

Metals in molluscs and associated bottom sediments of the southern Baltic

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ABSTRACT: The concentration of Zn, Cu, Pb, Cd, Ni, Co, Ag, Mn, Fe, Ca, Mg, K and Na in molluscs *Macoma balthica*, *Mya arenaria*, *Cardium glaucum*, *Mytilus edulis* and *Astarte borealis* from the southern Baltic was determined. The surface sediments and ferromanganese concretions associated with the molluscs were also analysed for concentration of these metals. Species- and region-dependent differences in the metal levels of the organisms were observed. The properties of molluscs analysed which have a tendency toward elevated biological tolerance of selected trace metals were specified. The interelement relationship between metal concentrations in the soft tissue and the shell was estimated and was discussed.

INTRODUCTION

The ability of marine mussels to biologically incorporate trace metals in their tissues has been well established; most of the studies have been carried out on the soft tissue (Amiard et al., 1986; Bloom & Ayling, 1977; Bryan, 1980, 1983; Coleman et al., 1986; Cossa et al., 1979; 1980; Davies & Pirie, 1980; Di Giulio & Scanlon, 1985; Farrington et al., 1983; Galloway et al., 1983; Gault et al., 1983; Goldberg, 1975; Goldberg et al., 1978, 1983; Hung et al., 1981, 1983; Johnson & D'Auria, 1980; Julshamn, 1981; Karbe et al., 1977; Langston, 1986; Martincić et al., 1984; Möller et al., 1983; Phillips, 1976a, 1976b, 1977b, 1977c, 1978; Popham & D'Auria, 1983a, 1983b; Ritz et al., 1982; Segar et al., 1971; Slabyj & Carpenter, 1977; Szefer, 1986; Szefer & Szefer, 1985; Szefer & Wenne, 1987). However, the number of articles on the concentration or distribution of metals in shell material, particularly relating the metal concentration in the soft tissue to that in the shell is scanty (Al-Dabbas et al., 1984; Bertine & Goldberg, 1972; Carriker et al., 1980b, 1982; Chow et al., 1976; Ferrell et al., 1973; Koide et al., 1982; Pilkey & Goodell, 1963, 1964; Sturesson, 1976, 1978; Wada & Suga, 1976).

In recent years, much attention has been paid to the chemical composition of marine organisms, especially of molluscs, and of the associated sediments (Luoma & Bryan, 1978, 1979; Luoma & Jenne, 1976a, 1976b, 1977; Langston, 1986; Thomson et al., 1984). The sediments at the water-sediment interface are more important to benthic invertebrates than the subsurface sediments because meiofauna lives above the reduced zone in sediment (Luoma & Bryan, 1981). Detritus-feeding organisms are exposed directly to sediment-bound metals, and the bioavailability of the latter depends to a significant extent upon the geochemical fraction with which a metal is associated in the bottom

substrate (Luoma & Jenne, 1976a). Compared to sediments, organisms exhibit a greater spatial sensitivity and a greater ability to concentrate some metals and therefore are often considered as bioindicators. Analysis of seawater and sediments are rarely carried out; the main disadvantage of the two non-biological monitoring methods is that neither allows for the estimation of the biological availability of the trace metals (Bryan, 1980).

The aim of the present paper was to examine species- and region-dependent variations of metals concentrations in some Baltic mollusc as well as in the associated bottom sediments.

MATERIALS AND METHODS

Samples were collected during the cruise of the research vessel "Oceania" in the southern Baltic in May 1987. The location of the sampling stations is presented in Figure 1. The organisms were caught using a bottom trawl, and were immediately sorted, in the ship laboratory, in respect to species, size (age), and the region where caught. The animals were dissected, and the soft tissue separated from the shell with a plastic spoon. The shells were carefully cleaned of foreign matter, especially of numerous specimens of *Balanus improvisus* that were fastened on the shells of *Mytilus edulis*. Groups of 5 to 33 specimens of similar size were pooled for analysis, weighed and dried at 105 °C. Sediment samples, collected with a grab sampler, and surface layers of sediments were frozen during transport to the laboratory. Then, 1-g subsamples (after drying at 105 °C) were used for the extraction of metals with 1M HCl according to the procedure recommended by Luoma & Bryan (1981). The organic matter content of sediments was

