

Conference Paper / Review

MARINE RADIOECOLOGY AND WASTE MANAGEMENT IN THE ADRIATIC

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This paper gives a review of marine radioecology research in the Adriatic area carried out by the Radiation Protection Unit of the Institute for Medical Research and Occupational Health. Measurements of radioactivity in the Adriatic started in 1963 as a part of an extended monitoring programme of radioactivity in Croatian environment. The main sources of radioactive contamination of the Adriatic Sea are the fallout from past nuclear weapon testing conducted in the atmosphere and the Chernobyl accident. In 2005, the activity concentrations of fission radionuclides were detectable at very low levels in all environmental samples collected on the Adriatic. The ^{90}Sr data obtained from long-term monitoring were used to estimate the upper limit of the Adriatic seawater turnover time, which turned out to be (3.4 ± 0.4) years. Detailed knowledge about seawater circulation, including the turnover time is essential for planning an overall communal and other wastewater management on the Adriatic coast. The paper concludes with the prospects for future marine radioecological investigations.

KEY WORDS: ^{137}Cs , ^{90}Sr , seawater turnover time

Interest in marine radioecology owes to a great extent to concerns raised about the impact of atmospheric nuclear weapons testing in the 1950s and 1960s on the marine environment, and it soon proved to be a powerful tool for assessing radioactive contamination and evaluating doses received by population. However, with the development of civil nuclear programmes and the increasing problems of overall waste management in coastal areas, marine radioecology addressed the new challenges as well (1).

In addition, investigations targeted to reduce radiological impact on the environment, revealed the multidisciplinary advantages of radionuclides as marine radiotracers and clocks for various processes. Radiotracer studies helped to understand water circulation, measure biological productivity, and track the flux and fate of dissolved and particulate matter in the oceans (1). Therefore, marine radioecology is closely connected not only with the assessment of

human health, but also with physical oceanography, basic ecology, ecotoxicology, geochemistry and biogeochemistry, as well as with other fields covering pollution by radioactive and some non-radioactive substances. All of these investigations and resulting protective measures and activities have had a direct impact on improving the overall quality of life.

Among the anthropogenic radionuclides present in global fallout, ^{137}Cs and ^{90}Sr have been regarded as the fission products of major potential hazard to living beings due to the unique combination of relatively long half-lives and chemical and metabolic properties resembling those of the potassium and calcium, respectively (2).

Consequently, investigations of radiostrontium and radiocaesium in environmental samples take a significant part in an extended, ongoing monitoring programme of radioactive contamination of human environment in Croatia, including the Adriatic (3-6).

This paper gives an overview of the ongoing radioecological studies in the Adriatic area carried out by the Radiation Protection Unit of the Institute for Medical Research and Occupational Health in the broad context of wastewater management.

CHARACTERISTICS OF THE ADRIATIC SEA

The Adriatic is a rectangularly shaped basin, oriented in a NW-SE direction, with the length of about 800 km and width varying from 102 km to 355 km (250 km in average). The northern half of the Adriatic can be divided into three sub-basins (7): 1) the northernmost shallow basin with the bottom sloping gently to the south and not going over 100 m in depth, then dropping quickly to 200 m just south of Ancona; 2) the middle Adriatic basin with three pits located along the transversal line of the Italian city of Pescara, one of which is known as the Jabuka Pit; and 3) the southern half of the Adriatic consisting of a basin called the South Adriatic Pit, which is separated from the middle basin by a 170 m deep Palagruža Sill. It is characterised by approximately circular isobaths, with a maximum depth of 1233 m in the centre (8).

Further south, the bottom rises toward the Strait of Otranto, forming the Otranto sill (780 m) that separates the Adriatic from the Ionian Sea. The Strait of Otranto is about 70 km wide and the maximum sill depth is only 780 m (9).

Due to geomorphological characteristics (small, semi-enclosed and relatively shallow sea, almost land-locked between the high mountains situated on Balkan and Italian Peninsulas) the Adriatic Sea, with its coastal areas is potentially more sensitive to pollution than the rest of the Mediterranean, including anthropogenic and natural radionuclides.

In addition, the geomorphological characteristics of the Adriatic in combination with the continuous influence of mid-latitude meteorological perturbations and the wind systems associated with them, especially in winter, play an important role in controlling the dynamics of its waters. In particular *bora*, a dry and cold wind blowing in an offshore direction from the eastern coast, which affects the wind-driven and thermohaline circulation and, what is the most important, is responsible for deep water formation processes. Namely, the Adriatic Sea being exposed to very low winter temperatures and violent episodes of *bora* has been identified as one of the regions of the

world oceans where deep-water formation processes take place. Meteorological conditions favourable to dense water formation cause rapid mixing of surface waters with the intermediate water layer. This dense water spreads through the Strait of Otranto, being an important component of the Eastern Mediterranean Deep Water (EMDW) and contributing to their ventilation (10).

