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## RADON SOURCES AND ACTION LEVELS

### *Abstract*

Radon is by far the largest contributor to population exposure because the dose received from radon and its progeny exceeds all other radiation sources. After smoking, it is the second most frequent cause of the lung cancer, therefore it is classified as Group 1 carcinogenic to humans. The soil under a building is the major source of indoor radon. An important contribution to indoor radon concentration comes from building materials whose composition includes volcano rocks as well as household water. The aim of this paper is to present a brief overview of indoor radon sources and prescribed permitted concentration levels in Bosnia and Herzegovina and the world. Finally, the hazardous effects on human health have been discussed.

*Keywords: radon, exposure, soil, building materials, water, lung cancer risk*

## ИЗВОРИ РАДОНА И АКЦИОНИ НИВОИ

### *Сажетак*

Радон је елемент који далеко највише доприноси изложености становништва јер доза примљена од радона и његових продуката превазилази све друге изворе зрачења. Послије пушења, радон је најчешћи узрочник рака плућа због чега је сврстан у прву категорију канцерогена за људе. Главни извор радона у унутрашњем простору је земљиште испод зграде. Значајан допринос концентрацији радона у унутрашњости потиче од грађевинских материјала у чији састав улазе вулканске стијене и воде за домаћинство. Циљ овога рада јесте преглед извора радона у унутрашњем простору и дозвољених концентрација радона у Босни и Херцеговини и свијету. На крају су размотрени штетни ефекти по здравље људи.

*Кључне ријечи: радон, излагање, земљиште, грађевински материјали, вода, ризик од рака плућа*

## 1. INTRODUCTION

Naturally occurring radioactive elements are spontaneously disintegrated in the process of radioactive decay, after which a nucleus with different properties is formed and the process is accompanied by the emission of the alpha particles, beta particles, or gamma rays. Most of the natural radioactive isotopes that contribute to the dose received by humans are members of one of the three radioactive decay chains (uranium  $^{238}\text{U}$  chain, thorium  $^{232}\text{Th}$  chain, and uranium  $^{235}\text{U}$  chain), [1]. The most important non-chain radionuclides that contribute to human exposure are potassium  $^{40}\text{K}$  and rubidium  $^{87}\text{Rb}$  [1].

The decay chain of the uranium 238 contains radium as one of the members of its series (Figure 1). By  $^{226}\text{Ra}$  alpha particle emission natural, noble gas radon is formed and released into the atmosphere. Radon is colorless and odorless gas without taste, chemically inert and radioactive, and cannot be detected by any single sense but an appropriate detection system. Under normal conditions it is gaseous, easily inhaled, and one of the densest gases at room temperature. All of the radon isotopes, radon ( $^{222}\text{Rn}$ ), thoron ( $^{220}\text{Rn}$ ), and acton ( $^{219}\text{Rn}$ ), are radioactive and formed as a result of the uranium, thorium, or actinium decay series. Since it has the longest half-life of 3.8232 days [2], the most significant among them is  $^{222}\text{Rn}$ , a member of the  $^{238}\text{U}$  decay series. The half-life for the thoron ( $^{220}\text{Rn}$ ) and acton ( $^{219}\text{Rn}$ ) is 55.6 s and 3.96 respectively, thus their impact on population exposure is negligible.

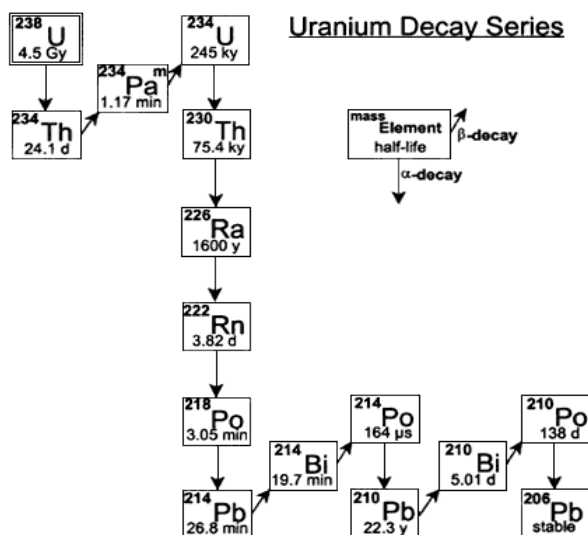


Figure 1. Uranium 238 decay series [2].

