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Activity distribution of ^{90}Sr and ^{137}Cs in the Adriatic surface sea-water and fallout along the Croatian coast

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The activities of ^{90}Sr in the surface waters of the Adriatic Sea were investigated twice a year over the period 1963–1990 at four locations. A tendency was noted for the maximum concentrations to occur in the first half of the year. This can be explained by reflection of common spring activity peak on surrounding land masses. The ^{90}Sr activity of the North Adriatic region was greater than the activities of south regions in both spring and autumn. The mean residence time of ^{90}Sr in the mixed sea-water layer was practically identical at all locations. The lower limit of ^{90}Sr sedimentation rate in the Adriatic Sea was estimated to be 2.4×10^{-8} ms^{-1}, the upper limit being 4.9×10^{-7} ms^{-1}. The Chernobyl nuclear accident did not affect the activity of ^{90}Sr in the Adriatic surface waters.

Distribucija aktivnosti ^{90}Sr i ^{137}Cs u površinskim vodama i radioaktivnim oborinama duž hrvatske obale Jadranskog mora

Aktivnost ^{90}Sr u površinskim vodama Jadranskog mora istraživana je dva puta godišnje u periodu 1963–1990 na četiri lokacije. Maksimum koncentracije ^{90}Sr pojavljuje se u prvom dijelu godine, što se može objasniti kao odraz uobičajenog proljetnog porasta aktivnosti na okolnom kopnu. Aktivnost morske vode sjevernog Jadrana veća je od aktivnosti južnog Jadrana i u proljeće i u jesen. Srednje vrijeme boravka ^{90}Sr u miješanom sloju mora na svim ispitivanim lokacijama je podjednako. Donja granica brzine sedimentacije ^{90}Sr u Jadranskom moru procijenjena je na 2.4×10^{-8} ms^{-1}, a gornja na 4.9×10^{-7} ms^{-1}. Nuklearna nesreća u Černobilju nije utjecala na aktivnost ^{90}Sr u površinskim vodama Jadranskog mora.

1. Introduction

The anthropogenic radioactive contamination of the sea originates primarily from atmospheric nuclear weapon testing. As about 70 percent of world surface is covered by water, artificial radionuclides enter the oceans primarily by deposi-

tion through the air-sea interface. Furthermore, the sea is the ultimate recipient of the run-off from the land masses. The study of marine radioactivity includes not only the monitoring and hazard assessment, but also studies related essentially to oceanographic investigations using the radionuclides as tracers, that have been introduced through fallout or waste disposal operations. Among the many radionuclides produced in the testing of nuclear weapons, ^{90}Sr has been the most intensively and extensively examined one, owing to its large fission yield, its bone–seeking character (due to biological similarity to calcium) and a moderately long physical half-life ($T_{1/2}$ = 29.0 years) which makes it the greatest fallout hazard to man. Another radionuclide which has been widely studied in connection with the fallout from nuclear test explosions is ^{137}Cs. Analysis of most environmental samples studied in the investigations of nuclear fallout before Chernobyl nuclear accident (26 April 1986) yielded $^{137}Cs/^{90}Sr$ activity ratios ranging between the values of about 1 and 3. Since both nuclides have inert gaseous precursors in their fission chains and generally similar non-refractory chemical characteristics, substantial fractionation from the time of their creation in nuclear burst is considered unlikely and all of the fallout entering the sea is assumed to carry approximately the same ratio of ^{137}Cs to ^{90}Sr. Measurements of the $^{137}Cs/^{90}Sr$ ratio in ocean water largely lie within 1.45–1.80 (Broecker, 1966; Kupferman et al, 1979). Thus, data for pre-Chernobyl ^{90}Sr activities can be tested by known ^{137}Cs activities. Some radionuclides settle in the sediments (plutonium, and to some degree ^{137}Cs), while others, e.g. ^{90}Sr, stay in solution in the sea. In local waters, contamination from peaceful uses of nuclear energy may predominate over fallout. For areas with short turn-over times, sea-water concentrations respond rapidly to variations in discharge rate and marine currents.

