# COMPARATIVE ANALYSIS OF GAMMA DOSE RATES MEASURED BY ION CHAMBER IN AND AROUND THE HISTORICAL SACRAL OBJECTS IN BOSNIA AND HERZEGOVINA

by

***Senada AVDIĆ*** [1*], ***Adnan BEGANOVIĆ*** [2], ***Armin LAGUMDŽIJA*** [3], ***Samra SADIKOVIĆ*** [4], ***Beco PEHLIVANOVIĆ*** [5], ***Damir DEMIROVIĆ*** [6], ***and Irma KADIĆ*** [7]

[1] Department of Physics, Faculty of Science, University of Tuzla, Tuzla, Bosnia and Herzegovina
[2] University Clinical Center Sarajevo, Sarajevo, Bosnia and Herzegovina
[3] State Regulatory Agency for Radiation and Nuclear Safety and Security of Bosnia and Herzegovina, Sarajevo, Bosnia and Herzegovina
[4] Department of Mathematics, Faculty of Science, University of Tuzla, Tuzla, Bosnia and Herzegovina
[5] Faculty of Educational Science, University of Bihac, Bihac, Bosnia and Herzegovina
[6] Faculty of Electrical Engineering, University of Tuzla, Tuzla, Bosnia and Herzegovina
[7] Institute for Public Health Federation of Bosnia and Herzegovina, Sarajevo, Bosnia and Herzegovina

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**The main objective of this study was to investigate the correlation between the indoor and outdoor ambient dose equivalent rates measured by the ion chamber inside and around the historical sacral objects at a few locations in Bosnia and Herzegovina. The investigated objects made of the traditional building materials were built in the Late Medieval, Post Medieval, and Ottoman Period of Bosnia and Herzegovina history. The LUDLUM Model 9DP instrument based on a pressurized ion chamber was selected for natural low level radiation measurements since the ionisation chambers have higher sensitivities than the other types of detectors. The detection capability of the LUDLUM Model 9DP pressurized ion chamber was examined in the laboratory conditions with a source of low activity and under natural environmental radiation conditions by measuring the indoor and outdoor dose rates. A weak positive correlation was found between the ambient dose equivalent rates inside the historical sacral objects and the dose rates outside the objects. The average evaluated value of the indoor to outdoor dose rate ratio of 1.07 for the studied historic objects is less than that obtained for the contemporary building materials such as concrete. No study on the indoor to outdoor dose rate ratio in Bosnia and Herzegovina measured by the LUDLUM 9DP dose rate meter based on an ion chamber has been conducted yet. In addition to direct measurements, the first gamma spectrometric analysis of a few samples of building materials from the Late Medieval period in Bosna and Herzegovina was performed. The results of the gamma analysis revealed almost uniform distribution of primordial radionuclides in the investigated samples. It was demonstrated that such materials had the reduced content of radioactive isotopes compared to the contemporary building materials and therefore they could have potential advantages in specific applications related to the environmentally sustainable architecture.**

*Key words: ambient dose equivalent rate, indoor/outdoor dose rate, pressurized ion chamber, gamma spectrometry*

## INTRODUCTION

Natural environmental radiation is subjected to temporal and spatial variations to a great extent and therefore there is a clear need in Bosnia and Herzegovina (B&H) for gamma radiation survey data acquired in a shorter time interval at various locations in addition to a few gamma radiation monitoring stations working in continuous mode at fixed locations. The LUDLUM Model 9DP pressurized ion chamber [1] as a radiation dosimeter of the compact size and higher sensitivity than the other types of radiation detectors was available for survey of indoor and outdoor ambient dose equivalent rates. For the purposes of direct measurements and routine radiation protection the International Commission on Radiation Units and Measurement (ICRU) has developed an *operational quantity* ambient dose equivalent H*(10) which can be defined as a measurable estimate of the effective dose for a potentially irradiated individual at the point of the

* Corresponding author; e-mail: aadeno@bih.net.ba

monitoring instrument taking into account a phantom approximation of the human body [2].

The dose rate measurements were carried out in one category of the historic sacral objects from the Late Medieval, Post Medieval, and Ottoman Period of B&H history. The historic objects were selected for investigation considering their similar building structure such as the number of bulkheads, ceilings and walls that were made mostly of the local traditional materials of simpler composition compared to the contemporary building materials.

