

REVIEW OF THE PRINCIPAL MECHANISM OF RADON IN THE ENVIRONMENT

DANIELA GURAU*, DORU STANGA, MITICA DRAGUSIN

“Horia Hulubei” National Institute for Physics and Nuclear Engineering, P.O.B. MG-6, 077125,
Magurele, Romania

*E-mail: daniela.gurau@ymail.com

Received June 4, 2014

The properties and behavior of ^{222}Rn and ^{220}Rn in the free atmosphere are presented. The emanation and transport in materials with the principal mechanism are discussed. The aerosols formation process and surface deposition are also very important in view of environment radioactivity evaluation being a very important task in the contingency planning for radiological and/or nuclear emergency.

Key words: radon, decay products, aerosols, emanation power, surface deposition.

1. INTRODUCTION

Life on Earth has emerged and developed in terms of natural radioactivity generated by existing natural radioactive isotopes in the environment and steady stream conditions of radiation from cosmic sources and the Sun. Living organisms that inhabit the planet, including humans, have adapted to a certain level of radiation named natural background. In nature, at any level of organization of living matter, every body its shape a space of existence called habitat. Within or between members of the same species are established biological interaction relationships, which underlie the formation and function of biotic communities. However, to ensure their biological survival, any biotic community must develop its own system of relationships with the abiotic environment around them. Natural ecosystems show not only a wide spatial distribution, but also a large temporal variability depending on many ecological interactions between different types of biotic communities and their natural environment. The interdependent relationships between different biological species lead to the establishment of biocenosis, which varies according to the specific of environmental conditions in which they were formed. The living organisms are involved in the supply chains, and participate to the redistribution of natural radioactive isotopes in the environment, without essentially affecting the natural background radiation. Man is the only creature that can dramatically alter the environmental radioactivity, through specific activities.

He can modify the local natural radioactivity by redistributing the natural radioactive isotopes during household or industrial activities. Also, by producing new radioactive isotopes, which do not naturally exist in the environment, the man can add a new component called artificial radioactivity, with all the implications that result. The natural radioactivity level is very variable in the world; there are significant variations on small areas. It is obvious that if a person in an area with low natural radioactivity lives in an area with higher natural radioactivity, there is some additional risk to it.

In the case of contingency planning guidelines for radiological and/or nuclear emergency, the evaluation of the environmental radioactivity level is a very important task that determines the right action to be followed. This program is carried out regularly and continuously under normal working conditions, that ensures the measuring of the radioactivity level in the environment for determining the background or the normal levels of radioactivity, and monitors the influence of natural radioactivity on the human body. Because a very large part of the population exposure to the natural radioactivity comes from inhalation of ^{222}Rn and ^{220}Rn decay products with short-lived, the authors analyse in this paper the properties and the behavior of ^{222}Rn and ^{220}Rn in the free atmosphere, very important to know for the radioactivity level evaluation. ^{222}Rn and ^{220}Rn concentrations have a height level in the atmosphere, and the radon background variation is the major factor that influences the detection capability for the environmental samples.

2. THE RADON RADIOACTIVITY

Radon, the chemical element, is part of the noble gas group with extremely low or totally without chemical reactivity. All the radon isotopes are called emanation and are radioactive with short half-life; ^{222}Rn has the longest period of 3.8 days. Because it is a noble gas, radon does not form chemical compounds and do not react with any other element. Radon gas formed by the decay of radium and has a tendency to migrate even at great distances to the atmosphere. Radon “emanate” from the air accumulated in the alveoli, fissures or cracks of the rocks of the mountains, the air in the soil, water, and so on, for this was named “emanation”. The separation of radon parent rock depends on its environment temperature, humidity and mineral structure. This problem of radon was first reported more than 120 years ago in the mines when was observed that most fraction of the population to natural radiation exposure comes from inhalation of short-lived decay product of ^{222}Rn and ^{220}Rn occurring in the free atmosphere and in higher concentrations in the air of rooms.