The Adriatic Sea as the northern part of the Mediterranean Sea, is a rather shallow and partially enclosed sea, under the strong impact of the Po River as the major source of fresh water in the area. The Adriatic Sea is generally characterized by low precipitation, high evaporation, low tidal action, low nutrient content, low suspended load, and low biological productivity. While these features result in hydrographical conditions quite different from those in other seas, the fundamental biogeochemical processes taking place in water columns are not to be considered different from other seas (UNEP 1991).

Investigations of the distribution and fate of natural, weapon-produced and reactor-released radionuclides in the Adriatic Sea have been conducted in the Department for Radiation Protection of the Institute for Medical Research and Occupational Health, as a part of an extended monitoring programme. Strontium investigations have been going on since 1963 and those of caesium and some other radionuclides from 1978. The results are published yearly (Popović, 1962–1976, Bauman et al, 1977–1990).

2. Material and methods

Sampling sites and their coordinates are given in Table 1. Samples of sea-water, 150 litres each, were collected twice a year (May and October, if feasible) 3 km from the shore, at a depth of 0.5 meters, at four sampling sites (towns of Rovinj, Rijeka, Split and Dubrovnik).

Table 1. Coordinates of sampling sites

Location	Latitude (N)	Longitude (E)	Altitude* (m)
Dubrovnik	42° 39'	18° 06'	15
Pula	44° 51'	18° 52'	25
Rijeka	45° 20'	14° 28'	50
Rovinj	45° 04'	13° 37'	
Split	43° 26'	16° 25'	
Zadar	44° 06'	15° 15'	2

* Altitude refers to fallout sampling station

A network of meteorological stations was used for fallout collection. Fallout samples were collected monthly in the town of Zadar. After the Chernobyl nuclear accident quarterly collection of the fallout also took place in the towns of Pula, Rijeka and Dubrovnik. The funnels which were used for fallout collection had a 1 m^2 area. Precipitation height was measured by Hellman pluviometer.

For the determination of strontium and caesium our own radiochemical methods were used (Bauman and Juras, 1971, Bauman, 1974). The radioactivity of ^{90}Sr was determined with a low background Geiger Miller counter PHILIPS. Counting time was 80000 seconds.

A gamma-ray spectrometry system based on a low-level ORTEC Ge(Li) detector (FWHM 1.82 keV at 1.33 MeV) coupled to a computerized data acquisition system (4096-channel pulse height analyzer and personal computer) was used to determine radiocaesium levels in the samples from their gamma-ray spectra. Samples were measured in cylindrical plastic containers of appropriate volume which were placed directly on the detector. Counting time depended on sample activity, but was never less than 10000 seconds.

One sigma counting error of measured activities in sea-water or fallout samples never exceeded 10%.

Efficiency calibration was carried out using sources provided by the International Atomic Energy Agency (IAEA) and World Health Organization (WHO).

Quality assurance and intercalibrations of radioactivity measurements in sea-water and fallout were performed through participation in the IAEA and WHO quality control programmes.

3. Results and discussion

3. 1. ^{90}Sr activities in surface sea-waters

^{90}Sr is produced in the fission reaction that occurs during the explosion of a nuclear device and as a by-product of energy production with nuclear fission reactors. Atmospheric detonations of nuclear devices began in the mid 1940s. The most active year in the history of atmospheric nuclear weapon testing was 1962. As a result, 1963 saw highest fallout activities on the Adriatic coast and the Adriatic surface waters (Popović, 1963).

Within the hemispheres ^{90}Sr deposition is far from uniform. The seasonal meteorological phenomena that give rise to substantial interchanges of air between the stratosphere and the troposphere cause the release of stratospheric debris into the troposphere. This release, its latitudinal distribution and the precipitation patterns on the ground combine to produce in late spring an activity maximum (spring peak) in the fallout and global distribution with higher values in the mid-latitudes (UNSCEAR 1982). Figure 1 shows mean monthly ^{90}Sr fallout activities in Zadar and Zagreb for the period 1962–1990. Similar patterns have been observed for ^{137}Cs (Franić, 1992). A significant correlation ($r = 0.76$) was found between the activities in Zadar and those in Zagreb.