The main goals of our study were to investigate the correlation between gamma dose rates inside and outside the studied objects measured with the LUDLUM 9DP dosimeter as well as to determine the average indoor to outdoor dose rate ratio for the historic objects.

Gamma spectrometric analysis of a few building material samples from the Late Medieval period of B&H history collected in accordance with the relevant institutions for protection of cultural monuments was carried out in addition to the direct measurements of ambient dose equivalent rates. The main purpose of the gamma analysis with an HPGe spectrometer was to evaluate gamma absorbed dose rates in air due to samples of the old building materials and to compare the obtained results with the ones associated with modern building materials.

Performances of the LUDLUM 9DP instrument were examined in the laboratory conditions with a radionuclide of low activity and in field conditions in order to gain more useful information that could be helpful for a clearer interpretation of the measured data. The results obtained during this study could be of great interest in further work on outdoor and indoor radiation level baseline information in B&H.

## MATERIALS AND METHODS

### Measurement equipment and locations

The direct measurements of ambient dose equivalent rates at a few locations in B&H were carried out by using the LUDLUM model 9DP pressurized ion chamber designed to measure gamma and X-ray energies above 25 keV. The instrument measures the ambient dose equivalent rate – H*(10) in the range from 0 to 50 $mSvh^{-1}$. Energy response of Model 9DP is flat within 25 % from 60 keV to 1.25 MeV. The volume of the chamber is $230 \cdot 10^{-6}$ $m^3$ with 99.89 % of nitrogen and 0.11 % of argon under pressure of $8.1 \cdot 10^5$ Pa

The gamma spectrometric analysis of a few historic building material samples was carried out to evaluate the gamma absorbed dose rate in air and to compare the calculated values with the dose rates from the modern materials. The approvals for the gamma spectrometric analysis of the Late Medieval building material samples were obtained from the authorities of the relevant institutions such as the National Museum of B&H, the Museum of Eastern Bosnia and the Institute for the Protection of Cultural Monuments of the Federation of Bosnia and Herzegovina (FB&H). The samples were extracted from the investigated objects during the previous restorations at four various historic sites (St Nicholas monastery, Bobovac fortress, Pocitelj fortress, and Soko fortress built in the 16$^{th}$ century, 14$^{th}$ century, 15$^{th}$ century, and 15$^{th}$ century, respectively). The gamma spectrometric analysis of the samples was performed at the Radiation Protection Center – the Institute for Public Health of FB&H in Sarajevo. The presence of primordial radionuclides such as $^{226}$Ra, $^{232}$Th, and $^{40}$K in the investigated samples was recorded by using an HPGe gamma spectrometer of the *n*-type with a 30 % relative efficiency at 1.33 MeV and an energy resolution of 1.9 keV at 1.33 MeV (ORTEC, USA).

Indoor and outdoor ambient dose equivalent rates were directly measured at a height of 1 m in the air above the ground surface at the following locations: Tuzla (44°32'19.15"N, 18°40'21.20"E) with 6 historic objects, Sarajevo (43°51'22.531"N, 18° 24'47.075"E) with 2 historic objects, Bihać (44°48'43.22"N, 15°52'02.67"E) with 1 historic object, Petrovo (44°35'48.92"N, 18°19'51.56"E) with 1 historic object, Jajce (44°20'31.31"N, 17°16'14.12"E) with 1 historic object, while the samples of the Late Medieval building materials were collected at the following sites: Počitelj (43°07'60.00"N, 17°43'59. 99"E), Gračanica (44°42'11.05"N 18°18'36.36"E), Bobovac fortress located near Vareš (44°09'51.98"N 18°19'41.99"E) and Petrovo. The map of B&H with the 11 measurement locations related to the selected historic objects and 4 sites of origin of the Late Medieval building material samples is given in fig. 1.

The direct measurements with the LUDLUM 9DP instrument include at least three measurement points inside the historic objects greater size whereas for the objects of smaller size one measurement point in the center of the building was considered as representative enough. The outdoor measurements were performed at least at two various measurement points a few meters away from each object. Measurements were repeated more times at each measurements point according to the evaluation of the number of repeated readings described below.

