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Uranium and thorium in muscle tissue of fish taken from the southern Baltic

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ABSTRACT: The determination of U and Th was carried out on pooled samples of muscle tissue of cod *(Gadus morhua)*, herring *(Clupea harengus)*, sprat *(Sprattus sprattus)* and some other species of fish caught in 1981 in the southern Baltic. The levels of U obtained in the present study are generally within the ranges presented by other authors for fish from different aquatic regions of the world. The concentrations of U are similar to those found for Th in Baltic fish analysed. The concentration factors $CF_{U(Th)}$, discrimination factors $DF_{U/Th}$ and Th/U ratios (by weight) were calculated and discussed. These coefficients were also compared with values determined in other Baltic organisms. Transfer of U and Th along a food chain from potential prey (diet) to fish as predator (consumer) was quantified with a transfer factor.

INTRODUCTION

The concentration and distribution of U and Th in various environmental components, especially in marine food such as plankton, macroalgae, corals, molluscs, crustacea and ducks, have been investigated by several authors (Amiel et al., 1973; Bangera & Patel, 1984; Blanchard & Oakes, 1965; Blanchard et al., 1967; Chassard-Bouchaud, 1982a, 1983; Chassard-Bouchaud & Escaig, 1984; Edgington et al., 1970; Flor & Moore, 1977; Gvirtzman et al., 1973; Hamilton, 1980; Hodge et al., 1979; Holm & Persson, 1980; Kharkar et al., 1976; Knauss & Ku, 1983; Koide et al., 1982; Livingston & Thompson, 1971; Miyake et al., 1970; Möller et al., 1983; Omura, 1976; Schroeder et al., 1970; Schwochau et al., 1976; Sugimura & Mayeda, 1980; Szefer, 1987; Szefer & Falandysz, 1983; Szefer & Wenne, 1987; Thompson & Livingston, 1970; Tsytsugina et al., 1973; Veeh & Turekian, 1968). A survey of the literature (on the basis of 74 articles) concerning the concentration, discrimination and distribution of U in marine biosphere has been presented by Szefer (1989). The migration of natural alpha-emitting radionuclides in the marine biosphere is an interesting problem from a radioecological and hygienic point of view. To anticipate effects of the causal release of radionuclides from nuclear power plants to the sea, it is important to understand the mechanisms by which stable isotopes like U and Th are concentrated and transported along the food chain of an ecosystem. Aquatic organisms are most likely to be affected by radionuclide release; hence data on their natural concentrations would be helpful as reference values (Edgington et al., 1970).

There is only limited information available on the concentration and distribution of U in marine fish (Aten et al., 1961; Chassard-Bouchaud, 1982b; Hamilton, 1972; Ichikawa &

Ohno, 1981; Pentreath et al., 1979; Tsytsugina et al., 1973); a wide range of levels of U in muscle of fish has been reported, i.e. from < 0.1 to 40 ng/g on a wet-weight basis. Analysis of ^{232}Th resulted in negative results; it was computed that this isotope was present in concentrations lower than 0.5 ng/g wet weight in muscle of North Sea fish (Pentreath et al., 1979). Thus, it is important to know to what degree U and Th are incorporated in respective Baltic organisms. The aim of this paper is to report the concentrations of U and Th in muscle tissue of some commercially utilized fish of the southern Baltic. In order to estimate the degree of concentration, discrimination and transfer for U and Th in the Baltic environment, we utilized and recalculated their mean levels obtained here, as well as those recorded previously for other representatives of biosphere.

