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²¹⁰Po Activity Concentration of Blood Samples after the Radon Inhalatoric Therapy

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One of the most effective treatments of asthma is the cave therapy. The hospital cave in Tapolca (Hungary) has been used for treatments for the past decades. In summer, the ²²²Rn concentration may even reach 20 kBq/m³ (17 times higher than winter); the role of radon in the treatment of asthma is still not clarified. Daughter elements of ²²²Rn (²¹⁰Pb; ²¹⁰Po) with longer half-lives may accumulate in the blood, increasing the radiation dose of the human body. The changes of the ²¹⁰Po concentration in blood samples taken from the patients were examined in winter and in summer. The samples were dissolved with HNO₃ and HCl, and the sources were prepared by spontaneous deposition on stainless steel. The ²¹⁰Po concentration was measured using a semi-conductor detector α -spectrometer. To increase measurement accuracy, ²⁰⁹Po tracer was added to the samples. In 63 samples the measured ²¹⁰Po activity-concentrations were between 0.035 and 0.800 mBq/g. After the therapy the ²¹⁰Po concentration in the blood of the patients was increased in all cases. Based on this result, it was possible to calculate the absorbed dose of the patient during the treatment, which ranged from 4.24 10⁻¹⁶ to 5.85 10⁻¹³ J/kg, the mean value being 1.04 10⁻¹³ J/kg.

Key words: ²¹⁰Po, cave therapy, dose assessment, blood

1. Introduction

²²²Rn is the gaseous radioactive product of the decay of the ²²⁶Ra. This radioactive noble gas (²²²Rn), together with its short-lived decay products, is responsible for more than half of the natural dose in the population¹⁾. In the 1970s, it

was realized that high radon concentrations may be found in caves. Environmental studies have revealed that a strong spatial variability of radon concentrations in a cave is often found²⁾ and the values measured in individual caves can differ significantly from each other³⁻⁶⁾. In the majority of caves studied, considerable seasonal variations, variations in different weather conditions, and diurnal variations were observed⁶⁻¹¹⁾.

Atmospheric pressure changes and the temperature differences between the atmosphere and the cave interior can result in spatial and seasonal variability of the radon concentration^{7, 12-14}.

High radon concentration causes a remarkable dose.

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Fig. 1. The hospital cave of Tapolca is located in the Balaton Highlands in Hungary

Most of the radiation dose is received in the tracheobronchial region of the lungs by inhaled ²²²Rn daughters and, in particular, by the short-lived α emitters ²¹⁸Po and ²¹⁴Po. The risk of exposure with high radiation dose exists in most caves^{15, 16}. On the other hand, caves are often used for curing bronchial and asthmatic diseases of children and adults in several places^{17.19}. Some ascribe the positive effect – for example, stimulation of the endocrine system and DNA repair²⁰– of cave treatment partly to radon¹⁹, however, the role of radon in the treatment of asthma is still not clarified ¹⁷.

The first cave in Tapolca was discovered in 1903. The lamellate limestone in which the tunnels are located was created by the Miocene Sea 13.7 million years ago. During the decades, the cave system functioned as a hospital and bomb shelter.

Therapeutic examinations commenced in the cave in the beginning of the 1960s, with the experimental treatments were starting in 1972. The cave was transformed as a hospital in 1982.

During the treatments in therapeutic caves with high radon concentrations, the level of the whole-body dose of the patient is an important question.

The cave therapy in the cave examined has been of great importance for decades¹³⁾ (Hospital Cave in Tapolca). The change of radon concentration in the cave shows a seasonal recurrence²¹⁾: the value of radon concentration in winter is approximately 1000 Bq/m³, whereas the average level of radon in the summer exceeds the value 10000 Bq/m³ and it often reaches 20000 Bq/m³.

The mean yearly temperature in the cave is 13.9° C (13.2-14.8) and the pressure is 1004 kPa (997-1014), which is considered to be very stable. The seven years' average of the radon concentration is 4.88 ± 0.14 kBq/m³.

Figure 2 shows the change of the monthly mean radon concentration (min-max. value) over seven years.

