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Comparative studies on radon seasonal variations in various undeground environments: Cases of abandoned Beshtaugorskiy uranium mine and Kungur Ice Cave

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ABSTRACT

It is well known that one of the most important risk factors in underground environment is the harmful effects of radon. The reasons for strong seasonal fluctuations in radon content in underground environments remain not fully understood. The purpose of this article is to improve existing ideas about this phenomenon. The article presents the results of a study of radon transport in two different underground spaces – the Beshtaugorskiy uranium mine (North Caucasus) and the Kungur Ice Cave (Middle Ural). We have used the direct measurements of the equilibrium equivalent concentration (EEC) of radon progeny in air, as well as the air flow velocity. A very wide range and strong seasonal variations in the radon levels have been recorded in both cases. The EEC has a range of 11–6653 by Bq m^{-3} and 10–89,020 Bq m^{-3} in the Kungur cave and the Beshtaugorskiy mine, respectively. It has been established that seasonal fluctuations in radon levels both in the mine and in the cave are caused by the same process – convective air circulation in the underground space due to the temperature difference between the mountain massif and the atmosphere (so called chimney effect). Overall, these results indicate that due to convective air circulation, underground spaces are periodically intensively ventilated with atmospheric air, and then, on the contrary, they are filled with radon-enriched air that seeps into caves or adits from rocks and ores. In both cases, the EEC of radon progeny exceeds the permissible level for the population and workers. The results of this study highlight the need for the development of measures to limit the presence of people in the surveyed underground spaces.

1. Introduction

The amazing natural environment of the caves, bizarre labyrinths and grottoes like the Hall of the Mountain King, underground lakes and rivers, unique rock paintings, and archaeological finds have always attracted scientists, tourists, and just adventurers. Recently, interest has also been growing in artificial underground structures created not by fabulous gnomes and trolls, but by mere mortals. Some of the former coal, silver, and other mines are becoming museums. In recent decades, due to the development of tourism around the world, more and more natural caves and artificial underground spaces are becoming available for public visits, and the number of visitors of existing underground tourist sites increases. In this regard, the question arises about the safety of tourists and specialists working in the underground space. And one of the most important risk factors is the harmful effects of radon, the radioactive noble gas (^{222}Rn), contained in the air of the underground environment.

Radon is a decay product of radium (^{226}Ra) in the natural chain of

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uranium (^{238}U), it is exhaled from the rock or soil and accumulated in these poor ventilated sites at subsurface (Alvarez-Gallego et al., 2015). Radon and its short-lived decay products make up a health hazard due to their carcinogenic effects (Zeeb and Shannoun, 2009). Epidemiological studies prove a significant increase in the risk of lung cancer for elevated indoor ^{222}Rn concentrations (Darby et al., 2005; Gaskin et al., 2018). In accordance with the ICRP recommendations radon activity concentration should not exceed 300 Bq m^{-3} or 1000 Bq m^{-3} in dwellings or workplaces respectively (ICRP, 2014). For underground facilities, the international standards recommend the limit of 1.5 kBq m^{-3} (IAEA, 2003; ICRP, 1993). The new dose conversion factors including for mines and tourists caves were recommended in the newest ICRP item (ICRP, 2017).

However, the actual values of radon concentration in caves and mines in many cases significantly exceed recommended limits. Numerous studies have found that radon concentrations in underground spaces fluctuate in a very wide range. In most natural caves, radon concentrations range from 0.5 to 30 kBq m^{-3} (Hakl et al., 1997; Cigna, 2003; Font et al., 2008; Briestenský et al., 2022; Ambrosino et al., 2019; Sainz et al., 2018; Perrier and Richon, 2010; Somlai et al., 2011). However, in some cases, radon levels can reach significantly higher values, e.g. 30–50 kBq m^{-3} in the Castanar cave in Spain (Alvarez-Gallego et al., 2015), 88 kBq m^{-3} in the Petralona cave in Greece (Papastefanou et al., 2003), 123 kBq m^{-3} in Shawan Cave in southwest China (Wang et al., 2019), or 155 kBq m^{-3} equilibrium equivalent radon concentration at Giants Hole in Derbyshire in England (Gunn et al., 1991). The radon concentration in caves depends on many parameters, including the radium content in rocks, the emanation coefficient, rock fracturing, ventilation regime, outside–inside temperature differences, soil moisture, the amount of precipitation and its infiltration to the cave environment, etc. (Gregorič et al., 2013; Smetanová et al., 2020). In artificial underground structures radon concentrations additionally depend on the type of ores mined and the presence of forced ventilation. Forced ventilation in non-uranium mines can reduce radon concentrations to 0.05–0.1 kBq m^{-3} (Font et al., 2008), while in unventilated parts of abandoned uranium mines, radon levels can reach 500 kBq m^{-3} (Fijałkowska–Lichwa, 2016). It is important to note that in mines that are currently open as museums, the radon levels can be reduced during working hours by means of forced ventilation. While in natural caves, forced ventilation is most often unacceptable due to the need to observe the natural heat and humidity regime in order to preserve natural and art monuments (stalactites, stalagmites, unique crystals, ice, rock paintings, etc.).

In underground spaces, significant fluctuations in radon concentration are observed throughout the year. Temporary fluctuations in the radon activity concentration occur in general due to variations of the difference in outdoor and indoor air temperatures, atmospheric pressure, as well as associated changes in the speed and direction of the airflow in the underground space (Ambrosino et al., 2019).

A. Pflitsch et al. (2010) distinguish (theoretically) two types of caves characterized by different types of air exchange and temperature regime. These are the so-called thermal and barometric caves. Barometric caves have either one entrance (dead-end caves) or have several entrances located at the same elevation. In such caves, the ventilation is mainly due to fluctuations in atmospheric pressure. An increase in atmospheric pressure leads to the entry of atmospheric air into the cave, and vice versa, a drop in atmospheric pressure leads to the flow of cave air outside. The effect is especially evident in large caves with small cross-section of entrances. These fluctuations do not have a clearly defined seasonal rhythm. Thermal caves have several entrances located at different elevations above sea level. The air exchange in such cases is caused by natural convection due to the difference between the inside and outside temperature (chimney effect), as a result of which the direction of airflows clearly depends on the season. In cold weather, the cave air is warmer than atmospheric. The air moves upwards and is discharged through the higher entrances, while cold atmospheric air is drawn into the cave in the lower entrances. During the warm period, cold cave air moves down and is discharged in the lower entrances, while warm atmospheric air is drawn into the upper entrances (Pflitsch et al., 2010).