MATERIALS AND METHODS

The fish was caught by pelagic krill and/or herring trawls in July 1981 during the cruise of the R.V. "Profesor Siedlecki" and R.V. "Birkut". The localization of the sampling sites (fisheries) with the codes is presented in Figure 1. The material has also been utilized previously to determine eight trace metals after their separation by an anion exchange procedure (Szefer & Falandysz, 1985). The U and Th fractions collected were combined respectively to obtain composite eluates equivalent to pooled samples weights of maximum ca 500 g (on a wet weight basis). The sample preparation and details of the

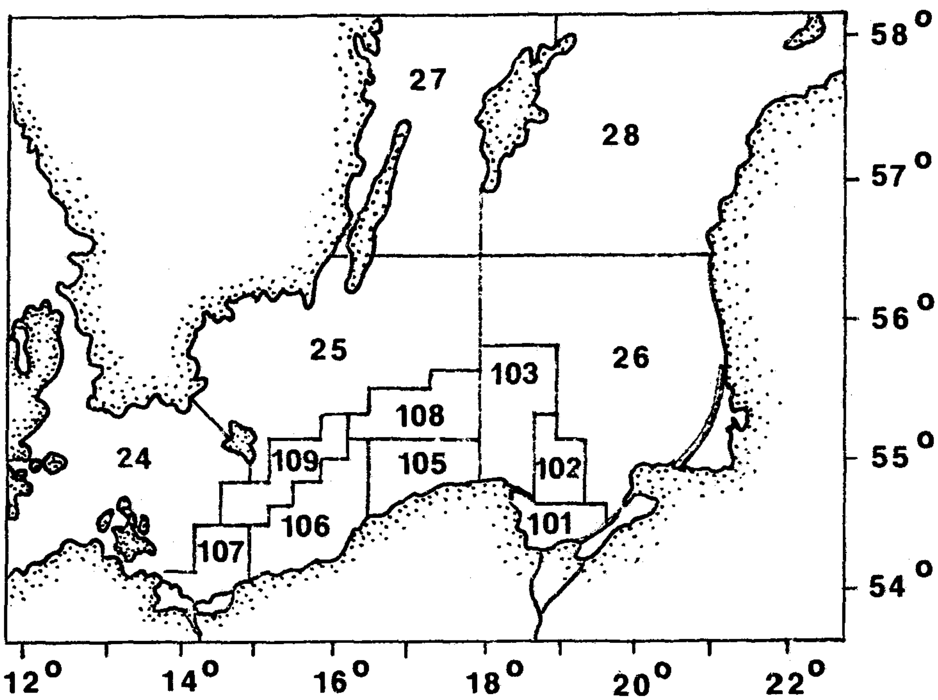

Fig. 1. Location of the sampling areas (fisheries) in the southern Baltic marked by the codes: 101 Gdańsk Bay, 102 Gdańsk Deep, 103 Władysławowskie fishery, 105 Ustecko-Łebskie fishery, 106 Kołobrzesko-Darłowskie fishery, 108 Słupsk Furrow fishery, 109 Bornholm S fishery

analytical procedure have been presented elsewhere (Szefer, 1987; Szefer & Falandysz, 1983, 1985). Briefly, ashed samples were converted into nitrates by evaporation with concentrated HNO_3. The nitrate salts were dissolved in 8 M HNO_3 and then passed through an anion exchanger Dowex 1 ×4 (NO_3^-) to adsorb Th, eluted with 10 M HCl. The eluent containing nitrates was evaporated to dryness and nitrates converted into chlorides by evaporation with concentrated HCl. The chloride salts were dissolved in 1 M HCl and aliquot was destined to determine some microelements by AAS method.

The remaining part of the solution was evaporated to dryness and dissolved with 9 M HCl. The solution was passed through an anion exchanger Dowex 1 × 8 (Cl^-) to isolate U from most of the other metallic ions which were analysed by AAS (Szefer & Falandysz, 1985). The U and Th were determined spectrophotometrically with Arsenazo III (Szefer, 1987; Szefer & Falandysz, 1983). The average limit of detection for U and Th was 0.20 ng/g w.w.