The main aim of this study is to determine the changes in the 210 Po concentration of the blood samples.

A treatment generally lasts for 1-3 weeks, and the patients spend 3 hours per day in the cave. During this study, the ²¹⁰Po concentration of the blood samples taken from the patients before and after the treatment was determined with the help of a semi-conductor (PIPS) detector α -spectrometer.



Fig. 2. Monthly average radon concentrations in a hospital cave (7 years' average)

²¹⁰Po is a naturally occurring α-emitter with a half-life of 138.3767 ± 0.002 days²²⁾ as a pure α-emitting radionuclide with a high specific activity of ~1.7 10¹⁴ Bq/g. The toxicity of Po is 5 times greater than that of Ra²³⁾. Therefore, Po is considered to be one of the most toxic naturally occurring radionuclides and one of the most important environmental radionuclides due to its wide distribution and potential for human radiation exposure through ingestion and inhalation²⁴⁾. ²¹⁰Po contributes a substantial part of the radiation dose to humans. Bulman et al.²⁵⁾ estimated that the dose due to ingestion of ²¹⁰Po was about 7% of the natural internal radiation dose. The main source of Po in the environment is Rn gas, which diffuses into the atmosphere from rocks and soil where it ultimately decays to Pb, Bi, and then to Po in the atmosphere. ²¹⁰Po attaches itself further electrostatically to aerosol particles²⁶⁾.

2. Materials and methods

2.1. Sampling

Blood samples were collected in the winter months (December-February) and in the summer months (July-August), as the greatest changes of radon levels were measured in these periods. A total of 63 (27 winter and 36 summer) blood samples were obtained from the cave hospital in Tapolca, including the blood samples of patients and those working in the cave. Blood was taken from the patients before and after the treatment (10 ml per sample) and only once from those working there. The samples were processed within 1-2 weeks.

2.2. Determination of 210 Po concentration 2.2.1. Sample preparation

During the sample preparation, 0.5 ml 20 mBq/ml ²⁰⁹Po tracer and 20-30 ml cc. HNO₃ were added to approximately 10 g blood sample, then the sample was evaporated to 5 ml by continuous and intensive stirring; this step (cc. HNO₃ adding) was repeated two or three times. After 3×20 -30 ml cc. HCl was added and the solution was evaporated to 5 ml by continuous and intensive stirring again. In order to fully digest the possible existing organic residues, a few drops of H₂O₂ were added to the solution carefully. Following the last

evaporation with cc. HCl, the sample was also evaporated to 5 ml with 3×20 -30 ml Millipore Q ($18M\Omega$) water to reduce its hydrochloric acid content. After the last evaporation, the final step was to prepare 100 ml stock solution with 0.5 mol/l of HCl.

2.2.2. Source preparation

Measurements of ²¹⁰Po were realized through its 5.30 MeV α -particle emission by semiconductor α -spectrometry after spontaneous deposition with ²⁰⁹Po yield tracer onto a stainless steel disc (0.5 mm thickness, Ø18 mm, polished KO-33 MSZ 9-11% Ni-Cr disc); the method is well described²⁷⁾. The sources were made from 50 ml stock solution. 50-100 mg ascorbic acid was added in order to reduce the concentration of Fe³⁺ ions present in the sample. Finally, the solution was continuously stirred in the deposition cell (see Fig. 3) at 80°C for 3 h. After the deposition, the disc was rinsed with Millipore Q water and it was left to dry.

2.2.3. α -spectrometric measurement

Measurement was carried out in a vacuum (0.2 torr) with a semi-conductor PIPS detector (Eurisys 19 keV resolution)



α-spectrometer (Canberra 7401 α chamber, Silena 9302 multichannel analyser). The spectrum analysis was carried out by EMCA 2000 software. The calibrations were made by certified ²³⁹Pu, ²⁴¹Am, and ²⁴⁴Cm sources. The minimal detectable activity (MDA) (95% confidence level) was 0.034 mBq/g with a counting time of 240000 sec. The chemical yield of ²⁰⁹Po ranged from 79.24% to 97.11%, with a mean value of 90.20%.