In the atmosphere there are a large number of radioactive nuclides that decay by emission of nuclear radiation. The main natural radionuclides that emit alpha and beta radiation are ^{222}Rn and its daughters ^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po , ^{210}Pb , ^{210}Bi ,

and ^{210}Po , and ^{220}Rn and its decay products ^{216}Po , ^{212}Pb , ^{212}Bi (^{212}Po , ^{208}Tl) belonging to uranium-radium radioactive series and thorium respectively (outside them, there are radionuclides that are produced by cosmic radiation and artificial radionuclides, in much smaller amounts). As a result of decay processes that occur in rocks appear to belong to the disintegration products of inert gas, ^{222}Rn and ^{220}Rn , which diffuse through the soil and into the atmosphere. The flow of ^{222}Rn and ^{220}Rn in soil depends on soil type and some meteorological factors. Being radioactive it can return in the atmosphere and can suffer a process of decay, giving rise to descendants that when are formed are positively ionized and can adhere to existing aerosols or form molecular complexes. In normal conditions regarding the concentration of aerosols in the air, most of the radioactive particles attach to aerosols. The big difference between the half-life of 3.8 days for ^{222}Rn and 55.8 second for ^{220}Rn it is important for escaping from the soil, the soil surface free distribution and the distribution in rooms. The decay products of ^{222}Rn are divided into two groups: products with short-lived and products with long-lived. The largest fraction of the activity concentration in the free atmosphere or at the soil surface is owned by ^{222}Rn , ^{220}Rn and their decay products with short-lived.

3. RADON EMANATION AND TRANSPORT IN MATERIALS

^{222}Rn and ^{220}Rn get into the atmosphere mainly by crossing the ground-air or building material-air interfaces. Contributions from other sources such as oceans or groundwater and natural gas are relatively small. Because in the soil, and in the majority of building materials that have soil in the composition, concentrations of ^{222}Rn and ^{220}Rn are found to be up to $1\text{E}+04$ times higher than in the atmosphere; there is a high concentration gradient between $^{222}\text{Rn}/^{220}\text{Rn}$ of these materials and the atmosphere. This gradient is constantly maintained by radionuclide generation from ^{238}U and ^{232}Th series and is responsible for continuous flow isotope of ^{222}Rn and ^{220}Rn in the atmosphere. The $^{222}\text{Rn}/^{220}\text{Rn}$ concentration in the atmosphere is governed by the source term, characterized by exhalation rate (the activity reported on surface and unit time) and atmospheric dilution processes that are affected by weather conditions. Besides of ^{222}Rn flux from soil, the building materials (which have soil in the composition) that contain ^{226}Ra are potential sources of isotopes of ^{222}Rn in the rooms. The ^{222}Rn flow from material occurs in two steps: the release from mineral grains in the material pores (a process called emanation) and the radionuclide transport by material pores in free atmosphere and rooms.

When radon decays in soil granules, the resulting atoms (isotopes of radon) must first get out from mineral grains in air pores. The fraction of radon coming out into the pores is known as power emanation coefficient or fraction. The emanation fraction is considered to be composed of: recoilless fraction and diffusion fraction. Due to the small diffusion coefficient of gases in the soil, it is

assumed that the majority of the emanation power fraction is made up of recoil processes. After α disintegration of Ra isotopes (^{226}Ra and ^{224}Ra), ^{222}Rn and ^{220}Rn atoms have kinetic energies of 86 keV and 123 keV and they are moving from the point of generation until the energy that is transferred to the material. The distance traveled is approximately from $4\text{E-}02\mu\text{m}$ up to $6\text{E-}02\mu\text{m}$ in granular material and $6\text{E+}01\mu\text{m}$ in the atmosphere [1, 2]. The influence of temperature is small [3], but few studies [4, 5, 6, 7] have shown that moisture has a large impact on the coefficient of emanation. The explanation of these phenomena is that the stopping distance of radon recoil is less in water than in air. An atom of radon that enters into a pore filled or partially filled with water has a higher probability of being stopped in pore volume, without being able to pass the pore and penetrate another grain.