### Distribution of experimental data

The stochastic nature of radiation and the detection process itself contribute to the uncertainty in nuclear measurements. In order to estimate inherent statistical fluctuations of the instrument used in this investigation 50 measurements of the ambient dose equivalent rate were performed inside the Atik


**Figure 1. The map of B&H with the measurement locations and sites of origin of the Late Medieval building material samples**

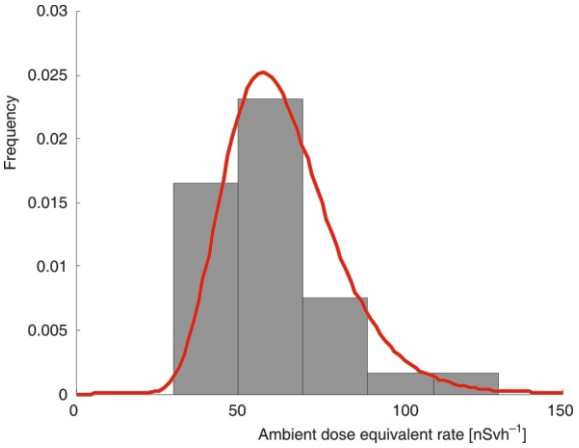


**Figure 2 Histogram and fitted log-normal distributions for indoor ambient dose equivalent rates measured by using the LUDLUM Model 9DP instrument**

mosque in Tuzla. It was demonstrated that the measured results in the range of 30-130 nSv$^{-1}$ follow log-normal distribution (given in fig. 2) with the geometric mean value of 61.9 nSvh$^{-1}$ and geometric standard deviation (GSD) of 1.3 nSv$^{-1}$.

## Coefficient of variation and the number of repeated readings

In order to investigate the response and sensitivity of the LUDLUM 9DP dosimeter used in this study the measurements of ambient dose equivalent rates were conducted at different distances (5 cm to 25 cm with a step of 5 cm) from a $^{137}$Cs radioactive source with a low activity of 0.12 MBq at the time of measurements in the Nuclear Physics Laboratory at the University of Tuzla. The coefficient of variation (CV) as a ratio of the standard deviation to the mean value was calculated for the experimental results obtained in the laboratory conditions. The CV represents a measure of relative variability of data associated with the precision of a measurement technique [3]. As can be seen in fig. 3(a) that the LUDLUM Model 9DP dose rate meter based on an ion chamber is more reliable for monitoring higher dose rates compared to environmental dose rate levels. It was found that the contribution from the $^{137}$Cs gamma source with activity of 0.12 MBq at the distance of 30 cm is not significantly different from the background radiation.

It is well-known that it is desirable to repeat measurements more times in a longer sampling interval in order to obtain more reliable results. However, since no unified protocol for all dose rate meters has been developed yet there is a need to examine the device used in our study individually. A procedure for selection of the number of manual readings required to obtain a representative mean value of the measured quantity was suggested recently [4]. The LUDLUM 9DP indoor ambient dose equivalent rate data set of 50 data points was analyzed with the goal of estimation of the number of repeated readings taking into account the relative margin of 5 % and the confidence level of 95 % regarding the two-tailed inverse of the Student's *t*-distribution [4]. Figure 3(b) shows dependence of the relative margin on the number of repeated readings and it is obvious that about three readings are enough to achieve the relative margin of less than 5 % for the