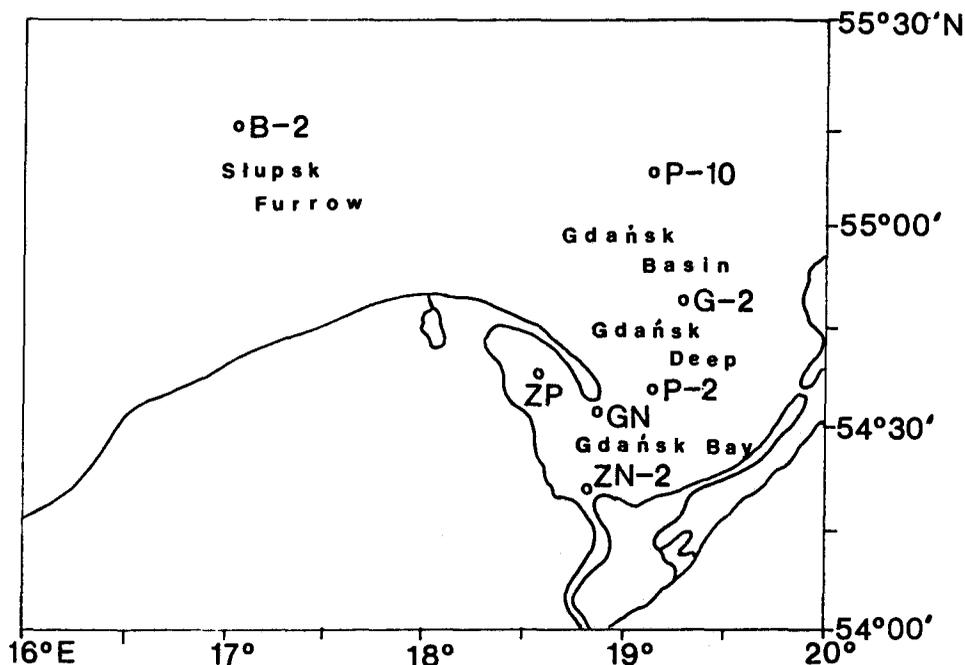


Fig. 1. Location of the sampling stations

determined from the weight loss on ashing at 400 °C to a constant weight. The ferromanganese nodules were taken by bottom trawl together with specimens of *Astarte borealis* (station B-2), and then dried at room temperature. Thin (~2 mm) portions of the surface and the subsurface of the disk nodules were scraped off. The first layer was brown, the second one was very dark brown. This material was used for the extraction of metals in 1M HCl in the same manner as described above for the sediment samples.

The biological material was treated with concentrated HNO₃ (30–50 ml) and left to rest a few days. Concentrated HClO₄ (2–3 ml) was added. Decomposition was accomplished at the lowest possible temperature. The dry residue was then converted to chlorides by evaporation with concentrated HCl and heated in 1M HCl to dissolve the salts. Such prepared solutions and both sediment and nodule filtrates were transferred into an acid-washed volumetric flask (25 ml). Cadmium, Zn, Pb, Cu, Co, Ni and Ag were determined directly from the solution by atomic absorption spectroscopy (AAS); Fe, Ca, Mg, K, Na and partly Mn determined after appropriate dilution. As the sample weight varied, the detection limit of the method used (expressed in µg g⁻¹) also varied. The shell results of Pb, Ag, Ni, Co and Cd were generally below the limit of detection, i.e. were lower than 0.1 µg ml⁻¹ for Pb and Ag, 0.04 µg ml⁻¹ for Ni, 0.02 µg ml⁻¹ for Co and 0.01 µg ml⁻¹ for Cd. Blank samples were routinely run through the analysis to check for contamination. In order to correct for broad band absorption in the case of both the shell and nodule materials, deuterium background correction was used. The standard addition technique was applied to control the data quality. The results obtained for both the soft tissue and shell, by analysis of the spiked material, varied depending on the metal added, its concentration and part of molluscs. The coefficients of variation of AAS measurements for metals in the soft tissue and the shell (in parentheses) were between 3.8 (0.7) and 13.4 (22.8) %. The average recoveries of metals ranged from 77 (70) % to 100 (100) % and were as follows: Zn – 93 (76) %; Cu – 94 (100) %; Pb – 100 (86) %; Cd – 99 (95) %; Ni – 97 (100) %; Co – 87 (70) %; Mn – 98 (87) % and Fe – 77 (94) %.

DESCRIPTION OF HABITATS

The Baltic as a shallow land-locked sea differs considerably from most seas and oceans. It is characterized by low salinity maintained by a high river inflow (457 km³ year⁻¹) in relation to the total capacity of the basin (21 500 km³). The limited exchange of the Baltic waters with saltier North Sea waters through the shallow Danish Straits continuously maintains a low salt content (Łomniewski et al., 1975).

The southern Baltic (92 795 km²) divides into the Arkona Basin, Pomeranian Bay, Bornholm Basin, Słupsk Furrow, Gdańsk Basin and other smaller bays. Gdańsk Bay offers favourable conditions for marine life and, hence, an abundance of zoobenthic organisms. Forty one species and three taxons are present in the bottom fauna of this area; the average percentage share of molluscs is estimated to be 93.7 % of the total biomass (Wenne & Wiktor, 1982).

