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POST-CHERNOBYL INVESTIGATIONS OF RADIOCAESIUM ACTIVITY CONCENTRATIONS IN ADRIATIC SEA PILCHARDS

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Investigations in the post-Chernobyl period (1986–2009) of radiocaesium activity concentrations in Adriatic pilchards are presented. Compared with pre-Chernobyl period, the Chernobyl nuclear accident caused increase of ^{137}Cs activity concentrations in pilchards. By fitting the measured ^{137}Cs activity concentrations to the theoretical curve was estimated to be 1.5 ± 0.4 y for 1986–90 and 5.8 ± 0.4 y for 1991–2009 and the bimodal behaviour for the ecological half-life of ^{137}Cs in pilchards has been observed. Estimated annual effective doses received by ^{134}Cs and ^{137}Cs intake due to consumption for an adult member of Croatian population are small. Collective dose for the 1986–2009 period was $4.9 + 0.3$ person-Sv. The observed ^{134}Cs/^{137}Cs activity ratio in pilchards was similar to the ratio that has been found in other environmental samples. The concentration factor for pilchards was roughly estimated to be 93.7 ± 39.2 l kg^{-1}, which is consistent with the values observed elsewhere.

INTRODUCTION

In addition to naturally occurring radionuclides, atmospheric nuclear weapons tests, resulting in global fallout and accidental or routine releases of radioactive material from various nuclear facilities are the main causes of man-made radioactive contamination that have entered the marine environment, including the Adriatic Sea. The main contributions of artificial radionuclides to the Adriatic marine environment, originate from global fallout from previous atmospheric nuclear weapons tests, fallout from the accident at the Chernobyl nuclear power plant and the river runoff, especially of the Po river.

Among man-made radionuclides, those of caesium and strontium, particularly ^{137}Cs and ^{90}Sr, are regarded as a particular hazard to organisms. This results from the unique combinations of relatively long physical half-lives (30.14 and 29.12 y, respectively) and the chemical and metabolic properties of these radionuclides, which closely resemble potassium and calcium, respectively. In aquatic environments both radionuclides tends to accumulate in fish, both through its diet and its gills.

The process of risk assessment both to the humans and biota, following the release of radioactive material to the environment requires the quantification of activity concentrations in environmental media and organisms. In the marine environment much of the research on the fate of radionuclides is therefore focused on determining their concentrations in sea water and along exposure pathways. But, in modern marine radioecology, it is broadly accepted that establishing environmental quality threshold values for overall coastal management requires more information on baseline levels than those currently available[1, 2]. The demand for contaminant monitoring is especially critical for enclosed or semi-enclosed basins, such as the Adriatic Sea, as well as water bodies characterised by long water residence time.

Therefore, investigations of the distribution and fate of natural, nuclear weapons-produced and reactor-released radionuclides in the Adriatic Sea marine environment, including marine organisms, have been conducted as a part of an extended and ongoing monitoring programme of radioactive contamination of the environment in Croatia[3–6]. Regular biomonitoring programme of radioactive contamination of pilchards (*Pilchardus sardina risso*), mussels (*Mytilus galloprovincialis*) and musky octopuses (*Ozaena moschata*) started after the Chernobyl accident. Marine radioecological investigations were described earlier in full detail[7].

Pilchards, being the predominant species in total sea–food catch in Croatia are potentially a major source of radioactive contamination in the human food chain posing a potential health hazard to the Croatian population through the direct irradiation and consumption of contaminated foodstuffs.

In Croatia, pilchards represent the majority of the total sea–food fish catch and consumption. During the period 1986–2009, it was ~16 340 ± 7335 tones annually[8]. As virtually all this catch is consumed in Croatia, it implies an annual average consumption of ~2.6 kg of pilchards per person. Radioactive contamination of pilchards in the Adriatic Sea by fission products, i.e. by ^{90}Sr and to a lesser extent by radiocaesium has been studied previously[9]. However, these investigations were focused on the pre-Chernobyl period. Presented in this study are long-term post-Chernobyl investigations of radiocaesium activity concentrations in Adriatic Sea pilchards.

