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RADIOACTIVITY OF FISSION PRODUCTS IN CISTERN WATERS ALONG THE CROATIAN COAST OF THE ADRIATIC SEA

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Abstract

Measurements of radioactive contamination of water samples from cisterns collecting rainwater from roofs etc. with fission products have been carried out along the Croatian coast of the Adriatic Sea since 1968. An exponential decline of radioactivity followed the nuclear moratorium. After the nuclear accident at Chornobyl, higher levels of ^{137}Cs and ^{90}Sr were detected again, cistern waters being the only environmental samples in Croatia in which elevated ^{90}Sr activities persisted for several years. For the pre-Chornobyl period, the mean residence time of ^{90}Sr in cistern waters being order of magnitude several years was similar to that calculated for fallout. Contrary, for the post-Chornobyl time, ^{90}Sr mean residence time was calculated to be considerably shorter, reflecting the tropospheric mean residence time. Therefore, the pre-Chornobyl and the post-Chornobyl mean residence times of ^{90}Sr in cistern waters reflect the mechanism by which strontium was released to the atmosphere. The annual dose for the critical adult population received from ^{90}Sr, ^{134}Cs and ^{137}Cs by drinking cistern water was estimated to be a few per cent of the dose from natural background radiation.

Introduction

Shortage of water has been a major problem for the population since ancient times. However, people learned how to create for themselves a near equivalent of permanent water, even on the sites where Nature was not kind enough to place a river or a spring. The first text mentioning water cisterns is the "Moabite Stone"text around 850 BC (1). Namely, at that time King Mesha of Moab was victorious in war and conquered a considerable territory east of the Jordan river. Than he ordered that every man must make a cistern in his house. However, the valuable device itself has been invented considerably earlier. For example, a large secret cistern has been found in Mycenae below the royal palace, but still within the citadel walls. Romans also built huge water reservoirs, sometimes bigger than 10,000 m^3, and cisterns (2), as they developed cement similar to the kind used today. However, the knowledge of making cement has been lost after the fall of Roman Empire and consequently the art of building cisterns as well.

Cisterns are artificial storage tanks for rainfall that has been collected from a roof or some other catchment area. Although usually located underground, cistern reservoirs may be placed at ground level or on elevated stands, either outdoors or within buildings. Cisterns are watertight and usually have smooth interior surfaces, enclosed lids, and are large enough to provide adequate storage. Therefore, they are excellent cumulative samplers of radioactive material introduced into the atmosphere by nuclear tests that have been conducted in the atmosphere and the release of the radioactive material from nuclear facilities. As rainwater lacks calcium, ^{90}Sr in cistern waters represents potential risk for population depending upon the water supplies from them.

This paper summarizes the measurement results of ^{90}Sr and ^{137}Cs in cistern water samples along the Croatian coast of the Adriatic sea for the period 1968 - 1996. It should be noted that the rain water from those cisterns is nowadays used mainly as technical water and for irrigating, since fresh water is available.

Material and Methods

Samples of cistern waters were taken once a year on twelve locations (if possible) along the Croatian coast of the Adriatic sea. Strontium has been investigated since 1968, and occasionally caesium. After the Chornobyl nuclear accident, caesium has been investigated regularly as well. Fallout samples were collected monthly in the town of Zadar.

For the strontium determinations were used radiochemical methods and the radioactivity was measured with a low background Geiger Müller counter. Counting time was 80,000 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 (4,096-channel pulse height analyser and personal computer) was used to determine radiocaesium levels in the samples from their gamma-ray spectra. Samples were measured in Marineli beakers which were placed directly on the detector. Counting time depended on sample activity, but was never less than 60,000 seconds.

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

Quality assurance and intercalibration of radioactivity measurements were performed through participation in the IAEA and WHO quality control programmes.

Results and Discussion

Nuclear tests conducted in the atmosphere and release of radioactive material from nuclear facilities cause the radioactive contamination of human environment. The fallout resulting from atmospheric dispersion of both short and long-lived radionuclides not only directly affects humans, but also enters the food chain through the plants and animals and water supplies. Among anthropotropic radioactive nuclides ^{90}Sr and ^{137}Cs have been regarded as the fission products of a great potential hazard, due to relatively long half-life and similarity in matabolic processes to calcium and potassium respectively.