Seasonal fluctuations in radon levels correlated with air temperature were registered in various underground spaces including caves, adits, tunnels etc. (Barbosa et al., 2010; Alvarez-Gallego et al., 2015; Tchorz-Trzeciakiewicz and Parkitny, 2015; Fijałkowska-Lichwa, 2014, 2020; Kleinschmidt et al., 2018; Pla et al., 2020; Ambrosino et al., 2020; Zafrir et al., 2020). Most often, the highest radon concentrations are observed during the summer, and the lowest in winter. For example, such fluctuations were found in six dead-end caves across Europe in a study by Briestenský et al. (2022). It is noteworthy that in all caves, very similar radon behavior has been identified (mean correlation factor R of 0.73 ± 0.06). At the same time, in the Castanar cave (the northwest of Spain), the opposite pattern of radon fluctuations was found with maximum values in late autumn – winter and minimum values in summer (Alvarez-Gallego et al., 2015). It is generally considered that low radon concentrations are observed in caves during intense ventilation time and high concentrations are usually associated with the radon accumulation in underground space for periods with low air exchange.

This paper presents the results of a periodic measurement of the air radon levels and the airflow velocity at two underground spaces, which differ in genesis, geometry, geological and climatic conditions. One of them is the natural Kungur Ice Cave, located in the Perm Region, the Middle Urals, and the other is the abandoned Beshtaugorskiy uranium mine in the Stavropol Territory, the North Caucasus. The Kungur Ice Cave is one of the few excursion sites in Russia where large-scale excursion events are held. Issues of ensuring the safety of people during work and excursions are crucial when exploring the cave. The abandoned Beshtaugorskiy uranium mine, located in the resort area of the Caucasian Mineral Waters, is the most promising underground structure from the point of view of the organization of an underground museum and radon therapeutic medical facility. The existing developed resort infrastructure and a large number of tourists visiting the resort contribute to this. The above conditions the choice of these underground spaces for research. The Russian legislation regulates the equivalent equilibrium concentration (EEC) of radon in the air, which should not exceed 310 Bq m^{-3} in workplaces or 1200 Bq m^{-3} in uranium mines. In this regard, EEC measurements were carried out in all cases. The radon activity concentration in the air was measured only at the Beshtaugorskiy mine.

The purpose of this work is to compare the seasonal radon variations and the changes in airflow in the natural cave and the abandoned mine. This will improve the understanding of the radon transport and air exchange patterns in the underground space. The results of this study can be used to estimate the radiation dose to personnel in the caves above, underground mountain museums and tunnels for radon therapy and to account for the seasonal patterns of radon transport.

2. Materials and methods

2.1. Geological setting and measurements in the area of the Beshtaugorskiy uranium mine

The abandoned Beshtaugorskiy uranium mine is located in the North Caucasus, Stavropol region, Caucasian Mineral Waters Resort near Pyatigorsk. A characteristic feature of the region is a group of separate mountains, the central and largest of them is Mt. Beshtau (1401 m a.s.l.), which is a complex multiphase intrusion of alkaline granite porphyries, trachytes, and liparites that are called in general as "beshtaunites". Uranium ore veins are confined to the northwestern system of faults which dissects magmatic massif (Mashkovtsev et al., 2010). The magmatic massif is adjoined by an elevated plain consisting of sedimentary deposits — limestone, marls, sandstones, and clays (Fig. 1).

The area is characterized by increased natural radioactivity not only

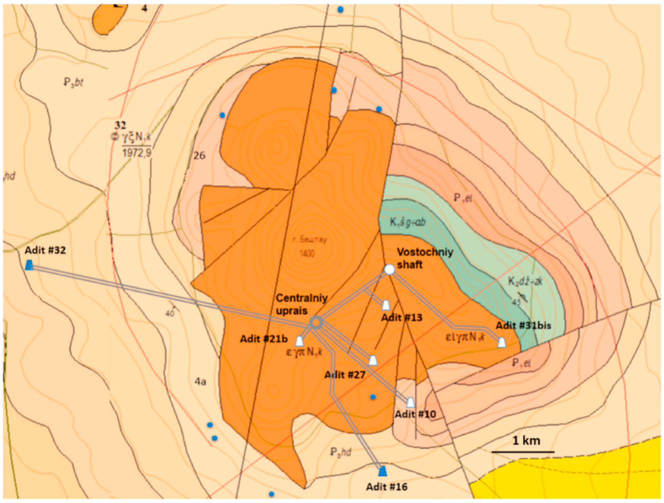

Fig. 1. Geological map of the Beshtau area with the schematic plan of the Beshtaugorskiy uranium mine. *Legend*: $K_1sg\text{-}ab$ – Cretaceous sandstone, $K_2\ dz\text{-}zk$ – Cretaceous limestone, $₽_3el$, $₽_3hd$ – Palaeogene marl, $₽_3bt$ – Palaeogene clays, $\varepsilon\gamma\pi N_1k$ – Neogene magmatic rocks.

due to the presence of uranium ores, but also due to the increased content of naturally occurring radionuclides in igneous rocks and their loose weathering products. The content of ^{226}Ra in beshtaunites varies in range between 181 and 280 Bq kg^{-1} that are correspond to the upper range of values characteristic of acidic igneous rocks (International Atomic Energy Agency, 2014). The values of the radon exhalation rate from surface at the Mt. Beshtau are 130–3000 mBq m^{-2} s^{-1} (Miklyaev et al., 2022). The region is one of the most radon hazardous territories in Russia (Lezhnin et al., 2011).

Starting in the late 1940s until the early 1980s, the uranium deposit was developed in the Beshtau. The uranium-producing mine was located in the southern parts of the mountain and consisted of 12 horizons, with the adits' mouths situated at elevations between 720 and 1100 m (Fig. 2). The levels were connected with two vertical shafts called Vostochniy (Western) and Centralniy (Central). The Vostochniy shaft was primarily purposed for ventilation and comes out to the surface level at an elevation of 1001 m. The Centralniy shaft was used as a transport one and does not come out to the surface.

From the 1990s until nowadays, the territory was rehabilitated in a number of phases. The entrances to most of the adits were blocked (either blasted or filled with cement plugs). The rock dumps were terraced and coated with protective clay shielding. Two adits at the lower level (#16 and #32) have not been decommissioned or blocked, and are currently used for technical purposes, including subterranean radon water exploitation and monitoring the radiation levels (Karpenko et al., 2009). However, the access to these adits is now restricted. At the same time, the entrances to some adits were not completely sealed. At the mouths of some adits, relatively small-diameter (Ø 0.8–1.5 m) holes, perhaps of suffusive origin, were discovered. These holes are usually located above the destroyed and buried adit entrances. Fig. 3 shows a typical hole. We found suffosion holes at the mouths of adits #10, 13, 21b, 27, 31bis and the Vostochniy shaft. Although it is still possible that there are more similar holes on the slopes of Beshtau. These openings are currently the main channels of interaction between the mine space and the atmosphere.