RESULTS AND DISCUSSION

As can be seen from Table 1 the mean levels of U in the muscle tissue of cod *(Gadus morhua)*, herring *(Clupea harengus)* and sprat *(Sprattus sprattus)* amounted to 0.30, 0.29 and 0.44 ng/g on a wet-weight basis, respectively. These species contained similar concentrations of Th, i.e. 0.41, 0.38 and 0.64 ng/g w.w., respectively. Among the other species analysed, the muscle levels of U and Th varied between 0.22–0.42 ng/g and 0.43–1.6 ng/g w.w., respectively. Elevated levels of U occurred in the whole specimens of stickleback *(Gasterosteus aculeatus)* (2.6 ng/g w.w.). The muscle-levels of U (BLD-0.65 ng/g w.w.) obtained in this study are comparable with the lowest values of 0.24 and 0.98 ng/g w.w. reported by other authors (Ichikawa & Ohno, 1981; Tsytsugina et al., 1973) for Japanese and Black Sea fish, respectively.

The ratio of Th/U (by weight) for *Gadus morhua, Clupea harengus* and *Sprattus sprattus* is near unity (on an average 1.37).

The mean levels of U and Th in fish and other Baltic organisms (Table 2) were utilized (after conversion: dry to wet weight) to calculate the concentration factor CF:

$$CF = (\text{ng U(Th)/g wet organism})/(\text{ng U(Th)/g seawater})$$

To calculate this factor, the average southern Baltic water data for U and Th were taken from articles of Szefer (1977) and Bojanowski & Szefer (1979). The salinity of surface water in this region was between 6.58 and 9.16‰ (on the average 7.7‰). The concentration of U ranged from 0.65 to 1.06 μg/l (on the average 0.87 μg/l) in open Baltic and from 0.26 to 1.06 μg/l (on the average 0.64 μg/l) in coastal water. The correlation between the U concentration and the salinity was 0.962; hence a dependence of the CF_U upon the salinity to be expected.

Table 3 presents values of CF_U and CF_{Th} for fish as well as for other representatives of Baltic biocenosis. These data reveal that the soft tissue of molluscs and whole specimens of *Mesidothea entomon* were characterized by maximum concentration of U and Th, respectively, whilst in fish the smallest amounts of the two elements were concentrated. The mean CF_U and CF_{Th} values for fish muscle were 0.5 and 10, respectively. For comparison, similar muscle value of CF_U (0.3) was estimated by other authors (Ichikawa & Ohno, 1981) for Japanese species of marine fish, i.e. for file fish *(Stephanolepis cirrhifer)*, horse mackerel *(Trachurus japonicus)* and mackerel *(Scomber japonicus)*. It is

Table 1. Mean concentration of U and Th in ng/g on a wet weight basis (± standard deviation) in muscle tissues of fish from the southern Baltic

