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TRACE ELEMENT DISTRIBUTION IN SEVEN MOLLUSC SPECIES FROM SARONIKOS GULF

KONCENTRACIJA ELEMENATA U TRAGOVIMA U SEDAM VRSTA MEKUŠACA U ZALJEVU SARONIKOS

CATHERINE PAPADOPOULOU AND GEORGE D. KANIAS

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INTRODUCTION

The need for more research on the kinds and effects of pollution as well as the need for basic research on the causes affecting the chemical composition of oceans is an increased function of time. This fact necessitates the systematic study of the ecological parameters of the marine ecosystem of Greece.

Pollution of the marine environment results from the industrial, domestic and radioactive wastes disposal. The radioactive wastes from different human activities reach the sea directly via rivers or carried by rain and air as shown in Figure 1 (Koch, 1960; Nishihara, 1967; Cottam, 1960; Jammet, 1962).

An important parameter for studying the marine pollution is the elementary composition of marine organisms. The ability of certain marine species to concentrate chemical elements from the environment in their bodies is of special importance. Due to this accumulation through the food chain, certain sea foods may become unsuitable for human consumption since the sea is the final receiver of several pollutants (Sitting, 1969).

Toxic elements such as Cu, Hg, and Pb contained in industrial discharges can be concentrated selectively by fish and marine invertebrates. Fatal poisoning accidents on the population of Japan, reported as »Minamata desease,« was caused by the excess of methyl mercury found in fish and shellfish of Minamata Bay (Irukayama, 1967; Sumino, 1968). High mercury levels were found in fish and birds in Sweden and Canada (Rissanen and Miettinen, 1972; Sprague and Garson, 1970). Recent work on the estimation of arsenic content in marine organisms confirmed that this toxic element can reach the higher trophic levels through the aquatic ecosystem (McBride and Wolfe, 1971; Leblanc and Jackson, 1973).

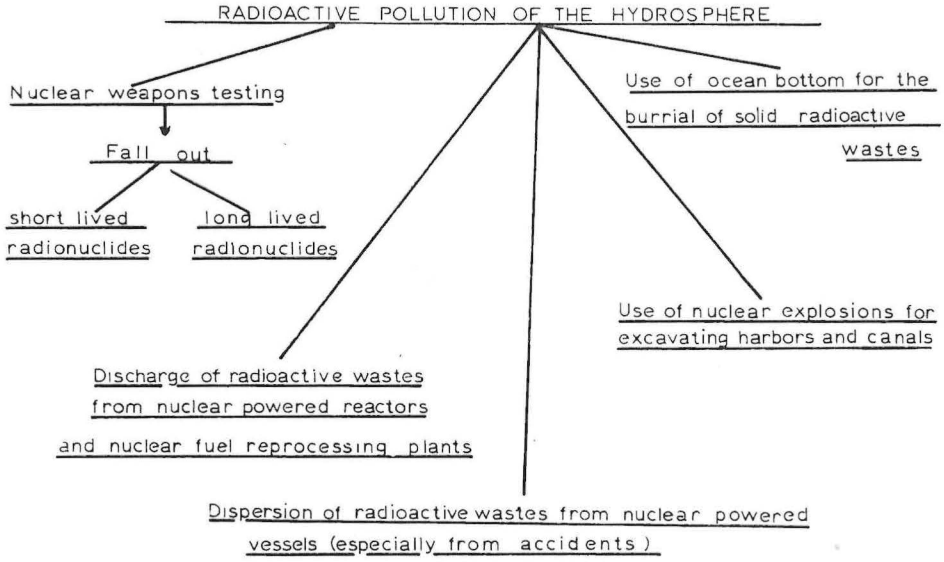

Fig. 1 — Dispersion of radioactive pollution in the hydrosphere.

Toxic element accumulation may have harmful effects on the organism itself. Several cases of damage to oyster and mussel populations due to such accumulation have been reported by Roskam (1965) and Ikuta (1967).

There are also special dangers to the seas associated with nuclear power plants located on sea shores because of the exchange of stable elements by their radioisotopes into the organisms (Polykarpov, 1966; Perkins, Nielsen, Ruesch and McColl, 1960).

The work done on the elementary composition of molluscs until 1953 has been summarized by Vinogradov (1953). Recently data on this subject have been collected by Goldberg (1965). Studies on the trace element composition of mollusc species up to 1965 have been reported by several authors (Hobben, 1967; Fukai, 1968; Robertson, Rancitelli and Perkins, 1968; Butterworth, Lester and Nickless, 1972; Papadopoulou, 1973).