Currently, Mt. Beshtau, including the territory of the former mine, is part of the State conservation reserve of a regional significance and is called the Beshtaugorskiy forest park. The locals of the surrounding settlements and the vacationers of the spa resorts like to hike and have picnics there. The nearby Mashuk Aqua-Term sanatorium complex plans to build Russia's first underground radon and climate therapy center in one of the adits of Beshtaugorskiy mine.

Periodic measurement of radon levels and environmental parameters were carried out at the mouths of adits # 10, 13, 21b, 27, 31bis and the Vostochniy shaft from March to September 2020. The measurement has been carried out at the holes of suffusion and human-made origin which have a pneumatic connection with the mine space. The measurement has been performed directly in the airflow blowing through the suffusion holes at 0.5 m above the holes. The following parameters have been measured at the mouths of the adits:

- Equilibrium equivalent concentration of radon progeny (*EEC*);
- Radon activity concentration in ambient air (C_{air});
- Ambient dose equivalent rate ($^*H\gamma$);
- Airflow direction and velocity (V_a).

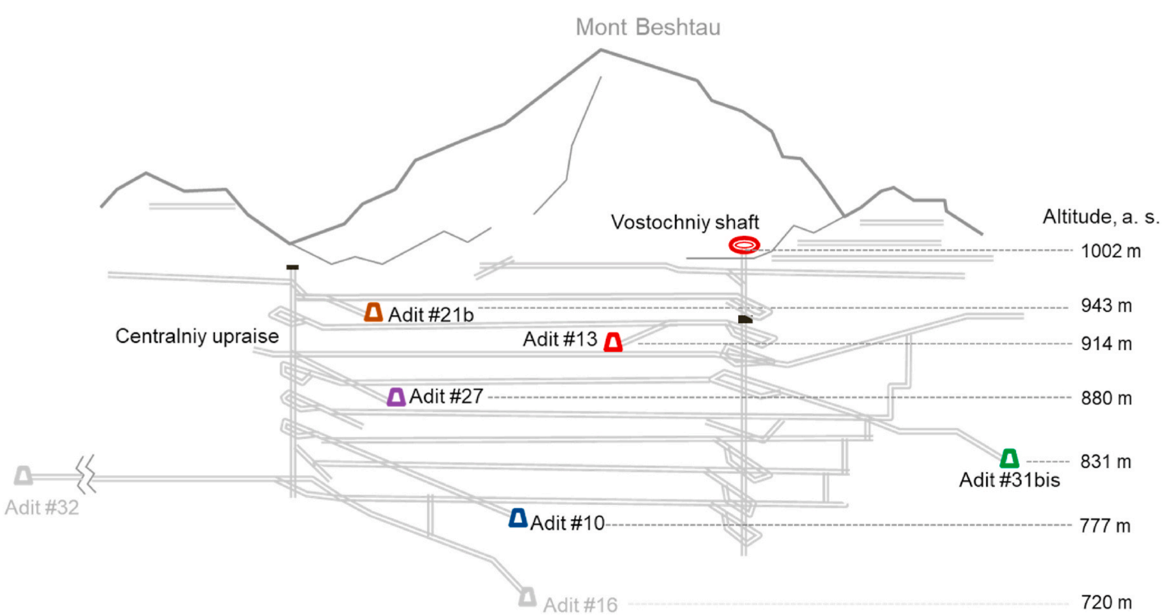

Fig. 2. Scheme of former Beshtaugorskiy uranium mine. *Legend*: The colored trapezoids show the adits with open holes where the radon measurement was carried out.

Fig. 3. A hole of likely suffusive origin above the mouth of adit #10.

The equilibrium equivalent concentration of radon progeny(EEC) and the radon activity concentration in ambient air has been measured at 0.5 m height from the ground using a Radon Aerosol Alpha Radiometer RAA-3-01"AlphaAERO", developed in Scientific and Technical center "Amplituda" (Russia). The estimation of EEC in the air is based on the alpha-spectrometry measurement of the activity of ^{218}Po and ^{214}Po deposited on a spectrometric aerosol filter through which air is pumped at a constant rate. A semiconductor spectrometric detector is used. The measurement and air pumping through the filter are carried out simultaneously. The measurement (pumping) time is at least 10 min. After each measurement, the filter is replaced with a new one, and the detector is checked by using the reference source of alpha radiation included in the kit. In addition to EEC, the radiometer also evaluates the air radon concentration and the equilibrium factor F by the ratio of polonium isotope activities. Despite the fact that this is a fairly rough estimate, the comparative measurement of air radon concentration using "AlphaAERO" and the charcoal method shows good convergence of the results. This allows us to use "AlphaAERO" to measure the air radon concentration. The measuring range of the EEC is 1–10^6 Bq m^{-3}; the measurement uncertainty does not exceed 30%. The radiometer "AlphaAERO" has successfully passed the metrological and climatic tests in Russia (Tsapalov and Kovler, 2018).

The airflow velocity has been determined using professional anemometers ADA AeroTemp and Testo 410–2. Airflow velocity measurement range is 0.4–20 m s^{-1}; error is $\pm$0.2 m s^{-1}; resolution is 0.1 m s^{-1}.

The ambient dose equivalent rate has been determined using the portable dosimeter DKG-07D "Drozd" based on Geiger-Muller detector (SPC DOZA, Russia). This instrument can detect the ambient dose equivalent rate from 10^{-1} up to 10^3 μSv h^{-1} in a wide energy range from 0.05 to 3.0 MeV. The error of measurement does not exceed 15% (2σ). The measurement has been carried out at the mouth of adits on the 0.5 m height from the ground.

The meteorological data have been taken from the nearest Mineralnye Vody weather station located 14 km northeast of the measurement site. A comparison of meteorological parameters (air temperature and humidity) measured at the site and received from the weather station shows acceptable agreement. The small differences are due to differences in the altitude and slope exposure.

All used equipment has passed mandatory periodic verification and/ or participated successfully in interlaboratory comparisons.