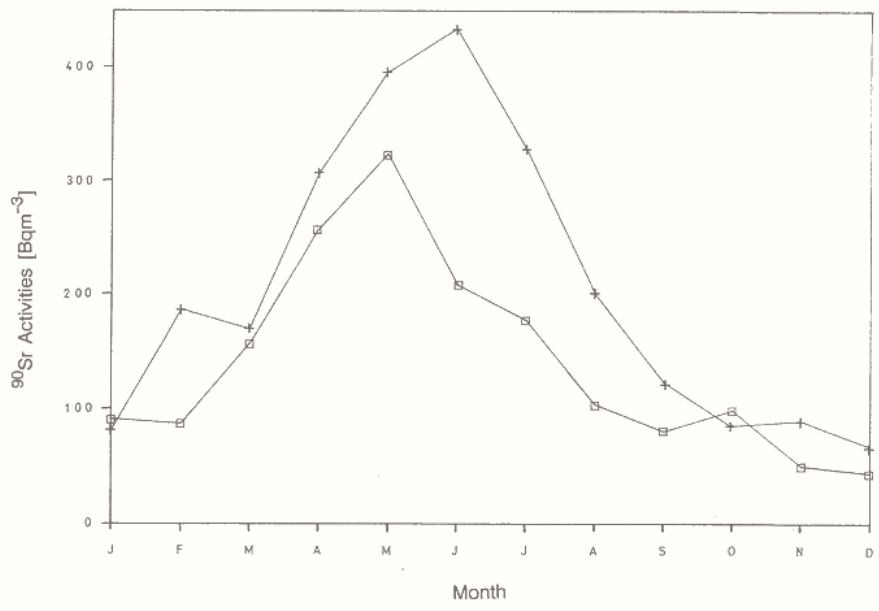

Figure 1. ^{90}Sr fallout activities throughout the year, during 1963–1990. □ – Zagreb, + – Zadar

While the spring peak of ^{90}Sr fallout on land, generally attributed to seasonal meteorological phenomena, has been extensively documented, on the sea neither direct measurements of ^{90}Sr in precipitation nor surface water samples seem to systematically reflect this annual fallout maximum at most sites.

In the Adriatic surface sea-waters the activity of ^{90}Sr is in a good correlation with fallout activity (i.e. surface deposit in Bqm^{-2}), the coefficient of correlation being 0.73. ^{90}Sr concentrations in the Adriatic Sea dropped exponentially from 14.8 ± 2.4 Bqm^{-3} in 1963 to 2.1 ± 0.1 Bqm^{-3} in 1990 (an average of four sampling sites). In the same period total annual surface deposit (in Zadar) fell by three orders of magnitude, from 636.3 ± 25.2 Bqm^{-2} in 1963 to 6.6 ± 0.6 Bqm^{-2} in 1990.

Figure 2 shows ^{90}Sr activity ratio between spring and autumn samples of surface waters at four different locations. Unfortunately, in the 1983–1986 period autumn samples of sea-water were not collected. At all locations the maximum ^{90}Sr concentrations tend to occur in the first half of the year. In the late 1980s the activity ratio (≈1) was approximately the same at all locations. But, in these samples (as well as in fallout), low ^{90}Sr activities (being essentially background variations), leading to greater uncertainties in activity ratios, were encountered.