A great part of the southern Baltic floor is covered by fine and medium-grained sands and aleurite-silty loams. Residual sediments such as coarse-grained sands, pebbles and, sometimes, fine-grained sands are formed as a result of selective bottom abrasion. Most of the Polish coast is formed of such unconsolidated sediments exposed to continuous movement and shifting because of the action of waves and currents. The zone far from

coastal accumulation of sands is covered with medium and fine-grained sands. Richest in organic matter are the silts from particular Baltic deeps, e. g. Gdańsk Deep.

Ferromanganese nodules are abundant in an area of Słupsk Furrow. According to Lomniewski et al. (1975), the floor of this region is, as a rule, covered by nodules at the depth range of 40 to 91 m. The bottom sediment accompanying ferromanganese nodules are mainly sands, gravels and clays. The nodules are characterized by different shape and magnitude; shapeless, granular and disk-nodules occur in the southern Baltic. The material analysed in the present study was discoid, and measured ~1.5–5 cm; fragments (to ~7 cm in length) of nodules were also found.

RESULTS AND DISCUSSION

Table 1 presents the concentration of metals in the soft tissue and shell of molluscs taken from the southern Baltic. Tables 2 and 3 give the results for a labile (soluble in 1M HCl) fraction of surface sediments and ferromanganese nodules taken at the same stations as the zoobenthic organisms.

Metals in molluscs

The metal concentrations obtained here (Table 1) are generally comparable with those observed in molluscs from other Baltic regions (Brzezińska et al., 1984; Brüggemann, 1981; Karbe et al., 1977; Möller et al., 1983; Tervo et al., 1980). There are some interspecies-dependent changes in the metal concentrations. Maximum levels of Zn and Cu were observed in the soft tissue and shell of *Macoma balthica* whilst both of these parts in *Mya arenaria* contained the highest levels of Fe and Mn. Tissue metals such as Cd and Ni were accumulated to the greatest extent by *Mytilus edulis* and *Cardium glaucum*, respectively. The data obtained here are in agreement with those reported previously by Brzezińska et al. (1984) for the southern Baltic molluscs; *Macoma balthica* accumulated particularly strongly Cu and Zn but Cd less efficiently than *Mytilus edulis*. Bryan (1980) recorded also significant bioavailability of these metals to East Looe Estuary molluscs; Zn and Cu had a maximum bioavailability to *Macoma balthica*, Ni to *Cerastoderma edule* (*Cardium edule*) and Cd to *Mytilus edulis*. It suggests that these molluscs as non-regulators incorporate quickly the trace metal levels from the environment because of their elevated biological tolerance and/or limited elimination with respect to the selected metals.

To characterize the region-dependent variations, the data concerning *Macoma balthica* taken in the same period but from different sites of the southern Baltic were compared. The highest levels of the trace metals analysed, except for Cu, occurred in the soft tissue of *Macoma balthica* from the Vistula estuary. This is to be expected since the area borders immediately on a highly urbanized and industrialized centre and hence is exposed to an anthropogenic flux of metals (Szefer, 1990a, 1990b; Szefer & Skwarzec, 1988). As far as the variations of concentrations in the shell are concerned, *Macoma balthica* from stations P-2 and ZN-2 contained maximum levels of Zn, Pb and Mn, Fe, respectively. Specimens of this species taken from station ZP incorporated the highest amount of Cu. The results obtained suggest that *Macoma balthica* and *Mytilus edulis* may be good bioindicators of metal pollution since, according to several authors (Bryan,

Table 1. The concentrations (mean \pm s. d., and range) of Zn, Cu, Pb, Cd, Ni, Co, Ag, Mn, Fe, Ca, Mg, K and Na (mg g⁻¹ dry wt.) in the soft tissue and shell of molluscs from the southern Baltic. T = soft tissue; S = shell; BLD = below limit of detection; NA = not analysed

