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^{90}Sr AND ^{137}Cs IN PILCHARDS FROM THE ADRIATIC SEA

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The biological concentration factors for the system pilchard/sea-water for ^{90}Sr and ^{137}Cs are presented and discussed. The ^{90}Sr concentration factor was constant, whereas the concentration factor for ^{137}Cs steadily increased after the Chernobyl accident. An UNSCEAR model was used to describe ^{90}Sr transfer from fallout deposition to pilchards. The transfer coefficient was calculated to be 1.85×10^{-3} Bq yr $kg^{-1}/(Bq\ m^{-2})$. The dose incurred by pilchard consumption was estimated for the Croatian population, the annual collective equivalent dose being less than 0.1 Sv.

Key terms: marine organisms, radioactive contamination, fallout, radiochemical methods, sea-water

Nuclear tests conducted in the atmosphere and releases of the radioactive material from nuclear facilities generated the radioactive contamination that has entered the oceans mainly through the air–sea interface. Furthermore, the oceans are the ultimate recipients of the run-off from the land masses.

Among anthropogenic radioactive nuclides ^{90}Sr is regarded as the fission product of great potential hazard to living beings because of the unique combination of its relatively long (29.12 years) half-life, the very energetic beta particle of its ^{90}Y daughter, and its general resemblance to calcium metabolic processes. Considering the long half-lives of ^{90}Sr and ^{137}Cs dose assessment requires understanding of their behaviour in the environment. In the marine environment much of the research on the fate of radionuclides is focussed on determining their concentrations in sea-water and marine organisms and along exposure pathways. Dose prediction models for humans depend crucially on the transfer coefficient values quantifying the movement of radionuclides through food chains. Pilchards (*Pilchardus sardina risso*) being the predominant species in total sea-food catch in Croatia are potentially a major source of radioactive contamination in the human food chain for the Croatian population.

MATERIAL AND METHODS

Samples of sea-water, 150 litres each, were collected from the Adriatic Sea twice a year (in May and October, if feasible). The sampling took place 3 km from the shore, at

a depth of 0.5 metres, at four sampling sites (towns of Rovinj, Rijeka, Split and Dubrovnik). Fallout samples were collected monthly in the town of Zadar. Fish samples were obtained commercially on the fish-markets in the above sampling towns, at the same time when sea-water was sampled. In the laboratory fish samples were oven-dried and ashed at 450 °C. For strontium and caesium determination radiochemical methods were used (1–3).

The ^{90}Sr radioactivity was determined by beta-counting its decay product, ^{90}Y, in a low-background anti-coincidence counter, whereas ^{137}Cs and other gamma emitters were measured directly by gammaspectrometry.

RESULTS AND DISCUSSION

^{90}Sr and ^{137}Cs activity concentrations in sea-water

The radioactive fallout resulting from large-scale nuclear weapon tests in the atmosphere conducted in the 1960s, followed by similar, but smaller scale tests in the 1970s and onwards, was the dominant route for the introduction of artificial radionuclides into the Mediterranean Sea until the nuclear accident at Chernobyl, former USSR on 26th April 1986. Therefore, the mean annual sea-water activity concentration of ^{90}Sr tends to be in correlation with the fallout activity (i.e. surface deposit in $Bq.m^{-2}$), the coefficient of correlation being 0.72 (4). The base-line levels of ^{90}Sr and ^{137}Cs in 1985 for the Adriatic Sea were around 3 and 5 Bq m^{-3} respectively.

The nuclear accident at Chernobyl did not increase ^{90}Sr activity concentrations in marine organisms to any significant extent, contrary to ^{137}Cs. Unlike the debris from the atmospheric testing of nuclear weapons, the radionuclides that originated from the Chernobyl accident were not released directly into the upper atmosphere. As the result of the release mechanism and the prevailing meteorological conditions at the time, the refractory components of the Chernobyl debris (e.g. ^{90}Sr) were deposited closer to the accident location than the more volatile constituents (i.e. radiocaesium) (5). Thus, ^{90}Sr did not become involved in the global dispersion processes, having been deposited to the Earth's surface within a period of a few days to a few weeks after the accident. In addition, the late spring and early summer of 1986 in the east Adriatic region were very dry (only 5 mm of precipitation in May 1986 against the 1963 – 1990 May average of 50 mm) leading to very low direct radioactive contamination of the Adriatic. Therefore, on the Croatian Adriatic coast elevated ^{90}Sr activities, which persisted for several years, were found only in cistern water samples (6).