2.2. Geological setting and measurements in the area of the Kungur Ice Cave

The Kungur Ice Cave is located on the northeastern outskirts of Kungur, Perm Region, on the right bank of the Sylva River (Mavlyudov and Kadebskaya, 2018). This cave has been known for more than 300 years and regularly visited by tourists for about 100 years. The entire cave is located within the southern slope of the Ice Mountain massif, which is a platform karst upland with absolute heights up to 200 m and is part of the denudation plain of the Middle Urals (Krasikov et al., 2022).The cave was formed in a sulfate-carbonate complex of lower Permian karst rocks. It is a one-level dead-end maze consisting of grottoes and narrow passages connecting them. At the moment, the known length of the cave is 8153 m. The area of the studied galleries is 63.8 thousand m^2, and their volume is 223.3 m^3. With an amplitude of 36.3 m, the average height of the galleries is 2.6 m (Krasikov, 2022). The cave has three entrances: one natural and two artificial (entrance and exit tunnels 40 and 101 m long), which are located at the foot of the Ice Mountain, at elevations of 123 m, 120 and 131 m a.s.l., respectively (Kadebskaya, 2004).

The Kungur Ice Cave is a reference system of air draft due to the chimney effect (Kungur Ice Cave, 2005). In winter, streams of warm cave air rise through vertical karst cavities, inaccessible to people, and discharge at the platform karst upland surface. While cold atmospheric air enters the cave through the entrance tunnels and many fissures at the foot of the mountain. In summer, the direction of the airflow changes to the opposite: colder airflows out of channels in the Ice Mountain slope, and warmer atmospheric air is sucked from surface through karst sinkholes and cracks (Kungur Ice Cave, 2005). Constant cooling of the entrance part of the cave due to air circulation contributes to the formation of cave ice of amazing shape, attracting tourists to the cave (Fig. 4). The schematic geological section of a karst massif with direction of seasonal cave air circulation and plan of the cave are shown in Figs. 5 and 6.

The cave is one of the few excursion sites where large-scale excursion activities are conducted. Issues of ensuring the safety of people during work and excursions are crucial when exploring the cave. Currently, research in the field of assessing the radiation situation of caves is gaining rapid pace around the world, which has also affected this cave

Fig. 4. "Icy rain" in the Kungur Ice Cave.

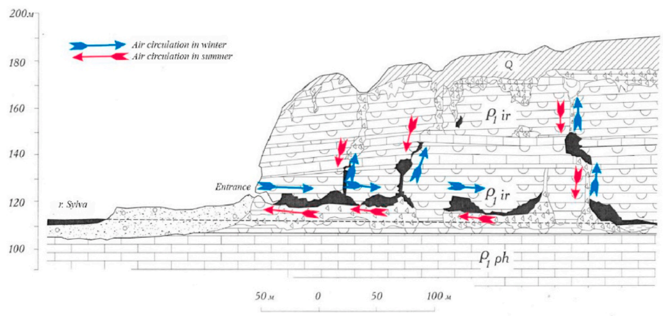

Fig. 5. Schematic geological section of a karst massif with direction of seasonal cave air circulation. *Legend*: P_1 *ph* - Permian dense non - karst limestone, P_1 *ir* - lower Permian sulfate-carbonate complex of Permian karst rocks, Q – quaternary deposits.

within the framework of safety. The first measurement of gamma dose rate in the Kungur Ice Cave were carried out in 1992 and they recorded increased dose rate 5–10 times higher than the background values on the surface (Ponosov and Kataev, 1992; Maksimovich et al., 2011). In 1999, the dose rate measurement was carried out over the entire area of the cave. The seasonal changes in the gamma dose rate taking maximum values in summer were recorded. It has been suggested that the reason for the increase in the gamma dose rate may be the daughter products of radon present in the form of aerosols (Krasikov et al., 2022). According to the results of the radon measurement in the cave for the period from 2003 to 2008, it has been confirmed, that the annual dose from the radon progeny can exceed the permissible level of 5 mSv/year. Measures were proposed to temporarily limit the presence of employees in the cave (Testov, 2003; Ponosov and Stepanov, 2003; Testov et al., 2008). In 2018–2019, radon concentrations were measured, but the studies were episodic (Blamykov, 2019). The content of radium-226 in rocks of the Kungur Ice Cave is 6–16 Bq kg^{-1} in limestone, gypsum, and anhydrites and 20–45 Bq kg^{-1} in dolomites and clays (Maksimovich et al., 2011).