Average ^{90}Sr in activities in cistern waters for the 1968-1996 period is presented at fig. 1.

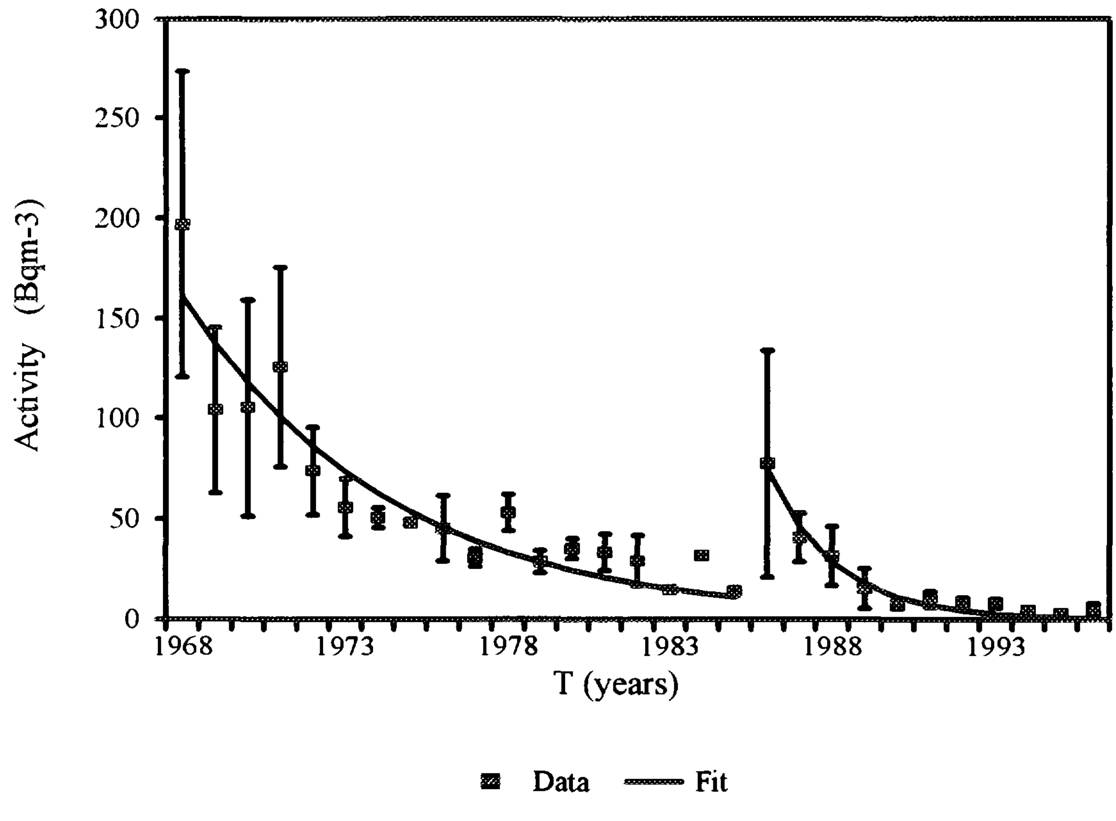

Figure 1
^{90}Sr activity in cistern waters in 1968-1996 period

An exponential decline of radioactivity followed the nuclear moratorium up to 1986 Chornobyl accident. Exponential decrease after the Chornobyl accident has a different slope. Generally, the Chornobyl reactor accident did not cause any significant increase in ^{90}Sr activity in most of the environmental samples in Croatia. Unlike the atmospheric testing of nuclear weapons, the radionuclides that originated from the Chornobyl 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 Chornobyl debris (e.g., ^{90}Sr) were deposited closer to the accident location than the more volatile constituents (i.e., radiocaesium) (3,4) Thus, ^{90}Sr was not subjected to the global dispersion processes, being 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 Croatia were rather dry, leading to relatively low direct radioactive contamination, which was especially true for the Adriatic region (5). However, due to large catchment areas that are used to collect rain water (mostly roofs, but also other specially built catchment areas) fallout radionuclides originated in Chornobyl were apparently "enriched" in cistern waters. Namely, dry fallout finally also entered the cistern reservoires. Therefore, the cistern waters were the only environmental samples in Croatia in which ^{90}Sr, attributed to the Chornobyl, persisted for several years after the accident. For comparison, in the city of Zadar, the total of 90.7 Bqm^{-2} of ^{90}Sr has been deposited in 1968. However, in 1986 (the year of the Chornobyl accident) has been deposited only 7.7 Bqm^{-2} compared to 6.6 and 6.2 Bqm^{-2} in 1985 and 1987 respectively. By function minimization, the data on ^{90}Sr activity in cistern water were fitted to the exponential function:

$$N(t) = N(0)e^{-kt} \qquad /1/$$

where:

N(t) is ^{90}Sr activity concentration in cistern water at time t,
N(0) is ^{90}Sr activity concentration in cistern water at zero time (years 1968 and 1986 respectively),
t is elapsed time and
k is constant.

The reciprocal value of the constant k is the mean residence time of ^{90}Sr in the cistern water, i.e., $T=1/k$. From equation /1/, T was found to be 6.3 and 2.1 years for 1968-1985 and 1986-1996 respective periods. However, the mean residence time for the individual cisterns is considerably different, depending on the rate of water use. The ^{90}Sr mean residence time in cistern waters for the post-Chornobyl period reflects the tropospheric mean residence time that is considerably shorter than the stratospheric mean residence time. Namely, the radioactive material from the damaged Chornobyl reactor, did not reach the stratosphere, which is especially true for the less volatile radionuclides.

Because of the fact that caesium is far more volatile than strontium, ^{137}Cs activities in cistern waters considerably varied from location to location. Therefore, correlation between ^{137}Cs activities in Zadar fallout and in ^{137}Cs activities in cistern waters was poor. Contrary, coefficient of correlation between fallout ^{90}Sr activities and ^{90}Sr in cistern waters has been fond to be fairly good, $r = 0.85$. In addition, average ^{137}Cs activity was an order of magnitude than ^{90}Sr activity.

The doses that would have received a hypothetical adult member of the public, consuming 1 liter of cistern water per day, are presented in Table 1. The dose-conversion used for calculation were 2.8×10^{-8} and 1.3×10^{-8} $SvBq^{-1}$ for ^{90}Sr and ^{137}Cs respectively. The doses being the order the magnitude of μSvy^{-1} pose no significant radiation risk.

Table 1
Estimated effective doses for an adult member of general public from consumption of cistern water

Year	Dose (μSvy^{-1})		
	^{90}Sr	^{137}Cs	Total
1968	2.01 ± 0.78	0.23 ± 0.05	2.24
1985	0.14 ± 0.02	not available	<0.14
1986	0.79 ± 0.58	7.73 ± 1.89	8.52
1996	0.04 ± 0.04	0.10 ± 0.07	0.14

Conclusions

Analysis of cistern water radioactivity is a valuable tool for assessment of environmental radioactive contamination, especially for the fission radionuclides. Estimated doses due to consumption of cistern water are small. However, the radioactivity should be regularly monitored.

In the era of Agenda 21, when we say we strive for sustainability and good husbandry with the earth's resources, a properly constructed and operated cistern can be a source of supplemental water to provide for water needs. This supplemental water supply can be very useful when sources of potable water are limited or expensive.

Literature

1. *Ross D.* "The Stele of Mesha" 13 February 1997.
   <http://www.owlnet.rice.edu/~malsoft/reli240b.html">
   (Site last visited on 10 September 1997.)
2. "The Roman Cistern"
   <http://www.agora.stm.it/J.Purtell/part51.htm>
   (Site last visited on 16 September 1997.)
3. *Aarkrog A.*, The radiological impact of the Chernobyl debris compared with that from nuclear weapons fallout. J. Environ. Radioactivity. 1998;**6**:151-162.
4. *Franic Z. and Bauman A.*, Activity of ^{90}Sr and ^{137} Cs in the Adriatic Sea, Health Phys. 1993;**64**:162-169.
5. *United Nations Environment Programme (UNEP), Mediterranean action plan*, Assessment of the state of pollution in the Mediterranean Sea by radioactive substances, UNEP, Athens, 1991.
6. *International Atomic Energy Agency (IAEA)*, International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, IAEA, Vienna 1996.