Species of mollusc and code of station	Part of the mollusc	No. of pooled samples (Total No. of specimens)	Zn	Cu	Pb	Cd	Ni	Co	Ag	Mn	Fe	Ca	Mg	K	Na		
<i>Macoma balthica</i>																	
ZP	T	6(140)	335 \pm 53 174-476	72.0 \pm 5.0 60.2-88.2	3.6 \pm 0.8 2.2-7.6	1.59 \pm 0.20 0.97-2.15	3.9 \pm 0.5 2.3-5.2	3.7 \pm 0.3 2.4-6.1	BLD-3.6	35.1 \pm 2.8 28.8-46.9	0.88 \pm 0.14 1.04-1.67	1.91 \pm 1.02 0.89-2.93	1.10 \pm 0.09 0.63-1.38	7.46 \pm 0.43 6.77-8.06	8.4 \pm 0.6 7.1-11.1	NA	
	S	10(140)	16.0 \pm 1.4 10.3-23.1	20.8 \pm 1.7 14.1-30.4	1.5 \pm 0.3 BLD-3.6	BLD	BLD	BLD	BLD	NA	11.3 \pm 0.8 7.5-16.8	0.28 \pm 0.03 0.13-0.43	NA	NA	NA	NA	NA
GN	T	11(141)	520 \pm 46 254-703	32.0 \pm 1.1 26.6-38.4	3.1 \pm 0.5 1.6-6.6	2.08 \pm 0.17 1.03-2.98	5.8 \pm 0.6 3.6-10.0	4.6 \pm 0.7 1.5-8.7	BLD-4.2	46.3 \pm 3.7 19.1-60.1	0.62 \pm 0.03 0.48-0.82	2.88 \pm 1.22 1.19-3.25	0.99 \pm 0.08 0.77-1.46	6.61 \pm 0.21 5.68-8.52	12.4 \pm 1.4 8.4-24.9	NA	NA
	S	16(141)	8.6 \pm 0.6 6.6-13.6	13.9 \pm 0.7 10.0-19.7	0.8 \pm 0.1 BLD-1.7	BLD	BLD	BLD	BLD	NA	18.0 \pm 0.8 13.8-24.4	0.10 \pm 0.01 0.06-0.14	NA	NA	NA	NA	NA
P-2	T	5(134)	415 \pm 20 300-471	97.8 \pm 8.7 80.7-128.6	3.7 \pm 0.5 2.1-5.1	1.73 \pm 0.12 1.43-2.11	3.1 \pm 0.4 2.2-4.2	3.5 \pm 0.4 1.9-4.2	3.8 \pm 0.5 2.8-5.2	25.4 \pm 2.5 17.5-32.1	0.87 \pm 0.05 0.58 \pm 0.03	2.28 \pm 0.07 2.19-2.32	0.95 \pm 0.15 0.62-1.36	8.02 \pm 0.34 6.92-8.96	10.4 \pm 3.0 5.0-17.8	NA	NA
	S	9(134)	29.4 \pm 2.1 20.2-36.0	15.2 \pm 0.9 10.9-18.6	2.6 \pm 0.3 1.6-4.3	BLD	BLD	BLD	BLD	NA	15.3 \pm 1.0 10.0-18.7	0.76 \pm 0.05 0.47-0.76	NA	NA	NA	NA	NA
ZN-2	T	2(33)	629 \pm 272 355-900	38.6 \pm 5.2 33.3-43.8	11.2 \pm 1.0 10.2-12.2	2.70 2.01-3.70	12.2 \pm 3.8 8.4-16.1	17.9 \pm 7.8 10.1-25.7	BLD	83.6 \pm 40.6 43.0-124.2	2.42 \pm 1.00 1.40-3.43	NA	0.60 \pm 0.28 0.33-0.88	4.20 \pm 2.59 1.61-6.79	7.7	NA	NA
	S	3(33)	18.5 \pm 3.1 12.5-22.7	16.6 \pm 1.3 15.4-19.2	1.9 \pm 0.4 BLD-2.4	BLD	BLD	BLD	BLD	NA	34.4 \pm 46.8 0.38-1.00	0.79 \pm 0.20 0.38-1.00	NA	NA	NA	NA	NA
<i>Mya arenaria</i>																	
ZN-2	T	5(62)	212 \pm 43 130-318	23.8 \pm 2.0 20.5-31.8	6.2 \pm 2.0 3.5-8.8	2.28 \pm 0.28 2.01-3.70	9.9 \pm 2.0 5.3-17.3	10.1 \pm 2.9 3.7-20.7	BLD	24.5 \pm 6.4 14.9-48.7	12.63 \pm 1.31 10.33-17.53	1.17 \pm 0.39 0.78-1.56	1.56 \pm 0.24 1.00-2.33	6.77 \pm 0.47 5.02-6.74	NA	NA	NA
	S	5(62)	13.9 \pm 1.5 8.8-17.4	2.5 \pm 0.7 0.6-4.4	1.9 \pm 0.6 BLD-3.8	BLD	BLD	BLD	BLD	NA	52.5 \pm 7.5 32.6-76.7	2.21 \pm 0.25 1.30-2.72	NA	NA	NA	NA	NA
<i>Cardium glaucum</i>																	
ZN-2	T	4(45)	98 \pm 8 83-114	24.3 \pm 2.3 17.4-27.5	7.8 \pm 0.1 7.6-8.0	5.71 \pm 1.51 3.04-9.90	3.96 \pm 7.0 30.0-59.8	5.7 \pm 1.9 3.3-11.4	BLD	60.4 \pm 5.3 47.4-71.8	2.23 \pm 0.35 1.73-3.25	NA	2.06 \pm 0.16 1.65-2.41	NA	NA	NA	NA
	S	7(45)	9.0 \pm 0.8 6.5-11.9	2.7 \pm 0.2 1.9-3.5	0.9 \pm 0.2 BLD-2.1	BLD	BLD	BLD	BLD	NA	24.4 \pm 40.0 63.0-136.0	0.88 \pm 0.07 0.67-1.18	NA	NA	NA	NA	NA
<i>Mytilus edulis</i>																	
GN	T	6(68)	125 \pm 14 91-167	13.4 \pm 1.2 11.0-16.3	2.5 \pm 0.4 1.3-3.5	7.14 \pm 0.79 4.97-9.54	5.1 \pm 0.6 4.2-7.3	2.8 \pm 0.5 1.7-4.4	BLD	73.4 \pm 11.1 48.1-117.1	0.39 \pm 0.04 0.24-0.47	2.19 \pm 0.93 1.27-3.11	2.28 \pm 0.19 1.78-3.00	10.25 \pm 0.40 9.02-12.21	25.3 \pm 3.8 15.4-32.9	NA	NA
	S	12(68)	9.5 \pm 1.2 5.1-17.7	3.2 \pm 0.2 1.9-4.7	1.0 \pm 0.2 BLD-2.0	BLD	BLD	BLD	BLD	NA	99.4 \pm 7.2 63.0-136.0	0.05 \pm 0.01 0.02-0.08	NA	NA	NA	NA	NA
<i>Astarte borealis</i>																	
B-2	T	5(64)	128 \pm 7 107-148	54.2 \pm 15.4 34.7-115.6	BLD	30.9 \pm 2.7 23.8-40.9	21.3 \pm 2.2 15.7-27.7	20.5 \pm 4.1 11.6-31.5	BLD	19.84 \pm 0.21 [*] 14.66-26.71	3.96 \pm 0.23 3.36-4.76	1.88	1.09 \pm 0.15 0.94-1.53	12.16 \pm 3.78 5.70-25.3	NA	NA	NA
	S	8(64)	97.0 \pm 9.7 56.0-131.0	5.4 \pm 0.6 3.4-7.8	9.9 \pm 1.2 5.3-13.4	0.44 \pm 0.06 0.15-0.60	12.7 \pm 1.5 8.4-17.4	14.8 \pm 2.8 8.3-19.8	NA	34.1 \pm 3.4 [*] 18.2-45.0	8.1 \pm 0.7 5.8-11.9	NA	NA	NA	NA	NA	NA

