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TRANSFER OF RADIONUCLIDES FROM SEAWATER, SEDIMENT AND MUD WITH DETRITUS TO THE MULLET SPECIES *LIZA AURATA* (MUGILIDAE)

Abstract

Cesium-137, ^{40}K, ^{226}Ra and ^{232}Th in whole *Liza aurata* individuals, its muscles and skeleton, as well as in seawater, surface sediment and mud with detritus, have been measured. The activity concentrations were used to calculate concentration factors (CFs) for transfer of these radionuclides from seawater, sediment and mud with detritus to fish tissues. It follows from the results that accumulation of ^{137}Cs in an edible part in comparison with whole fish accumulation is about 3 times higher. The highest CFs were – thorium and radium CF for skeleton (from seawater) with an average of 371.5 and 303.1, respectively, and then thorium and cesium CF for muscles (again, from seawater) with an average of 59.6 and 44.9, respectively. All CFs (from seawater, sediment and mud with detritus) show accumulation of radium in skeleton more than 20 times higher than in the muscle of *L. aurata* species.

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TRANSFER RADIONUKLIDA IZ MORSKE VODE, SEDIMENTA I MULJA SA DETRITUSOM U VRSTU CIPOLA *LIZA AURATA* (MUGILIDAE)

Izvod

Cezijum-137, ^{40}K, ^{226}Ra i ^{232}Th mjereni su u cijelim jedinkama vrste cipola *Liza aurata,* u njenim mišićima i skeletu, kao i u morskoj vodi, površinskom sedimentu i mulju sa detritusom. Koncentracije aktivnosti korišćene su za proračune koncentracionih faktora (KF) za transfer ovih radionuklida iz morske vode, sedimenta i mulja sa detritusom – u tkiva ribe. Iz rezultata slijedi da je akumulacija ^{137}Cs u jestivom dijelu oko 3 puta veća od akumulacije u cijeloj jedinki. Najveći KF bili su – KF torijuma i radijuma za skelet (iz morske vode) sa srednjim vrijednostima 371.5 i 303.1, respektivno, a zatim KF torijuma i cezijuma za mišiće (ponovo iz morske vode) sa srednjim vrijednostima 59.6 i 44.9, respektivno. Svi KF (iz morske vode, sedimenta i mulja sa detritusom) pokazuju da je akumulacija radijuma u skeletu više od 20 puta veća od njegove akumulacije u mišiću vrste *L. aurata*.

INTRODUCTION

Radioactivity in fish is usually analyzed to estimate doses to fish consumers, for which the radionuclide activities in the edible parts of fish are needed. On the other hand, radionuclide transfer to fish tissues is important for understanding isotope dynamics in the aquatic environment.

Thus, radiologically important isotopes ^{137}Cs and ^{226}Ra (as significant contributors to dose rates) have been already measured in different marine environments, as well as in some fish species (for example, [1, 2, 3]). Concentration factors (CFs) for transfer of radium from water to fish were also determined [4, 5] and showed considerable ability for fish to accumulate this radionuclide from water even when it was present in very small concentration. However, radionuclides uptake, distribution and bioaccumulation in different fish species are not well known.

Therefore, a few important radionuclides (^{137}Cs, ^{40}K, ^{226}Ra and ^{232}Th) in the mullet species *Liza aurata* Risso, 1810 (Mugilidae) from the South Adriatic Sea were analyzed, together with their activity in seawater, surface sediment, and mud with detritus. This fish species is consumed by the local population in Montenegro, and it is one of six mullet species that occur in the South Adriatic Sea.