Species, locality (fishery)	No.	U	No.	Th
Cod *(Gadus morhua)*				
Gdańsk Bay 101*	4 (6)**	0.21 ± 0.01	6	0.38 ± 0.06
Gdańsk Deep 102	3 (11)	0.25 ± 0.06	11	0.63 ± 0.05
Władysławowskie (Northern part) 103	4 (8)	0.30 ± 0.07	8	0.33 ± 0.05
Władysławowskie (Southern part) 103	3 (8)	0.30 ± 0.06	6	0.32 ± 0.07
Kołobrzesko-Darłowskie 106	4 (6)	0.24 ± 0.00	6	0.40 ± 0.11
Słupsk Furrow 108	4 (6)	0.49 ± 0.09	10	0.34 ± 0.06
Bornholm S 109	4 (10)	0.32 ± 0.06	10	0.39 ± 0.03
Average	26 (55)	0.30	55	0.41 ± 0.06
Herring *(Clupea harengus)*				
Gdańsk Bay 101	5 (60)	0.25 ± 0.06	9 (60)	0.34 ± 0.06
Gdańsk Deep 102	1 (10)	0.21	2 (20)	0.30
Władysławowskie 103	2 (20)	BLD+−0.29	5 (35)	0.44 ± 0.12
Ustecko-Łebskie 105	2 (20)	BLD−0.65	3 (30)	0.37 ± 0.14
Kołobrzesko-Darłowskie 106	2 (30)	0.28	5 (30)	0.34 ± 0.09
Słupsk Furrow 108	1 (30)	BLD	4 (30)	0.55 ± 0.01
Bornholm S 109	1 (10)	BLD	2 (30)	0.22
Average	14 (180)	0.29	30 (235)	0.38
Sprat *(Sprattus sprattus)*				
Gdańsk Bay 101	1 (45)	0.44	3 (45)	0.82 ± 0.06
Kołobrzesko-Darłowskie 106	1 (15)	BLD	1 (15)	0.34
Słupsk Furrow 108	–	–	1 (15)	0.67
Bornholm S 109	1 (15)	BLD	1 (15)	BLD
Average	–	–	6 (90)	0.64
Flounder *(Platichthys flesus)*				
Gdańsk Bay 101	1 (8)	0.22	1 (8)	0.43
Kołobrzesko-Darłowskie 106	–	–	1 (9)	0.53
Garfish *(Belone belone)*				
Władysławowskie 103	1 (2)	0.30	2 (2)	1.30
Kołobrzesko-Darłowskie 106	1 (2)	0.42	2 (2)	0.63
Eel *(Anguilla anguilla)*				
Gdańsk Bay 101	–	–	1 (1)	0.66
Whiting *(Merlangus merlangus)*				
Bornholm S 109	–	–	1 (1)	1.60
Salmon *(Salmo salar)*				
Ustecko-Łebskie 105	–	–	1 (1)	0.50
Greater sand eel *(Hyperoplus lanceolatus)*++				
Gdańsk Bay 101	1 (10)	0.44	2 (10)	1.20
Stickleback *(Gasterosteus aculeatus)*++				
Gdańsk Bay 101	1 (16)	2.60	1 (16)	1.0

* Code of the fishery
** Pooled samples; number of samples and total number of fish (in brackets)
+ BLD – below limit of detection
++ Whole fish

Table 2. Concentrations of U and Th (μg/g on a dry weight basis) in various representatives of southern Baltic biocenosis*

Representative of biocenosis	U	Th
Seaweed	0.23	0.24
Zooplankton	0.14	0.19
Molluscs (whole)	0.04	0.06
Crustacea (whole):		
Mesidothea entomon	0.03	0.30
Fish (muscle)	0.0013	0.0018

* Data taken or recalculated from Szefer (1987), Szefer & Wenne (1987) and the present paper

Table 3. Mean concentration factors (CF) and discrimination factors (DF) for U and Th in various representatives of southern Baltic biocenosis. To calculate the CF and DF factors, Baltic water data were taken from articles of Szefer (1977) and Bojanowski & Szefer (1979)

Biocenosis component	CF_U	CF_{Th}	$DF_{U/Th}$
Seaweed*	40	300	0.13
Zooplankton**	30	600	0.05
Molluscs+			
soft tissue	60	570	0.10
shell	30	670	0.04
whole	35	600	0.05
Crustacea+			
Mesidothea entomon	10	1200	0.01
Fish++			
muscle	0.5	10	0.05
whole	0.8	17	0.05

* Data calculated from Szefer (1987)
** Unpublished data; dominant components of mesozooplankton – *Pseudocalanus elongatus*, nauplii *Copepoda*, *Evadne nordmanii* and *Temora longicornis* (Szefer et al., 1985)
+ Data calculated from Szefer & Wenne (1987)
++ Data calculated using values obtained in the present paper

emphasized that the CF_U value obtained here is in the same order of magnitude as that reported in IAEA Technical Reports Series No. 247 (1985).

To determine the degree of discrimination of U in respect to Th in Baltic organisms, the discrimination factor DF was estimated according to the formula:

$$DF = (C_U/C_{Th})_c/(C_U/C_{Th})_s$$

where $(C_U/C_{Th})_c$ and $(C_U/C_{Th})_s$ are the concentration ratios of U to Th in organisms and seawater, respectively.