* mg g⁻¹ dry wt

Table 2. The concentrations (mean \pm s. d., and range) of Zn, Cu, Pb, Cd, Ni, Co, Ag, Mn, Fe, Ca, Mg, K and Na (mg g^{-1} dry wt.) in a soluble in 1M HCl fraction of surface sediments of the southern Baltic. Results are mean values of triplicate analysis. BLD = below limit of detection

Code of station	Organic matter (%)	Zn	Cu	Pb	Cd	Ni	Co	Ag	Mn	Fe	Ca	Mg	K	Na
ZP	9.8	68.4 \pm 1.7	26.9 \pm 1.5	62.1 \pm 2.6	1.5 \pm 0.2	8.0 \pm 1.2	6.4 \pm 1.0	BLD	190 \pm 5	10.1 \pm 0.5	10.6 \pm 4.5	3.96 \pm 0.13	1.54 \pm 0.02	2.3 \pm 0.1
		65.6-71.4	24.6-29.6	56.9-65.3	1.2-1.9	5.9-10.0	4.9-8.2	BLD	182-199	9.1-10.9	5.4-19.5	3.71-4.17	1.51-1.58	2.1-2.4
GN	6.8	18.6 \pm 1.1	6.5 \pm 0.5	5.7 \pm 0.5	0.5 \pm 0.2	2.6 \pm 0.2	1.0 \pm 0.2	BLD	30.5 \pm 1.7	0.80 \pm 0.03	6.9 \pm 0.9	0.64 \pm 0.03	0.52 \pm 0.08	4.9 \pm 0.5
		17.3-20.8	5.6-7.1	5.2-6.2	0.3-0.7	2.1-3.2	0.8-1.4	BLD	28.4-33.9	0.77-0.86	5.4-8.4	0.60-0.69	0.41-0.68	4.4-5.3
P-2	11.6	66.6 \pm 0.4	26.2 \pm 0.2	75.0 \pm 1.2	1.6 \pm 0.1	10.5 \pm 0.3	6.4 \pm 0.7	BLD	96.5 \pm 1.4	0.87 \pm 0.02	0.84 \pm 0.02	0.12 \pm 0.00	0.59 \pm 0.01	7.7 \pm 0.1
		66.1-67.3	25.9-26.7	72.5-76.4	1.4-1.8	10.2-11.2	4.7-6.7	BLD	93.8-98.2	0.83-0.90	0.80-0.88	0.11-0.12	0.56-0.62	7.7-7.8
ZN-2	0.65	6.7 \pm 0.6	4.3 \pm 0.1	5.7 \pm 0.1	BLD	1.9 \pm 0.5	1.3 \pm 0.3	BLD	16.0 \pm 0.9	0.40 \pm 0.01	5.1 \pm 0.3	0.16 \pm 0.02	0.29 \pm 0.05	15.7 \pm 0.6
		5.5-7.5	4.1-4.5	5.3-6.1	BLD	1.3-2.4	0.7-1.6	BLD	14.4-17.3	0.38-0.41	4.6-5.7	0.13-0.19	0.24-0.34	15.1-16.2
B-2	1.53	28.7 \pm 0.6	9.2 \pm 0.3	19.4 \pm 0.6	1.1 \pm 0.1	3.9 \pm 0.9	2.3 \pm 0.1	BLD	916 \pm 1	1.06 \pm 0.01	9.6 \pm 1.6	1.25 \pm 0.09	0.72 \pm 0.06	2.0 \pm 0.1
		27.6-29.6	8.6-9.7	18.3-20.5	1.0-1.2	2.1-4.9	2.1-2.4	BLD	915-917	1.05-1.08	6.5-11.4	1.16-1.34	0.61-0.82	1.9-2.1
P-10	-	74.0 \pm 4.1	27.7 \pm 2.0	53.6 \pm 4.9	1.3 \pm 0.3	6.8 \pm 0.3	6.3 \pm 0.7	BLD	85.1 \pm 4.8	1.57 \pm 0.07	13.1 \pm 7.7	3.19 \pm 0.14	2.47 \pm 0.11	15.8 \pm 0.1
		68.6-81.9	25.0-31.5	44.4-60.8	1.1-1.5	6.3-7.4	5.1-7.4	BLD	76.3-92.6	1.49-1.64	2.7-28.2	2.94-3.43	2.32-2.69	15.7-16.0
G-2	-	98.2 \pm 0.5	30.1 \pm 0.2	71.6 \pm 5.7	1.6 \pm 0.1	7.4 \pm 1.1	6.7 \pm 0.1	BLD	233 \pm 3	6.73 \pm 0.16	9.5 \pm 3.9	4.09 \pm 0.10	2.57 \pm 0.17	0.70 \pm 0.02
		97.3-99.1	29.7-30.5	62.9-82.4	1.4-1.7	5.8-9.4	6.6-6.8	BLD	255-249	6.41-6.94	2.4-15.7	3.98-4.29	2.33-2.90	0.69-0.73