Additional radiocaesium activity released in the atmosphere by the Chernobyl nuclear accident caused the ^{137}Cs spring activity peak in fallout to be more intensive (Bauman et al, 1977–1990; Franić, 1992). As in the case of ^{90}Sr the

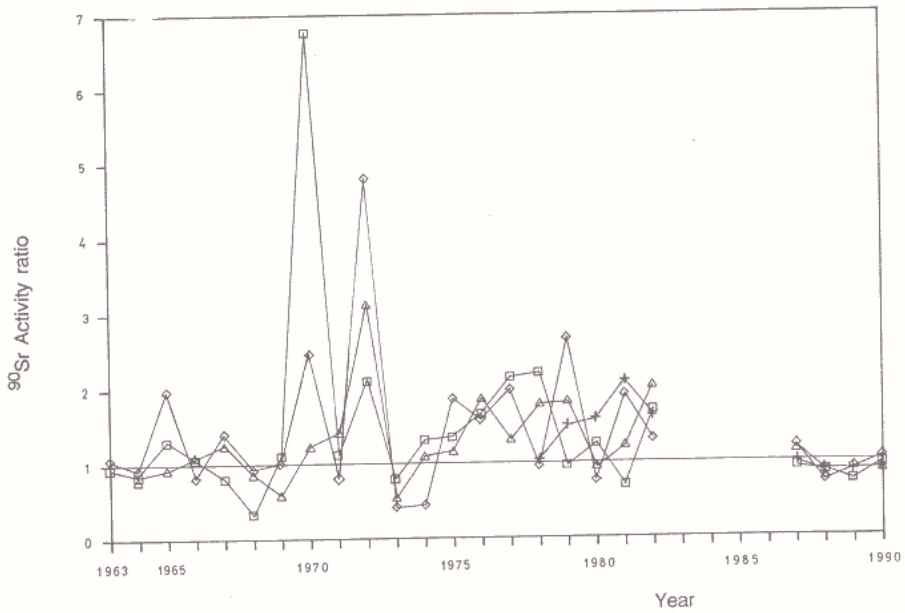

Figure 2. ^{90}Sr activity ratio in spring and autumn samples of the Adriatic surface waters, at four locations. □ – Rovinj, + – Rijeka, ◇ – Split, Δ – Dubrovnik

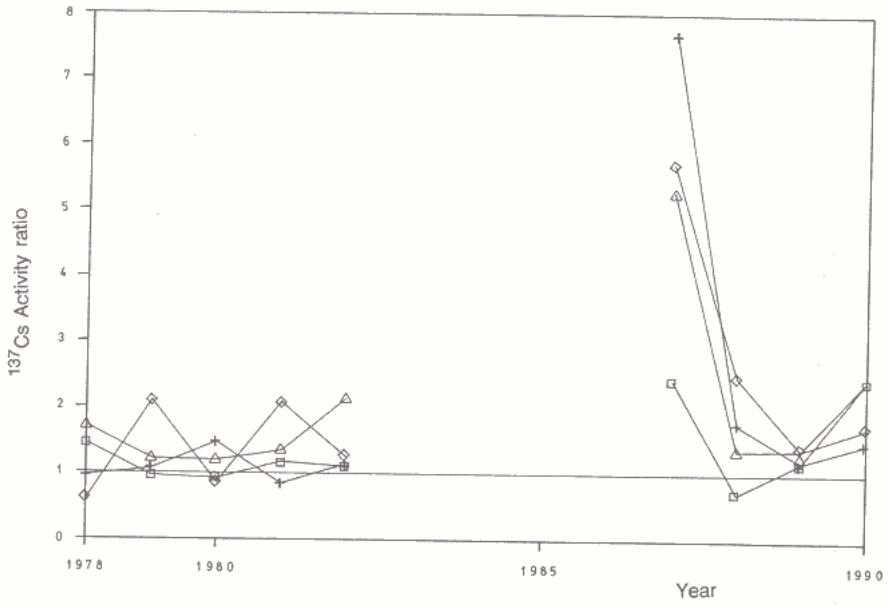

Figure 3. ^{137}Cs activity ratio in spring and autumn samples of the Adriatic surface waters, at four locations. □ – Rovinj, + – Rijeka, ◇ – Split, Δ – Dubrovnik