Saronikos Gulf was chosen by the authors as a suitable area for the collection of data on the concentration of trace elements in molluscs.

METHODS AND MATERIALS

Individuals of seven mollusc species were collected during October 1969 from the coastal waters of the west part of Saronikos Gulf between Megara and Salamis Island from a maximum depth of 12 m (Figure 2). The species included *Mytilus galloprovincialis, Meretrix chionae, Ostrea edulis, Venus verrucosa, Ensis ensis, Tapes decussatus* and *Glycymeris glycymeris*. Seawater samples were collected from the same area and depth as the molluscs.

The molluscs were washed with sea- and distilled water to remove external mud and other materials. Three groups of 60 and one group of 10 individuals of about the same size for each species were chosen. The groups of ten animals were used for the »whole body« analysis and the other groups for the »various parts« or organ analysis.

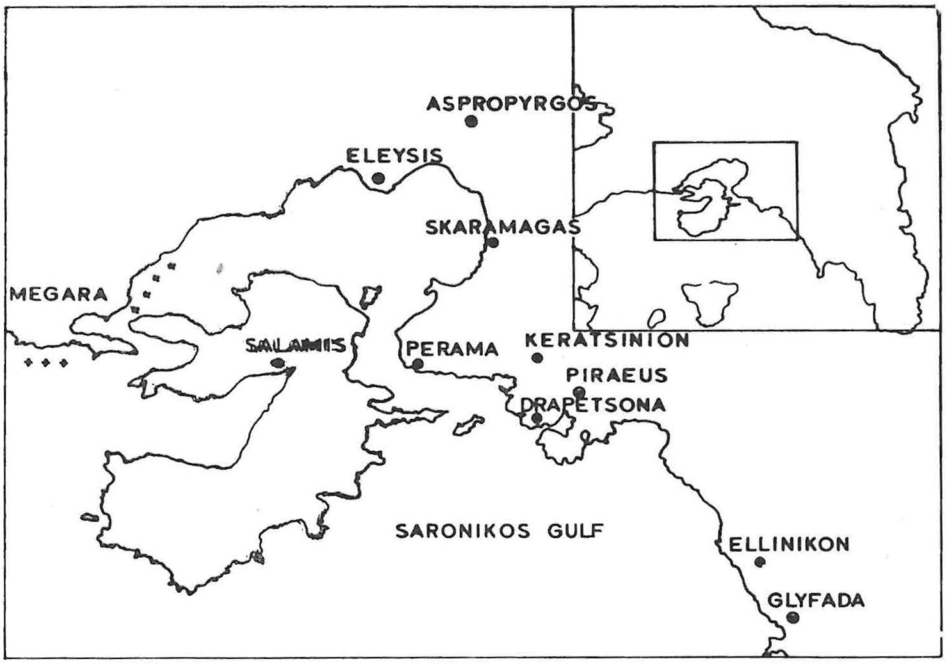

Fig. 2 — Saronikos Gulf showing area of collection of molluscs (+) for trace element content.

After the removal of the shell and dissection of the different parts, the groups were dried at 75°C for 20 hours. The dry matter was homogenated by pulverization and then used for preparing the samples for the analysis. A part was ignited to 550°C and the ashed material was used for Ba, Sr, Cu, Eu, V, Zn and Au determination.

For neutron activation analysis, the samples were prepared by sealing 50 to 250 mg of matter into 1 ml polyethylene or quartz tubes depending on the irradiation time. For the spectrophotometric analysis, the samples were prepared by wet ashing 100 to 200 mg of dry matter with high purity sulfuric and nitric acids and diluted with tridistilled water.

All neutron irradiations of the samples were performed in the reactor of Democritos Nuclear Research Center in the thermal neutron flux of about 1.5×10^{13} n/cm^2/sec. For the short time irradiation, the pneumatic tube transfer system was used. The measurements of radiation energy of radioisotopes produced were made by a 75 × 75 mm Na (Tl) crystal connected with a 400 channel pulse height analyser. A Perkin-Elmer model 303 atomic absorption spectrophotometer was also used.

