Radiation and physical parameters of kimberlites and host rocks of the Lomonosov diamond deposit (Russian North-West)

E U Yakovlev, A V Puchkov

N. Laverov Federal Center for Integrated Arctic Research of the Ural Branch of the Russian Academy of Sciences, 109, Severnoi Dviny Emb., Arkhangelsk, 163000, Russia

E-mail: Evgeny.yakovlev@fciarctic.ru

Abstract. In this paper, experimental studies of rocks were carried out to assess the main radiation and physical factors affecting the formation of the radon field over the kimberlite pipes of the Arkhangelsk diamondiferous province (for example, the Lomonosov diamond deposit). For various types of rocks, the following was studied: porosity, density, radium-226 activity, free radon activity, radon production rate, emanation coefficient. The research results showed that the main factors in the formation of the radon field are porosity and the coefficient of emanation. These parameters directly affect the amount of free radon. In addition, we calculated the radon production by various types of rocks, represented by vent kimberlites, tuffaceous-sedimentary rocks of the crater, enclosing and overlying sediments. Regression analysis showed that the radon production rate in rocks reflects the formation of a radon field under conditions of occurrence of a kimberlite body. The studied radiation and physical parameters showed that the greatest amount of radon in a free state is produced by rocks of the near-pipe space, represented by the enclosing Vendian V2 deposits and characterized by high values of the emanation coefficient, specific activity of radium, the radon production rate, and porosity. At the same time, the formation of free radon in the tube body is limited due to the low values of porosity, emanation coefficient, and the radon production rate.

1. Introduction

The purpose of this paper was to study the radiation and physical parameters of kimberlites overlying and enclosing rocks of the Zolotitskiy kimberlite field of the Arkhangelsk diamondiferous province. Measurements and calculations of the following parameters were carried out: activity of radium-226, radon-222 in a free state, emanation coefficient, radon production rate, average and true density, porosity. The assessment of the main factors causing the formation of increased activity of radon over the tubes has been carried out.

2. Materials and methods

The sampling of rocks was carried out in the quarries of the kimberlite pipes of the diamond deposit named after M.V. Lomonosov. We took 30 rock samples weighing up to 2 kg each, represented by different types of sediments: overlying Quaternary (Q) and Carboniferous rocks (C2), enclosing Vendian rocks (V2), tuffaceous sedimentary rocks of the crater (iD3-C2) and zerlovoy kimberlites facies (iD3-C2). After delivery to the laboratory, rock samples were dried in a BINDER E28 drying oven at a...
temperature of 105 °C to an air dry state. The determination of the radiation and physical parameters of rocks was carried out in the laboratory of environmental radiology. The samples were processed for 226Ra and 222Rn analysis at the laboratory of ecological radiology, which complies with the accreditation criteria for testing laboratories established in ISO/IEC 17025.

3. Gamma spectrometry and radiometric (emanation) measurements
To determine radionuclide radium-226 in rocks, we used a semiconductor gamma-spectrometric complex with nitrogen cooling ORTEC with a GEM 10 P4-70 detector (Ametek Ortec, USA) completed with lead shielding. The resolution of the gamma spectrometer along the 1.33 MeV (60Co) line is 1.75 keV, the relative efficiency is 15%. Measurement geometry is a 1-liter Marinelli vessel (counting sample). The activity of the radium-226 radionuclide, taking into account the accumulation, was determined from the radionuclide lead-214 (351.93 keV with a quantum yield of 35.60%) and bismuth-214 (609.32 keV with a quantum yield of 45.49%, 1120.29 keV with a quantum yield of 14.92%, 1764.49 keV – the yield is 15.3%).

The determination of the volumetric activity of radon was carried out using a measuring complex for monitoring radon, thoron and their daughter products "Alfarad plus" (SPC DOZA, Russia). Measurement geometry is a 5-liter plastic cylinder (counting sample).

The radon emanation coefficient was determined by the gamma spectrometric method according to the following formula (1):

$$K_{Rn} = \left(1 - \frac{A_{Ra226\text{(non-equilibrium)}}}{A_{Ra226\text{(equilibrium)}}}\right) \times 100,$$

where: $A_{Ra226\text{(non-equilibrium)}}$ is the specific activity of 226Ra (in a non-equilibrium state), determined as the average value of the results of the first and last measurements (unsealed), Bq∙kg$^{-1}$; $A_{Ra226\text{(equilibrium)}}$ - specific activity of 226Ra (in equilibrium state), determined as the average value of the results of the last 5 measurements in a sealed state, Bq∙kg$^{-1}$.