Contrary to ^{90}Sr, high radiocaesium (i.e. ^{137}Cs and ^{134}Cs) activities persisted in the Adriatic surface waters for several years after the accident, contaminating marine organisms as well. ^{137}Cs activity concentrations in marine organisms dropped exponentially, the turnover times being 2.2, 2.0 and 2.4 years in pilchards, mussels (*Mytilus galloprovincialis lamk.*) and poulps (*Ozoena moschata lamk.*) respectively (7).

Concentration factors

In contact with a variety of organisms living in the marine environment, the radionuclides introduced into the sea undergo various biological processes and enter complicated marine food-webs. As the major route of man's exposure to artificial radionuclides from the marine environment is through ingestion of contaminated marine organisms, the levels of radioactive contamination of those organisms are key factors for radiopro-

tection of the marine environment. Biological concentration factors are defined as the ratio of activity concentration, usually on a wet basis, of radionuclide in biological material to the activity concentration in the ambient sea-water. Thus the concentration factor for radionuclide k is:

$$CF^k\ (L\ kg^{-1}) = \frac{A^k\ organism\ (Bq\ kg^{-1})}{A^k\ sea\text{-}water\ (Bq\ L^{-1})} \qquad /1/$$

Since the partitioning of radionuclides between biological materials and the ambient sea-water is achieved through the complicated combination of biological processes such as assimilation, excretion, grazing, specific absorption, ion-exchange etc., it is practically impossible to understand the precise mechanism of the partitioning. Therefore, concentration factors do not make any reference to the partitioning mechanisms, simply representing the ratios of radionuclide activity concentrations between biological material and ambient water (8). Nevertheless, if the concentration factor of a specific radionuclide is known for a given marine organism, the level of the radionuclide activity concentration in that organism could be predicted and controlled, based on its activity concentration in the ambient water, which is far more easily determined.

Concentration factors for pilchards are given in Table 1.

Table 1. *Biological concentration factors (L kg^{-1}) for pilchards*

Year	^{90}Sr	^{137}Cs	^{40}K
1969	43.3±6.6	-	-
1971	42.9±6.6	-	-
1975	39.0±6.2	-	-
1980	39.8±6.3	58.7± 3.8	9.7±1.6
1986	-	30.0± 2.7	8.4±1.3
1987	-	42.2± 3.2	7.8±1.3
1988	35.7±6.0	50.0± 3.5	11.6±1.7
1989	46.1±8.1	114.7± 8.7	9.9±1.5
1990	49.4±0.3	188.8±17.1	8.4±0.0
1991	41.4±1.9	149.8± 6.1	9.6±1.5
Average	42.2±5.2	90.6±56.6	9.3±1.2

Although the ^{90}Sr activity concentration in sea-water was found to be greater in samples collected in May than in those collected in October, the absence of a significant variation in ^{90}Sr mean residence time for different locations, the average being 12.96±0.67 years, implies a similarity of oceanographical factors at individual locations concerned (4). Consequently, the ^{90}Sr concentration factor for pilchards tends to be fairly constant.

As our analysis of pilchards included the bones (pilchards are usually consumed with bones), and ^{90}Sr is known to be a bone-seeker owing to its biochemical similarity to calcium, the corresponding concentration factor could be expected to be greater than the one determined for the fish flesh/sea-water system. For comparison, the ^{90}Sr concentration factor for fish flesh in the Yellow Sea is 15.0±12.7 (9).

The concentration factor for ^{137}Cs in sea-water after the Chernobyl nuclear accident steadily increased as the ^{137}Cs activity concentration decreased, much faster than in pil-

chards. The concentration factors for ^{90}Sr and natural ^{40}K are relatively constant. While the concentration factor for ^{40}K in pilchards is of the order of 10, for the fresh-water species it is 10^3– 10^4 since under ambient conditions the biological systems regulate tissue concentrations of potassium (thus ^{40}K as well) to a constant level.

^{90}Sr transfer from fallout to pilchards

Provided ^{90}Sr concentration factors for the system pilchards/sea-water (Table 1) and the average ^{90}Sr sea-water activity concentrations are known (10, 11), it is possible to reconstruct ^{90}Sr activities in pilchards.