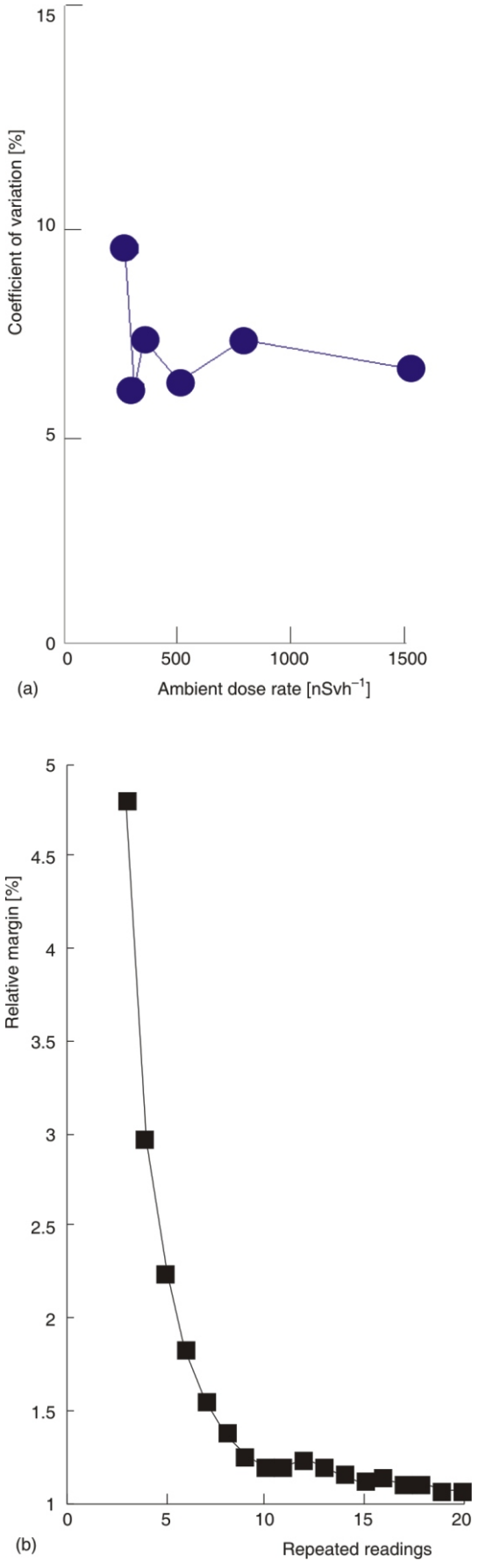


**Figure 3. The CV as a measure of relative variability of ambient dose rates (a) and the relative margin as a function of the number of repeated readings (b)**

dose rate measurements using the LUDLUM 9DP dose rate meter.

## RESULTS AND DISCUSSION

### Gamma spectrometric analysis of the historic building material samples

In addition to direct measurements, gamma spectrometric analysis of a few building material samples from the Late Medieval period of B&H history was carried out. Since indoor radiation is mainly determined by the building material itself there was a clear need to examine the radioactive isotope concentration in the traditional materials in order to compare the obtained results with the radioactivity content of the modern building materials.

The presence of primordial radionuclides such as $^{226}$Ra, $^{232}$Th, and $^{40}$K in the investigated samples (St Nicholas monastery, Bobovac fortress, Počitelj fortress, and Soko fortress) was detected by using an HPGe gamma spectrometer. The spectra were collected and processed using a multi-channel analyzer (DSPEC jr 2.0, ORTEC) and the GammaVision software (ORTEC). It was applied the standard procedure for the sample processing was applied [5, 6]. The acquisition time of 86400 seconds was long enough to reduce the counting error. The gamma dose rates in air (nGyh$^{-1}$) due to gamma radiation from the building material samples were estimated taking into account the measured values of specific activity of natural radioisotopes $^{226}$Ra, $^{232}$Th, and $^{40}$K and the parameter values proposed in [7]

$$D_{\mathrm{R}} \quad 0.92A_{\mathrm{Ra}} \quad 1.1A_{\mathrm{Th}} \quad 0.048A_{\mathrm{K}} \qquad (1)$$

where $A_{\mathrm{Ra}}$, $A_{\mathrm{Th}}$, and $A_{\mathrm{K}}$ are the specific activities (Bq kg$^{-1}$) of the primordial radionuclides.

The results obtained for the specific activity of radioisotopes in the investigated historic samples are presented in tab. 1. The reported uncertainty of the measured results was calculated from counting statistics, *i. e.*, taking into account the number of counts under the gamma peak with energy of interest after the correction for background counts. In order to correspond approximatelly to a confidence level of 95 %, the reported uncertainty was based on a coverage factor $k = 2$. The same table includes the calculated absorbed dose rates in air with the uncertainties obtained by using the error propagation procedure. The specific activities of the studied medieval samples were found to be less than the average activity concentrations for a block made of concrete (40 Bqkg$^{-1}$, 30 Bqkg$^{-1}$, and 400 Bqkg$^{-1}$ for radium, thorium and potassium, respectively) and close to the lower limit (within $1\sigma$) of the range for the contemporary materials from the EU member states [7]. The gamma analysis results re-

**Table 1. The specific activities of primordial radionuclides for the historic building material samples and estimated absorbed dose rates in air**

| Radionuclide | Specific activity [Bqkg$^{-1}$] Mean 2* standard deviation | | | |
|---|---|---|---|---|
| | St. Nicholas Monastery 16$^{th}$ century | Bobovac fortress 14$^{th}$ century | Počitelj fortress 15$^{th}$ century | Soko fortress Gračanica 15$^{th}$ century |
| $^{40}$K | 66.78 23.36 | 216 74 | 6.87 3.16 | 24.37 9.66 |
| $^{226}$Ra | 8.97 3.38 | 16.42 6.06 | 24.53 8.84 | 22.79 8.30 |
| $^{232}$Th | 11.62 3.26 | 22.31 6.24 | 1.18 0.34 | 0.48 0.14 |
| Absorbed dose rate [nGyh$^{-1}$] | 26.38 5.10 | 56.93 10.64 | 24.42 8.14 | 23.44 7.66 |

vealed almost uniform distribution of $^{226}$Ra, $^{232}$Th, and $^{40}$K radionuclides in the investigated samples taken from the Late Medieval objects in B&H.