As it is known, ^{137}Cs is a fission product with a half-life of 30.1 y, which decays to ^{137}Ba via β^-–decay accompanied by the 661.657 keV γ-ray, with an intensity 85.1 %. Naturally occurring ^{40}K decays by electron capture and β^+–decay to stable ^{40}Ar (followed by the 1460.83 keV γ-ray, with an intensity 10.7 %), but also by β^-–decay to stable ^{40}Ca (89.3 %). Radium-226 originates from the radioactive series of ^{238}U. Its chain contains eight daughter radionuclides (^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po, ^{210}Pb, ^{210}Bi, ^{210}Po), which decay via α– and β^-–decays, mostly accompanied by emission of γ–rays. This chain ends with the stable isotope of lead – ^{206}Pb. The radioactive series of ^{232}Th (to the stable lead ^{208}Pb) contains ten daughter radionuclides (^{228}Ra, ^{228}Ac, ^{228}Th, ^{224}Ra, ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi, ^{208}Tl, ^{212}Po), whose α– and β^-–decays are followed by emission of γ-rays [6].

In the present study, radionuclide activity measurements have been performed on five whole *L. aurata* individuals, and additionally – on two muscles and skeletons.

MATERIALS AND METHODS

Sampling and sample preparation

Fish material was collected in the South Adriatic Sea (Boka Kotorska Bay, area of Tivat), using the trawl net. Seven *L. aurata* individuals were selected on the basis of taxonomic characteristics [10].

The sample 1, with total length of 35.6 cm and fresh mass of 0.258 kg, was measured by the PRIPYAT-2M spectrometer without both any preparation and establishing radioactive equilibrium between radium and thorium and their decay products, which causes a detection of the 609 keV and 2615 keV γ-rays, enables determination of ^{214}Bi and ^{208}Tl activity, respectively (but not ^{226}Ra and ^{232}Th). Then, the sample was homogenized, hermetically closed in 0.5L Marinelli beaker and measured (after 38 days) by both the HPGe detector (ORTEC – GEM-40190, relative efficiency – 40 %) and spectrometer PRIPYAT-2M (described below). This time period (of sealed storage) is enough to minimise emanation of the radium and thorium progenies ^{222}Rn and ^{220}Rn, respectively.

The samples 2, 3, 4 and 5 (total length – 29.6, 29.5, 30.1 and 23.5 cm, respectively; fresh mass – 0.182, 0.194, 0.185 and 0.105 kg, respectively) have been hermetically closed in plastic vessels, and radionuclide activities were measured by the PRIPYAT-2M spectrometer after radioactive equilibrium was established. In all cases, whole fish individuals were measured over 5000 s real measuring time (live times > 4950 s). As an illustration, *L. aurata* whole individual is shown in Fig. 1.

Figure 1. *L. aurata* from the South Adriatic Sea

The rest two samples (6 and 7, with total length of 31.9 and 34.6 cm, and fresh mass of 0.231 and 0.265 kg, respectively) were measured to determine ^{137}Cs, ^{40}K, ^{214}Bi and ^{208}Tl activity, and then were dissected to separate muscles and skeletons. These samples were hermetically closed and measured over 10 000 s live measuring time. Figs. 2 and 3 show muscles and skeleton of *L. aurata*, respectively.

30L seawater was also sampled in the Boka Kotorska Bay (area of Tivat), evaporated, hermetically sealed in 1L Marinelli beakers for 38 days, and measured using the same method as in the case of the other samples (live measuring time – 10 000 s), as well as the sediment (sampled in