In view of entering into the atmosphere, $^{222}\text{Rn}/^{220}\text{Rn}$ will pass through the material pores. In this condition only a fraction of it will get out to the surface, because the rest of it will be decayed. Two principal mechanisms are very important for the ^{222}Rn transportation into air: the diffusion length and the convection flow. The exhalation rate from soil depends on the emanation fraction, the radium activity mass concentration of the material, the density of the material, the decay constant of the radon isotope and the diffusion length of isotopes in soil; and it is expressed in $\text{Bqm}^{-2}\text{s}^{-1}$. Many external factors can influence the diffusivity and through this the rate of exhalation; increasing the atmospheric pressure decreases the rate of exhalation, while increasing the wind speed and temperature can enhance it. In addition, these parameters may forecast convection due to a flow of air in the pore space, and thus changing the radon flow in the soil. Previous studies have shown that the exhalation rate is governed by the diffusion length. The transport of radon isotopes from buildings constructions and so the exhalation rate can be influenced by the convection current, especially those determined by atmospheric pressure.

4. AEROSOL FORMATION BY RADON DECAY PRODUCTS

Aerosols produced by the ^{222}Rn and ^{220}Rn decay products are generated in two steps. After their appearance, through decay of ^{222}Rn , the radionuclides freshly generated quickly reacts (in less than 1s) with trace gas and vapor from the air and are becoming small particles, called clusters, or radionuclide “unattached” having a diameter from 0.5 to 5 nm. In addition to cluster formation, these radionuclides attach to existing aerosol particles in the atmosphere in 1 to 100 seconds, forming the radioactive aerosol of ^{222}Rn and ^{220}Rn decay products. Most of the newly formed clusters with disintegration products are positively charged and have a high mobility [8]. The mobility is characterized by the diffusion coefficient, which controls mainly the radioactive aerosol formation by attaching and deposition on

surfaces and in the lungs. Since 1956, a number of studies concerning the diffusion coefficient were made with partially conflicting results [9, 10]. It was found, however, that the aerosols formation depend on the experimental conditions, *i.e.* humidity and gaseous traces in the air. Excepting the rapid neutralization reactions and clustering, the decay products of ^{222}Rn attaches to existing aerosol particles. The attachment distribution can be characterized by the number of particles attached per unit volume, and the activity size per unit volume. These numbers are different because the aerosol attachment is made according to the diameter of the products.

5. THE SURFACE DEPOSITION AND THE TURBULENT MIXING TRANSPORT

The removal processes from the atmosphere, such as wet and dry atmospheric deposition, have a significant influence on radon and thoron decay products concentration. An important parameter, especially in buildings is the radionuclide deposition on the walls and furniture. The wet deposition of aerosol particles is governed mainly by physical processes of sedimentation by gravity, impact due to inertial forces, interception and Brownian diffusion. The mechanisms of particles surface deposition from a turbulent air flow can be divided into two categories: transport to surface (determined mainly by the turbulent diffusion) and aerosols precipitation on surfaces (depend on the particle diameter, structure and surface roughness). The flow to the surface of the particles in equilibrium can be described by a sum of three terms: the flow fraction with turbulent diffusivity, the Brownian diffusivity fraction and the sedimentation fraction [11]. In most cases, in turbulent air flow, the flow fraction with turbulent diffusivity is much higher than the Brownian diffusivity fraction and the sedimentation fraction. The deposition can be characterized by the introduction of the deposition rate that is a fraction between the number of particles deposited per surface unit and time, and the concentration of particles (depending on the particle diameter and the height above the surface of the particles) [12]. ^{222}Rn and ^{220}Rn formed in the soil are released into the atmosphere. After exhalation, the radioactive gases and the decay products are distributed in the troposphere especially through the turbulent mixing air characterized by the turbulent diffusion. The turbulent diffusion coefficient varies with the altitude in accordance with vertical variations of wind speed and atmospheric stability. The concentrations profiles are influenced by the radionuclide decay constant and by the wet or dry removal of aerosols from atmosphere.