The results in Table 3 show that analysed organisms preferentially discriminate Th over U (all $DF_{U/Th}$ values lower than unity), i.e. they exhibit a stronger ability to adsorb and/or take up Th as compared to U (with respect to seawater).

The U and Th transfer along the trophic levels of the food chain, i.e. from potential prey (diet) to fish (consumer) was quantitatively estimated by the transfer factor TF (Amiard et al., 1980):

$$TF = \frac{C_c}{C_p}$$

where C_c and C_p mean the U and Th concentration (expressed on dry weight basis) in consumer and diet, respectively.

Helpful information concerning the food habits of fish analysed in the calculations was applied. According to the data obtained by Rutkowicz (1982) and Cięglewicz et al. (1972), cod *(Gadus morhua)* feed on benthic invertebrates, e.g. molluscs and crustacea. Specimens of *Gadus morhua* (length body > 45 cm) caught in spring in the southern Baltic contained in their alimentary tracts mainly specimens of herring *(Clupea harengus)*, sprat *(Sprattus sprattus)* (59.5 percentage by weight) and *Mesidothea entomon* (4.7 %). Greater mass contribution of *Mesidothea entomon* (13 %) was found in the food content of small specimens of cod *(Gadus morhua)* (15–25 cm). The food content of herring *(Clupea harengus)* and sprat *(Sprattus sprattus)* consisted mainly of zooplanktonic organisms *Temora longicornis* and *Pseudocalanus elongatus* (Cięglewicz et al., 1972).

Bearing the above-mentioned information in mind, the degree of transfer of U and Th along the trophic levels was estimated (Table 4). Approximately equal or lower than unity TF values suggest that there is no biomagnification of these elements along the food chain steps leading to fish from their potential diet.

Table 4. Mean transfer factors (TF) for U and Th in fish with respect to lower trophic levels in southern Baltic ecosystem

Trophic relation Consumer – potential prey	TF_U	TF_{Th}
Gadus morhua – *Clupea harengus*	1.30	1.40
Gadus morhua – *Mesidothea entomon*	0.05	0.01
Gadus morhua – molluscs	0.04	0.04
Clupea harengus – zooplankton	0.01	0.01
Sprattus sprattus – zooplankton	0.01	0.01
Hyperoplus lanceolatus – zooplankton	0.02*	0.03*

* Values calculated using the concentration of U and Th in whole fish

LITERATURE CITED

Amiard, J.-C., Amiard-Triquet, C., Metayer, C., Marchand, J. & Ferre, R., 1980. Etude du transfert de Cd, Pb, Cu et Zn dans les chaînes trophiques neritiques et estuariennes. I. Etat dans l'estuaire interne de la Loire (France) au cours de l'été 1978. – Wat. Res. *14*, 665–673.

Amiel, A. J., Miller, D. S. & Friedman, G. M., 1973. Incorporation of uranium in modern corals. – Sedimentology *20*, 523–528.

Aten, A. H. W., Dalenberg, J. W. & Bakkum, W. C. M., 1961. Concentration of uranium in sea fish. – Hlth Phys. *5*, 225–226.

Bangera, V. S. & Patel, B., 1984. Natural radionuclides in sediment and in arcid clam (*Anadara granosa* L.) and gobiid mudskipper (*Boleophthalmus boddaerti* Cuv. & Va.). – Indian J. mar. Sci. *13*, 5–9.

Blanchard, R. L. & Oakes, D., 1965. Relationship between uranium and radium in coastal marine shells and their environment. – J. geophys. Res. *70*, 2911–2921.

Blanchard, R. L., Cheng, M. H. & Potratz, H. A., 1967. Uranium and thorium series disequilibria in recent and fossil marine molluscan shells. – J. geophys. Res. *72*, 4745–4757.