Figure 2. Muscles of *L. aurata*

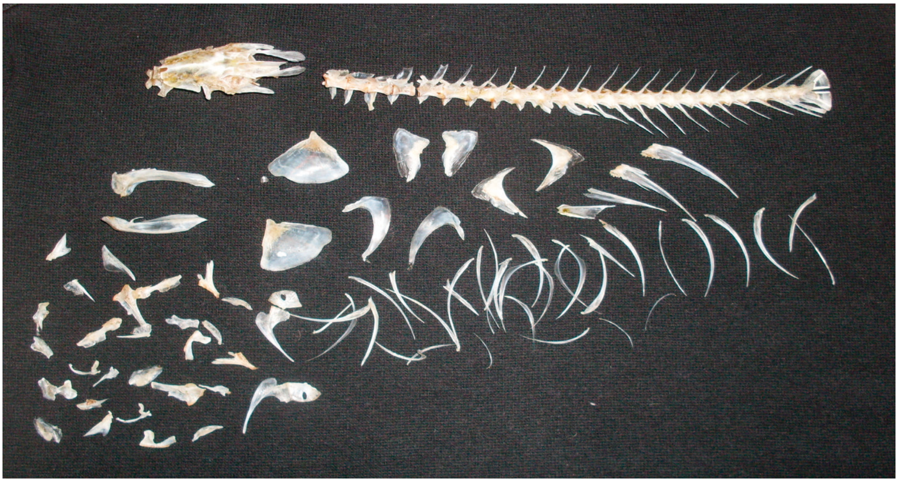

Figure 3. Skeleton of *L. aurata*

the Tivat area) and mud with detritus (sampled in the Sveti Stasije area), dried at room temperature before being weighed (1.345 and 1.078 kg, respectively), hermetically sealed in 1L Marinelli beakers and measured over 2000 s real time.

Gamma-spectrometry

The six-crystal γ-spectrometer PRIPYAT-2M with outer dimensions of 250 cm x 145 cm x186 cm, and mass of 4200 kg, has a measuring chamber shaped as a cube with sides of 17.5 cm. At each face of the cube is placed a NaI(Tl) crystal with diameter of 15 cm and a thickness of 10 cm. The sensitive volume is sufficient to accommodate samples of different nature, with an arbitrary shape and a volume of up to 5 dm^3.

The system has an energy resolution of 10.5 % for the ^{137}Cs – 662 keV line, total solid angle of ~ 0.7x4π sr, i. e., measuring geometry close to 4π, the resolution time for coincidences of 40 ns, and the multiplicity of coincidences – from 2 to 6, which means registering double, triple, four-, five- and six-fold coincidences.

All spectrometer characteristics (measuring geometry of 4π, six detectors and the software PRIP) enable an independence on sample geometry, i. e., measurements without preliminary sample preparation and calibration measurements for different sample geometries [7, 8], as well as registration of cascaded γ-rays. All necessary calculations are carried out using the PRIP software which has wide service possibilities – calibration, data acquisition, processing, presentation and archiving, etc.

A measurement of ^{137}Cs, ^{40}K and decay products ^{226}Ra and ^{232}Th by this spectrometer is based on using the counting mode in which non-coincidence and double coincidences spectra are produced simultaneously [9]. The thorium activity in a sample is determined by the 2615 keV (γ-ray in the ^{232}Th chain which follow β^-–decay of ^{208}Tl to ^{208}Pb, with an intensity 99 %) photopeak in the spectrum of double coincidences, and then, in the same spectrum, the radium activity by the 609 keV photopeak (γ-ray in the ^{226}Ra chain, which follow β^-–decay of ^{214}Bi to ^{214}Po, with an intensity 46.1 %). The cesium activity is determined by the 662 keV photopeak in the non-coincidence spectrum, whilst potassium activity – by the 1460 keV photopeak in the non-coincidence spectrum, as well.

Calculation of CFs

The results of radionuclide activity measurements are used to calculate CFs as follows [11]:
for seawater to fish,

$$CF_1 = \frac{\text{Bq kg}^{-1}\text{ of wet wt fish}}{\text{Bq L}^{-1}\text{ of seawater}}; \tag{1}$$

from sediment to fish,

$$CF_2 = \frac{\text{Bq kg}^{-1}\text{ of wet wt fish}}{\text{Bq kg}^{-1}\text{ of dried sediment}}; \tag{2}$$

and from mud with detritus to fish,

$$CF_3 = \frac{\text{Bq kg}^{-1}\text{ of wet wt fish}}{\text{Bq kg}^{-1}\text{ of dried mud with detritus}}. \tag{3}$$

RESULTS AND DISCUSSION

The radionuclide activity concentrations in *L. aurata* from the South Adriatic Sea are given in Table 1.