Table 1 shows that the calculated gamma dose rates in air for the studied samples are near the lower boundary of the worldwide values in the interior of buildings ranging from 20 to 200 nGyh$^{-1}$ [8]. The results of the gamma spectrometric analysis of the available historic samples indicate that the traditional building materials reduce the values of gamma absorbed dose rates when compared to the modern building materials. It could be their potential advantage for specific applications as a complementary material in environmentally sustainable architecture [9].

## Evaluation of the indoor to outdoor dose rate ratio in the historic objects

The several historic public objects in B&H with the similar building structure were selected for investigation of the detection capacity of the available portable dose rate meter for measurements of natural radioactivity. The sacral objects were chosen because they have the similar building construction related to the number of walls, bulkheads and ceilings. Table 2 contains the basic information on the studied objects and the indoor and outdoor ambient dose equivalent rates for each object measured by using the LUDLUM 9DP device. The reported uncertainty was obtained according to [10] in order to get a confidence interval of 95 % for the geometric mean values.

Figure 4 shows the indoor and outdoor histograms for the experimental data aqcuired in and around of all the objects being examined. It shows a somewhat broader indoor distribution with the shift to higher dose rate values compared to the outdoor distribution.

Figure 5(a) indicates a positive correlation between the LUDLUM 9DP outdoor and indoor ambient dose equivalent rates measured around and inside the historic objects, respectively. Since the construction materials of the historic objects are supposed to have the same geological composition as the surrounding soil a high positive correlation between the mentioned quantities could be expected. However, taking into account that the LUDLUM 9DP dose rate meter is not

**Table 2. Geometric mean (GM) and the uncertainty for the indoor and outdoor ambient dose equivalent rates measured inside and around the historic objects, respectively**

| Objects | Building material/Built in | LUDLUM 9DP dosimeter GM GSD [nSvh$^{-1}$] | |
|---|---|---|---|
| | | Indoor | Outdoor |
| Atik mosque Tuzla | Brick/1888 | 104.2 1.7 | 97.4 1.7 |
| Hajji Hasan mosque Tuzla | Brick/1548 | 161.3 1.5 | 114.9 2.0 |
| Husein Caus mosque Tuzla | Clay, wood/1701 | 133.2 1.7 | 96.9 1.5 |
| Mejdan mosque Tuzla | Clay, wood/1644 | 140.8 1.5 | 115.6 1.5 |
| Turali-begova mosque Tuzla | Stone/1572 | 101.2 1.7 | 81.6 2.0 |
| St.Peter and Paul Monastery Tuzla | Stone, marble/1986 | 97.5 1.7 | 98.0 2.0 |
| St. Nicholas Monastery Ozren | Sandstone, limestone/1284-1316 | 95.1 1.7 | 116.5 1.3 |
| The Jewish Museum-The oldest synanogue in B&H, Sarajevo | Stone, marble/1581 | 54.5 2.0 | 66.2 1.3 |
| Brusa Bezistan Sarajevo | Stone, wood/1551 | 61.4 1.7 | 109.3 1.5 |
| Catacombs (underground church) Jajce | Stone/14$^{th}$ century | 67.2 1.5 | 89.0 1.5 |
| Fethija mosque Bihać (former church) | Stone/13$^{th}$ century | 130.4 1.7 | 123.4 2.6 |

suitable for natural radioactivity measurements with a high reliability as it was presented in fig. 3(a), the adjusted R-Squared of 0.27 for the investigated data indicates a weak positive correlation. The result obtained is in accordance with the LUDLUM 9DP performances and limitations.