To assess the ^{90}Sr transfer from fallout to pilchards, several functions were tested, but the best fit was obtained by applying the function recommended by *UNSCEAR* (12) for food:

$$A_k(t) = b_1\dot{U}_k(i) + b_2\dot{U}_k(i-1) + b_3\sum_{m=1}^{\infty} e^{-\mu m}\,\dot{U}_k(i-m) \qquad /2/$$

where:

$A_k(t)$ is the activity concentration of radionuclide *k* in food (pilchards), the unit for $A_k(t)$ being Bq kg^{-1},

$\dot{U}_k(i)$ is the fallout deposition rate of radionuclide *k* in year *i* (Bq m^{-2} yr^{-1}),

$\sum e^{-\mu m}$... is the cumulative fallout deposit for radionuclide *k* as the result of deposition in previous years (Bq m^{-2} yr^{-1}),

μ^{-1} is the effective mean life of available ^{90}Sr in food (pilchards),

b_1, b_2, b_3 are the factors which can be derived from reported data by regression analysis. The unit is Bq yr kg^{-1}/(Bq m^{-2}).

Equation /2/ assumes the chain model for the transfer of radionuclides between environmental compartments, linking the input to the atmosphere to the dose in man:

Input → Atmosphere → Earth's surface → Diet → Tissue → Dose

0 1 2 3 4 5

The physical meaning of terms in model /2/ is as follows: the first term (rate factor) describes direct deposition and transfer, the second term (lag factor) describes contamination through fallout from previous year, and the third term (sea factor) reflects the contamination from fallout deposition accumulated from all preceding years, the exponential describing the combined physical decay and any other decrease in availability of considered radionuclide due to various other processes (like sedimentation dilution etc.).

Table 2 shows ^{90}Sr activities in samples of the Adriatic sea-water, fallout and pilchards. As ^{90}Sr activity concentrations for pilchards were not systematically measured, the missing data were reconstructed multiplying ^{90}Sr activity concentrations in sea-water for respective years by the average ^{90}Sr concentration factor given in Table 1.

Table 2. *^{90}Sr activity in the Adriatic sea-water, fallout and pilchards*

Year	Sea-water Bq m^{-3}	Fallout Bq m^{-2}	Pilchards Bq kg^{-1}
1963	14.84±2.37 *	813.4±9.0 **	**0.63±0.13** ***
1964	11.56±1.68	804.8±9.0	**0.49±0.09**
1965	13.72±2.86	290.5±5.4	**0.58±0.14**
1966	10.24±1.76	200.2±4.5	**0.43±0.09**
1967	6.56±1.94	69.9±2.6	**0.28±0.09**
1968	13.53±4.12	106.9±3.3	**0.57±0.19**
1969	10.43±1.94	78.4±2.8	0.45±0.10
1970	5.42±2.70	79.8±2.8	**0.23±0.12**
1971	4.72±0.75	61.8±2.5	0.20±0.04
1972	3.05±1.18	45.5±2.1	**0.13±0.07**
1973	3.05±1.18	14.1±1.2	**0.13±0.05**
1974	1.93±0.52	56.1±2.4	**0.08±0.02**
1975	2.80±0.60	40.9±2.0	0.12±0.03
1976	2.71±0.73	62.8±2.5	**0.11±0.03**
1977	2.55±0.79	58.3±2.4	**0.11±0.04**
1978	2.92±0.88	31.9±1.8	**0.12±0.04**
1979	4.38±1.65	20.0±1.4	**0.19±0.07**
1980	3.78±0.61	24.5±1.6	0.16±0.03
1981	3.37±0.99	41.4±2.0	**0.14±0.05**
1982	2.20±0.87	10.1±1.0	**0.09±0.04**
1983	1.10±0.30	5.8±0.8	**0.04±0.01**
1984	2.43±0.91	9.9±1.0	**0.10±0.04**
1985	2.66±0.17	6.6±0.8	**0.11±0.02**
1986	2.08±1.52	7.7±0.9	**0.09±0.07**
1987	2.87±0.48	6.2±0.8	**0.12±0.03**
1988	2.74±0.31	13.3±1.2	0.12±0.02
1989	2.48±0.32	1.8±0.4	0.11±0.02
1990	2.14±0.09	6.6±0.8	0.09±0.01
1991	2.19±0.11	5.3±0.7	0.09±0.01