MATERIALS AND METHODS

Pilchard samples were obtained directly from fishermen, in the cities of Rijeka (45° 20′ N, 14° 28′ E), Zadar (44° 06′ N, 15° 15′ E) and Split (43° 26′ N, 16° 25′ E).

Fallout samples were collected in the city of Zadar. Samples of sea water were also collected in the cities of Rijeka and Split, as well as in Rovinj (45° 04′ N, 13° 37′ E), Plomin (45° 08′ N, 14° 12′ E), Kaštela (43° 33′ N, 16° 21′ E) and Dubrovnik (42° 39′ N, 18° 06′ E) usually twice a year (spring and autumn).

A map showing the locations of sea water and fish sampling sites is shown in Figure 1.

From each site, at least 5 kg of pilchards were obtained at the same time when sea water was sampled. The samples were taken in spring or early summer, in order to observe possible reflections of common spring and well documented activity peak in sea water and fallout[10] and its transfer to pilchards.

Fish samples were dried in an oven and then washed in a muffle furnace at 450°C for 24 h. The ^{137}Cs activity concentrations in pilchards are reported on a wet weight basis.

Regarding fallout samples for rainwater collection, funnels of 1 m^2 collection area were used. Precipitation was measured by a Hellman pluviometer.

For radiocaesium determination in fallout and sea water the gamma-spectrometric method was used, while trace level measurements were performed by direct, low-background high-resolution Ge gamma spectrometry as a routine gammaspectometrical technique that was accredited by Croatian Accreditation Agency—HAA[11]. All samples were gammaspectrometrically analysed using HPGe (full width at half maximum of 2.24 keV at 1.33 MeV ^{60}Co and relative efficacy of 74.2 % at 1.33 MeV) and/or Ge(Li) detector (resolution 1.78 keV on 1.33 MeV ^{60}Co, relative efficiency 16.8 %) coupled to a data acquisition system. The samples were measured in Marinelli beakers of 1 l volume.

Quality assurance and intercalibration measurements were performed through participation in an International Atomic Energy Agency (IAEA), World Health Organization and Joint Research Centre international intercalibration programmes, which also include the regular performance of blank and background as well as quality control measurements.

Counting times depended on sample activities and ranged from 10 000 to 250 000 s, typically being 80 000 s.

RESULTS AND DISCUSSION

^{134}Cs and ^{137}Cs activity concentrations in the Adriatic Sea

In the year 1985, ^{137}Cs activity concentrations in the Adriatic Sea water was measured to be 4.3 ± 0.3 Bq m^{-3}. However, after the nuclear accident at the Chernobyl nuclear power plant in Ukraine on 26 April 1986, fallout from highly radioactive atmospheric plumes originating from the damaged nuclear reactor has been spread and transported all over Europe. However, due to the prevailing meteorological conditions at the time after the accident, Croatia was only partially affected by the edge of one of the plumes which was especially true for the Adriatic coast[12]. Consequently, in 1986, the total surface deposition of ^{137}Cs measured in the fallout collected in the city of Zadar was 2360 Bq m^{-2}[4], which 75.1 % of the total measured ^{137}Cs surface deposit for the overall post-Chernobyl period (i.e. 3143.0 Bq m^{-2}). That fallout, entering the sea water through air-sea interface in addition to river runoff caused elevated radiocesium levels in the Adriatic Sea water and essentially constant levels of ^{137}Cs activity concentrations in the Adriatic Sea water in the pre-Chernobyl period of around 4 Bq m^{-3} increased by two orders of magnitude, i.e. to 117.3 ± 31.7 Bq m^{-3}.