Samples of whole fish individuals 6 and 7 did not show precise photopeaks at the energy of 662 keV (i. e., ^{137}Cs was not detected precisely), but their muscles had ^{137}Cs activity concentrations as presented in Table 1. Radium-226 and ^{232}Th activities for the same samples reported in Table

1, are in fact activities of ^{214}Bi and ^{208}Tl, respectively (radioactive equilibrium has not been established in these samples).

As mentioned above, the sample 1 was also measured by the spectrometer PRIPYAT-2M without any preparation and establishing radioactive equilibrium, and measured ^{214}Bi and ^{208}Tl activity was (0.43±0.09) and (0.77±0.14) Bq kg^{-1}, respectively. After had been homogenized and hermetically sealed, it was measured by this (the results are presented in Table 1), as well as by the HPGe spectrometer – (0.62±0.09) and (1.67±0.15) Bq kg^{-1} (for ^{226}Ra and ^{232}Th, respectively). Differences between results obtained by two spectrometers were less than 12 %.

Cesium-137 was detected in two whole individuals only (3 and 4) and its activity is found to be significantly lower than cesium activity concentration found in sprat from the Baltic Sea (5.09 Bq kg^{-1}, measured in 2008) [12]. Potassium-40 activity in whole *L. aurata* ranged from 53.24 (sample 6) to 121.15 Bq kg^{-1} (sample 3), while ^{226}Ra activity ranged from 0.58 Bq kg^{-1} (sample 1) to 1.97 Bq kg^{-1} (sample 3). These values are slightly higher than those reported for whole benthic fish from Puget Sound, Washington, USA (where maximum measured radium activity was 0.75 Bq kg^{-1} [4]), or for sprat from the Baltic Sea (0.7 Bq kg^{-1} [12]). Thorium-232 activity in *L. aurata* was also found to be relatively low, i. e., lower than 1.9 Bq kg^{-1}.

Table 1. Radionuclide activities in L. aurata

L. aurata	^{137}Cs, Bq kg^{-1}	^{40}K, Bq kg^{-1}	^{226}Ra, Bq kg^{-1}	^{232}Th, Bq kg^{-1}
1	–	95.1±7.39	0.58±0.11	1.47±0.42
2	–	75.05±6.81	0.66±0.05	1.51±0.45
3	0.23±0.04	121.15±8.47	1.97±0.46	0.97±0.23
4	1.35±0.32	85.22±12.56	1.29±0.11	1.02±0.18
5	–	116.95±13.90	1.90±0.28	1.88±0.45
6	–	53.24±1.03	1.43±0.13[1]	0.92±0.28[2]
7	–	82.41±4.71	0.83±0.18[3]	0.64±0.11[4]
Muscle – 6	2.47±0.18	101.35±10.31	2.39±0.37	4.54±0.98
Muscle – 7	2.02±0.15	129.86±13.79	2.14±0.87	7.39±1.52
Skeleton – 6	–	32.05±2.55	29.91±5.61	54.2±5.63
Skeleton – 7	–	58.39±5.78	18.6±2.56	20.07±3.14

[1] ^{214}Bi; [2] ^{208}Tl; [3] ^{214}Bi; [4] ^{208}Tl

Moreover, cesium activity in muscle (as an edible part) was found to be higher than, for example, in fish from the Brazilian Coast (from 0.1 to

0.3 Bq kg^{-1} [13]), but also lower than in muscle/flesh of the Baltic Sea fish species (5.1 Bq kg^{-1} in herring, 5.0 Bq kg^{-1} in plaice, and 7.09 Bq kg^{-1} in cod – sampled in 2008 [12]). Here reported ^{137}Cs activity concentrations in muscles were higher than those found in whole individuals. An average concentration in muscles was about 2.8 times higher than that in whole individuals. Previous research of some fish species showed that accumulation of ^{137}Cs in edible parts in comparison with whole body accumulation is much higher (for example, 80 % for *Murrel* fish [14]). ^{40}K was detected in both muscle samples and results are in accordance with ones obtained for flesh of the Baltic Sea fish species (112 – 122 Bq kg^{-1} in herring, 93.6 – 102 Bq kg^{-1} in plaice, and 107 – 129 Bq kg^{-1} in cod [12]).

