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Exposure to Natural Radioactivity from Thermal Waters in Croatia

G. Marović, J. Senčar

Institute for Medical Research and Occupational Health, Radiation Protection Unit, HR-10000 Zagreb, Ksaverska cesta 2, Croatia

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In this century, harmonization among radiation, human life and the ecosystem will be a very important issue. The growth of the natural radiation background on a global scale, exposure to doses above the natural background of all the biota on the Earth and involvement into the area of radiation exposure of growing number of people, all this made the problem of radiation safety greatly important.

Public perception of the needs for response and protection is influenced by many factors and elevated prolonged exposures due to natural radiation sources are usually ignored by society. Typically, the claim for protection is generally stronger when the source of exposure is a technological by-product or in exposure situations when a large degree of damage occurs infrequently rather than when it is considered to be of natural origin. Several types of exposure situations that have been identified so far, in which members of public may find themselves, were included into a carefully elaborated system of radiological protection and require implementation of appropriate protective actions. On the other hand, there are numerous situations of prolonged exposures, which are integrated into the human habitat and cannot be kept under control. Though excluded from the scope of regulations on radiological protection, these exposures to natural radioactivity, should not be ignored and objective assessment of possible health risk should be regularly monitored (ICRP 2000; UNSCEAR 2000; Gonzáles 2000).

Thermal waters are known as valuable natural resources of a country. They contain certain degree of natural radioactivity attributable to the elements of the uranium and thorium natural decay series. Among these elements, the most radiotoxic and most important is radium which exists in several isotopic forms (^{226}Ra and ^{228}Ra) (Iyengar 1990; Molinari and Snodgrass 1990).

Croatia, abundant in thermal water springs on its relatively small area, may serve as a good example of the natural radioactivity sources present in human habitat used without any restriction by the numerous population from all over the country and by majority of local inhabitants throughout their lifetime (Marović et al. 1996). As part of the national survey of natural radioactivity level in the environment the Radiation Protection Unit of the Institute for Medical Research and Occupational Health in

Correspondence to: G. Marović

Zagreb has been regularly monitoring the presence of natural radioactivity in Croatian thermal spring waters. The focus of attention was mainly the content of radium in the samples of thermal spring water from different locations in Croatia, the spas and health resorts. These waters are mainly used for bathing and recreational purposes, except for the waters from five locations used also for drinking.

Considering the great radiotoxicity of ^{226}Ra (a half-life of 1600 years) and ^{228}Ra (a half-life of 5.8 years), their presence in water require particular attention. When radium is taken into the body, its metabolic behaviour is similar to that of calcium and an appreciable fraction is deposited in bone, the remaining fraction being distributed almost uniformly in soft tissue (Wrenn et al. 1985; Iyengar 1990; Rowland 1993; Sidhu and Breithart 1998).

Our study deals with ^{226}Ra and ^{228}Ra concentrations in thermal spring water from the selected Croatian spas as a source of radiation doses that might be received by the patient or tourist visiting a spa. Our aim was also to calculate the radiation dose originating from drinking mineral spa water. On the basis of the obtained data the ^{226}Ra and ^{228}Ra activity ratio was calculated for each investigated spa.

MATERIAL AND METHODS

Samples of thermal spring water (50 L) were evaporated to 1 L, packed and left until they reach radioactive balance. The samples of tap water were also collected 1 L per day, united in the monthly sample and evaporated.

The presence of ^{226}Ra and ^{228}Ra in groundwater was investigated in samples of thermal spring waters taken directly from the springs or wells. The samples were collected at irregular intervals over the period of 10 years, variably for different locations. Determination of radioactive contamination required analyses of water samples from different areas, at 9 sampling sites, the spas and health resorts (waters from 5 locations are also used for drinking).

For the purpose of comparison, the content of radium was also calculated in the samples of municipal tap water in the city of Zagreb.

The collected water samples were gammaspectrometrically analysed in the laboratory using Ge(Li) Ortec detector (resolution 1.78 keV on 1.33 MeV 60Co, relative efficiency 16.8%) coupled to multichannel analyser system and a personal computer. All samples were measured in Marinelli beaker, volume 1L. Measurement time was 80.000 - 170.000 sec.

RESULTS AND DISCUSSION

The study of thermal spa waters in Croatia included only selected most popular spas clustered in north-west part of the country (Figure 1).