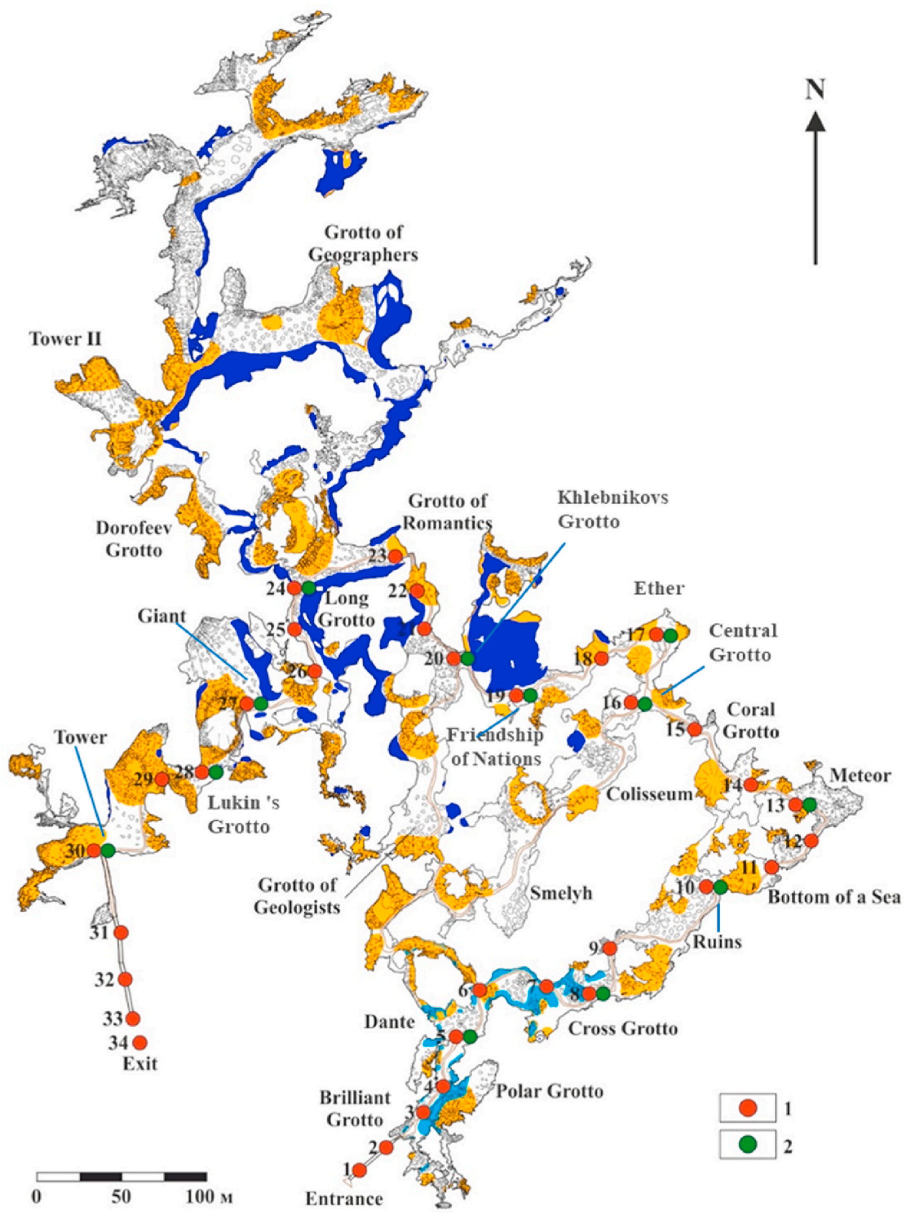

Fig. 6. Kungur Ice Cave plan with location of gamma dose rate (1) and radon progeny EEC (2) measurement points. *Legend*: Dark blue areas – lakes, light blue areas – ice, light orange areas – clay deposits. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

This article presents the results of the systematic radon measurement conducted in the Kungur Ice Cave since 2021. Monitoring is carried out using two express methods. The former is a method for "instantaneous" measuring the EEC of radon progeny, the latter is the measurement of the ambient dose equivalent rate. In addition, the airflow velocity has been measured.

To measure the ***EEC of radon progeny***, the aerosol radiometer RAA-10 has been used. The measurement of the radon progeny activity has been carried out on aerosol filters AFA-RSP-10 after pumping 45 L of air. The mode is automatic, 5-min alpha-spectrometric. The error of measurement does not exceed 30%. The device is in a stable position at the height of 1.5–1.7 m from the grotto platform of the cave. The radon measurement is performed at 12 control points in summer (July) and winter (January). Control points are located directly on excursion sites in the largest grottoes of the cave (see Fig. 6).

The airflow velocity has been determined using professional anemometers: ADA AeroTemp and Testo 417. The airflow velocity measurement range is 0.3–20 m s^{-1}; the error is ±0.1 m s^{-1}; the resolution is 0.01 m s^{-1}.

As part of radiological monitoring, the measurement ***of the ambient dose equivalent rate*** in the grottoes and passages of the Kungur Cave has been carried out three or four times a season. The measurement of the gamma dose rate is carried out at 34 control points, for which three dosimeters are used: the RADEX RD1503 dosimeter, the MKS-01CA1 dosimeter-radiometer, and the SOEKS 112 portable dosimeter. The error of measurement does not exceed 15%.

All used equipment has passed mandatory periodic verification and/or participated successfully in interlaboratory comparisons.

3. Results

3.1. The results of research at the Beshtaugorskiy uranium mine

The results of regular measurement of radon levels and airflow velocity at the adit mouths of the former Beshtaugorskiy uranium mine are presented in Table 1.

In Table 1, the following notations are taken:

Vflow – velocity of airflow at the mouth holes: (+) – direction from the mine, (–) – direction into the mine (the measurement error is ±0.2 m s^{-1});

Rn – radon activity concentration in air at the mouth holes (the uncertainty does not exceed 30%); EEC – equivalent equilibrium concentration of radon progeny in air at the mouth holes (the uncertainty does not exceed 30%);

*H(10)/dt–ambient dose equivalent rate (the uncertainty does not exceed 15%);

"–" – not measured.

The measurement" shows that significant seasonal variations in radon levels are observed at the adit mouths. It should be noted that in adits located at different elevations, the pattern of variations is different. Adit # 21b and the Vostochniy upraise located at the higher elevations show high radon concentrations (16,320–383,220 Bq m^{-3}) and EEC

Table 1
The results of regular measurement of radon levels as well as airflow direction and velocity at the adit mouths of the former Beshtaugorskiy uranium mine.

Adit #, Altitude	Parameter	05 Mar	18 Mar	5 Apr	17 Apr	30 Apr	20 May	29 May	19 Jun	18 Jul	29 Sep
Vostochniy vent upraise, 1002 m a.s.l.	Vflow, m s^{-1}	+1.5	+1.5	+0.5	–	+0.2	–	−0.4	−0.4	–	–
	Rn, Bq m^{-3}	47,602	65,591	16,320	–	1236	–	50	17	–	–
	EEC Bq m^{-3}	16,129	13,937	4112	–	92	–	10	15	–	–
	*H(10)/dt μSv h^{-1}	1.52	2.81	1.97	–	1.3	–	1.54	1.86	–	–
Adit # 21b, 943 m a.s.l.	Vflow, m s^{-1}	+0.8	+3.0	+2.5	+1.0	+2.0	−1.0	−4.4	−3.7	−5.0	−3.8
	Rn, Bq m^{-3}	147,602	173,226	383,220	178,644	274,073	233	50	26	20	35
	EEC Bq m^{-3}	15,900	46,332	47,350	60,101	33,864	83	10	23	15	28
	*H(10)/dt μSv h^{-1}	2.9	12.0	15.0	12.5	10.5	0.99	0.93	0.75	0.59	0.45
Adit # 13, 914 m a.s.l.	Vflow, m s^{-1}	−2.1	+1	–	−0.4	0	+3.3	−4.4	−3.7	−5.0	−3.8
	Rn, Bq m^{-3}	12	1620	–	79	253	2200	40	52	32	31
	EEC Bq m^{-3}	8	201	–	41	87	1600	22	20	20	28
	*H(10)/dt μSv h^{-1}	0.39	0.82	–	0.64	0.57	1.0	0.41	0.5	0.59	0.45
Adit # 27, 880 m a.s.l.	Vflow, m s^{-1}	−1.9	−5.3	−1.1		−0.4	+0.4	+0.4	+1.7	+2.1	−1.1
	Rn, Bq m^{-3}	112	52	–		386	69,000	–	98,000	112,000	184
	EEC Bq m^{-3}	50	44	–		175	36,000	–	12,210	42,500	132
	*H(10)/dt μSv h^{-1}	0.56	0.55	0.58		0.54	14.7	6.68	6.67	11.64	0.52
Adit # 31bis, 831 m a.s.l.	Vflow, m s^{-1}	–	–	−1.4	−1.0	−1.9	+1.6	+1.2	+3.6	–	−1.3
	Rn, Bq m^{-3}	–	–	67	49	13	–	18,244	180,000	–	12
	EEC Bq m^{-3}	–	–	58	18	12	–	16,390	35,600	–	11
	*H(10)/dt μSv h^{-1}	–	–	0.62	0.73	0.63	2.28	1.98	7.6	–	0.66
Adit # 10, 777 m a.s.l.	Vflow, m s^{-1}	−3.9	−6.8	−2.4	−1.5	−3.0	+4.0	+4.0	+6.1	+8.4	+4.6
	Rn, Bq m^{-3}	51	120	25	23	16	393,636	326,515	594,685	233,559	226,515
	EEC Bq m^{-3}	18	57	22	17	14	34,097	28,335	38,778	89,020	28,335
	*H(10)/dt μSv h^{-1}	0.60	0.56	0.60	0.60	0.65	7.04	5.59	10.79	18.6	5.8