For the determination of Cu, Zn, Mn, Ba, Sr, Sc, Eu and the detection of the rare earths the ion exchange separation procedure given by Hadzistelios and Papadopoulou (1969) was applied. For the determination of V, the extraction method given by Fukai and Meinke (1959) was applied. This method was modified by the authors only for the chemical yield determination (Papadopoulou, Hadzistelios and Grimanis, 1972). For the determination of Au, the method of Hummel (1957) was applied. For the determination of As, Sb, and Hg the combined ion exchange method of Hadzistelios and Grimanis (1969) was applied.

The amount of the radioactivity emitted by the radioisotopes Co^{60} and Cr^{51} at the characteristic energy region was measured for the samples and standards. The concentration was estimated and compared according to Covell (1959). Atomic absorption spectroscopy was used for the determination of Ni, Mg, Mo and Cs.

The seawater samples were processed by applying the same analytical procedures as in the case of mollusc samples. The only modification was in the treatment of the sample before irradiation (Papadopoulou, Hadzistelios and Grimanis, 1972).

RESULTS AND DISCUSSION

The distribution of trace elements in the 7 species of mollusc is presented in Tables 1 through 7. The content given represents the mean value of three duplicate determinations performed in three different groups of the same species.

Table 1. Concentration of trace elements (ppm) in *Mytilus galloprovincialis* from Saronikos Gulf. Standard error ± 10%. X = below limits of detection.

Elements	Whole body	Muscle	Mantle and Gills	Stomach and Intestine	Gonads	Foot	Byssus
Dry							
As	25.5	29.8	34.8	28.0	35.5	26.8	0.61
Sb	0.14	0.16	0.31	0.14	0.044	0.104	0.15
Hg	0.21	0.48	0.43	0.20	0.48	0.44	X
Co	19.4	33.0	3.67	35.2	37.4	33.3	75.4
Cr	7.8	5.2	3.1	33.4	7.7	32.6	14.1
Ni	39	55.6	26	82	58	38	58
Mg	5484	3888	5082	6158	3567	9294	1380
Cs	—	—	—	—	—	—	—
Mo	X	X	X	23	X	X	X
Ash							
Cu	45.1	14.6	50.1	83	26.5	36.3	12.8
Zn	872	353	830	948	649	372	28
Mn	135	73.9	142	48	85	33.5	118
Sr	262	61	314	153	23	422	435
Ba	X	X	X	X	X	X	26
Eu	0.13	X	0.16	0.23	0.08	0.12	X
Au	0.54	—	—	—	—	—	—
V	2.3	—	—	—	—	—	—

Table 2. Concentration of trace elements (ppm) in *Venus verrucosa* from Saronikos Gulf. Standard error ± 10%. X = below limits of detection.

Elements	Whole body	Muscle	Mantle and Gills	Stomach and Intestine	Gonads	Foot
Dry						
As	15.1	6.89	3.68	8.7	12.3	9.7
Sb	0.037	X	0.020	0.045	0.031	0.024
Hg	0.022	0.019	0.019	0.024	0.016	0.016
Co	1.71	2.38	3.63	3.29	4.61	5.27
Cr	4.7	X	4.7	5.2	7.7	3.8
Ni	—	—	—	—	—	—
Mg	7280	5050	8980	6977	6902	5487
Cs	—	—	—	—	—	—
Mo	X	X	X	X	X	X
Ash						
Cu	73.8	78.1	85.4	51.7	58.7	2498
Zn	249	106	231	267	330	79
Mn	29.4	24.3	79	38.2	45.7	134
Sr	347	209	722	100	104	3077
Ba	27	33	58	X	X	512
Eu	0.11	0.08	0.63	X	X	0.75
Au	0.24	—	—	—	—	—
V	1.5	—	—	—	—	—

Table 3. Concentration of trace elements (ppm) in *Glycymeris glycymeris* from Saronikos Gulf. Standard error ± 10%. X = below limits of detection.

Elements	Whole body	Muscle	Mantle and Gills	Stomach and Intestine
Dry				
As	7.32	5.62	3.35	5.64
Sb	0.098	X	0.260	0.062
Hg	0.016	0.017	0.053	0.17
Co	0.52	X	2.32	2.62
Cr	4.9	11.0	9.2	X
Ni	14	X	27	X
Mg	5968	6097	7132	7537
Cs	X	X	X	X
Mo	X	X	X	X
Ash				
Cu	356	334	275	115
Zn	963	619	1158	966
Mn	44.9	42.3	46.7	53.7
Sr	1706	425	530	3791
Ba	23	X	32	X
Eu	0.06	X	0.11	0.06
Au	0.16	—	—	—
V	9.3	4.5	6.8	14.5

Table 4. Concentration of trace elements (ppm) in *Ensis ensis* from Saronikos Gulf. Standard error ± 10%. X = below limits of detection.