Radon is the most significant natural source of ionizing radiation because it has been disintegrated into radioactive, short-lived alpha particles emitter: bismuth, polonium, and plumb. The biological effects of radiation in a certain medium are described by the equivalent dose. Since the equivalent dose for the alpha particles is higher than for the other ionizing radiation (radiation weighting factor is 20 times higher than for gamma and X-rays), alpha particles produce greater biological harm than do beta particles and gamma rays. Passing through the biological structures, ionizing alpha particles can cause irreparable damage to DNK or produce chemically active free radicals.

According to UNSCEAR (2008) annual global average dose received by humans from natural sources is 2.4 mSv (80%) while humans on average receive 0.6 mSv (20%) from artificial sources. It can be seen in Figure 2 that the average dose due to exposure to radon is 1.26 mSv. Dose from radon and its decay products for the average person exceeds all other sources together: radon and its progenies, mostly  $^{218}\text{Po}$  and  $^{214}\text{Pb}$ , contribute about 50% to the total annual dose from the natural sources [3]. While alpha particles and radon may lead to internal exposure, external exposure occurs due to the emission of gamma rays produced in the decay chains or in  $^{40}\text{K}$  decay process.

Uranium and radium naturally occur in soils, rocks, and groundwater. As a result of mixing with human-inhaled air or drinking-water radon can enter the human body. Radon has been classified by International Agency for Research on Cancer (IARC) as Group 1, carcinogenic to humans [4]. After smoking, it is the second most important cause of lung cancer in many countries, while the probability of cancer rises relative to smoking habits [3] because the process of attaching decay products to the atmospheric particles is more pronounced in the smoking rooms regarding nonsmoking.

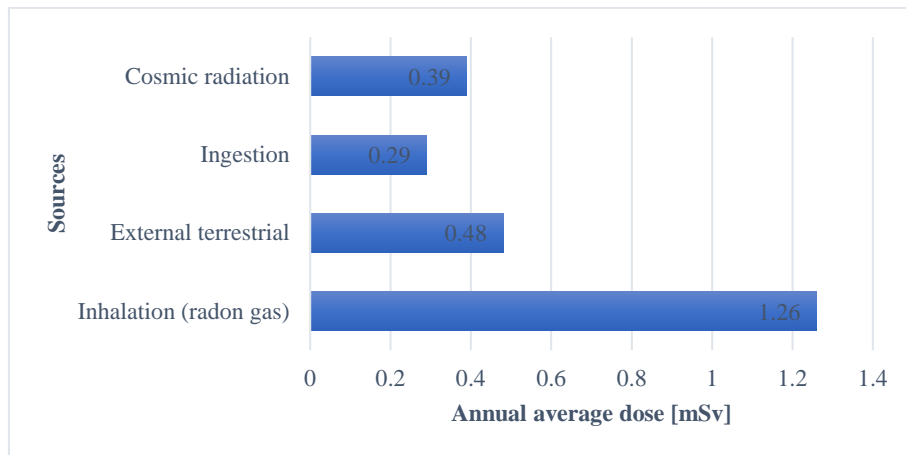


Figure 2. Radiation sources and their contribution to the annual average dose.

Nowadays, radon levels in the interior spaces have been evaluated in most countries, parameters that affect concentration have been analyzed as well as procedures that should have to be taken in order to reduce the level below prescribed values.