The indoor to outdoor dose rate ratio is generally due to terrestrial radiation while in our study the contribution of secondary cosmic radiation was included in the LUDLUM 9DP signal. However, it was a common procedure applied by other authors as well [11]. The evaluated indoor to outdoor ratios obtained with the LUDLUM Model 9DP instrument for all the stud-

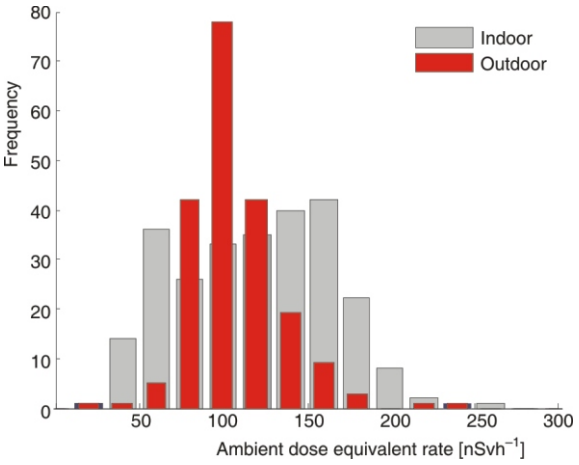


**Figure 4. Indoor and outdoor histograms for the experimental data acquired in and around all the studied objects**

ied historic objects ranged from 0.56 0.01 to 1.65 0.02 with the average value of 1.07 0.20. The evaluated ratio range of 0.56-1.65 is within the lower part of the ratio range for dwellings ranged from 0.6 to 2.3 [8]. Most of the obtained ratio results are within the range of 0.8-1.2 which can be attributed to houses made of construction materials with nearly the same radioactivity content as the surrounding soil [12 and references therein].

## CONCLUSIONS

The results of our study represent the first attempt of experimental determination of the indoor to outdoor dose rate ratio by using the LUDLUM 9DP portable dose rate meter based on an ion chamber. The ambient dose equivalent rate measurements were performed in and around the historic objects made of the traditional building materials mainly from the Late Medieval, Post Medieval, and Ottoman Period of B&H history. Detection capabilities of the available instrument were investigated in laboratory conditions with a source of low activity and it was found that the instrument is more reliable for measuring a higher dose rate level when compared to low level environmental radiation. The results of our study indicated that there was a weak positive correlation between the indoor and outdoor data for the historic objects. It was experimentally verified that the shielding capability of light construction materials such as the traditional building materials is lower than that for the modern materials. The obtained average dose rate ratio of 1.07 is within the ratio range of 0.8-1.2 for houses made of materials with radioactivity content similar to that of the local soil.

The gamma spectrometric analysis of a few building material samples from the Late Medieval Period in B&H history performed using an HPGe spec-