Numerous literature data tend to support the model predictions of Jacobi and Andre [13]. The calculation presented in their paper offer information regarding the isotopes and their decay products variations with height. Depending on temperature, temperature gradient and wind speed were materialized different types

of weather conditions. The difference between the radon concentrations during the day and the night is small. Large variation between the radon and radon progeny concentrations during the day is determined by the turbulent diffusion changes in the atmospheric layer from the soil surface. The exhalation rate modification with the radionuclide concentration change during one day is usually small.

6. ESTIMATION OF RADON AND THORON CONCENTRATION

Many literature data reveal some methods for the assessment of the activity concentration of ^{222}Rn and ^{220}Rn activity concentration [14, 15, 16, 17]. A few years ago, Sima [18] presented the structure and the applications of the AMERATHOR (Active Methods for Radon and Thoron decay products) program, dedicated to evaluate the complete information available about the activity of a filter. The software can be used also for assessing the expected quality of a measurement protocol and for optimizing it.

Based on the basis process description of the generation and behavior of the ^{222}Rn , ^{220}Rn and their decay products, the activity concentration of these radionuclides in the free atmosphere and in the residual air can be analyzed. The ^{222}Rn and ^{220}Rn decay products concentrations from the free atmosphere are in very wide limits, depending on the nature of the rocks, ground state and the diffusion conditions from atmosphere. By vacuuming air, the radioactive aerosols of ^{222}Rn and ^{220}Rn decay products are retained on filters. From the quantitative point of view, the accumulation processes on filters can be obtained by solving a system of differential equations for each radioactive series:

$$\begin{cases} \frac{dN_1}{dt} = D\eta C_1 - \lambda_1 N_1 \\ \frac{dN_i}{dt} = D\eta C_i + \lambda_{i-1} N_{i-1} - \lambda_i N_i \end{cases} \quad (1)$$

where N_i is the number of i type nuclei retained on filters, D is the air flow, η is the retention factor of particles on filters, C_i is the number of i type nuclei from the unit volume of air, λ_i is the i nuclide decay constant. Resolving this equations system permit the concentration assessment of nuclei from filters for different moments during the collection time, and considering $D = 0$ and other initial conditions, it can be possible the determination of the ^{222}Rn and ^{220}Rn concentration from air.

The variety concentrations of Rn isotopes from atmosphere depend on year periods, height from the ground soil, the meteorological conditions when the aspiration is made. ^{222}Rn and ^{220}Rn movement in the atmosphere after their exhalation from soil is mainly determinated by the turbulent diffusion and is limited only by the radioactive decay. The activity concentration at a certain height

can be changed by a factor up to 100, according to different conditions of vertical mixing, assuming a constant rate of radon isotopes exhalation. ^{222}Rn and ^{220}Rn concentrations decrease with altitude. Because of the short half-life, ^{220}Rn concentration is very small at height greater than 1m from ground soil and atmospheric conditions with low turbulent on vertical. The decay products of ^{222}Rn with sort-lived are in equilibrium with the ^{222}Rn , excepting the surface soil area. The height of the mixing layer depends on the turbulent mixing conditions in the surface layer of soil. For low turbulences, the difference between the ^{222}Rn decay products concentration and ^{222}Rn is relatively high at ground level, but rapidly decreases with height increasing. Along with increased turbulences, the deviation from the radioactive equilibrium with ^{222}Rn is small at ground level and increases at higher altitudes. The activity concentrations of ^{220}Rn decay products (^{212}Pb and ^{212}Bi) are approximately constant in the boundary layer.

7. CONCLUSIONS

The principal characteristics of ^{222}Rn and ^{220}Rn and of their decay products were reviewed and analyzes on this paper. They are very important to follow in the radioactivity environmental evaluation program because of their relatively high contribution of radiation dose due to inhalation, being an important task in the radiological and/or nuclear emergency situation. The ^{222}Rn decay products with half-life can be uniformly distributed in the body being able to estimate the dose on each organ; on the other hand, the ^{220}Rn decay products can be embedded in certain organs, in which the products can specifically interact, resulting in irradiation of these organs only. Thus, it is more difficult to estimate radiation in the body due to ^{220}Rn decay products.