## 2. RADON INSIDE OF BUILDINGS

The term Indoor Air Quality (IAQ) is used to refer the air quality inside of building depending on concentration of the pollutants and thermal conditions that affect a health, comfort and performances of inhabitants in all types of buildings. One of the most significantly indoor air contaminants is radon. Long term exposure to radon and other hazardous substances in air results in some chronic effects, most notably lung cancer. As noted early, radon can enter house moving up from the ground soil and rocks, from building materials and from ground water. According to UNSCEAR soil contribute about 80% to indoor radon concentration, building materials with 12%, while the rest is from the water and air from the outside [5].

### 2.1. SOIL UNDER A BUILDING

Concentration of the uranium/radium in the soil under a building is determined by concentration of these elements in the rocks from which the soil was formed. Soil is considered as the main source of indoor radon concentration.

Radon formation and pathways to the air include processes of emanation, transport and exhalation [2]. Emanation is defined as the process of radon releasing the grain that contains  $^{226}\text{Ra}$  and escapes into the interstitial space between the grains when radium decays [2]. After that, radon atoms can be moved through the process of diffusion or convective fluid flow before reaching the surface. Exhalation represents releasing gas into the atmosphere from the ground surface.

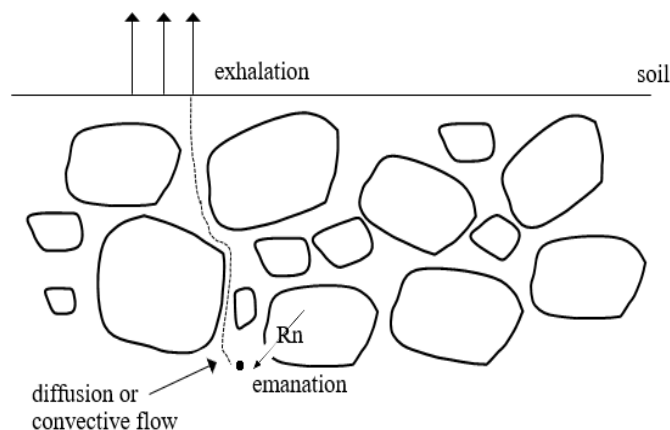


Figure 3. Process of radon transport in the soil.

Temperature difference between the air inside a building and the air outside creates the pressure difference in the way that the air pressure inside is usually lower than the pressure in the soil around the foundations and basements floor slab. The existence of the air pressure difference leads to the process of convective fluid flow. The pressure differences of the air inside and outside of the building can also be affected by wind [6]. The quantity that represents the ability of a material to acts as a barrier to gas movement through it when pressure gradient exists is named permeability.

The existence of the radon concentration differences between low-concentration regions, such as the interior of the building, and higher-concentration area of the soil or building materials, causes the process of diffusion throughout which the radon atoms are moved in the direction opposite of concentration increasing gradient, from the place of higher radon concentration to the lower concentration place [6]. Furthermore, the diffusion is dominant transport mechanism process so the convective component of radon transport usually is not considered [2, 7]. The value of radon diffusion coefficient for materials represents the ability of gas movement through the material when concentration gradient exists and it is proportional to permeability.

Regarding capability to act as a protection from radon, the diffusion coefficient, diffusion length, and thickness of different materials are generally determined, although in some cases the permeability coefficient is also calculated.

The amount of radon gas that reaches the interior depends on the diffusivity of the substructure and permeability of soil [6, 8]. As a result, higher radon concentrations can be found in buildings without basements [9]. To ensure reliable protection, the thickness of insulating material on the ground floor must satisfy the following condition [10]:

$$d \geq 3l \quad (1)$$

where  $d$  (m) is the thickness of the material, while  $l$  (m) represents radon diffusion length in the material and can be calculated as:

$$l = \sqrt{\frac{D}{\lambda}} \quad (2)$$

$D$  (m<sup>2</sup>/s) is radon diffusion coefficient and  $\lambda$  (1/s) is radon decay constant. Diffusion length primarily depends on the material species. According to some new research, condition (1) is too strict and only a small number of waterproofing materials can satisfy this condition (less than 5.5%) despite the fact that they represent a good protection [11]. Instead condition (1), it is more relevant to calculate the radon resistance value for each material and use an approach that is based on exponential radon distribution in the sample [11].