(4112–60,101 Bq m^{-3}) in March and April. While during the warm period from May to September the radon concentrations and EEC are 17–50 Bq m^{-3} and 10–28 Bq m^{-3} respectively. At the lower adits # 10, 27 and 31b, on the contrary, the low radon activity concentrations (13–120 Bq m^{-3}), and EEC (14–50 Bq m^{-3}) are observed in March and April. In warm period, radon concentration and ECC vary from 18,244 to 594,685 Bq m^{-3} and 12,210 to 89,020 Bq m^{-3} respectively.

It is established, that these variations are associated with the periodic changes in airflow from the holes. At the mouths of upper adits, warm air blows from the mine to the surrounding atmosphere in winter (from February to April), and in summer (from May to September) air moves from the atmosphere into the mine. At the mouth of bottom adits, the opposite pattern is observed: in summer, relatively cold mine air blows from the mouth holes to the atmosphere, while in winter the air moves through the holes from the atmosphere to the mine space. The direction of air movement in the adit mouths reverses in spring and autumn, when there is an inversion of the ratio between the temperature in the mine and in atmospheric air. Very high radon concentrations and EEC in the open air are observed around the adits' mouths during the periods, when the mine air blows from the holes. High concentrations of radon progeny cause an increase in the values of the gamma dose rate (see Table 1).

Fig. 7 shows that the adits characterized by "winter" radon anomalies are located above the elevation of 900 ± 20 m a.s.l., and the adits characterized by "summer" anomalies are below this level. The adit mouths, located at an elevation close to 900 m (adit #13), are characterized by the absence of a clear seasonal periodicity of air movement direction, as well as relatively low values of air radon concentration.

The airflow velocity and direction and hence the air radon concentration at the mouth holes correlate with the ambient air temperature (Fig. 8). This correlation is positive for the lower horizons (Pearson correlation factor R = 0,96) and negative for the upper horizons of the mine (Pearson correlation factor R = −0,95). The temperature threshold at which the air movement in the mine stops and changes direction is +11.5 °C. This temperature corresponds to the average annual temperature in the mine.

The open-air radon concentration and the gamma dose rate in the areas adjacent to the adits mouths' during the discharge of mine air reach 6*10^{5} Bq m^{-3} and 15–19 μSv h^{-1}, respectively. This exceeds the world average (UNSCEAR, 2000) and local background values (Miklyaev et al. 2022) by dozens or hundreds of times. Extremely high values of ambient dose rate are explained, apparently, by the discharge of radioactive aerosols with the mine air flow, and their settling on the surrounding soils and plants. Typically, this should not have a significant effect on gamma radiation as the aerosols are still too dispersed. However, with such an extremely high EEC, the amount of radioactive aerosols is sufficient to cause dose rate increase. The Pearson correlation coefficient between dose rate and EEC is 0.78.

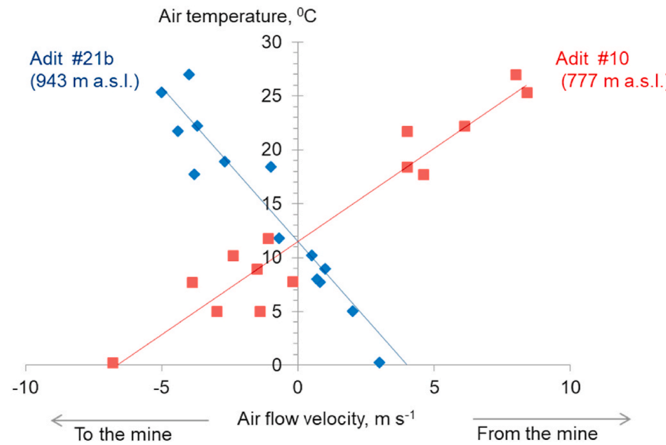

Fig. 8. The relationship between the airflow velocity at the mouth holes (adits ##10,21b) and the air temperature (temperature data taken from the Mineralnye Vody weather station located 14 km northeast of the measurement suite).

The gamma dose rate and the open-air radon concentration decrease drastically with an increasing distance from the holes and do not exceed the background values at a distance of 10–15 m. These narrowly localized, but very high values of the gamma dose rate and the air radon concentration can pose a threat to the health of people visiting the Beshtaugorskiy forest park close to the adit mouths.

3.2. The results of research on the Kungur Ice Cave

The results of the radon EEC and the gamma dose rate measurement in the Kungur Ice Cave is shown in Fig. 9. The figure clearly demonstrates the seasonal fluctuations of the radon EEC and the dose rate in the cave grottoes. In winter, extremely low EEC values (11–56 Bq m^{-3}) were recorded. At the same time, some increase in radon concentration from the entrances to the inner part of the cave is well traced. In summer, the radon EEC exceeds winter values almost by 100 times and varies from 1100 to 6653 Bq m^{-3}. The exception is the Central Grotto, where the