We also calculated the radon production rate per unit volume, $P$ (Bq∙m$^{-3}$∙h$^{-1}$), using the following formula (2) [1]:

$$P = \lambda K_{Rn} A_{Ra226} \rho_b,$$

where: $\lambda$ is decay constant for radon (2.1·10$^{-6}$ s$^{-1}$); $\rho_b$ is the bulk density, kg·m$^{-3}$.

4. Assessment of density and porosity parameters
The determination of the parameters of the average (bulk) and true density of the selected samples was carried out according to the methods below, based, among other things, on the works [1-2]. In this work, the average (bulk) density is used as a physical quantity determined by the ratio of the mass of the material to the entire volume occupied by it, including pores and voids. In this case, the concept of true density in the calculation does not include the presence of pores and voids.

The porosity of rock samples, $V_{pore}$, %, was determined by calculation based on the preset values of the true and average density according to the formula (3):

$$V_{pore} = \left(1 - \frac{\rho_b}{\rho}\right) \times 100,$$

where: $\rho_b$ is the average density of the rock, g·cm$^{-3}$; $\rho$ is the true density of the rock, g·cm$^{-3}$.

5. Results
In general, the research results are presented in Table 1, in which the following designations are used: $A_{Ra226}$ — specific activity of radium-226 under sealing conditions; $K_{emanation}$ - coefficient of emanation; AVdens - average density; TRdens - true density; Porosity - porosity. Also, these designations are used further in the text of the paper.
Based on the results of measurements of the above parameters, we calculated the radon production rate from different types of rocks. This parameter is one of the most important for the quantitative assessment of radon in its free state [1, 3]. The calculation results are shown in Table 2.

Table 1. Radiation and physical characteristics of samples

| Radiation and physical characteristics, range / average | Overlapping rocks | Kimberlites | Host rocks | Tuffaceous sedimentary rocks |
|--------------------------------------------------------|-------------------|-------------|------------|-------------------------------|
| $\text{Ra}_{226}$                                    | 15.88 - 30.21     | 12.42 - 31.46 | 16.05 - 63.32 | 11.45 - 48.4                 |
| $\text{K}_{\text{emanation}}$                         | 11.09 - 24.91     | 1.76 - 10.67  | 6.19 - 29.13  | 9.82 - 34.13                 |
| $\text{AV}_{\text{dens}}$                            | 1.83 - 2.03       | 1.74 - 2.35   | 1.47 - 2.19   | 1.36 - 1.90                  |
| $\text{TR}_{\text{dens}}$                            | 2.38 - 2.94       | 1.83 - 2.37   | 2.06 - 2.7    | 2.12 - 2.54                  |
| Porosity                                              | 18.47 - 32.65     | 0.46 - 4.92   | 4.72 - 40.96  | 13.21 - 36.74                |

Table 2. The calculation results of the radon production rate

| Radon production rate, Bq*m$^3$*h$^{-1}$, range/mean | Overlapping rocks | Kimberlites | Host rocks | Tuffaceous sedimentary rocks |
|------------------------------------------------------|-------------------|-------------|------------|-------------------------------|
|                                                     | 38.88 - 59.12     | 9.84 - 18.96 | 37.19 - 132.03 | 33.97 - 139.57               |

We have identified a wide range of the radon-$^{222}$ emanation coefficient in the studied rock samples (in the range from 3 to 40%). The lowest values of the emanation coefficient in the range from 1 to 8%, porosity from 0.46 to 4.91%, radium-$^{226}$ activity from 12.42 to 15.89 Bq∙kg$^{-1}$ and the radon production rate are characteristic of kimberlites of the vent facies of pipes, represented by autolith breccia. These results are confirmed by previous studies [4-8].