Elements	Whole body	Muscle	Mantle and Gills	Stomach and Intenstine	Foot
Dry					
As	15.0	7.4	10.3	13.8	14.7
Sb	0.058	0.017	0.016	0.034	0.079
Cr	12.4	5.6	8.3	6.3	16.9
Hg	2.36	1.51	21.5	0.60	0.97
Co	0.51	0.53	0.32	0.36	16.0
Ni	X	X	X	12	37
Mg	589	462	725	691	1955
Cs	X	X	X	X	73
Mo	X	X	X	21	X
Ash					
Cu	109	99	114	123	157
Zn	277	175	222	519	840
Mn	149	164	268	196	76
Sr	174	54	63	189	X
Ba	—	—	—	—	—
Eu	0.06	0.005	X	0.41	0.13
Au	0.30	—	—	—	—
V	8.2	3.4	12.1	16.5	6.2

Table 5. Concentration of trace elements (ppm) in *Meretrix chionae* from Saronikos Gulf. Standard error ± 10%. X = below limits of detection.

Elements	Whole body	Muscle	Mantle and Gills	Stomach and Intestine	Gonads	Foot
Dry						
As	58.8	41.2	50.0	37.3	32.5	36.0
Sb	0.37	0.194	0.142	0.125	0.69	0.11
Hg	0.073	0.20	X	0.059	0.038	X
Co	14.2	10.8	37.7	29.6	26.1	20.3
Cr	4.1	8.5	4.9	18.8	6.5	13.3
Ni	26	28	60	63	66	47
Mg	6721	6579	3927	4155	2945	5756
Cs	—	—	—	—	—	—
Mo	—	—	—	—	—	—
Ash						
Cu	84	52.7	64.8	153	114	136
Zn	669	446	308	161	557	741
Mn	528	262	157	1237	255	59.2
Sr	783	418	394	1290	456	382
Ba	101	34	X	70	25	X
Eu	0.058	0.089	0.040	0.061	0.108	0.060
Au	—	—	—	—	—	—
V	9.2	7.4	2.5	11.8	6.6	2.5

Table 6. Concentration of trace elements (ppm) in *Ostrea edulis* from Saronikos Gulf. Standard error ± 10%. X = below limits of detection.

Elements	Whole body	Muscle	Mantle and Gills	Stomach and Intestine	Gonads	Foot
Dry						
As	83	113	130	151	192	111
Sb	0.52	0.88	0.65	0.71	0.80	0.30
Hg	0.32	0.74	X	0.42	0.20	X
Co	0.73	1.71	1.54	2.24	X	X
Cr	12.5	9.7	27.6	4.7	X	X
Ni	15	18	X	20	X	X
Mg	4515	7770	18,670	5226	1895	1088
Cs	X	X	X	X	X	54
Mo	X	X	X	21.0	X	X
Ash						
Cu	1609	368	590	518	370	398
Zn	10,710	4853	9787	17,080	2777	4112
Mn	30.9	11.8	18.6	12.1	5.9	7.6
Sr	372	155	144	158	97	52
Ba	X	X	X	X	X	X
Eu	0.22	0.16	0.07	0.05	X	0.05
Au	0.46	—	—	—	—	—
V	0.31	—	—	—	—	—

Table 7. Concentration of trace elements (ppm) in *Tapes decussatus* from Saronikos Gulf. Standard error ± 10%.

Elements	Whole body
Dry	
As	56.0
Sb	0.10
Hg	0.29
Co	22.5
Cr	18.1
Ni	45
Mg	4276
Cs	—
Mo	—
Ash	
Cu	103
Zn	827
Mn	6.40
Sr	322
Ba	—
Eu	0.11
Au	1.88
V	3.24

Based on the trace element content, the selective accumulation by a certain organ of the organism can be estimated. The accumulating capacity of the same organ in different species for a certain element is given in Figure 3. The specific accumulation of certain elements in the organism can serve as a significant factor for the prediction of the pollution levels in the sea. This property of the organisms is expressed by the »concentration factors« (Polykarpov, 1966) presented in Table 8.