3. Results and discussion

3.1. Samples taken in winter

The ²¹⁰Po concentration values of blood samples taken during the winter (low) radon concentration treatments are shown on Figure 4. The ²¹⁰Po concentrations of the blood samples were between 0.035 and 0.379 mBq/g. It can be clearly seen on the figure that the amount of radon daughter elements (²¹⁰Po) can get into the human body during the 3-hour-long daily treatment and may cause a significant increase even with a lower (1000 Bq/m³) radon concentration. The average increase of ²¹⁰Po concentration of the blood sample pairs was 0.069 mBq/g.

3.2. Samples taken in summer

For the blood samples taken during the summer months (with high radon concentration), ²¹⁰Po values were measured from 0.035 to 0.800 mBq/g (Fig. 5). The average changes after the treatment was 0.162 mBq/g.

3.3. Comparison of the results in summer and winter months

²¹⁰Po concentration values measured at the beginning of treatments during the summer were not higher compared to the winter data (average in winter: 0.068 mBq/g, altered between 0.035 and 0.212 mBq/g; average in summer: 0.089 mBq/g, altered between 0.035 and 0.270 mBq/g). However, the average ²¹⁰Po concentration after the treatment in winter



Fig. 4. Winter ²¹⁰Po concentration in the blood samples.





was 0.139 mBq/g (0.052-0.379 mBq/g), whereas in summer the average was 1.8 times higher at 0.251 mBq/g (0.040-0.800 mBq/g). The average radon concentration in the cave was 10 times higher in summer than in winter.

Figure 6 shows that the degree of the increase in Po concentration was between the values 1.25 and 2.5 in most cases, both in the winter and in the summer. Although the highest increase is 6.4 times during the winter, values increased over more than 10 times were detected by even 3 cases during the summer.

4. Dose calculation

The effective dose of ²¹⁰Po could not be calculated as there

were no available data concerning other tissues of the body. This way, the absorbed dose of the blood tissue could be calculated using the available data.

During the calculation, we took into consideration that the α -particle generated during the decay of the ²¹⁰Po isotope gives all of its energy to the blood tissue (based on LET calculation²⁸⁾).

$$D=E(\alpha) \cdot K \cdot C$$

where

D: absorbed dose (J/kg) E: α particle energy (5.3·10⁶ eV) K: coefficient (1.6·10⁻¹⁹ J) C: ²¹⁰Po concentration (Bq/kg)



The absorbed dose of the blood tissue is in relationship with the increase of 210 Po concentration.

The calculated values altered between $1.61 \cdot 10^{15}$ and $2.28 \cdot 10^{13}$ J/kg in the case of the winter samples (average: $5.88 \cdot 10^{14}$ J/kg), and altered between $4.24 \cdot 10^{16}$ and $5.85 \cdot 10^{13}$ J/kg in the case of the summer samples (average: $1.38 \cdot 10^{13}$ J/kg) (Fig. 7).

5. Conclusion

As a part of an experiment series, the ²¹⁰Po concentration of the blood tissue was studied during therapeutic treatments carried out in the cave hospital in Tapolca.

During the measurements, the increase of ²¹⁰Po concentration in the blood samples of 63 patients was observed. During the 3-hour-long daily cave therapeutic treatments, a significant ²¹⁰Po increase could be detected even by relatively small radon concentrations; the average ²¹⁰Po concentration was between 0.035 and 0.379 mBq/g in the winter months and between 0.035 and 0.800 mBq/g in the summer months. Expectedly, the average ²¹⁰Po increase in the summer months. It may have several causes as the dependencies of the physical properties (weight, height), gender and age of the patients were not reviewed in this paper.

The absorbed dose of the blood tissue was calculated from the increase of the ²¹⁰Po concentration, which was minimal even in the summer months: when the average value was $1.38 \cdot 10^{-13}$ J/kg (altered between $4.24 \cdot 10^{-16}$ and $5.85 \cdot 10^{-13}$ J/kg).

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126