After reaching peak levels they exponentially decreased ever since[10]. In addition, in the Adriatic Sea environment, the presence of ^{134}Cs (half–life of 2.06 y) was detected for the first time. Namely, ^{134}Cs is not produced in significant amounts in nuclear explosions of fission weapons. ^{134}Cs is, however, found in reactor inventories due to long irradiation times of reactor fuels. Therefore, the presence of this radionuclide in the environment clearly indicated that the nuclear accident occurred.

Consequently, elevated levels of both ^{137}Cs and ^{134}Cs were detected soon after the Chernobyl accident detected in marine biota, including pilchards. However, in 1986, the average value of ^{134}Cs activity concentration that has been observed in pilchards was 1.5 ± 0.7 Bq kg^{-1}.

Figure 1. Map showing the sea water, fallout and fish sampling sites.

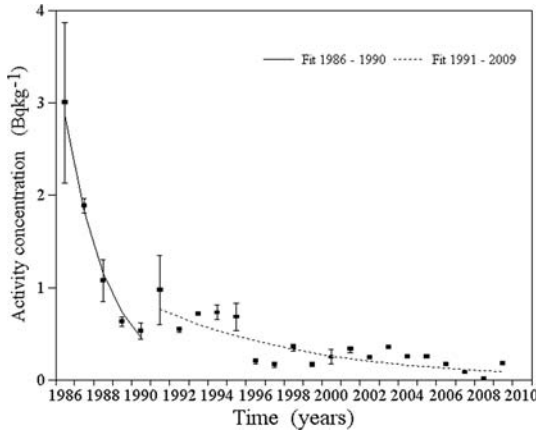

Figure 2. Observed and modelled ^{137}Cs activity concentrations in pilchards as a function of time during 1986–90 and 1991–2009 respective periods. The solid exponential curves indicate the slope from a least-square fitting with correlation coefficient $r^2=0.99$ and $r^2=0.74$ for respective periods.

Regarding ^{137}Cs, in 2009 its activity concentration in the Adriatic Sea water was measured on pre-Chernobyl levels, i.e. 2.3 ± 0.2 Bq m^{-3}.

Effective ecological half-life of ^{137}Cs in pilchards

The highest post-Chernobyl ^{137}Cs activity concentration recorded in pilchards was 3.00 ± 0.87 Bq kg^{-1} in 1986, while in 2009, the value was only 0.18 ± 0.01 Bq kg^{-1}. The latter value is comparable with the ^{137}Cs activity concentration found in the late 2000s for freshwater fish, especially carps[13]. For comparison, pre-Chernobyl ^{137}Cs activity concentration in pilchard, i.e. in the year 1985 was ~0.2 Bq kg^{-1} while the activity concentration of ^{90}Sr was 0.09 Bq kg^{-1}[4, 9].

To study the time course of the ^{137}Cs activity concentrations and to assess the effective ecological half-life of ^{137}Cs in pilchards, measurement data have been graphed as a function of time. The distribution shows a rapid decrease during first several years after the Chernobyl accident while after ~5 y, this decrease was significantly less rapid. The similar pattern has been observed for some other foodstuffs in Croatia and elsewhere[14, 15].

A first order kinetic equation was used to parameterise time changes in the ^{137}Cs activity concentrations in pilchards. Therefore, for the respective periods, 1988–90 and 1991–2009, the measured data were fitted to the following exponential function:

$$A_{pil}(t) = A_{pil}(0)e^{-kt} \quad (1)$$

where $A_{pil}(t)$ is time-dependent activity concentration of ^{137}Cs in pilchards (Bq kg^{-1} wet weight), $A_{pil}(0)$ initial activity concentration of ^{137}Cs in pilchards (Bq kg^{-1} wet weight) and $\ln(2)/k = T_{1/2,eff}$ effective (observed) ecological half-life of ^{137}Cs in pilchards (years).

