Natural Radioactivity of Thermal Springs in Croatia

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INTRODUCTION

The Republic of Croatia is one of the richest countries in thermal and mineral springs in the world and abounds in spas and health resorts. Besides the curative powers of thermal springs, these waters are known to contain certain degree of natural radioactivity attributable to the elements of the uranium and thorium natural decay series. The most radiotoxic and most important among these elements is radium, which is a known carcinogen and exists in several isotopic forms. The predominant radium isotopes in groundwater are $^{226}$Ra, an alpha emitter with a half-life of 1600 years, and $^{228}$Ra, a beta emitter with a half-life of 5.8 years (1,2,3).

Considering the great radiotoxicity of $^{226}$Ra and $^{228}$Ra, their presence in water and associated health risk require particular attention. It is known that even small amounts of radioactive substances may produce a damaging biological effect and that ingested and inhaled radiation can be a serious health risk (4). When radium is taken into the body, its metabolic behavior is similar to that of calcium and an appreciable fraction is deposited in bone, the remaining fraction being distributed almost uniformly in soft tissues (5).

Important aspect of radium protection is the prevention of its entry into human body, the critical pathway being ingestion through food chain or drinking water (6). Although environmental aspects of radioactivity and its possible effects on population have been a subject of interest for over several decades in Croatia, no reports have so far attempted to estimate thermal spring waters as possible routes of exposure to radioactivity.

The spas are widely popular for medical therapy, tourism, recreation, rehabilitation and mineral water drinking cures. Considering this popularity and habit of Croatian population to use beneficial effects of thermal spring waters it is of interest to determine the degree of radioactivity that may be absorbed by bathing and drinking practices. With this in view, the Radiation Protection Unit of the Institute for Medical Research and Occupational Health in Zagreb has conducted a research programme in order to estimate dose contribution from thermal spring waters to total exposure from the natural radiation.

The aim of this paper was to determine $^{226}$Ra and $^{228}$Ra concentrations in thermal spring waters from the selected spas in order, to estimate the radiation doses received by patients or tourists during their stay in the spa, and to calculate the radiation dose originating from drinking spring waters with higher natural radionuclide content. On the basis of the obtained data the $^{226}$Ra and $^{228}$Ra activity ratio was calculated for each investigated spa.

MATERIAL AND METHODS

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 from 3 to 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).

The collected water samples were gammaspectrometrically analysed in the laboratory using Ge(Li) Ortec detector (resolution 1.78 keV on 1.33 MeV $^{60}$Co, 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 sec.

RESULTS AND DISCUSSION

Specific activities of $^{226}$Ra and $^{228}$Ra by separate locations are shown in Figures 1 and 2, respectively. As can be seen, $^{228}$Ra concentrations varied between 80.4 and 4550.0 Bqm$^{-3}$, and $^{228}$Ra concentrations varied between 27.0 and 2890.0 Bqm$^{-3}$. These values show wide ranges of $^{226}$Ra and $^{228}$Ra activities in investigated thermal springs, which suggests 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 more regular basis.
Regarding the concentration of these two isotopes no generic correlation was found. In our study, in the majority of cases the concentration ratio of $^{226}\text{Ra}$ exceeded that of $^{228}\text{Ra}$, ranging from 0.5 to 8.5. At location no. 3 $^{226}\text{Ra}$ specific activity was as many as 93 times higher than that of $^{228}\text{Ra}$. Some authors maintain that there should be no reason to expect a correlation between the concentration of these two isotopes because $^{226}\text{Ra}$ and $^{228}\text{Ra}$ originate from two separate naturally occurring decay series ($^{238}\text{U}$ and $^{232}\text{Th}$). Besides, the geological and solubility properties of radium isotopes and their parent elements are completely different. Their actual occurrence in thermal
waters is thus determined by several factors such as 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 – rocks, soil and ore deposits, etc. (7,8,9)

All Croatian spas under study have been used for curative baths or recreational purposes, but spring waters at locations 4, 5, 6, 8 and 9, recommended for drinking, are usually included in medical cure.

In Croatia radium environmental pollution control is related to the regulations associated with the consumption of drinking water. Generally, measures should be taken if the activity of radioactive contaminants detected in water exceeds the permissible levels as set up by a country's legislation. The current guideline activity concentration for radium, as recommended by the World Health Organization, is 1 BqL⁻¹ (10). In Croatia, health-based acceptable level of exposure from drinking water is set to 1000 Bqm⁻³ as the maximum permissible level for radium in drinking water. Guideline activity refers exclusively to municipal drinking water supplies and there is no regulation that would refer to the occurrence of radioactivity in other water categories, such as thermal and mineral waters, and to possible contribution by routes of exposure other than ingestion. This implies that natural mineral water has not been included so far in the existing Croatian legislation on drinking water. Consequently, thermal spring water has not been taken into consideration as a distinct category of water even though the analyses indicated that the content of radium in thermal water is higher than in other categories of water in Croatia.

When waters with radium content are consumed, incorporation of radium involves a long-term hazard. 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 daily doses of 0.2–2.0 litres. The estimated dose of incorporated Ra 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 (11). 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 3 shows the estimated effective dose received by individuals undergoing thermal-water drinking cure over the period of 4 weeks at investigated spas.

According to ICRP recommendations (12) 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. This supports the conclusion that in Croatia 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 and that other sources of exposure are also taken into account.
REFERENCES