In regard to edible tissue, detected radium activity concentrations in *L. aurata* were found to be higher than in the flesh of Baltic Sea fish (0.057 Bq kg^{-1} in cod [12]), or 0.26 Bq kg^{-1} – in muscle of fish in the Cauvery River – India [5] , and 1.4 Bq kg^{-1} – in muscle of herring from Quirke Lake in Canada [3]. On the other hand, in some organs (in particular in bones) of the other aquatic (or semi-aquatic) species significantly higher radium activity concentrations were determined (e. g. 466.6 Bq kg^{-1} – in otters *Lutra canadensis*[15]).

At the same time, ^{232}Th activities in muscles were found to be significantly higher than radium ones, and its values for *L. aurata* are, for example, in accordance with findings for fish from lakes near the city of Elliot Lake, Ontario – Canada, where ^{232}Th activity in all body tissues was found to be below 20 Bq kg^{-1} [16].

Moreover, no one skeleton showed ^{137}Cs activity above minimum detectable one and, again, thorium activities are found to be higher than radium ones. However, this radium activity in *L. aurata* skeleton, although higher than in fish from Cauvery River – Indija (1.3–2.8 Bq kg^{-1} [5]), is comparable with radium activity in herring from Quirke Lake in Canada (an average of 18 Bq kg^{-1} [3]).

Radionuclide activity concentrations in seawater, sediment and mud with detritus are reported in Table 2.

The measurements of seawater sample showed level of ^{137}Cs in area of Tivat higher than, for example, in the Arabian Sea (<0.03 Bq L^{-1} [17]), but lower than in some areas of the Baltic Sea [18]. Radium-226 activity is found to be in accordance with that in the Arabian Sea (<0.4 Bq L^{-1}), as well as ^{232}Th (measurements of ^{228}Ra (^{232}Th decay product) in the Arabian Sea showed level <0.1 Bq L^{-1} [17]).

Table 2. Radionuclide activity concentrations in seawater, sediment and mud with detritus

Sample	^{137}Cs, Bq kg^{-1}	^{40}K, Bq kg^{-1}	^{226}Ra, Bq kg^{-1}	^{232}Th, Bq kg^{-1}
Seawater (in Bq L^{-1})	0.05±0.02	7.91± 0.65	0.08±0.02	0.10±0.03
Sediment	1.16 ± 0.22	102.5 ± 7.11	7.32±0.75	8.61±0.82
Mud with detritus	4.74±0.12	175.12 ± 10.36	10.35±1.66	11.71±0.50

Cesium-137 activity concentration in sediment sample is also found to be in accordance with findings for the Arabian Sea (below 1.3 Bq kg^{-1}), while ^{226}Ra activity is found to be lower than there (from 16.9 to 31.7 Bq kg^{-1} [17]), and than in the Baltic Sea sediments (from 10 to 100 Bq kg^{-1} [18]). Additionally, ^{226}Ra and ^{232}Th activity in the Tivat area sediment are found to be lower than corresponding values in the sediments of Irish Sea (23.9 and 36.7 Bq kg^{-1}, respectively [19]) or Indian Ocean (13.8 and 26.7 Bq kg^{-1}, respectively [20]).

The levels of all radionuclides in dry mud with detritus sample were found to be higher than in the surface sediment. This is important to know because mullet (including *L. aurata*) feed on detritus, mud and various organic materials from the Sea floor [21].

The results of CFs calculations (Eqs. 1, 2 and 3) are presented in Table 3.