Table 3. The concentrations (mean \pm s. d., and range) of Zn, Cu, Pb, Cd, Ni, Co, Ag ($\mu\text{g g}^{-1}$), Mn and Fe (mg g^{-1}) in a soluble in 1M HCl fraction of ferromanganese nodules of Słupsk Furrow, the southern Baltic. Results are means of triplicate analysis. BLD = below limit of detection

Layer of nodule	Zn	Cu	Pb	Cd	Ni	Co	Ag	Mn	Fe
Surface top	165 \pm 3	22.7 \pm 1.3	26.7 \pm 3.3	BLD	33.3 \pm 2.5	19.2 \pm 0.6	BLD	17.5 \pm 1.3	171 \pm 7
	161–171	20.3–24.9	20.2–31.1		30.0–38.0	18.3–20.3		15.0–19.2	160–185
Subsurface top	167 \pm 3	15.2 \pm 0.8	BLD	BLD	16.2 \pm 1.3	15.1 \pm 2.0	BLD	14.6 \pm 1.9	156 \pm 5
	163–173	13.8–16.6			13.8–18.1	11.9–18.6		12.0–18.5	146–161

1980; Goldberg et al., 1978; Phillips, 1980), among the properties required of such organisms are that they should be easily recognised, widely distributed, common, accessible, sensitive to locally dependent variations of the trace metals, available at all times of year, relatively stationary and sufficiently tolerant of low salinity. The latter property is very important for estuarine and near estuarine areas, represented here by the Vistula estuary. It is noteworthy that all these requirements for the two molluscs investigated are entirely satisfied. Moreover, specimens of *Macoma balthica* and especially *Mytilus edulis* often reach a relatively substantial weight and length of shell; hence, their suitability in the preparation of pooled samples composed of not numerous specimens belonging to the same or similar size (age) population. The age-dependence of shell length in *Macoma balthica* from the Gdańsk Bay was described by the logarithmic curve and the von Bertalanffy equation (Wenne & Klusek, 1985). A relation between size and age of *Mytilus edulis* and *Cardium glaucum* from this area has also been presented previously (Barron & Wołowicz, 1981; Wołowicz, 1984).

Bearing in mind that one species only, i. e. *Astarte borealis*, was taken at station B-2, it is impossible to determine both the interspecies- and region-dependent variations of metal concentrations. The significantly elevated levels of tissue metals (Cd, Co, Mn and Fe) as well as shell metals (Zn, Pb, Cd, Co, Mn and Fe) in this species originated probably from ambient ferromanganese nodules. This material covered closely the surface area of the shell, and contained high concentrations of the metals, significantly higher than those in the ambient surface sediments (Tables 2 and 3). Moreover, fine-grained fragments of nodules accompanied numerous specimens of *Astarte borealis* during each and every haul.