* Annual average of four locations (Rovinj, Rijeka, Split and Dubrovnik)
** Total annual fallout (surface deposit) measured in Zadar, reported as ± one sigma counting error
*** Reconstructed activity concentrations with estimated total error in bold

Regression analysis gives the following values:

$b_1=1.00 \times 10^{-4}$, $b_2=2.98 \times 10^{-4}$, $b_3=2.46 \times 10^{-4}$ Bq yr $kg^{-1}/(Bq\ m^{-2})$ and $\mu=0.1568\ yr^{-1}$, the coefficient of correlation being 0.70. Thus, the effective mean life of ^{90}Sr in pilchards is $1/\mu=6.4$ years. The ^{90}Sr activity concentration in pilchards and the fit obtained by equation /2/ are shown in Figure 1.

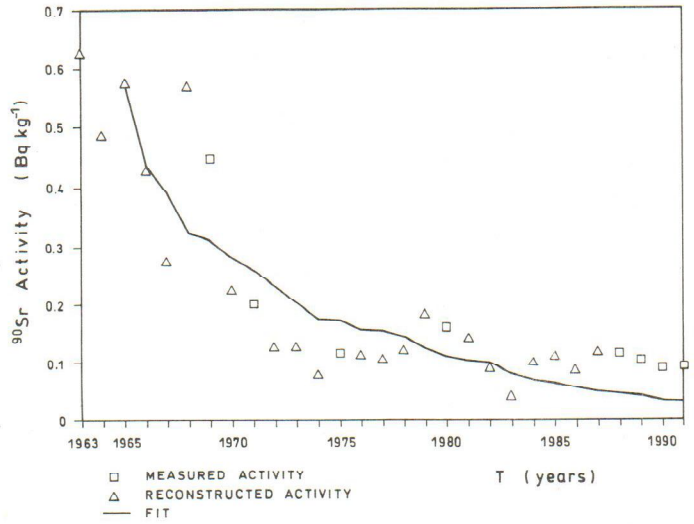

Figure 1. *^{90}Sr activity concentration in pilchards in the Adriatic Sea (Bq kg^{-1})*

The transfer between successive steps in the pathway chains is described by transfer coefficients, which relate infinite time integrals of activity concentration in the relevant compartments. Thus, the transfer coefficient from fallout deposition (compartment 2) to diet (compartment 3) is given by equation (12):

$$P_{23} = \frac{\int_0^\infty A_k(t)dt}{\int_0^\infty U_k(t)dt} \qquad /3/$$

where:
$A_k(t)$ is the activity concentration of radionuclide *k* in food, i.e., pilchards (Bq kg^{-1}) and $\dot{U}_k(t)$ is the fallout deposition rate of radionuclide *k* (Bq m^{-2} yr^{-1}).

As for values of $A_k(t)$ and $\dot{U}_k(t)$ assessed on the yearly basis the integration can be replaced by summation, the combination of equations /2/ and /3/ leads to:

$$P_{23} = b_1 + b_2 + b_3 \frac{e^{-\mu}}{1 - e^{-\mu}} \qquad /4/$$

P_{23} for ^{90}Sr in pilchards was assessed to be 1.85 x 10^{-3} Bq yr kg^{-1}/(Bq m^{-2}). That means that with each Becquerel deposited on an area of 1 m^2 of sea by fallout, the activity concentration of one ton of pilchards increases by 1.85 Bq. For comparison, the transfer coefficient P_{23} for total diet was estimated to be 4 x 10^{-3} Bq yr kg^{-1}/(Bq m^{-2}) (12).

From investigations of radioactive fallout in Denmark, Farøe Islands and Greenland, the mean transfer coefficient to fish was estimated to be 1.0 x 10^{-4} Bq yr $kg^{-1}/(Bq\ m^{-2})$, i.e. it was about 20 times lesser (13). However, these investigations were performed for various fish species, not only for surface fish such as pilchards. And, as mentioned earlier, we have analysed pilchards together with the bones, the critical organ for ^{90}Sr accumulation.

Dose incurred by pilchard consumption

Only about 1-1.5% of total human food comes from seas and oceans although seas occupy 71% of the Earth's surface. In Croatia, despite its being a maritime country, the percentage is even lower. It is estimated that in Zagreb, capital of the Republic of Croatia fish consumption is about 5 kg yr^{-1} per person (14). As total catch of marine organisms in Croatia is only about 20 000 tons (15), the average per caput consumption is much lesser. For comparison, in neighbouring Italy the annual fish catch in the Adriatic Sea is about 100 000 tons (15). The fraction of pilchards in total fish catch in Croatia in the past few decades has steadily increased and nowadays it is more than 75% (15, 16). From that quantity about 50% is used for human consumption, the rest is being either exported or used as animal food.