Water exchange pattern between the Adriatic and the greater Mediterranean through the Strait of Otranto, suggests an inflow along the eastern and outflow along the western coast. Within the outflowing current portion of the Otranto Strait area, cold water exits the Adriatic in the surface layer of the western shelf. On the other hand, in the western continental margin there is the cold outflowing vein of the dense water formed in the Adriatic (9).

Surface currents are responsible for the transport of a substantial portion of marine pollutants and for the freshwater dispersion. The circulation regime varies seasonally and inter-annually in response to changes in the heating and wind regimes. The winter circulation is characterised by a prevalence of warmer Mediterranean inflow, reinforced by southerly winds. In the summer, there is a stronger outflow of fresher and warmer Adriatic water along the western coast, supported by etesian winds blowing from the northwest (9).

RADIOECOLOGICAL MONITORING PROGRAMME IN THE ADRIATIC

Speaking of marine radioecology in Croatia, limited measurements of the Adriatic radioactivity have been carried by the Radiation Protection Unit of the Institute for Medical Research and Occupational Health since 1963 (3), as a part of an extended monitoring programme of radioactivity in Croatian environment. From then on, spring and autumn samples were taken to measure ^{90}Sr and ^{137}Cs activity concentrations in the Adriatic surface seawater on four locations: Rovinj, Rijeka, Split and Dubrovnik (4-6). Continuous fallout samples are collected in Zadar, and high-volume air sampling is conducted using appropriate air filters. In addition, samples of cistern waters, which proved to be excellent fallout collectors, are collected annually from 5 to 20 cisterns along the coast (11).

In addition, some marine organisms regarded as good bioindicators of anthropogenic radionuclides

Table 1 Samples, locations, sampling intervals and specific analyses performed in the radioecological monitoring programme in the Adriatic

| Species or sample | Location | Sampling frequency | Analysis |
|----------------------------------|--|--|---|
| Sea water | Rovinj, Plomin Bay, Rijeka, Kaštela, Split Dubrovnik | Spring and autumn | gammasspectrometry, ⁹⁰ Sr, ²²⁶ Ra |
| Fallout | Pula, Rijeka, Zadar, Dubrovnik | Continuous sampling, Analysis of 6-months aliquots | gammasspectrometry, ⁹⁰ Sr (only in Zadar) |
| Cistern waters | Bale, Doli, Marina, Pag | Spring | gammasspectrometry |
| Absorbed dose rate in air | Zadar | Continuous sampling, Analysis 4 times per year | gammasspectrometry |
| Bioindicator organisms | | | |
| <i>Ozaena moschata</i> | Rovinj, Plomin, Kaštela, Split Dubrovnik | Spring | gammasspectrometry, |
| <i>Mytilus galloprovincialis</i> | Zadar, Dubrovnik | Spring | gammasspectrometry, ⁹⁰ Sr |
| <i>Sardina pilchardus</i> | Zadar | Spring | gammasspectrometry, ⁹⁰ Sr |

Results are published annually (3-6)

are collected once a year, such as mussels (*Mytilus galloprovincialis*), octopuses, (*Ozaena Moschata*) and pelagic fish, pilchards (*Sardina pilchardus*) in particular (12).

Collected samples are analysed according to procedures given by HASL-300 Environmental Measurement Laboratory (EML) (13), the International Atomic Energy Agency (IAEA), and the World Health Organization (WHO).

Quality assurance and intercalibration of radioactivity measurements were performed through participation in IAEA, WHO, EC and other intercomparison programmes and proficiency tests. Our Radiation Protection Unit has recently started the process of accreditation according to the standard HRN EN ISO/IEC 17025:2005 (General requirements for the competence of testing and calibration laboratories.) Table 1 shows the programme of radioecological investigations in 2005.

RADIOACTIVE CONTAMINATION OF THE ADRIATIC

Time-evolution, levels and inventories of man-made radioactivity have been investigated well in the greater Mediterranean Sea. Extensive reviews of artificial radionuclide inputs into the Mediterranean before Chernobyl have been reported by the UNEP (14) and Holm (15). Papucci and Delfanti (16) and Delfanti et al. (17) analysed existing and new data on

¹³⁷Cs distribution in the Eastern Mediterranean. The main sources of ¹³⁷Cs in the Mediterranean area were the fallout from past nuclear weapon testing and the Chernobyl accident in 1986.