Relation between thickness and material permeability is given through the equation [12]:

$$k = Pd \quad (3)$$

$k$  is permeability coefficient (m<sup>2</sup>/s),  $P$  is transmittance or the speed of radon flow (m/s). For the lower, medium and high values of diffusion length materials are marked as radon-tight, radon less permeable and radon permeable.

Materials that are homogeneous are likely to have lower permeability coefficient, while heterogeneity indicates higher value of permeability and diffusion coefficient. According to some researches [12] insulating materials such as foil thermo-vapor barrier, the insulation film under the foundation are found to be the best protection against soil radon gas. In this way, installing good waterproofing materials provides building protection from radon.

Problems with determining and comparing materials permeability/diffusion coefficient in different countries are caused by non-establishing unique experimental procedures which may lead to several orders of magnitude value variability for the same material. The new ISO/TS 11665-13 (2017) standard may lead to the improvement in this field [11].

## 2.2. BUILDING MATERIALS

Building materials used for construction like concrete, brick, cement, sand, and gravel may have increased concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, as well as <sup>40</sup>K, and they are usually denoted as naturally occurring radioactive materials or NORM. These elements reflect the geology of the places from which raw materials are taken. Despite the fact that the soil under a building is considered as the main source of radon in closed space, in some of the building materials made from granite, slate, or volcano rocks, the content of radionuclides is higher than in soils [13]. The high content of radionuclides can pose a risk not only because of radon but also from gamma radiation, particularly on the upper floor where contribution to exposure from building material is dominant.

In order to estimate building material's hazardous effects, especially since it is well known that individuals spent an average of 80% of their time indoors, activity concentrations, radium equivalent activity index, gamma index, absorbed dose, and annual effective dose from  $^{236}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  have been determined in the number of papers [7, 14, 15, 16]. Many studies reported that standardly used materials are safe for construction because obtained values are within permitted limits. World-wide activity concentrations of these radionuclides in building materials are 50 Bq/kg 50 Bq/kg and 500 Bq/kg respectively. Some of the research indicate that granite and siporex are not recommended in poorly ventilated rooms [7].

Radium equivalent activity index allows comparing measured activity concentrations of radionuclides since materials have different concentration of them and its recommended maximum value is 370 Bq/kg [17]. Gama index, the quantity that takes into account the different exposure contributions of different elements, should be less than 1 for bulk materials and less than 6 for superficial material, in which case annual effective dose from exposure to gamma radiation from radionuclides in building materials will be less than 1 mSv.

Radionuclide activity concentrations analysis of building materials used in Bosnia and Herzegovina has shown that values of all quantities are below prescribed standards [15]. Generally, in Bosnia and Herzegovina, insufficient attention is paid to the issue of materials radioactivity and only a few studies have been done. Also, a legal framework for radon protection, which is in the process of extension and improvement, does not clearly define the activities which could involve NORM [18] and only provide concentration limits (Table 1) for the three radionuclides depending on indoor and outdoor usage of building material in architectural engineering.

Table 1. Concentration limits for indoor and outdoor building materials in BiH [19].

Radionuclide	Indoor concentration limit (Bq/kg)	Outdoor concentration limit (Bq/kg)
$^{226}\text{Ra}$	300	400
$^{232}\text{Th}$	200	300
$^{40}\text{K}$	3000	5000

### 2.3. RADON IN WATER

Household water as the need for life may be used either from the surface or ground. Like building materials and soil, water contains dissolved naturally occurring radioactive elements: uranium, thorium, and actinium. Radon from water poses a danger to humans in two ways: it may be released into indoor air and attached aerosol, or can be ingested into the body through consumption which passes through the wall of the stomach. If the water comes from a surface water source, then most of the radon will be released into the atmosphere before reaching supply or home. Concerns about radon may be in the case if the underground water is used directly from the place of its origin. Underground water is moved through rocks and materials containing uranium (radium) contributing to high concentration of radionuclides in water. The concentration may be especially increased in volcanically active areas.