Per caput dose incurred due to fish (pilchard) consumption depends on the average activity concentration of a radionuclide and on the quantity consumed. The dose can be expressed as:

$$H_c = f\ C \sum_k D_{cf}(k) A_k \qquad /5/$$

where:
H_c is the mean annual collective equivalent dose (man Sv yr^{-1}),
f = 0.5 fraction of total fish catch used for human food in Croatia,
C total annual catch of pilchards in Croatia (1.5 x 10^7 kg yr^{-1}),
$D_{cf}(k)$ dose conversion factor for radionuclide *k*.i.e. equivalent dose per unit input, which converts the ingested activity concentration to equivalent dose; $D_{cf}(^{90}Sr)$=3.5 x 10^{-8} Sv Bq^{-1} and $D_{cf}(^{137}Cs)$=1.3 x 10^{-8} Sv Bq^{-1} (17) and
A_k average activity concentration of radionuclide *k* in pilchards (Bq kg^{-1}).

For the Croatian population (4.5 x 10^6 inhabitants), the annual collective equivalent dose, H_c, due to ^{90}Sr and ^{137}Cs ingestion by pilchard consumption, estimated for 1991 when the ^{90}Sr and ^{137}Cs activity concentrations were ≈ 0.1 Bq kg^{-1} and 0.7 Bq kg^{-1}, was 0.094 Sv yr^{-1}; 0.026 Sv was due to ^{90}Sr and 0.068 Sv to ^{137}Cs. Thus, the average annual equivalent dose for a member of Croatian population is 21.0 nSv (5.8 nSv due to ^{90}Sr and 15.2 nSv due to ^{137}Cs). As activity concentrations of long-lived fission products radionuclides, ^{90}Sr and ^{137}Cs, in other marine organisms do not significantly differ from those in pilchards, doses incurred by sea-food consumption are small.

Even though the critical population (fishing communities along the Croatian coast of the Adriatic) consumes far more sea-food than average, doses are still small.

To assess the collective dose due to ^{90}Sr for the Croatian population in the 1963 – 1991 period the integrated activity concentration of pilchards from Table 1 (6.12 Bq kg^{-1} yr) should be multiplied by the total annual catch of pilchards, the fraction of catch used for human food and dose conversion factor which leads to collective effective dose of 1.61 man Sv and average dose per person of 0.36 μSv.

CONCLUSION

Generally, after the Chernobyl nuclear accident the activities of fission radionuclides, ^{90}Sr and ^{137}Cs in pilchards in the Adriatic Sea have been relatively low, amounting to about one per cent of the naturally occurring ^{40}K activity. After the Chernobyl accident the concentration factors for ^{90}Sr and ^{40}K in the pilchards/sea-water system have been constant, while those for ^{137}Cs still vary.

The transfer coefficient for pilchards is of the same order of magnitude (10^{-3}) as for other food. Doses to the Croatian population, incurred by pilchard consumption are small, thus marine food is of secondary importance in respect to human intakes of ^{90}Sr and ^{137}Cs.

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Sažetak

^{90}Sr i ^{137}Cs U SRDELAMA JADRANSKOG MORA

Prikazani su i razmotreni biološki koncentracijski faktori za sustav srdele/morska voda za ^{90}Sr i ^{137}Cs. Koncentracijski faktor za ^{90}Sr je konstantan, dok je koncentracijski faktor za ^{137}Cs i dalje u porastu nakon nuklearne nesreće u Černobilju.

UNSCEAR-ovim modelom opisan je transfer ^{90}Sr od radioaktivnih oborina do srdela. Transferni koeficijent procijenjen je na $1,85 \times 10^{-3}$ Bq god $kg^{-1}/(Bq\ m^{-2})$. Procijenjena je godišnja kolektivna doza za hrvatsku populaciju zbog konzumacije srdela i ona je manja od 0,1 Sv.

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Ključne riječi: morski organizmi, morska voda, radioaktivna kontaminacija, radioaktivne oborine, radiokemijske metode