd content in the mass of *Hylacomium splendens*. By using this method it was found that in southern Scandinavia the cadmium content exceeded  $1 \mu\text{g/g}$  dry matter, whereas in the north it did not reach  $0.1 \mu\text{g/g}$  dry matter [32]. A comparison of the cadmium levels with those found in specimens stored in herbaria revealed a two-fold increase in the cadmium concentration in the air over the past 50 years.

In soil the Cd/Zn ratio oscillates between 1:500 and 1:5000, whereas in sea water it amounts to 1:168 [19].

The cadmium level of the sea water is approx.  $0.1 \mu\text{g dm}^{-3}$ , but in unpolluted inland waters it reaches  $1 \mu\text{g dm}^{-3}$ . In highly populated industrialized countries the cadmium level in rivers and lakes increases up to  $10 \mu\text{g dm}^{-3}$ .

In the distant Baltic Sea waters the cadmium level is  $0.1\text{--}0.5 \mu\text{g dm}^{-3}$ , but in the region of discharge of sewage into the Gulf of Bothnia it jumps up to  $10.2 \mu\text{g dm}^{-3}$  [18, 30].

The polluted air and water contributed to an increase in the cadmium levels in soil, which ranges widely from  $1 \text{mg kg}^{-1}$  on uninhabited barren land to  $100 \text{mg kg}^{-1}$  in industrialized regions.

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## PROBLEM ZAGROŻENIA ŚRODOWISKA MORSKIEGO METALAMI CIĘŻKIMI

### Streszczenie

Przedstawiono źródła, drogi przenikania, wpływ i efekty działania metali ciężkich na ekosystem morski, na żywe zasoby morza i zdrowie człowieka. Zebrano najnowsze dane dotyczące przenikania rtęci, ołowiu, miedzi, cynku i kadmu do powietrza, wód powierzchniowych i gleby, na tle ich produkcji w Polsce i na świecie.

Wykazano, że długi okres, jaki upływa od wprowadzenia do środowiska pierwiastka metalicznego do uzyskania odpowiedzi ekosystemu, utrudnia ustalenie zależności przyczynowo-skutkowej i ochronę ekosystemu.

Podano ważniejsze chemiczne i biologiczne przekształcenia pierwiastków metalicznych oraz ich organicznych i nieorganicznych połączeń na różnych poziomach troficznych w środowisku morskim.

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