Metals in surface sediments

As can be seen from Table 2, minimum amounts of Zn, Cu, Ni, Mn, Fe and K as well as organic matter were found in a labile fraction of sediments from the station ZN-2 (the Vistula estuary). The levels of Pb, Cd and Co were also low, similar to those in sediments of the station GN. Since the Vistula estuary sediments are represented by coarse-grained sands mixed with very small amounts of organic matter, the total levels of the trace metals in these sediments were, therefore, very low. According to Boström et al. (1981) and Krishnaswami & Sarin (1976), trace metals may be concentrated in particulate matter by biological activities. It concerns especially metals such as Zn, Cu, Pb and Cd, as postulated by Di Giulio & Scanlon (1985). The authors observed a significant correlation between the concentration of these four metals and organic matter in surface sediments

from the Chesapeake Bay. Such dependence may be used to normalize samples in order to distinguish the variability of sediment samples reflecting background metal levels and samples influenced by industrial contamination (Di Giulio & Scanlon, 1985). It should be kept in mind that the Vistula estuary sediments contain a high amount of acid-insoluble matter, essentially composed of an admixed trace metal-poor and silicate-rich dilutant. The concentrations recalculated to an acid-insoluble free fraction or to organic matter content showed that sediments, like mollusc soft tissue, from the Vistula estuary concentrated most trace metals. On the other hand, the analysis of the estuarine sandy sediments for total metal concentrations is insufficient for pollution control because of too high a percentage share of silicate dilutant. Moreover, the concentration of metals in sediments, unlike that in organisms, might not represent the time-integrated value of the biologically available metals.

Metals in ferromanganese nodules

The concentrations of Mn, Fe, Zn and Cu in a soluble in 1M HCl fraction of ferromanganese nodules of Słupsk Furrow were smaller but within the same order of magnitude compared with those in nodules from the Gulf of Bothnia, Baltic Sea (Boström et al., 1982). The significantly lower results obtained here than those given by Suess & Djafari (1977) for western Baltic nodules may be a result not only of natural variations but also of the different analytical methods used. Suess & Djafari provided bulk chemical composition data whilst results given in the present study concern an acid leachate fraction. Bearing in mind that, for example, Mn, Cd, Pb, Co, Ni, Cu and Zn are preferentially concentrated in a major phase of ferromanganese nodules such as Mn-oxides (Li, 1982), lower values for Zn, Cd, Pb, Cu and Co in an acid-soluble fraction than in bulk material may be explained by incomplete dissolution of Mn-matrix in cold 1M HCl during 2 h. It is noteworthy that the concentrations of Cu, Co and particularly Pb and Ni were higher in the surface layers of the nodules than in the deeper layers. It suggests that these metals are of anthropogenic origin and/or are more available to efficient leaching from surface parts by 1M HCl.

Relationship between soft tissue and shell metals

In order to determine the relation between both the shell and tissue concentrations of metals, we calculated the ratio (FR) of metal content and metal concentration in these parts of molluscs (Table 4). The concentration of metals in almost every case is higher in the dry soft tissue than in the shell. However FR values regarding the ratio of metal content in shell to metal content in the dry soft tissue were generally ≥ 1 for Cu, Pb, Mn and Fe; the $FR < 1$ was recorded in principle for Zn and Cd. Koide et al. (1982) also obtained the FR values < 1 for Zn and Cd in *Mytilus edulis* from the West and East Coast of U.S.A.

The correlation coefficients (r) for metal between its concentration in the soft tissue and in the shell of *Macoma balthica* are presented in Table 5. Since a higher number of pooled samples of shell than of soft tissue was analysed, the weighted average of the shell metal concentrations attributed to the corresponding tissue data was computed before correlation analysis, in order to obtain the same number of both. Regarding Mn concen-

Table 4. Ratio (FR) of metal content in the shells to the dried soft tissues of the southern Baltic molluscs, calculated by dividing the total amounts of metal in the shell sample by those in the soft tissue sample; ratio of metal concentrations (in $\mu\text{g g}^{-1}$ dry wt.) is given for comparison in parentheses

	Zn	Cu	Pb	Cd ^a	Ni ^a	Co ^a	Mn	Fe
<i>Macoma balthica</i>								
ZP	0.2(0.05)	1.2(0.3)	1.7(0.4)	<0.08(<0.02)	<1.1(<0.3)	<0.5(<0.1)	1.3(0.3)	1.1(0.3)
GN	0.1(0.02)	3.0(0.4)	1.6(0.2)	<0.07(<0.01)			2.6(0.4)	1.0(0.2)
P-2	0.3(0.07)	0.7(0.2)	2.9(0.7)	<0.08(<0.02)			2.4(0.6)	2.7(0.7)
ZN-2	0.2(0.03)	2.4(0.4)	0.6(0.1)				2.7(0.5)	1.8(0.3)
<i>Mya arenaria</i>								
ZN-2	0.3(0.07)	0.4(0.1)	1.1(0.3)				0.8(0.2)	0.7(0.2)
<i>Cardium glaucum</i>								
ZN-2	1.5(0.09)	1.9(0.1)	1.9(0.1)				8.5(0.5)	6.8(0.4)
<i>Mytilus edulis</i>								
GN	0.4(0.08)	1.2(0.2)	2.1(0.4)				7.3(1.4)	0.6(0.1)
<i>Astarte borealis</i>								
B-2	24(0.76)	2.9(0.1)	>32(>1.0)	0.32	19	23	56(1.8)	65(2.0)
^a FR values for Cd, Ni and Co were < 1								