In the immediate period after the Chernobyl accident, i.e. 1986–90 the observed effective ecological half-life of ^{137}Cs in pilchards was ~1.5 y (r=0.99, $P(t)$ <0.01 for three degrees of freedom). However, the effective ecological half-life of ^{137}Cs in pilchards increased to 5.8 y for the 1991–2009 period (r=0.74 with $P(t)$ <0.001 for 17 degrees of freedom). The observed and modelled ^{137}Cs activity concentrations in pilchards are shown in Figure 2.

In order to obtain the standard deviation of $T_{1/2,eff}$, Monte Carlo simulations were performed. To ensure conservative estimates, as well as to simplify calculations, a uniform distribution has been assumed over the $A \pm 2\sigma$ value of ^{137}Cs activity concentrations in pilchards for respective years, although a normal distribution would be more realistic. For each year a random value was generated over the interval $[A-2\sigma, A+2\sigma]$ and a $\ln(2)/k$ value estimated by fitting Equation (1) to the stochastically generated data. This process was repeated 500 times and 500 values for $\ln(2)/k = T_{1/2,eff}$ were thus obtained. From this data set, the mean values and standard deviations for $T_{1/2,eff}$ were calculated to be 1.52 ± 0.39 y for 1986–90 period and 5.76 ± 0.37 y for 1991–2007 period.

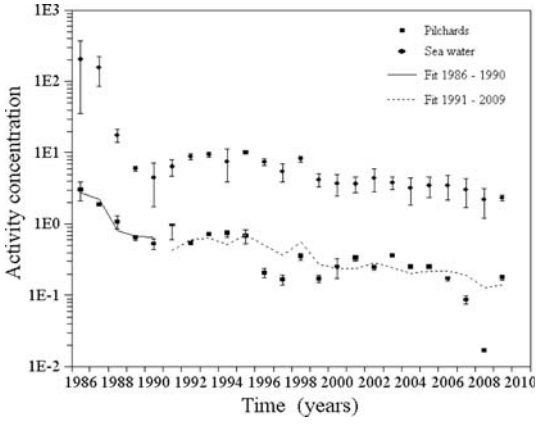

Figure 3. Observed and modelled ^{137}Cs activity concentrations in pilchards (Bq kg^{-1}) as a function of ^{137}Cs activity concentrations in the sea water (Bq m^{-3}) during 1986–90 and 1991–2009 respective periods.

Similar bimodal behaviour with comparable ^{137}Cs ecological half-lives has been observed for the Adriatic *Mytilus galloprovincialis* mussels[16], but also for other foodstuffs[15]. Regarding other seas, these results, especially in the latter period, are in good agreement with the literature for the ecological half-life of ^{137}Cs in other fish, e.g. Yamada and Nagaya[17] reported effective half-life for (unspecified species) of sea fish to be ~7 y. However, Morita and Yoshida[18] reported effective ecological half-lives of ^{137}Cs for various fishes in Japanese waters in the range of 18.1 y for fish living in open ocean sea waters to 13.5 y for coastal fishes.

For comparison, observed ecological half-lives for ^{137}Cs in carps in Croatia were found to be 1.05 ± 0.06 and 5.05 ± 0.32 y, which are slightly lesser than the effective ecological half-lives observed in pilchards for the respective 'fast' and 'slow' ^{137}Cs decreasing periods[13, 14].

The effective ecological half-life results from biological elimination of ^{137}Cs from fish as well as radioactive decay. In order to estimate the 'true' ecological half-life the effective constants k should be corrected for radioactive decay. Therefore constant k from Equation (1) can be written as:

$$k = \lambda + k_R \quad (2)$$

where $\ln(2)/\lambda$=30.14 y is the physical half-life of ^{137}Cs and $\ln(2)/k_R = T_{1/2,e}$ is ecological half-life for ^{137}Cs.

From Equation (2), the 'true' ecological half-lives for ^{137}Cs in pilchards $T_{1/2,e}$ were found to be 2.09 ± 0.69 and 6.98 ± 0.53 y, which are slightly higher than the effective ecological half-lives for the respective 'fast' and 'slow' periods.