Table 3. Concentration factors for *L. aurata*

L. aurata	Cs, CF$_1$	Cs, CF$_2$	Cs, CF$_3$	K, CF$_1$	K, CF$_2$	K, CF$_3$	Ra, CF$_1$	Ra, CF$_2$	Ra, CF$_3$	Th, CF$_1$	Th, CF$_2$	Th, CF$_3$
1	–	–	–	12.03	0.93	0.54	7.25	0.08	0.06	14.7	0.17	0.12
2	–	–	–	9.49	0.73	0.43	8.25	0.09	0.06	15.1	0.17	0.13
3	4.6	0.19	0.05	15.32	1.18	0.69	24.62	0.27	0.09	9.7	0.11	0.08
4	27	1.16	0.28	10.77	0.83	0.49	16.12	0.18	0.12	10.2	0.12	0.09
5	–	–	–	14.78	1.14	0.67	23.75	0.26	0.18	18.8	0.22	0.16
6	–	–	–	6.73	0.52	0.30	12.62	0.14	0.10	9.2	0.11	0.08
7	–	–	–	10.42	0.80	0.47	6.62	0.07	0.05	6.4	0.07	0.05
Muscle – 6	49.4	2.13	0.52	12.81	0.99	0.58	17.87	0.19	0.14	45.4	0.53	0.39
Muscle – 7	40.4	1.74	0.43	16.42	1.27	0.74	10.37	0.11	0.08	73.9	0.86	0.63
Skeleton – 6	–	–	–	4.05	0.31	0.18	373.87	4.1	2.89	542	6.29	4.63
Skeleton – 7	–	–	–	7.38	0.57	0.33	232.5	2.55	1.80	201	2.33	1.71

For those whole fish where ^{137}Cs was detected, the average CF from water was 15.8. The average CFs$_2$ (from sediment) was 0.67, and the average CFs$_3$ (from mud with detritus) – 0.16.

It is important to point out that ^{40}K is the largest source of natural radioactivity in animals and people. It is found in considerable amount in *L. aurata* (whole individuals and muscles) as well, i. e., significantly higher than in seawater, somewhat lower than in mud with detritus, and comparable with that in sediment. In skeleton, its activity concentration is found to be lower than in sediment and mud with detritus. However, many aspects have to be taken into account in considering potassium CFs, and that will be performed in a further research. So, here presented results (with an average of 11.4 for CF$_1$, 0.87 for CF$_2$, and 0.51 for CF$_3$) should be seen as an illustration, only.

Concentration factors CF$_1$ for ^{226}Ra (five whole individuals) showed an average of about 16. The CF$_2$ and CF$_3$ for this radionuclide have average values of about 0.18 and 0.1, respectively. Concentration factors CF$_1$ for ^{232}Th (whole individuals) showed an average of 14. The CF$_2$ and CF$_3$ for the same radionuclide have average values of about 0.16 and 0.12, respectively.

By considering CFs for *L. aurata* muscles and skeleton, thorium and radium CF$_1$ for skeleton were found to be the highest (an average of 371.5 and 303.1, respectively) and then thorium and cesium CF$_1$ for muscles (59.6 and 44.9, respectively). In regards to the radium, CFs$_2$ and CFs$_3$ also show accumulation of radium in skeleton more than 20 times higher than in the muscle of *L. aurata* species. Alhough only two muscle and skeleton samples were analyzed, taking into account an average value of CFs, it can be concluded that thorium CFs (from seawater, sediment and mud with detritus) showed accumulation of thorium in skeletons more than 7 times higher than in muscles of this fish species.

CONCLUSIONS

Concentration factors of radioisotopes for the mullet species *L. aurata* from the South Adriatic Sea, have been determined for the first time. The results presented here can be useful in a further research, in which fish, seawater, sediment and mud with detritus sampling should be performed along the whole Montenegro Coast. Radionuclide activity concentrations and CFs for other fish (including the other mullet species) should be determined and compared to obtain more detailed picture of radionuclides transfer from this marine environment to fish tissues.

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