According to Environmental Protection Agency for drinking water (EPA), radon concentration level in water that shouldn't be exceeded is 11.1 Bq/l, while WHO has a restricted level to 100 Bq/l which corresponds to annual effective dose of 0.1 mSv/y [20, 21]. Despite the fact that this country still doesn't have established regulations regarding radon levels in drinking water, in Bosnia and Herzegovina analysis has been performed for drinking water in the Tuzla area. Obtained results indicate that the radon activity concentration has not exceeded the value of 100 Bq/l [22].

Higher risk from radon in water occurs in places with thermal and mineral water spa, where the radon rich water has been used for therapeutic purposes. In spa areas, elevated radon concentrations are found both, indoor and outdoor. One such example is Niška Banja spa in Serbia, which represents an area of high activity due to the specific geology. Activity concentrations in some water samples were from  $24.5 \pm 2.4$  Bq/l to  $648 \pm 38$  Bq/l which exceeded EPA recommendation as well as WHO upper level [23] while high concentrations were also measured in hotel spas and houses. Measurements of radon activity concentrations in spas of B&H had shown that prescribed levels were not exceeded and all concentrations were below 11.1 Bq/l [24].

### 2.4. PARAMETERS THAT AFFECT RADON LEVEL

Apart from geology and building materials characteristic, it should be noted that radon level concentrations is influenced by several other parameters: ventilation, temperature, humidity,

pressure, seasons. Dwellings that are closed all day or during the weekend are expected to have higher average radon concentrations than if the same dwelling were well ventilated all the time.

A number of studies have shown the impact of temperature on radon concentration variation. In the winter period, the outdoor temperature is significantly lower than indoor temperature leading to higher outdoor pressure. This temperature ratio impacts the pressure difference and causes gas flow and infiltration to indoors. This effect is enhanced during the winter, while reverse flow may occur in the summer [25]. As a result, a radon level variation during the seasons was observed.

A study in China has shown the impact of rain on radon concentration. As it was suggested, a lower concentration during the rainy day was influenced by lower outdoor pressure, humidity and soil moisture that had limited pathways [26].

Due to the number of influencing parameters, it is advisable performing measurements for one year on all ground premises.

### **3. RECOMMENDED RADON LEVELS**

#### **3.1. INDOOR RADON LEVEL**

A recommended level or action level represents annual average radon activity concentration above which it is necessary to conduct measure in order to reduce radon concentration. By the World Health Organization, annual average indoor radon concentration reference level is  $100 \text{ Bq/m}^3$ ; if this level cannot be reached, then the chosen reference level should not exceed  $300 \text{ Bq/m}^3$  [3]. International Commission on Radiological Protection (ICRP) has recommended reference level of  $300 \text{ Bq/m}^3$  for homes and workplaces.

Countries in the region have different national reference levels. In Serbia, the national reference level for indoor radon concentration for new buildings is  $200 \text{ Bq/m}^3$ , for the existing buildings is  $400 \text{ Bq/m}^3$ , while  $1000 \text{ Bq/m}^3$  is the upper level recommended for workplaces. A new national radon survey has shown that in 3% of all measurements the level of  $400 \text{ Bq/m}^3$  has been exceeded [27]. The required annual average radon activity concentrations which represent upper limit in Montenegro currently have the same values as in Serbia. As a candidate to EU membership, in both countries, the required action level is  $300 \text{ Bq/m}^3$ . This level is defined for homes and workplaces according to the EU directive from 2013 [28]. Croatia has also defined an action level of  $300 \text{ Bq/m}^3$  according to the EU directive.