^{137}Cs activity concentrations in pilchards, sea water and fallout

Contrary to the magnitude of the caesium accumulation in freshwater fish that is related to potassium concentrations[19], sea water fish exhibit different radiocaesium uptake kinetics discussed elsewhere[20]. Namely, fish living in sea water are faced with a water loss and a possible surplus of electrolytes, normally eliminated via the chloride cells in the gills. To compensate for water losses, sea water fish are constantly drinking, adding an alternative route of entry for caesium. Apart from these two possible uptake routes for waterborne caesium, fish, either in freshwater or sea water, may also take up radiocaesium via their diet[21].

Therefore, it can generally be expected that ^{137}Cs activity concentration in pilchards as pelagic, i.e. migratory fish species, will be in good correlation with ^{137}Cs activity concentrations in sea water, the fact that the Adriatic Sea water is very well mixed[22] being a contributing factor.

In both respective periods, i.e. 1986–90 and 1991–2009, the observed ^{137}Cs activity concentrations in pilchards are in very good correlation with ^{137}Cs activity concentrations in the sea water (r=0.97, $P(t)$ <0.01 for 3 degrees of freedom for immediate post-Chernobyl period and r=0.71 with $P(t) < 0.001$ for 17 degrees of freedom for the latter period). ^{137}Cs activity concentrations in pilchards modelled from sea water data are presented in Figure 3.

As it has been previously been demonstrated that ^{137}Cs activity concentrations in the Adriatic Sea water are highly influenced by ^{137}Cs activity concentrations in fallout[10] it could be expected that, consequently, ^{137}Cs activity concentrations in pilchards are also correlated with that in fallout.

Indeed, in both respective periods, i.e. 1986–90 and 1991–2009, the observed ^{137}Cs activity concentrations in pilchards are in very good correlation with ^{137}Cs activity concentrations in the sea water (r=0.92, $P(t) < 0.01$ for 3 degrees of freedom for immediate post-Chernobyl period and r=0.79 with $P(t) < 0.001$ for 17 degrees of freedom for the latter period). ^{137}Cs activity concentrations in pilchards modelled from sea water data are presented in Figure 4.

Such fair correlation leads to greater significance of radioecological sensitivity (Rs) of pilchards as a useful tool that helps to compare sensitivities of various environmental samples to radioactive contamination.

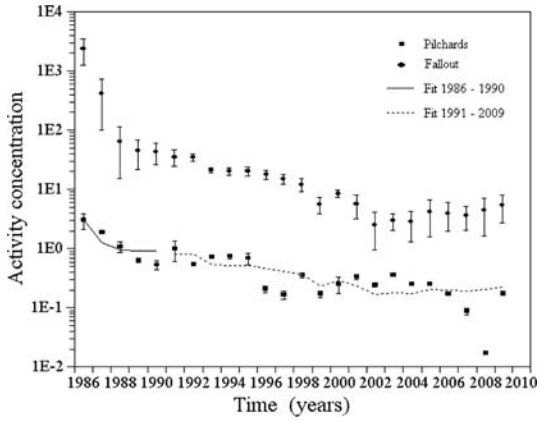

Figure 4. Observed and modelled ^{137}Cs activity concentrations in pilchards (Bq kg^{-1}) as a function of ^{137}Cs activity concentrations in fallout (Bq m^{-2}) during 1986–90 and 1991–2009 respective periods.

Radioecological sensitivity

Rs is defined as the infinite integral of activity concentrations of a particular radionuclide in a given environmental sample to the integrated deposition. Rs is sometimes also called the transfer coefficient from fallout to sample and in the case of food samples is equivalent to United Nations Scientific Committee on the Effects of Atomic Radiation transfer coefficient P_{23}[23]. Mathematically, P_{23} is defined as follows:

$$P_{23} = \frac{\int_0^\infty A(t)\mathrm{d}t}{\int_0^\infty \dot{U}(t)\mathrm{d}t} \quad (3)$$

where $A(t)$ is the activity concentration of given radionuclide (Bq kg^{-1}) in food and $\dot{U}(t)$ fallout deposition rate of this radionuclide (Bq m^{-2} y^{-1}).