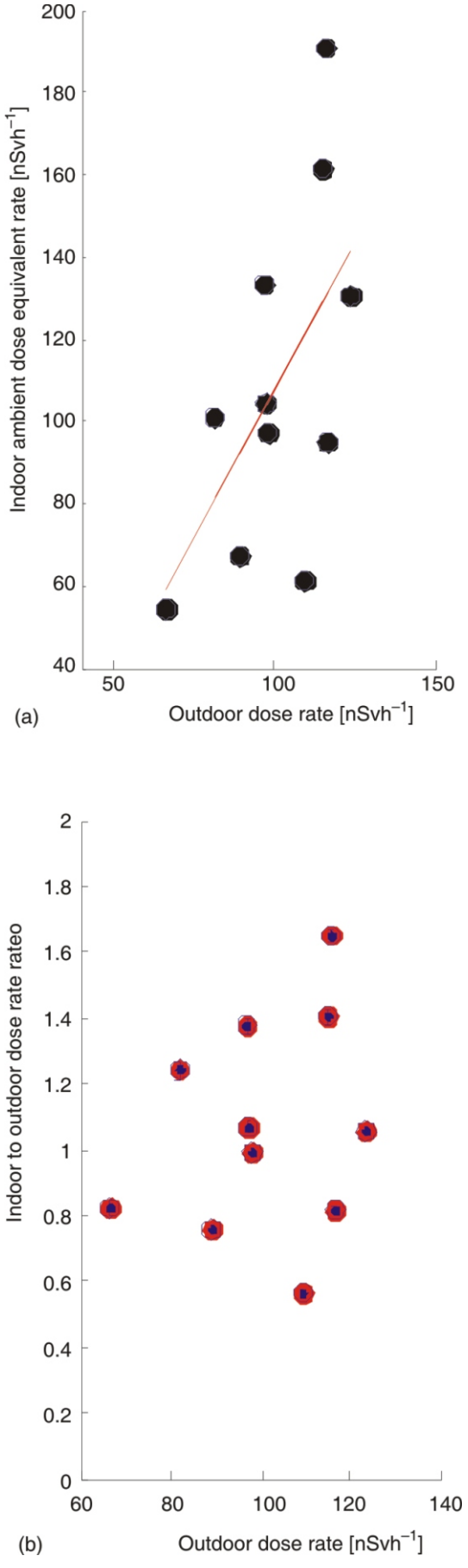


**Figure 5. Correlation between the indoor and outdoor measured dose rates (a) and their ratio (b)**

trometer has shown that the measured specific activities were close to the lower limit of the range for the contemporary materials from the EU member states. Such traditional materials therefore could have a potentional for specific applications in environmentally sustainable architecture.

This paper deals with the first indoor and outdoor dose rate measurements in B&H performed using the portable LUDLUM 9DP instrument based on an ion chamber as well as with the first gamma spectrometric analysis with an HPGe spectrometer of a few building material samples from the Late Medieval Period in B&H history. The data acquired in our study and their analysis represent a significant stage in developing the project on the indoor and outdoor dose rate database in B&H for different categories of buildings.

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## AUTHORS' CONTRIBUTIONS

S. Avdić, I. Kadić, D. Demirović, S. Sadiković, and B. Pehlivanović performed the experiments in the laboratory with a LUDLUM 9DP device and an HPGe spectrometer. All the co-authors conducted the field measurements at different locations. A. Beganović and A. Lagumdzija together with the other co-authors participated in analysis and discussion of the results presented, and in the preparation of figures, tables and text of the manuscript.

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**Сенада АВДИЋ, Аднан БЕГАНОВИЋ , Армин ЛАГУМЏИЈА, Самра САДИКОВИЋ, Бећо ПЕХЛИВАНОВИЋ, Дамир ДЕМИРОВИЋ, Ирма КАДИЋ**

## КОМПАРАТИВНА АНАЛИЗА ЈАЧИНЕ ГАМА ДОЗЕ МЕРЕНЕ ПОМОЋУ ЈОНИЗАЦИОНЕ КОМОРЕ УНУТАР И ИЗВАН ИСТОРИЈСКИХ САКРАЛНИХ ОБЈЕКАТА У БОСНИ И ХЕРЦЕГОВИНИ

Главни циљ овог истраживања био је да се испита корелација између јачине просторног еквивалента дозе гама зрачења измерене јонизационом комором унутар и изван историјских сакралних објеката на неколико локација у Босни и Херцеговини. Испитивани објекти направљени од традиционалних грађевинских материјала саграђени су у касном средњовековном и отоманском историјском периоду. Инструмент LUDLUM Модел 9DP који се темељи на јонизационој комори под притиском одабран је за мерења природне радиоактивности ниског нивоа јер јонизационе коморе имају већу осетљивост од других врста детектора. Способност детекције јонизационе коморе LUDLUM Модел 9DP испитана је у лабораторијским условима користећи извор ниске активности и у условима природног радиоактивног зрачења на неколико локација унутар и изван селектованих објеката. Утврђена је слаба позитивна корелација између брзине просторног еквивалента дозе гама зрачења унутар и изван историјских сакралних објеката. Просечна вредност односа унутрашње и спољашње јачине просторног еквивалента дозе гама зрачења од 1.07 за проучаване историјске објекте мања је од оног добијеног за савремене грађевинске материјале као што је бетон. Још није спроведена слична студија о односу јачине гама дозе унутар и изван историјских објеката у Босни и Херцеговини на основу експерименталних података добијених помоћу јонизационе коморе. Поред директних мерења јачине гама дозе, извршена је прва гама спектрометријска анализа у Босни и Херцеговини неколико узорака грађевинских материјала из касног средњовековног периода. Резултати гама анализе показали су готово униформну дистрибуцију примордијалних радионуклида у истраживаним узорцима. Установљено је да такви материјали имају смањени садржај радиоактивних изотопа у поређењу са савременим грађевинским материјалима и стога могу имати потенцијалне предности у специфичним апликацијама везаним за еколошки одрживу архитектуру.

*Кључ речи: јачина амбијенталне еквивалентне дозе, јачина унутрашње/спољашње дозе, јонизациона комора под притиском, гама спектрометрија*