However, data on radioactivity in the Adriatic are quite deficient, except for ⁹⁰Sr and ¹³⁷Cs activities in surface seawater on a few locations close to the coast, gathered for monitoring purposes. Nevertheless, these data enabled investigations of the long-term behaviour of fallout radionuclides (⁹⁰Sr and ¹³⁷Cs) in the Adriatic Sea (18), originating from atmospheric nuclear tests.

Atmospheric nuclear explosions had been conducted since 1945 and were especially intensive in 1960s, i.e., before nuclear moratorium became effective. Regardless, similar, but smaller tests were continued by the Chinese and French in the 1970s and afterwards. Therefore, the activity concentration of fission products in most environmental samples should be in good correlation with fallout activity (i.e., surface deposit in Bqm⁻²), which proved to be true for the Adriatic seawater (18).

From 1963 to 1986, ¹³⁷Cs activity concentrations in surface water regularly decreased, reflecting the decrease of the atmospheric input and vertical transport processes. The Chernobyl accident caused ¹³⁷Cs activity concentration to peak again in seawater, decreasing ever since, while ⁹⁰Sr levels were unaffected (4-6), as the latter, being much less volatile than radiocaesium isotopes, did not reach Croatia in significant quantities.

Namely, as a consequence of the release mechanism and meteorological conditions, the refractory components of the Chernobyl debris such as ^{90}Sr were deposited closer to the accident location than the more volatile constituents like radiocaesium (19). Thus ^{90}Sr was not distributed globally, but deposited on the Earth's surface within a few days to a few weeks from the accident. In addition, the late spring and early summer of 1986 in Croatia were rather dry, leading to relatively low direct radioactive contamination of environment, which was especially true for the Adriatic region (18). Consequently, the nuclear accident at Chernobyl did not cause any major increase in ^{90}Sr activity in environmental samples in Croatia, the only exception in the Adriatic being the cistern waters due their large catchment area which makes them efficient fallout collectors (11).

In 2005, baseline activity concentrations for ^{137}Cs and ^{90}Sr in the Adriatic Sea were very low, about (2 to 3) Bq m^{-3} , which are essentially background variations. The upper limits of ^{137}Cs activity concentrations in pilchards were about 0.25 Bq kg^{-1} , in octopuses 0.1 Bq kg^{-1} and in mussels 0.13 Bq kg^{-1} . However, in 2005, the ^{137}Cs activity concentrations in cistern waters were still a few Bq m^{-3} (6).

TURNOVER TIME OF THE ADRIATIC SEAWATER

A combination of long-term marine observations, measurements of activity concentrations of anthropogenic radionuclides in the Adriatic seawater, and mathematical modelling made it possible to estimate the Adriatic seawater turnover time using fallout ^{90}Sr as an intrinsic radioactive tracer of water circulation.

The turnover time of the Adriatic Sea water, i.e. the time needed for the spontaneous exchange of the entire volume of the Adriatic seawater with the Ionian Sea is very important. Namely, this information is essential for any risk assessment dealing with environmental and health implications of releases of radioactive and non-radioactive pollutants into the sea. This is especially important in the context of the overall communal and other wastewater management on the Adriatic coast.

Earlier results on the volume transport of seawater through the Strait of Otranto, which were obtained from very limited and sporadic measurements of currents, led to the estimate of turnover time of approximately five years (20). However, as the measured values of

volume transport of seawater on certain spots had been in range of $2 \times 10^3 \text{ m}^3 \text{ s}^{-1}$ to $5 \times 10^5 \text{ m}^3 \text{ s}^{-1}$ (20), this called for further investigation.

Therefore, water exchange between the Adriatic and the Ionian Sea through the Strait of Otranto was the subject of a series of subsequent experimental investigations and more recently of numerical studies partially presented by *Cushman-Roisin et al.* (9). The turnover time of the Adriatic seawater can easily be calculated from the data on water fluxes through the Strait of Otranto, by calculating the annual water mass flowing through the strait and dividing it by the total volume of the Adriatic sea.

Literature data for Adriatic seawater turnover time range from 0.7 years to 5 years, although recent estimates obtained from direct measurements of currents in the Strait of Otranto are on the order of one year (9). For comparison, the turnover time for the Black Sea is around 500 years (21).

However, these results are based on the water flow measurements, which depend on meteorological conditions, time of the year, and a number of other environmental parameters that fluctuate naturally.