maximum ^{137}Cs concentration in sea-water occurred in the first half of the year (Figure 3). As the Adriatic Sea is a closed water body under a strong influence of the surrounding land masses, this might reflect a common spring peak on land rather than to be a systematic phenomenon. For a reliable detection of the spring activity peak in the Adriatic surface sea-water, monthly specific activity monitoring is called for at several locations. As in the 1990s ^{90}Sr concentrations have been too low and the radiochemical preparation of both ^{90}Sr and ^{137}Cs is too complicated as well as time consuming and costly, tritium would be ideal tracer for detecting the spring activity peak in sea-water. Substantially, all airborne tritium, both natural and bomb produced, combines to form tritiated water in the stratosphere, which is subsequently brought down into the troposphere where both precipitation and molecular exchange serve to transfer tritium into the surface ocean (sea) water. Such investigations are in progress in the Department for Radiation Protection.

Figure 4 shows ^{90}Sr activity ratio between the north (Rovinj and Rijeka) and the south (Split and Dubrovnik) Adriatic surface waters in spring and autumn. The activity of North Adriatic is greater in both seasons. As at any given latitude the deposition is affected by the amount of rainfall, higher precipitation in the North Adriatic region compared to the South Adriatic brings down more activity either directly by rainfall or by scavenging and washout processes.

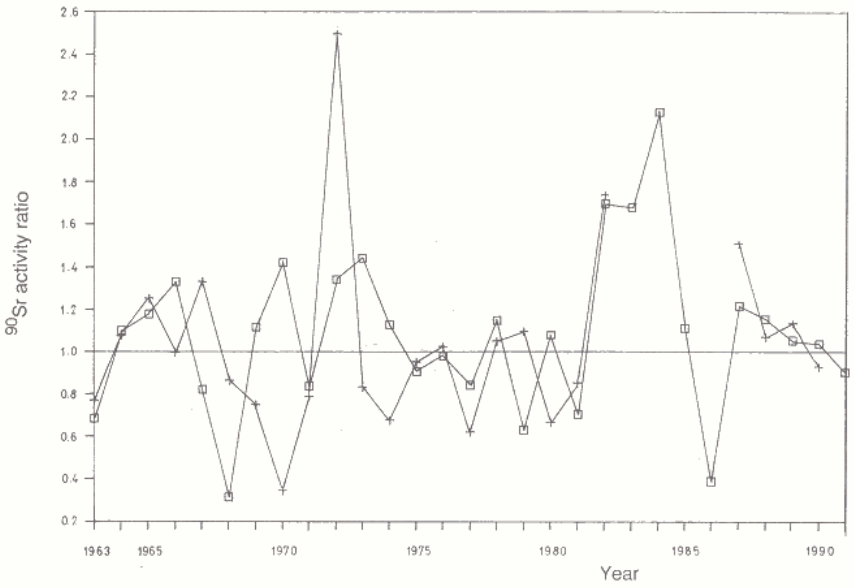

Figure 4. ^{90}Sr activity ratio between the North and South Adriatic surface waters.
□ – Spring, + – Autumn

The additional mechanism which may give rise to elevated ^{90}Sr activity in the North Adriatic basin can be the constant impact of the Po River which delivers to the Adriatic Sea not only the radionuclides which have escaped from two nuclear power plants but also the fallout from the northern Italy and southern Alps. The measurements of water profile activity in front of the Po River estuary (44° 45'N, 12° 44'E) indicate that the activity peak is at a depth of 15 meters (Jelisavčić 1989). Also, the radioactive contamination along the Italian coast is much higher in comparison with values along the Croatian coast (CNEN 1963–1966, Bauman et al. 1980). This contamination is spreading by diffusion, vertical mixing and convection. It can be expected that the wind-curl induced circulation at the surface and bottom of the North Adriatic (Kuzmić and Orlić 1987) stimulates the transport of radioactive contamination and other pollution towards the Croatian coast.