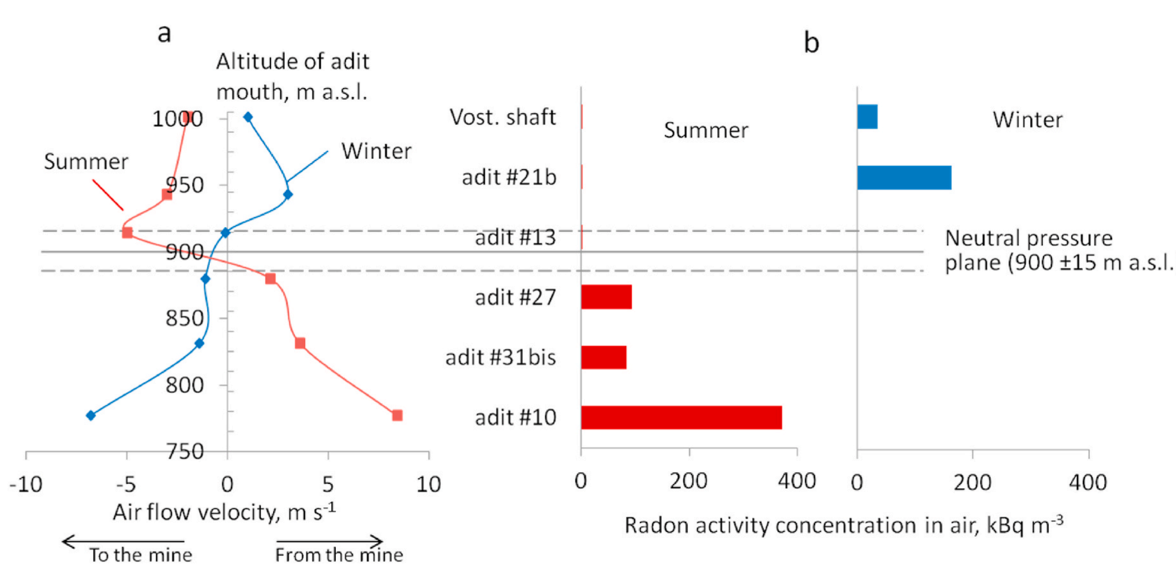

Fig. 7. The seasonal average values of the airflow velocity (a) and radon activity concentration in the air (b) at the mouth holes depending on the altitude of the adits' mouths.

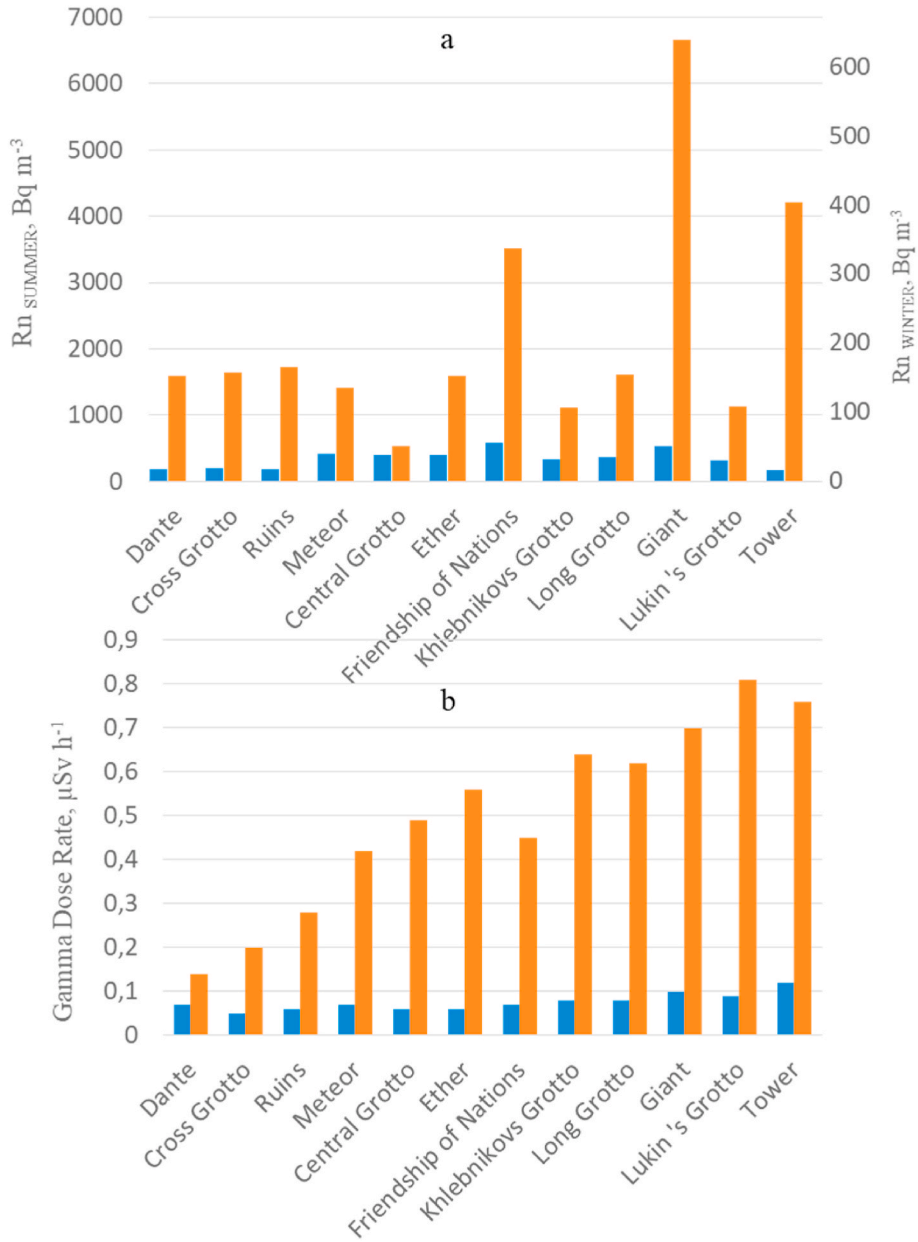

Fig. 9. The results of the radon EEC measurement in January and July 2021 (a) and the seasonal average values of the ambient dose equivalent rate (b) in the grottos of the Kungur Ice Cave. *Legend*: orange - summer, blue - winter. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

summer EEC values are 138 Bq m^{-3}. Taking into account the high summer average dose rate in this grotto (see Fig. 9b), it is most likely due to a measurement error or a random fluctuation of radon concentration at the measurement moment. The maximum values of the radon EEC during the summer period have been recorded close to the exit tunnel in the grottos Giant (6653 Bq m^{-3}) and Tower (4214 Bq m^{-3}), as well as in the central part of the cave in the Friendship of Nations grotto (3520 Bq m^{-3}).

The values of the ambient dose equivalent rate during the year range from 0.03 to 0.96 μSv h^{-1}, while seasonal changes are also well expressed. The summer average values (0.12–0.89 μSv h^{-1}) are significantly higher than the winter ones (0.03–0.15 μSv h^{-1}). In summer, the dose rate values are steadily increasing along the tourist route from the entrance tunnel to the exit which, apparently, is due to the peculiarities of air exchange in the cave. The Pearson correlation coefficient between ambient dose rate and EEC is 0.70.

As can be seen from Fig. 9, despite the general trend of increasing the radon EEC and the gamma dose rate from the entrance tunnel to the exit, there is no strong correlation between these values. This is most likely due to the fact that the EEC of radon measurement was carried out only twice, in July and January, while the seasonal average values of the gamma radiation dose rate were obtained from the results of averaging monthly monitoring data in the cave.