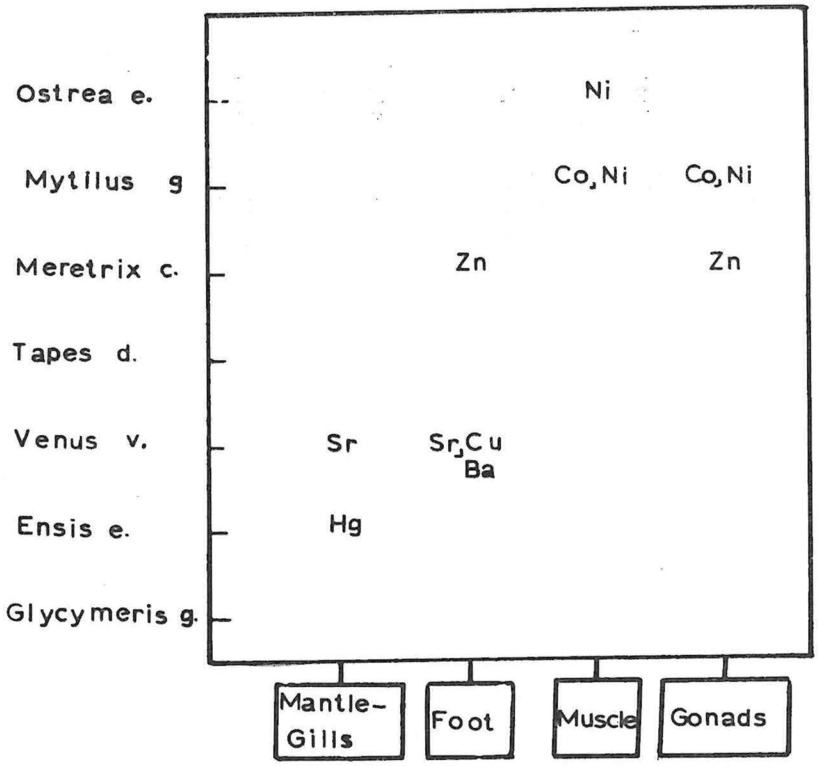

Fig. 3 — Selective concentration of trace elements in organs of different molluscs

The choice of an organism as an »indicator« for a certain pollutant can be made by means of the calculated concentration factors. For the toxic element pollution, *Ostrea edulis* and *Ensis ensis* are good indicators for evaluating As, Zn, Cu, Sb, Cr and Hg, Cr, V respectively. *Meretrix chionae* is a preferable organism for Ni, Mn, As, Sb and V, while *Tapes decussatus* is the best indicator for Cr and Ni when compared with *O. edulis, E. ensis,* and *Glycymeris glycymeris.*

Table 8. Concentration factor for trace elements in relation to the weight of the whole body for seven molluscs from Saronikos Gulf.

Elements	*Venus verrucosa*	*Glycymeris glycymeris*	*Ensis ensis*	*Tapes decussatus*	*Meretrix chionae*	*Ostrea edulis*	*Mytilus galloprovincialis*
As	660	400	1000	3200	3700	5000	1280
Sb	5	18	13	20	80	110	24
Hg	70	200	18,300	1930	530	2300	1200
Cu	200	1100	370	260	480	6700	100
Zn	200	950	300	660	1200	14,300	680
Mn	300	550	2000	60	12,200	520	1300
Sr	1.0	6.0	0.6	0.1	5	1.6	0.7
Ba	103	100	—	—	800	—	—
V	80	570	550	160	1000	25	110
Au	530	390	810	3700	—	1500	1000
Co	2600	990	1200	45,000	31,900	1500	51,700
Cr	14,600	19,000	58,000	72,000	18,200	53,600	27,400
Ni	—	1780	—	6060	3800	2100	4500
Mo	—	—	—	—	—	—	—

Radionuclide pollution of the marine environment resulting from thermonuclear reactions can be estimated by indicators like *Tapes decussatus* and *Mytilus galloprovincialis* for Co^{60}, *Meretrix chionae* for Ba^{140} and V and *Glycymeris glycymeris* for Sr^{90}. V is also found in *Ensis ensis* and *G. glycymeris* but to a lesser extent. This is not produced as a fission product but it can be replaced by Nb^{95}.