Acknowledgements. This work was supported by the European Regional Development Funds and co-financed by the Government of Romania - Ministry of Regional Development and Public Administration, in the framework of project "EMERSYS Toward an integrated, joint cross-border detection system and harmonized rapid responses procedures to chemical, biological, radiological and nuclear emergencies", MIS-ETC code 774.

REFERENCES

1. A.B. Tanner, *Radon migration in the ground: A review*, In: The natural radiation environment (ed. by J.A.S. Adams, W.M. Lowder), University of Chicago Press 161–190 (1964).
2. A.B. Tanner, *Radon migration in the ground: A supplementary review*, In: Proceedings natural radiation environment III (ed. by T.F. Gesell, W.M. Lowder), Technical Information Center, US Department of Energy, Washington D.C., CONF-780422, 1 5–56 (1980).
3. P.M.C. Baretto, *^{222}Rn emanation characteristics of rocks and minerals*, in "Radon in uranium mining", IAEA panel proceedings, Wien 129–148 (1975).

4. V.L. Shashkin, M.I. Prutkina, *On the mechanisms of emanation of radioactive minerals and ores*, *Atomnaya Energiya* **29** 41 (1970).
5. K. Megumi, T. Mamuro, *Emanation and exhalation of radon and thoron gases from soil particles*, *Journal of Geophysical Research* **79** 3357–3360 (1974).
6. K.P. Strong, D.M. Levins, *Effect of moisture content on radon emanation from uranium ore and tailings*, *Health Physics* **42** 27–32 (1982).
7. J.G. Ingersall, *A survey of radionuclide constants and radon emanation rates in building materials used in the U.S.*, *Health Physics* **45** 363–368 (1983).
8. J. Porstendörfer, T.T. Mercer, *Influence of electric charge and humidity upon the diffusion coefficient of radon decay products*, *Health Physics* **37** (2) 191–199 (1979).
9. O.G. Raabe, *Measurement of the diffusion coefficients of RaA*, *Nature* **217**, 1143–1146 (1968).
10. J.W. Thomas, P.C. Leclare, *A study of the two-filter method for radon-222*, *Health Physics* **18** (2) 113–122 (1970).
11. S.K. Friedlander, H.F. Johnstone, *Deposition of suspended particles from turbulent gas streams*, *Industrial and Engineering Chemistry* **49** 1151–1156 (1957).
12. A.C. Chamberlain, *Aspects of travel and deposition of aerosols and vapour clouds*, Atomic Energy Research Establishment, Harwell, England, HP/R 1261, 1955.
13. W. Jacobi, K. André, *The vertical distribution of radon-222 and radon-220 and their decay products in the atmosphere*, *Journal of Geophysical Research* **68** 3799–3814 (1963).
14. O. Sima, *Natural radioactivity of aerosols*, In: Proceedings of the annual scientific conference of the Institute for Meteorology and Hydrology, Part I (in Romanian), Bucharest, 361–380 (1978).
15. C. Dovlete, *Determination of the gamma emitting daughters of the atmospheric ^{222}Rn and ^{220}Rn* , (in Romanian) *Studii si Cercetari de Fizica* **35**(4) 333–338 (1983).
16. S. Sonoc, *On the determination of ^{218}Po concentration in aerosol samples by gamma spectrometry*, (in Romanian) *Studii si Cercetari de Fizica* **41**(9) 851–857 (1989).
17. S. Sonoc, O. Sima, *Optimal method for environmental radon and thoron daughters determination by alpha spectrometry*, *Radiation Protection Dosimetry* **45**(1/4) 51–52 (1992).
18. O. Sima, *Comprehensive software for the assessment of ^{222}Rn and ^{220}Rn decay products based on air sampling measurements*, *Applied Radiation and Isotopes* **67**(5) 867–871 (2009).