Determination of radon concentration for the whole territory in Bosnia and Herzegovina has not been completed yet and only local surveys were performed. An old regulation prescribed an action level of  $1000 \text{ Bq/m}^3$  for workplaces. State regulatory agency for radiation and nuclear safety works on new regulation that will include the new reference level (probably  $300 \text{ Bq/m}^3$ , in order to unify with some countries of the region and the European Union), monitoring radioactivity, as well as new legal framework for NORM [18].

The indoor radon measurements are generally performed in dwellings, while kindergartens and schools are sometimes chosen as location. The first investigation on the indoor radon and thoron and their decay products concentration in 25 primary schools of Banja Luka was performed from May 2011 to April 2012. The obtained geometric mean concentrations were  $99 \text{ Bq/m}^3$  and  $51 \text{ Bq/m}^3$  for radon and thoron gases respectively [29]. A new study on radon concentration in the school indicates a high spatial variability of radon concentration so it was recommended to measure it in all ground level rooms [30].

#### **3.2. OUTDOOR RADON LEVEL**

The average outdoor radon level is usually low, less than  $100 \text{ Bq/m}^3$  and does not represent a risk to the population because it gets diluted. Outdoor radon activity concentration in B&H was measured at 92 locations and mean activity concentrations were in range of 15 to  $38 \text{ Bq/m}^3$  [31]. Elevated activity concentrations in some areas were conditioned by the technological processing of coal and raw material.

### **4. CANCER RISK**

Radon progenies attached to the aerosols might enter the lungs by breathing, deposit on lung epithelial and increase the risk of lung cancer. From the respiratory tract, these particles may be transported to the gastrointestinal tract, to lymph nodes, or transferred by fluid (blood and lymph) to other tissues. Water ingesting is another way to enter and circulate through the body. Radon ingested by water enters the stomach from where it can enter the bloodstream.

Ionizing alpha particles emitted by deposited short-lived decay products of radon ( $^{218}\text{Po}$  and  $^{214}\text{Po}$ ) can interact with biological tissues and deliver large localized radiation doses. Even a single alpha particle can cause major genetic damage to a cell. Depending on the average radon concentration, the percentage of all lung cancers related to radon is estimated to be in the range of 3% to 14% while long-term exposure to radon concentration that is increased by  $100 \text{ Bq/m}^3$  increases lung cancer risk by about 16% [3].

Relation between radon progeny concentration in the air and the risk of lung cancer was obtained in the study on lung cancer mortality among uranium miners and other coworkers exposed to very high levels of radon progeny. Radon-related increase in mortality caused by other cancer types has also been researched but clear evidence that radon cause other than lung cancer has not been found yet.

## 5. CONCLUSION

In this paper, an overview of radon sources, parameters that affect radon levels, risks for human health and action levels for B&H and surrounding countries have been presented. It can be concluded that radon gas present in the buildings and homes comes mainly from the soil. After the soil, radon concentration may be increased by radon contained in the building materials and water. On the other hand, outdoor radon concentration is negligibly low compared to indoor and usually does not represent a risk for human health.

As there is a clear evidence that elevated radon concentration increases the risk of developing lung cancer, it is necessary to implement the measures in order to reduce the concentration when the prescribed radon levels are exceeded.

Waterproof materials act as a barrier between the soil and indoor space (materials that have small permeability) and can be used to reduce indoor radon concentration. It is advisable to perform an adequate control and measurements, especially in dwellings and workspaces located on the ground floor and basement levels for which is expected to have a higher radon concentration. Increasing ventilation can also be used as a measure.

From the available researches, it can be concluded that prescribed radon levels and radionuclides concentrations in B&H are underestimated. However, previous researches have shown that in B&H hazardous levels of radon concentrations are not exceeded, either indoors or outdoors neither in water.

In the future, it is necessary to conduct more detailed research and establish stricter regulations and values for radon levels.

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