The exchange of the radioisotopes, which are derived from nuclear wastes in water, with the corresponding stable elements absorbed selectively by some organisms, cause another form of pollution. This kind of water pollution can be estimated by induced radioctivity in certain mollusc species because of the selective absorption. Thus *Ostrea edulis* can be used for the detection of $Zn,^{65}$ $As,^{76}$ $Cu,^{64}$ Cr^{51} and $Sb,^{122,\ 124}$ while *Mytilus galloprovincialis* is the best for Co^{60} and *Meretrix chionae* for Mn^{54} detection. *Tapes decussatus* preferentially, or *Ensis ensis,* are good indicators for $Cr.^{51}$

The upper part of Saronikos Gulf around Elefsis Bay is a region of heavy pollution. The east part of this area from the town of Elefsis to Keratsini Bay is an industrial area where oil refineries are located in addition to other factories. On the contrary, the west part from Elefsis toward Megara is still favorable for fisheries and mariculture and is considered as non-polluted. Thus from the element content of indicator organisms selected periodically from the above area, one can estimate the alternation caused by pollutants in this region.

SUMMARY

The interest for the knowledge of the trace element content in the living organisms in recent years stems from the fact that certain elements play an important role in various physiological functions. Furthermore, the gradually increasing environmental pollution, resulting from industrial, domestic and radioactive waste disposal to the sea, dictates the necessity for a detailed study of the marine environment in order to protect the ecosystem and man.

This work is concerned with the study of the trace element distribution in the whole bodies and individual organs of seven mollusc species from the Saronikos Gulf. The mollusc species *Mytilus galloprovincialis, Meretrix chionae, Ostrea edulis, Venus verrucosa, Ensis ensis, Tapes decussatus, Glycymeris glycymeris* have been selected because they are common in the Greek sea and they consist the pathway in the food chain between the lower marine organisms and the man.

Modern analytical methods such as neutron activation analysis and atomic absorption spectroscopy were used for the detection and determination of the elements. The following elements were determined: As, Sb, Hg, Cu, Zn, Mn, Sr, Ba, Eu, Co, Cr, V, Au, Ni, Mg, Mo and Cs. The elements La, Ce, Sm, Gd, Tb, Ho, Er, Yb, Lu, Ga and Sc could not be detected. These elements have been chosen for the following reasons: Pollution effects, correspondence of these stable isotopes to the hazardous fission products of waste disposal radionuclides and biological importance.

To find the distribution of these elements, determination was carried out on various parts of each organism and on each whole body.

Concentration factors of the above elements have been found for the seven mollusc species by analysing the environmental water.

Discussion on the results is made in the light of the distribution of the elements in the individual organs of each organism, and the variation in accumulation of the elements in the corresponding organs of the seven species. On the basis of the concentration factors, some species identified as «bioindicators» for the determined trace elements are given.

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KONCENTRACIJA ELEMENATA U TRAGOVIMA U SEDAM VRSTA MEKUŠACA U ZALJEVU SARONIKOS

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KRATAK SADRŽAJ

Iznose se rezultati studija elemenata u tragovima u sedam vrsta mekušaca u zaljevu Saronikos, kako u čitavim organizmima, tako i u pojedinačnim organima. To su bile slijedeće vrste: *Mytilus galloprovincialis, Meretrix chionae, Ostrea edulis, Venus verrucosa, Ensis ensis, Tapes decussatus, Glycymeris glycymeris.* One su odabrane jer su uobičajene u grčkim vodama i predstavljaju u lancu ishrane vezu između nižih morskih organizama i čovjeka.

Modernim analitičkim metodama determinirani su slijedeći elementi: As, Sb, Hg, Cu, Zn, Mn, Sr, Ba, Eu, Co, Cr, V, An, Ni, Mg, Mo i Cs. Elementi La, Ce, Sm, Gd, Tb, Ho, Er, Yb, Lu, Ga i Sc nisu mogli biti otkriveni. Ovi su elementi odabrani zbog efekata polucije i reakcije ovih stabilnih izotopa na slučajne produkte fiksiranja ili raspodjele otpadnih radionukleida, te biološke važnosti.

Faktori koncentracije gornjih elemenata dobiveni su za ovih sedam vrsta moluska analizom okolne vode.

U diskusiji rezultata je razmatrana raspodjela elemenata u pojedinačnim organima i akumulacija elemenata u odgovarajućim organima navedenih sedam vrsta. Na osnovi faktora koncentracije, neke su vrste identificirane kao »bioindikatori« za određene elemente u tragovima.