As for values of $A(t)$ and $\dot{U}(t)$ assessed on the yearly basis the integration can be replaced by summation, for the overall observed period, i.e. from 1986 to 2009, the value of P_{23} for ^{137}Cs in pilchards can be easily calculated to be 4.4×10^{-3} Bq y kg^{-1}/(Bq m^{-2}). That means that with each Becquerel deposited by fallout on an area of one square meter of the Adriatic Sea surface the activity of 1 ton of beef increases approximately by 4.4 Bq.

However, as in the year 1986, direct ^{137}Cs deposition was very high, it affects the overall result. Therefore, when the year 1986 is excluded from analysis, P_{23} for ^{137}Cs in pilchards for 1987–2009 period is calculated to be 1.4×10^{-2} Bq y kg^{-1}/(Bq m^{-2}).

To put the obtained values into perspective, the ^{137}Cs transfer coefficient P_{23} for total diet was estimated to be approximately 1.2×10^{-2} Bq y kg^{-1}/(Bq m^{-2}) for the 1962–79 period in New York (reference location for a northern hemisphere) and 8.0×10^{-3} Bq y kg^{-1}/(Bq m^{-2}) for the 1963–73 period in Argentina (southern hemisphere)[23].

^{134}Cs to ^{137}Cs activity ratio in pilchards

The presence of ^{134}Cs (half-life of 2.06 y) in the Croatian environment was detected for the first time detected in May 1986. The estimated amount of caesium released after the reactor explosion at Chernobyl was 3.7×10^{16} Bq of ^{137}Cs (13 % of total reactor inventory) and 1.9×10^{16} Bq of ^{134}Cs (10 % of total reactor inventory[24]). Thus, the initial value for the ^{134}Cs:^{137}Cs activity ratio in May 1986 was 0.51. Radioecological studies of environmental samples showed that activity ratio has not been altered during the passage of the radioactive plume

from Chernobyl to other geographical areas in Europe.

As the half-life of ^{137}Cs (30.14 y) is about 15 times longer than that of ^{134}Cs, the ^{134}Cs:^{137}Cs activity ratio $R(t)$ should decrease, in a predictable way, due to differential radioactive decay, according to the following relationship:

$$R(t) = \frac{(1.9 \times 10^{16})}{(3.7 \times 10^{16})} \times e^{\ln(2) \times t \times (1/T_1 - 1/T_2)} \qquad (4)$$

where t is the time elapsed after the Chernobyl accident and T_1 and T_2 are the physical half-lives for ^{137}Cs and ^{134}Cs, respectively.

In 1986, the observed ^{134}Cs:^{137}Cs activity ratio in pilchards was 0.47 ± 0.07 that was slightly lower than the theoretical prediction of 0.51. However, in 1987, the ratio was higher than predicted, the ratio being 0.53 ± 0.04, approaching theoretical value ever since (Figure 5). These variations can be explained by a number of environmental factors that naturally fluctuate.

Similar ratios, decreasing according to Equation (4), have been found in most other environmental samples[4]. In 1990, contamination of pilchard by ^{134}Cs originating from the Chernobyl accident was detectable only at a very low level and afterwards was below the detection limit.

Concentration factors

Radionuclides once introduced into water systems, including oceans and seas, undergo various biological processes and may enter complicated food-webs. The level of radioactive contamination of aquatic biota by specific radionuclides is usually defined in terms of a concentration factor (CF). For particular radionuclide, CF is defined as the ratio of activity concentration in a biological material to the activity concentration in the ambient sea water:

$$\mathrm{CF}_m \frac{A_\mathrm{m}(\mathrm{biota})}{A_\mathrm{m}(\mathrm{water})} \qquad (5)$$

where CF_m is the concentration factor of radionuclide m for a specific organism (l kg^{-1}) A_m (biota) is the activity concentration of radionuclide m in particular organism (Bq kg^{-1} wet weight) and A_m (water) is the activity concentration of radionuclide m in sea water (Bq l^{-1}).