Tuffaceous-sedimentary rocks are distinguished by a wider range of radiation and physical parameters. The specific activity of radium-$^{226}$ in these rocks varies from 11.45 to 48.40 Bq∙kg$^{-1}$, the emanation coefficient is in the range from 9.82 to 34.13%, and the porosity is in the range from 13.20 to 36.74%. However, a somewhat different picture is observed in the distribution of the studied parameters in the host Vendian and overlying Quaternary and Carboniferous rocks. Most of these rocks are characterized by significant levels of porosity (up to 41%). The results of calculating the porosity are in good agreement with the data given in [4] on the study of the host rocks of the Arkhangelsk diamondiferous province (from 8 to 40%). The host rocks are distinguished by a high level of specific activity of radium-$^{226}$ - up to 63.32 Bq∙kg$^{-1}$, and the overlying rocks with a high level of emanation coefficient - up to 28%. Considering the high values of the specific activity of radium, the emanation coefficient, the radon production rate and porosity, the enclosing rocks of the near-pipe space stand out among the other types of rocks, which is associated with their structural and geological features.

To study the features of radon emanation in rocks, correlation and regression analyzes of the main radiation and physical parameters of the samples were carried out. A significant correlation is observed in rocks for the emanation coefficient and activity of radon in a free state ($r = 0.709$), the coefficient of emanation and porosity ($r = 0.753$), volumetric activity and porosity ($r = 0.691$).
The specific activity of radium-226 in rock samples has no significant correlations with any of the parameters. This fact suggests that the parameter of the content of radium-226 in rocks is not decisive for the formation of a radon field. The lack of relationships between the content of radium-226 and the volumetric activity of radon is probably associated with the form of the presence of radium-226 in the crystal lattice of the minerals that make up the rocks [9-12].

Based on the results of measuring the volumetric activity of radon in a sealed container and calculating the radon production rate, we built a regression model, which is a linear function of the dependence of two parameters (dependent variable and regressor) and is characterized by regression coefficients (slope, coefficient of determination).

Figure 1. Regression model of the relationship between the volumetric activity of radon and the radon production rate.

The coefficient of determination shows that the change in the volumetric activity of radon (dependent variable) by 87.9% is described by the independent variable (regressor) - the radon production rate, which indicates a sufficient justification for the choice of this model. This model more clearly predicts the distribution of radon based on the results of calculating the radon production rate.

6. Conclusion
The results of this work showed that to characterize the territory according to the distribution of radon, it is advisable to use a complex of two parameters - the activity of radium-226 in soils and rocks, their emanation coefficient and porosity, which in turn determines the radon production rate. Based on the results of the study of radiation and physical parameters, it was revealed that the main source of radon observed in the soil air above the kimberlite pipes is the enclosing Vendian rocks of the near-pipe space. This conclusion confirms the applicability of emanation methods for prospecting kimberlite pipes in the territory of the Arkhangelsk diamondiferous province.

Acknowledgments
The reported study was funded by RFBR according to research project No. 20-35-70060 «Investigations of the conditions for increased radon emanation in the sedimentary cover of areas of kimberlite magmatism (by the example the Arkhangelsk diamondiferous province)».

References
[1] Pereira A, Lamas R, Miranda M, Domingos F, Neves L, Ferreira N, Costa L 2017 J. Environ. Radioact. 166 270–277
[2] Soniya S R, Abraham S, Khandaker M U 2021 Radiation Physics and Chemistry 178
[3] Ye Y, Wu W, Feng S, Huang C 2018 Building and Environment 144 66-71
[4] Zinchuk N N, Zinchuk M N 2014 *Collection of scientific works of UkrGRRI* 2 97-108
[5] Burmistrov A, Boguslavskiy M 2009 *Moscow University Bulletin* 6 26-33
[6] Burmistrov A, Garanin K, Starostin V, Yuzhakov L 2005 *Geology of diamonds - present and future* pp 762-772
[7] Arbuzov S, Rihvanov L 2009 *Geochemistry of radioactive elements* pp 315
[8] Kiselev G P, Danilov K B, Yakovlev E Yu, Druzhinin S V 2016 *Bulletin of the Kamchatka Regional Association «Educational and Scientific Center»* 30 (2) 43-53
[9] Chauhan R P, Chakarvarti S K 2002 *Radiat. Meas.* 35 143-146
[10] Chauhan R P, Nain M, Kant K 2008 *Radiat. Meas.* 43 445-448
[11] Eakin M, Brownlee S J, Baskaran M, Barbero L 2016 *Geochem. Cosmochim Acta* 184 212-226
[12] Thu H N P, Thang N V, Hao L C 2020 *J. Environ. Radioact.* 2016