Table 5. Correlation coefficients (r) for metals between their concentrations in the soft tissue and in the shell of *Macoma balthica* from the southern Baltic

Station	No. of pooled samples	Zn	Cu	Mn	Fe
ZP	6	0.44	0.77 ^b	0.71 ^b	0.03
GN	11	-0.50	0.59 ^b	0.40	0.31
P-2	5	-0.76 ^b	-0.79 ^b	0.94 ^a	0.36
Altogether ^c	24	0.15	0.22	0.78 ^a	0.37
^a p < 0.01					
^b p < 0.05					
^c The metal concentrations for two samples from station ZN-2 were additionally included					

trations, the r value between the soft tissue and the shell is estimated to be 0.78. Only a weak correlation between shell and soft tissue concentrations for 24 samples of *Macoma balthica* is obtained for Zn and Cu, although greater correlation coefficients (positive or negative) for the two metals were found in samples from particular stations. On the other hand, there is strong covariance between concentrations of shell metals, unlike tissue metals, for the following assemblages: Zn-Cu, Cu-Fe, Zn-Fe and partly Fe-Mn (Table 6). According to Koide et al. (1982), the reason for strong shell correlation presumably relates to differences in the biochemical behaviour of the metals in the period between the uptake by the organism and the release to the environment or to the shell. Moreover,

Table 6. Correlation coefficients (r) between metal concentrations in the shell and in the soft tissue of *Macoma balthica* from the southern Baltic

Metal pair	Correlation coefficient				
	Station	ZP (n=7)	GN (n=11)	P-2 (n=6)	Altogether (n=27) ^c
Shell					
Zn-Cu		0.75 ^b	0.49	0.94 ^a	0.35 ^b
Cu-Fe		0.90 ^a	0.50	0.88 ^a	0.79 ^a
Zn-Fe		0.86 ^a	0.06	0.78 ^b	0.87 ^a
Zn-Mn		-0.11	0.38	0.95 ^a	0.09
Cu-Mn		-0.45	0.27	0.91 ^a	-0.13
Fe-Mn		-0.12	0.57 ^b	0.76 ^b	0.38 ^b
Soft tissue					
Zn-Cu		0.83 ^a	0.20	0.38	-0.27
Cu-Fe		-0.23	0.45	0.64	-0.10
Zn-Fe		0.04	-0.31	-0.06	0.48 ^a
Zn-Mn		-0.09	0.02	-0.65	0.56 ^a
Cu-Mn		0.43	-0.09	-0.25	-0.49 ^a
Fe-Mn		-0.06	0.13	0.12	0.10
^a p < 0.01 ^b p < 0.05 ^c The metal concentrations for three samples from station ZN-2 were additionally included					

factors such as a longer biological half-life in the shell than in the soft tissue and perhaps a relatively uniform pumping of metal from soft tissue to shell paralleling shell growth may also be responsible for the stronger correlations between metal concentrations in the shell than in the soft tissue. Therefore, Koide et al. (1982) suggest that the shell material as whole life integrator of metals may be a better recorder for environmental metal levels than soft tissue. This suggestion is supported by markedly higher concentrations of metals in shells found near industrialized and populated areas than in regions devoid of anthropogenic activity (Koide et al., 1982). Although use of hard parts as well as soft tissues of molluscs as recorders of metallic pollutions is clearly promising, further research must be performed, according to projectors of „Mussel Watch“ (Goldberg et al., 1978; Phillips, 1977a, 1980), on the effects of various factors on the metal levels in shell. These include ontogenetic metal variations (Carriker et al., 1982), the interaction of mineral elements in seawater and shell (Carriker et al., 1980b), the effect of weathering on the elemental composition of shells (Rosenberg, 1980), mineralogy of the shell and heterogeneous distribution of metal in shell layers (Carriker et al., 1980a; Rosenberg, 1980; Wada & Suga, 1976), the proportion of metals adsorbed to the surface and incorporated into the shell matrix (Phillips, 1980), the influence of environmental metal variations on the active incorporation of metals during shell formation (Pilkey & Harriss, 1966). Moreover, the effect of other environmental parameters such as temperature and salinity (Pilkey & Goodell, 1963) require further study.

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