By modelling ^{90}Sr activity concentrations in the Adriatic Sea, the upper limit for the Adriatic seawater turnover time was estimated to be (3.4 ± 0.4) years (22), which is in excellent agreement with the results obtained by different methods. As this result is obtained using long-term data, it can be regarded as a sort of mean value, averaged over a few decades. It should be noted, however, that the mean residence time for ^{90}Sr in the Adriatic Sea was estimated to about 13 years (18) for the period 1963-1991.

Reliable information on the Adriatic seawater turnover time is especially important in the context of potential pollution by runoff that contain agricultural, industrial, and other waste products that are carried into surface waters by rainfall and melting snow, eventually ending in the Adriatic Sea. The long-term average runoff rate along the Adriatic coast is $(5500 \text{ to } 5700) \text{ m}^3 \text{ s}^{-1}$, the Po River carrying 28 % of the total runoff alone, i.e., $(1540 \text{ to } 1600) \text{ m}^3 \text{ s}^{-1}$ (9, 23, 24). It can easily be calculated that on annual scale, total runoff corresponds to annual addition of about 1.3 m thick water layer over the whole basin, which is approximately 0.5 % of the total Adriatic sea volume.

Reliable information on the Adriatic seawater turnover time is also extremely important for planning and managing industrial activities, including maritime transport such as the recently proposed project "Družba Adria".

FUTURE RADIOECOLOGICAL INVESTIGATIONS

Utilising the radiotracer techniques, marine radioecology can address many issues related to the inventories and the dispersion i.e. transfer pathways of pollutants delivered by runoff to the Adriatic Sea.

Future radioecological investigations in the eastern part of Adriatic will be focused on investigations of radiological and non-radiological characterisation of seawater, targeting in particular natural and some fission radionuclides. As many coastal tracers, particularly those of radium, show significant concentration changes due to the ballast water exchange, these investigations are related to the development of a quantitative tool that would reliably discriminate between exchanged, interchanged and non-exchanged ballast water tanks.

In return, this can be the basis of efficient and relatively cheap permanent monitoring of whether ballast water exchange procedures utilised by oil tankers and other ships and freighters trafficking in the Adriatic are compliant with domestic and international regulations and Croatian requirements.

In addition, future research will focus on:

- Studies of inventories and transfer pathways of radionuclides in some marine organisms (ecological residence time of fission and related radioecological assessment)
- Sediment studies from the sea floor, including the areas of Palagruža Sill and Jabuka Pit, and one transect in the north Adriatic. This is important for the studies of radionuclide translocation, age evaluation, sedimentation rate and even for detecting past events such as earthquakes. Sediment analyses will provide deeper insights into the re-mobilisation of naturally occurring radioactive material (NORM) and into the transport mechanisms of radioactive and non-radioactive contamination.
- Models and parameters for radioactive and non-radioactive pollution in the Adriatic
- Field gammaspectrometric and absorbed dose rate measurements in air and seawater on and around the volcanic islands of Jabuka, Brusnik, and Svetac.
- Studies of the impact of radionuclides on marine ecosystems and risks related to human exposure

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Sažetak

RADIOEKOLOGIJA MORA I UPRAVLJANJE OTPADOM NA JADRANU

Dan je pregled i opis radioekoloških istraživanja jadranskog područja koja provodi Jedinica za zaštitu od zračenja Instituta za medicinska istraživanja i medicinu rada. Mjerenja radioaktivnosti na Jadranu započela su godine 1963. kao dio proširenog programa monitoringa radioaktivnosti okoliša u Hrvatskoj. Glavni izvori radioaktivne kontaminacije Jadranskog mora jesu fallout (radioaktivni ispadak) prouzročen atmosferskim probama nuklearnog oružja te čornobiljska nesreća. Godine 2005. koncentracije aktivnosti fisijjskih radionuklida u uzorcima iz okoliša skupljenim na Jadranu bile su detektabilne na vrlo malim razinama. Podaci za ⁹⁰Sr dobiveni dugoročnim monitoringom poslužili su za procjenu gornje granice boravka morske vode u Jadranskom moru, koja je procijenjena na (3,4±0,4) godine. Detaljna saznanja o cirkulaciji morske vode, uključujući i srednje vrijeme boravka morske vode izuzetno su potrebna pri planiranju cjelokupnog upravljanja komunalnim i drugim otpadom na jadranskoj obali. Prikazana su planirana buduća radioekološka istraživanja.

KLJUČNE RIJEČI: ¹³⁷Cs, ⁹⁰Sr, srednje vrijeme boravka morske vode

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