It should be noted that CFs do not have any mechanistic basis, simply representing the ratios of radionuclide activity concentrations between biological material and ambient water. However, if the CF of a specific radionuclide is known for a given organism, the level of the radionuclide activity concentration in this organism can be very easily predicted, based on its activity concentration in the ambient water. It should be noted that the use of CFs assumes equilibrium between organisms and water, which may not be the case in the real environment due to complex inputs of radionuclides and the variability in parameters influencing activity concentrations in abiotic compartments.

The calculated value of CF for ^{137}Cs in the Adriatic Sea pilchards for the overall post-Chernobyl period is 93.7 ± 39.2 l kg^{-1}. A similar CF value of 90.6 ± 56.6 l kg^{-1} has been estimated for the 1986–91 period in previous preliminary investigations[9].

Generally, CFs values estimated for the Adriatic Sea pilchards correspond with other reported fish CF values, e.g. in a comprehensive database published by IAEA[25], CF for fish was recommended to be 100 l kg^{-1} and Fisher *et al.*[26] reported a mean CF for ^{137}Cs of 146 ± 68 for various pelagic fish species collected in the Kara and Barents Seas.

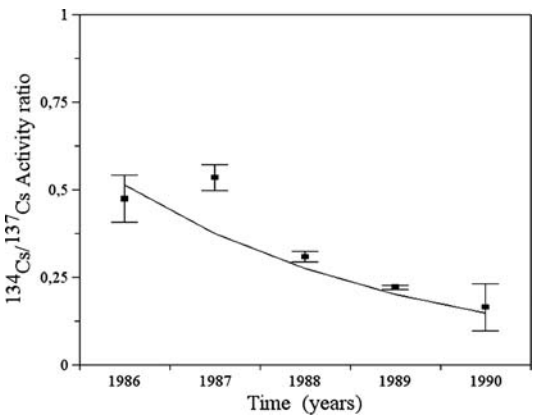

Figure 5. Observed and predicted ^{134}Cs to ^{137}Cs activity concentration ratio in pilchards.

Dosimetry

Due to the relatively high contribution of ingestion dose to total dose received by the Croatian population after the nuclear fallout[27], a reliable knowledge of ingestion dose is of particular importance. Data on activity concentrations of ^{134}Cs and ^{137}Cs in pilchards allow us to estimate the doses incurred by consumption of this fish species. Dose conversion factors, i.e. effective dose per unit intake via ingestion for adult members of the public, are 1.3×10^{-8} Sv Bq^{-1} and 1.9×10^{-8} Sv Bq^{-1} for ^{137}Cs and ^{134}Cs, respectively[28]. As the ratio of dose conversion factors for ^{137}Cs and ^{134}Cs is $\asymp 0.7$, it implies that ingestion of ^{134}Cs contributes $\sim$30 % more to

the dose, compared with ingestion of the same activity concentration of ^{137}Cs.

In the Republic of Croatia, overall consumption of fish, including fresh water and sea species, is relatively small, being between 7 and 8 kg per year per person[8]. The collective effective dose incurred due to food consumption over a specific time period depends on the activity concentration within the food and on the quantity of that food which is consumed. The dose can be expressed as:

$$E = C \sum_{m} D_{\mathrm{m}}^{\mathrm{cf}} A_{\mathrm{m}} \qquad (6)$$

where E is the effective dose in Sv, C is the total annual *per capita* consumption of food, D_m^{cf} is the dose conversion factor for radionuclide m, i.e. effective dose per unit intake, which converts the ingested activity to effective dose and A_m is the mean annual specific activity of radionuclide m in food (Bq kg^{-1}).