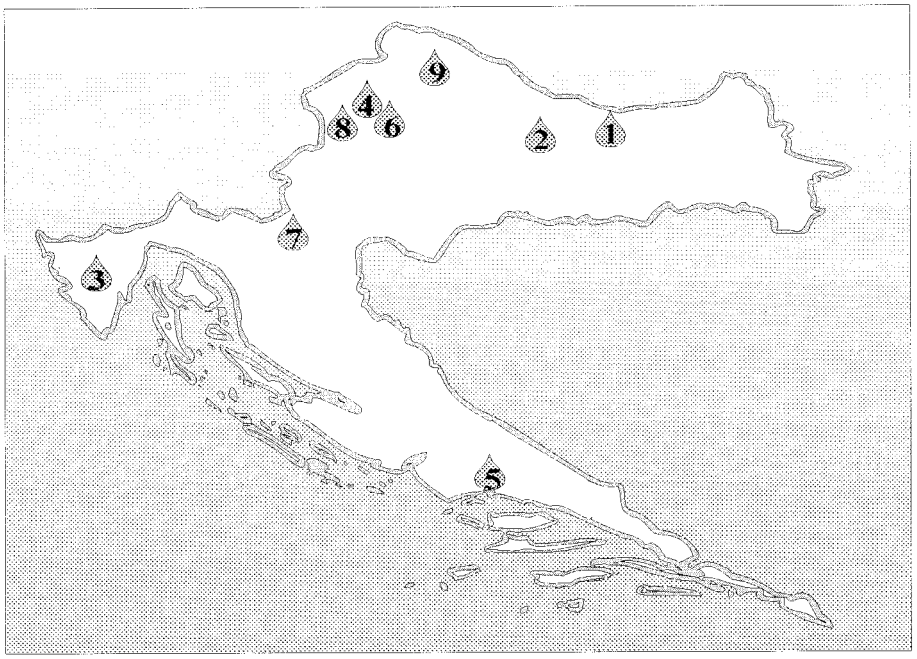

Figure 1. Locations of selected thermal springs in the Croatia

It provides summarized data on ^{226}Ra and ^{228}Ra content over a ten-year period, which reveal that individual springs differ by chemical composition, degree of mineralization, sulphur content, temperature, level of radioactivity and other characteristics.

Specific activities of ^{226}Ra and ^{228}Ra were calculated by separate locations (Figure 2). The data show that concentrations of ^{226}Ra varied between 80.0 and 4600.0 Bqm^{-3}, and of ^{228}Ra between 27.0 and 2890.0 Bqm^{-3}. Wide ranges of ^{226}Ra and ^{228}Ra activities suggest that a valid and reliable conclusion on the observed differences could be inferred by investigating them over a longer period of time and on a regular basis. The final column on the figure shows the content of these radionuclides found in municipal tap water of the city of Zagreb. This was done for the purpose of comparison because 1/4 of the total Croatian population lives there.

In most cases the concentration ratio of ^{226}Ra exceeded that of ^{228}Ra, ranging from 0.5 to 8.5, which may lead to a conclusion that no generic correlation could be obtained regarding the concentration of these two isotopes (Figure 3).

At location no. 3, ^{226}Ra specific activity was as many as 93 times higher than that of ^{228}Ra. According to some authors no correlation should be expected between the concentration of these two isotopes because ^{226}Ra and ^{228}Ra originate from two separate naturally occurring decay series (^{238}U and ^{232}Th). Besides, the geological and

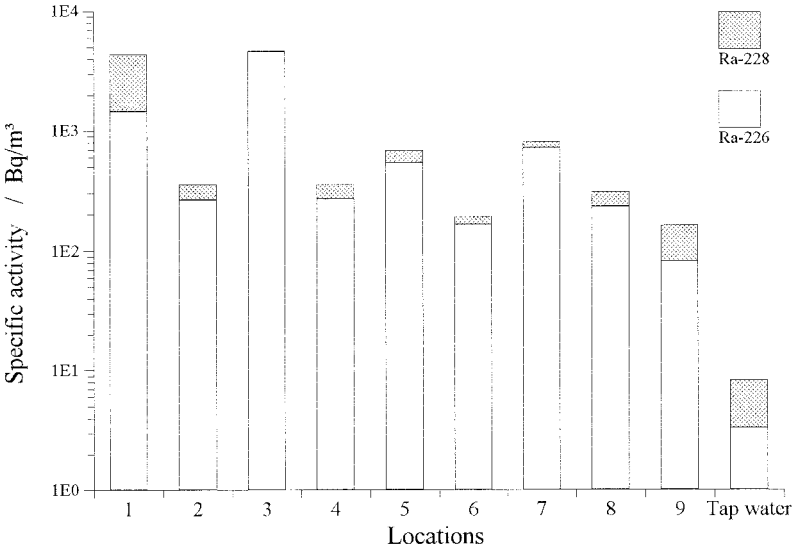

Figure 2. ^{226}Ra and ^{228}Ra specific activities in thermal springs of Croatia