Bojanowski, R. & Szefer, P., 1979. Uranium in the Baltic water. – Stud. Materiały Oceanol. *26*, 253–274 (in Polish, with an English abstract).

Chassard-Bouchaud, C., 1982a. Concentration de l'uranium par les Crustacés. Etude structurale, ultrastructurale et microanalytique par émission ionique secondaire et par spectrographie des rayons X. – C. r. Acad. Sci., Paris (Sér. 3.) *294*, 919–924.

Chassard-Bouchaud, C., 1982b. On the occurrence of uranium, ruthenium, cesium and thulium in diseased fish: a microanalytical ion microscopy study. – Electronmicroscopy *3*, 405–406.

Chassard-Bouchaud, C., 1983. Cellular and subcellular localization of uranium in the crab *Carcinus maenas*: a microanalytical study. – Mar. Pollut. Bull. *14*, 133–136.

Chassard-Bouchaud, C. & Escaig, F., 1984. Uptake, storage and excretion of uranium by *Mytilus edulis*. A structural, ultrastructural and microanalytical study by secondary ion emission and X-ray spectrometry. – J. Phys. (Suppl. au Nr 2) *45*, 545–548.

Cięglewicz, W., Załachowski, W., Szypuła, J., Krzykawski, S. & Krzykawska, I., 1972. Food habits of commercial fish of the southern Baltic. In: Marine ecosystems. Ed. by K. Siudziński, MIR, Gdynia, *2*, 363–450.

Edgington, D. N., Gordon, S. A., Thommes, M. M. & Almodovar, L. R., 1970. The concentration of radium, thorium and uranium by tropical marine algae. – Limnol. Oceanogr. *15*, 945–955.

Flor, T. H. & Moore, W. S., 1977. Radium/calcium and uranium/calcium determinations for Western Atlantic reef corals. – Proc. 3rd int. Coral Reef Symp. Miami *2*, 555–561.

Gvirtzman, G., Friedman, G. M. & Miller, D. S., 1973. Control and distribution of uranium in coral reefs during diagenesis. – J. sedim. Petrol. *43*, 985–997.

Hamilton, E. I., 1972. Concentration of uranium in man and his diet. – Hlth Phys. *22*, 149–153.

Hamilton, E. I., 1980. Concentration and distribution of uranium in *Mytilus edulis* and associated materials. – Mar. Ecol. Prog. Ser. *2*, 62–73.

Hodge, V. F., Koide, M. & Goldberg, E. D., 1979. Particulate uranium, plutonium and polonium in the biogeochemistries of the coastal zone. – Nature, Lond. *277*, 206–209.

Holm, E. & Persson, B. R. R., 1980. Behaviour of natural (Th, U) and artificial (Pu, Am) actinides in coastal waters. In: Marine radioecology (Proc. 3rd NEA Seminar., Tokyo, 1979). OECD, Paris, 237–243.

IAEA, 1985. Sediment K_ds and concentration factors for radionuclides in the Marine Environment. – Tech. Rep. Ser., IAEA *247*, 1–73.

Ichikawa, R. & Ohno, S., 1981. Uranium content in marine fish. – Bull. Jap. Soc. scient. Fish. *47*, 289.

Kharkar, D. P., Thomson, J., Turekian, K. K. & Forster, W. O., 1976. Uranium and thorium decay series nuclides in plankton from the Caribbean. – Limnol. Oceanogr. *21*, 294–299.

Knauss, K. & Ku, T.-L., 1983. The elemental composition of decay-series radionuclide content of plankton from the East Pacific. – Chem. Geol. *39*, 125–145.

Koide, M., Lee, D. S. & Goldberg, E. D., 1982. Metal and transuranic records in mussel shells, byssal threads and tissues. – Estuar. coast. Shelf Sci. *15*, 679–695.

Livingston, H. D. & Thompson, G., 1971. Trace element concentrations in some modern corals. – Limnol. Oceanogr. *16*, 786–796.