Based on the statistical data for the catch of pilchards in Croatia 1986–2009 period (16 340 $\pm$ 7335 tones annually) the same consumption rate was assumed for the period 1986–2009 as well. However, it should be noted that such consumption is overestimation since certain fraction of total catch is exported as well as used as animal feed.

Therefore, a conservative estimated of the collective effective dose due to ^{137}Cs and ^{134}Cs ingestion by pilchard consumption for the Croatian population (4.5×10^6 inhabitants) during 1986–2009 is 4.9 ± 0.3 person-Sv. Of this value 3.7 ± 0.3 person-Sv can be attributed to ^{137}Cs and 1.3 ± 0.3 person-Sv to ^{134}Cs. The majority of the overall dose was incurred during the 1986 (1.1 and 0.8 person-Sv for ^{137}Cs and ^{134}Cs, respectively).

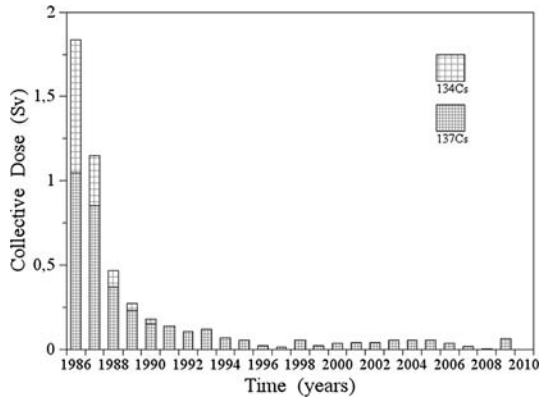

Figure 6. Estimated annual collective effective doses due to ^{134}Cs and ^{137}Cs intake due to consumption of pilchards by the Croatian population.

The annual effective collective doses are shown in Figure 6.

It should be noted that the doses were not corrected to take into account inedible parts of the fish such as bones and scales since pilchards are frequently eaten whole. Nevertheless, these are rather small doses, since for the 1986–2009 period per capita dose is 1.1 μSv. However, the doses are small not only because of low activity concentrations of radiocaesium in pilchards, but also due to the quite low consumption of fish in Croatia. In summary, it can be argued that consumption of pilchards, as most represented sea water fish in Croatian diet, is not a critical pathway for human intake of radiocaesium from the environment.

CONCLUSIONS

A relatively rapid decrease in ^{137}Cs activity concentrations in pilchards was observed for the immediate post-Chernobyl period (1987–1990), the effective ecological half-life of ^{137}Cs in pilchards within this period being approximately 1.5 y. However, approximately 5 y after the accident, this rate of decrease slowed significantly and the effective ecological half-life of ^{137}Cs in pilchards increased to 5.8 y, the same pattern being observed for some other fish and other foodstuffs.

The CF for pilchards (wet weight) was estimated to be 93.7 ± 39.2 l kg^{-1}, a value which is in reasonable agreement with fish CF values observed elsewhere. However, the knowledge about CFs for various organisms is important in the context of ICRP's 2007 recommendations regarding protection of the environment from exposure to ionising radiation. Namely, CFs are valuable tools for the assessments of the environmental impact of radiation on wildlife.

The activity concentrations of ^{137}Cs in pilchards became quite low within a few years after the Chernobyl nuclear accident, while ^{134}Cs activity concentrations were generally below detection limit after 1990. As ^{137}Cs activity concentrations in pilchards are in good correlation with ^{137}Cs activity concentrations in the Adriatic Sea water, which is, itself, influenced by fallout, the similar decreasing pattern of ^{137}Cs activity concentrations has been observed in sea water and in fallout.

Consequently, only very low doses have been received by the general population as a result of pilchard consumption, as predominant sea water fish species in the Croatian diet, which is consistent with quite low Rs for pilchards which was estimated to be 4.4×10^{-3} Bq y kg^{-1}/(Bq m^{-2}).

Generally, it can be concluded that pilchard consumption was not a critical pathway for the transfer of radiocaesium to humans after the Chernobyl accident.

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