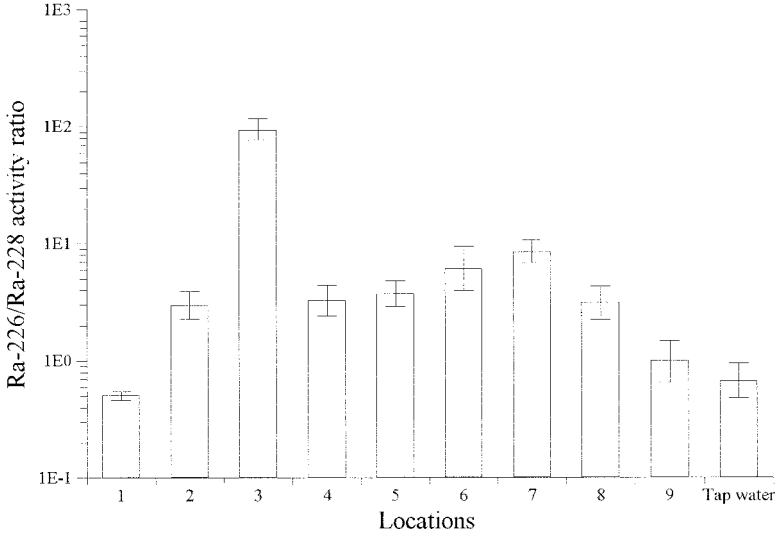

Figure 3. ^{226}Ra and ^{228}Ra activity ratio in thermal springs of Croatia

solubility properties of radium isotopes and their parent elements are completely different. Their actual levels in thermal waters are determined by the geological formation from which the spring flows, the geochemistry of the parent nuclides, and the interaction between the groundwater aquifers and radium-bearing materials. (Kriege and Hahne 1982; Molinari and Sondgrass 1990; Michel and Moore 1980)

All the investigated spas are popular tourist places and health resorts generally used for curative baths or recreational purposes. Located in rather densely populated area they are often crowded with visitors and local population especially during summer. Typical guests are patients who visit them, sometimes repeatedly, within their specific therapeutic or convalescent programme and usually spend there several weeks. During the past ten years the spas became increasingly important due to the fact that recent war in Croatia left numerous disabled and injured people for whom restoration of their health or improvement of their physical condition require the stay in a spa. In addition to the use of thermal waters for curative bathing, spring waters at certain locations (no. 4, 5, 6, 8 and 9) are recommended for drinking and make part of the medical treatment. Although the consumption of mineral water is considered beneficial, being rich in primordial radionuclides, it is also a source of natural radiation exposure.

Existing Croatian legislation deal with thermal spring water as a category of municipal potable water and not a distinct category of water, even though the analyses indicated that content of radium exceeds the values determined in other categories of water in Croatia. Following current national legislation on drinking water, natural mineral water should be controlled twice a year.

When waters with radium content are consumed, incorporation of radium involves a long-term hazard. Epidemiological studies conducted in the world so far gave sufficient evidence of association between exposure to radium and adverse health effects (Sidhu and Bretihart 1998).

The activity of radionuclides in potable water delivered to a human depends not only on intake but also on metabolic and dosimetric considerations. The guideline activity concentrations assume an intake of total radioactive material from the consumption of 2 litres of water per day for 1 year. For radionuclides ^{226}Ra and ^{228}Ra permissible activity concentrations in municipal drinking water, as recommended by the World Health Organization, is 1 BqL^{-1} (WHO 1993). When estimating the dose related to the radium content of the consumed spring waters, it has to be understood that these waters do not constitute a part of continuous drinking water supply, but are applied as medicinal waters for well defined periods and in given amounts. Therapeutic application usually takes several weeks and includes daily doses of 0.2-2.0 litres. The estimated dose of incorporated radium in a drinking cure was calculated as based on the recommendations of ICRP on condition that during a 4-week drinking cure the patient consumes 1 litre/day (ICRP 1979). In our study in consideration were taken only the samples of thermal spring water from the spas where therapy involved drinking cure lasting 4 weeks. Figure 4 shows the estimated effective dose received