3. 2. ^{90}Sr sedimentation rate

The observed effective half-life, T_{eff}, of ^{90}Sr (which is longer than the real effective half-life since fresh fallout each year replaces some of the decayed/removed activity) in the mixed layer of the Adriatic Sea in 1963–1990 period was quite constant, 6.85±0.31 years (Franić and Bauman, 1993). No significant variations in half-lives, and relatively constant ^{137}Cs/^{90}Sr activity ratios (1.65±0.41, 1.54±0.36, 1.55±0.28, 1.48± 0.40 for Rovinj, Rijeka, Split and Dubrovnik, respectively) for the period before the Chernobyl accident, imply

similarity of oceanographical factors at individual locations concerned. This is in spite of the heterogeneity of the fallout deposition patterns over the Adriatic region. The mean residence time, T_m, of ^{90}Sr (radiological half-life T_r = 29 years) in the mixed layer, calculated from equation

$$T_m = \frac{1}{\ln(2)} \frac{T_{eff}\, T_r}{T_r - T_{eff}}, \tag{1}$$

is 12.96±0.67 years. Using mean residence time, the sedimentation rate (vertical velocity) of ^{90}Sr can be estimated. The thickness of the mixed layer (near surface waters in seas and oceans), a region in which rapid mixing occurs as a result of wind action, varies geographically from 10 to 200 meters, mean value being d = 100 meters (Eisenbud 1973). Then, the sedimentation rate of ^{90}Sr can be calculated from equation:

$$v_v = \frac{d}{T_m} \tag{2}$$

The estimated lower limit of sedimentation rate of ^{90}Sr in the Adriatic Sea is 2.4×10^{-8} ms^{-1}, the upper limit being 4.9×10^{-7} ms^{-1}. These, relatively low values can be explained by low suspended load, resulting in low sedimentation rate characteristic of the Mediterranean. For comparison, vertical velocities for the South Pacific, Carribean Sea and North Atlantic are 3.0×10^{-7} and 2.9×10^{-6} ms^{-1} respectively (Broecker et al. 1966).

3.3. Impact of the Chernobyl nuclear accident

A major source of ^{90}Sr deposition on a global scale during 1986 was ^{90}Sr released into the atmosphere following the explosions and subsequent fire at the nuclear power reactor station at Chernobyl, former U.S.S.R. The additional activity attributed to the Chernobyl accident was very soon detected in sea-water and on the Adriatic coast (especially ^{137}Cs, ^{134}Cs and ^{131}I), although radioactive contamination was low. While relatively high ^{90}Sr activities, which persisted for several years, were determined only in closed systems, such as cistern waters (Franić et al. 1992), elevated concentration in sea-water (4.7±0.4 Bqm^{-3}) was found only in Dubrovnik. This can be explained by a heavy rain in Dubrovnik in May 1986. In the same month, at other locations, activities varied from 1.1±0.1 Bqm^{-3} in Rijeka to 1.3±0.1 Bqm^{-3} in Split. Unlike debris from the atmospheric testing of nuclear weapons, the nuclides 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, e.g. ^{137}Cs (Aarkrog 1988). Besides, most of ^{90}Sr was deposited during a period from several days to a few months,

having little impact on sea-water activity. For comparison, ^{137}Cs and ^{134}Cs persisted in the Adriatic surface waters for several years after the accident.

The base-line levels of ^{90}Sr and ^{137}Cs in the Adriatić surface water in 1990 were around 3 and 5 Bqm^{-3} respectively, as they used to be in the pre-Chernobyl period, leading the $^{137}Cs/^{90}Sr$ activity ratio to approximately the same value as in 1985.

4. Conclusion

The ^{90}Sr activities tend to show their maximum in the first half of the year. Yet, it cannot be said that the Adriatic surface water reflects systematically the spring fallout activity maximum. Generally, the non-uniformity of observed seasonal ^{90}Sr variations in the fallout over the oceans and also in surface waters as compared to the common spring activity peak on land is not yet clearly understood. The activity in the North Adriatic region is greater than the activity in south regions. This can be readily explained by higher precipitation levels in the North Adriatic as well as by the Po River impact. The heterogeneity of fallout deposition patterns did not influence the mean residence time of ^{90}Sr in the mixed layer, which was about 6.9 years.

The Chernobyl nuclear accident did not affect the activity of ^{90}Sr in the Adriatic surface waters.

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