Miyake, Y., Sugimura, Y. & Mayeda, M., 1970. The uranium content and the activity ratio $^{234}U/^{238}U$ in marine organisms and sea water in the western North Pacific. – J. oceanogr. Soc. Jap. *26*, 123–129.

Möller, H., Schneider, R. & Schnier, Ch., 1983. Trace metal and PCB content of mussels *(Mytilus edulis)* from the southwestern Baltic Sea. – Int. Revue ges. Hydrobiol. *68*, 633–647.

Omura, A., 1976. Thorium and protactinium isotopes in some present-day hermatypic corals and their implications to dating. – Trans. Proc. palaeont. Soc. Japan *101*, 271–290.

Pentreath, R. J., Lovett, M. B., Harvey, B. R. & Ibbett, R. D., 1979. Alpha-emitting nuclides in commercial fish species caught in the vicinity of Windscale, United Kingdom, and their radiological significance to man. Biological implications of radionuclides released from nuclear industries. IAEA, Vienna, *2*, 227–245.

Rutkowicz, S., 1982. Encyklopedia Ryb Morskich. Wydawnictwo Morskie, Gdynia. 663 pp.

Schroeder, J. H., Miller, D. S. & Friedman, G. M., 1970. Uranum distribution in recent skeletal carbonates. – J. sedim. Petrol. *40*, 672–681.

Schwochau, K., Astheimer, L., Schenk, H. J. & Schmitz, J., 1976. Verfahren zur Gewinnung von Uran aus dem Meerwasser. – Ber. Kernforschungsanl. Jülich *1415*, 38–45.

Sugimura, Y. & Mayeda, M., 1980. The uranium content and the activity ratio $^{234}U/^{238}U$ in sea water in the Pacific Ocean. In: Isotope marine chemistry. Ed. by E. D. Goldberg, Y. Horibe & K. Saruhashi. Uchida Rokakuho Publ. Co., Tokyo, 211–246.

Szefer, P., 1977. Investigations on the uranium and thorium contents of Vistula estuary water, also inshore water of the Gulf of Gdańsk. – Stud. Materiały Oceanol. *17*, 379–392 (in Polish, with an English abstract).

Szefer, P., 1987. Concentration of uranium and thorium by some Baltic seaweeds and lake plants. – Mar. Pollut. Bull. *18*, 439–442.

Szefer, P., 1989. Uranium and its isotopes in marine biocenosis. – Stud. Materiały Oceanol. (in Polish, with an English abstract). (In press)

Szefer, P. & Falandysz, J., 1983. Uranium and thorium content in long-tailed ducks (*Clangula hyemalis* L.) from the Gdańsk Bay. – Sci. Total Environ. *29*, 269–276.

Szefer, P. & Falandysz, J., 1985. Trace metals in muscle tissue of fish taken from the southern Baltic. – Z. Lebensmittelunters. – Forsch. *181*, 217–220.

Szefer, P. & Wenne, R., 1987. Concentration of uranium and thorium in molluscs inhabiting Gdańsk Bay, Baltic Sea. – Sci. Total Environ. *65*, 191–202.

Szefer, P., Skwarzec, B. & Koszteyn, J., 1985. The occurrence of some metals in mesozooplankton taken from the southern Baltic. – Mar. Chem. *17*, 237–253.

Thompson, G. & Livingston, H. D., 1970. Strontium and uranium concentrations in aragonite precipitated by some modern corals. – Earth Planet. Sci. Lett. *8*, 439–442.

Tsytsugina, V. G., Risik, N. S. & Lazorenko, G. E., 1973. Concentration and microdistribution of uranium in marine organisms under natural conditions. In: Artificially and naturally produced radionuclides in aquatic biocenosis. Ed. by G. G. Polikarpov. Naukova Dumka, Kiev, 58–94.

Veeh, H. H. & Turekian, K. K., 1968. Cobalt, silver, and uranium concentrations of reef-building corals in the Pacific Ocean. – Limnol. Oceanogr. *13*, 304–308.