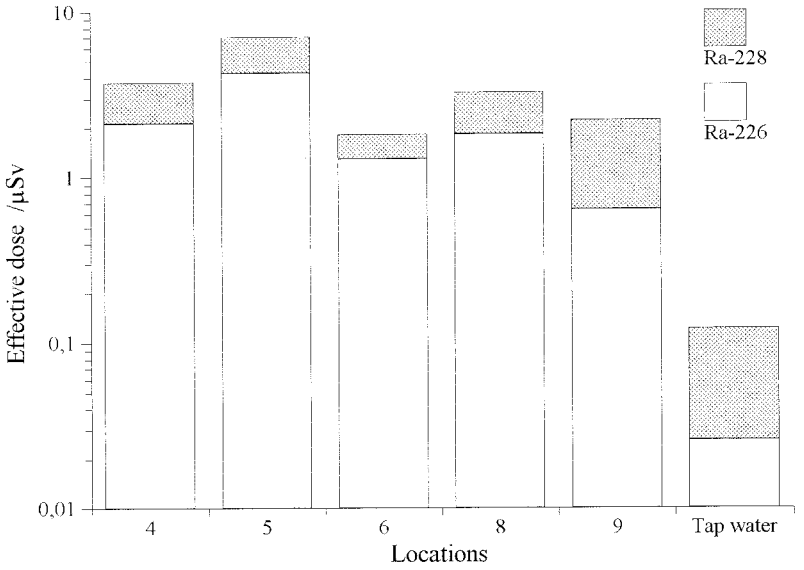

Figure 4. Effective dose received by individuals during a 4-week thermal-water drinking cure

by individuals undergoing thermal-water drinking cure over the period of 4 weeks at investigated spas.

The estimated received dose is compared to that received by consuming tap water only, over the same period. The equivalent dose received from a drinking cure over a 4-week period was estimated to be as much as 5 times higher than the dose received from drinking tap water over a period of 1 year (1.6 ± 0.3 μSv per year).

According to ICRP recommendations (ICRP Publication 60 1991) mineral water represents a considers commodity for public consumption containing radioactive substances. Today, the use of bottled water is on the increase as a healthier alternative to tap water (Gans 1985; Pip 2000) because it is believed it contains fewer contaminants. Radioactivity is measured only rarely in bottled water. The limit for public exposure should be expressed as an effective dose of 1 mSv in a year. The doses obtained by our study are significantly below that recommended dose for all categories of water. Therefore it might be concluded that thermal spring waters can be used without any restrictions (for bathing, drinking for medical therapy, recreation and rehabilitation), on the assumption that the 5-year average does not exceed 1 mSv per year. Taking into account disparity of the population groups that use these waters, ranging from resident population (supposedly a lifelong use) to the occasionally visiting tourists, the actual health risk might be most reliably determined on basis of epidemiological studies which remain to be conducted in future.

REFERENCES

Gans I (1985) Natural radionuclides in mineral waters. Sci Total Environ 45:93-99

Gonzáles AJ (2000) International harmonization of radiation protection and safety standards: The rule of the United Nations system. In: Harmonization of radiation, human life and the ecosystem. Proceedings 10th International Congress of The International Radiation Protection Association. Hiroshima

ICRP International Commission of Radiological Protection (1979) Limits for intakes of radionuclides by workers. ICRP Publication 30 (Supplement to Part 1). Pergamon Press, Oxford.

ICRP International Commission of Radiological Protection (1991) 1990 Recommendations of the International Commission on Radiological Protection. ICRP Publication 60. Pergamon Press, Oxford

ICRP International Commission of Radiological Protection (2000) Protection of the public in situations of prolonged radiation exposure. ICRP Publication 82 Pergamon Press, Oxford

Iyengar MAR (1990) The natural distribution of radium. In: The environmental behaviour of radium. IAEA Technical Reports Series No. 310.Vol.1. IAEA 1:59-128

Kriege LB, Hahne RMA (1982) ^{226}Ra and ^{228}Ra in Iowa drinking water. Health Phys 43:543-559

Marović G, Senčar J, Franić Z, Lokobauer N (1996) Radium-226 in thermal and mineral springs of Croatia and associated health risk. J Environ Radioactivity 33:309-317

Michel J, Moore WS (1980) ^{228}Ra and ^{226}Ra Content of groundwater in Fall Line aquifers. Health Phys 38:663-67

Molinari J, Snodgrass WJ (1990) The chemistry and radiochemistry of radium and the other elements of the uranium and thorium natural decay series. In: The environmental behaviour of radium. IAEA Technical Reports Series No. 310.Vol.1. IAEA 1:1-56

Pip E. (2000) Survey of bottled drinking water available in Monitoba, Canada. Environ Health Perspect 108: 863-866

Rowland RE (1993) Low-level radium retention by the human body: a modification of the ICRP Publication 20 retention equation. Health Phys 65:507-513

Sidhu KS, Breithart MS (1998) Naturally occurring radium-226 and radium-228 in water supplies of Michigen. Bull Environ Contam Toxicol 61:722-729

UNSCEAR United Nations Scientific Committee on the Effects of Atomic Radiation (2000). Sources and effects of ionizing radiation. United Nations New York

WHO World Health Organization (1993). Guidelines for drinking-water quality. Recommendations. WHO Geneva

Wrenn ME, Durbin PW, Howard B, Lipsztein J, Rundo J, Still ET, Willis DI (1985) Metabolism of ingested U and Ra. Health Phys 48:601-633