Based on the existing ventilation regulations, the natural airflow in the cave is regulated: in warm weather (at air temperatures above 0 °C), the ventilation mode is used in the cave, in which the airlock doors are closed in the entrance and exit tunnels. Air exchange in the atmosphere is carried out through numerous cracks in the rock and the Old entrance. In cold weather (at temperatures below 0 °C) – the doors of the entrance lock are open, and the doors of the exit lock are closed (Krasikov and Kazantseva, 2019).

The results of the airflow velocity measurement show that in winter the airflow is directed from the entrances deep into the cave. The airflow velocity in the cave varies from 0.5 to 3.85 m s^{-1}, which is consistent with the previous data (1–4 m s^{-1}) (Kungur Ice Cave, 2005). In general, the airflow velocity gradually decreases with distance from the entrance, while the highest values are recorded in the narrows and corridors, and the lowest, in the grottoes.

The measurement carried out in the summer has shown that the air movement toward entrances is felt, but it is so small that its velocity is practically not recorded by the instruments, i.e., less than 0.3 m s^{-1}. According to the previous data (Kungur Ice Cave, 2005), the air velocity in the cave in summer ranges from 0.1 to 0.2 to 0.4 m s^{-1}, reaching the maximum of 1.2 m s^{-1}. The highest airflow rates have been recorded in narrow passages and tunnels. As shown in V. Lukin's studies (Kungur Ice Cave, 2005), the direction and velocity of air movement in the cave depend on the temperature of the outside air (Fig. 10). The temperature threshold at which the air movement in the cave stops is +5 °C. This temperature corresponds to the average annual temperature in the cave. At a higher temperature, the air moves to the entrance and exit tunnels, at a lower temperature, it enters the cave through the tunnels (Kungur Ice Cave, 2005).

4. Discussion

The results obtained show that significant seasonal radon variations are observed both at the abandoned Beshtaugorskiy uranium mine and in the Kungur Ice Cave despite the drastic differences in the geological structure, climatic conditions, radium and radon concentrations, and the origin of these underground spaces. Moreover, in both cases, the EEC of radon progeny exceeds the permissible level not only for the population (310 Bq m^{-3}), but even for the workers of uranium mines (1200 Bq m^{-3}) in some periods.

The pattern of seasonal changes in the direction and the velocity of air movement at the adits of the Beshtaugorskiy mine corresponds to the phenomenon of natural ventilation caused by the convective air circulation due to the temperature difference (chimney effect).

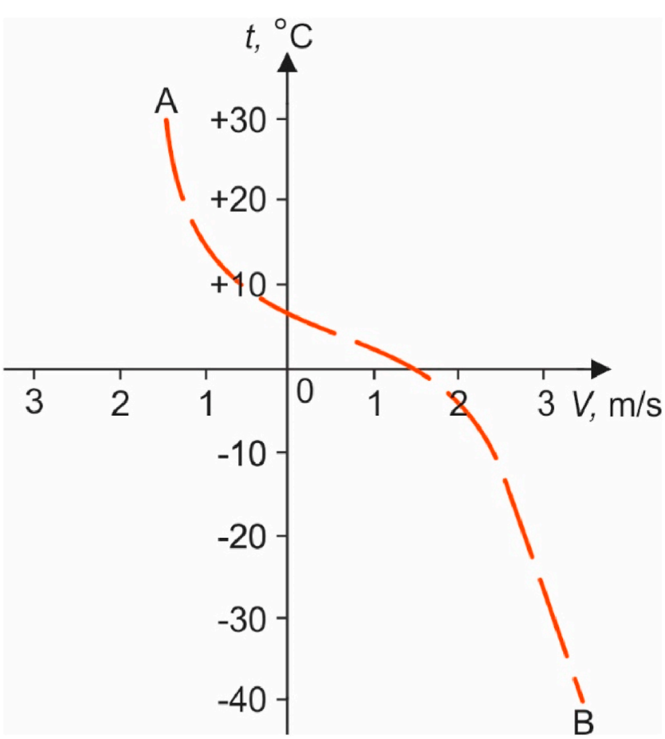

Fig. 10. Dependence of direction and speed of air stream at the Kungur Ice Cave entrance on the surface air temperature. *Legend*: A - summer air draught, and B - winter air draught (Kungur Ice Cave, 2005).

In the case of the Beshtaugorskiy mine, the general peculiarities of air movement in the system of underground passages are clearly visible. The temperature difference generates a pressure difference in the mine. In winter, the mine air is warmer than the outside one, which leads to an uprising of mine air to the top, creating a higher pressure at the higher elevations and a lower pressure at the lower ones. In summer, an inverse pressure distribution pattern is created. The level where the pressure inside the rock massif is the same, as the outside atmospheric pressure at the same elevation, is called the neutral pressure plane (NPP). In winter, radon-rich mine air discharges outward above the NPP and atmospheric air leaks inward to the mine below the NPP. In summer, cold mine air is released to atmosphere below the NPP, while the atmospheric air is sucked into the mine above the NPP. The position of the neutral pressure plane on Mt. Beshtau corresponds to an altitude of about 900 m a.s.l. (Fig. 7). The Beshtaugorskiy mine is a complicated dynamic system, so the vertical location of the NPP may change depending on the wind and atmospheric temperatures. Respectively, the adits located near the altitude of 900 m a.s.l. periodically turn out to be either above or below the NPP. On such sites, radon release can occur either in summer or in winter, depending on the current position of the NPP.

It should be noted that convective air circulation determines many parameters of the microclimate of underground spaces, including the temperature regime. In the adits located below the NPP, a lower mean temperature compared to the environment should be expected. Because cold mine air is discharged through them in summer, and cold air from the atmosphere is drawn in winter, which leads to the adit's cooling. On the contrary, in the adits of the upper horizons, the mean temperature may be higher than in the environment, due to the discharge of relatively warm mine air in winter and the entry of warm air from the atmosphere in summer.

As mentioned above, in the Kungur Ice cave, the air movement is also determined by the chimney effect. It is problematic to determine the exact elevation of the neutral pressure plane in the Kungur cave. However, the data obtained showed that the cave is entirely located below the NPP. The features of air exchange and the behavior of radon in the Kungur Cave generally correspond to those in the lower adits of the Beshtaugorskiy mine. In both cases, the underground space is intensively ventilated by atmospheric air in winter, which leads to a sharp decrease in the concentrations of radon and EEC. And in summer, radon-enriched air enters the tunnels and the grottoes seeping from above through the strata of rocks and sediments.

However, there are also differences due to both the different content of uranium and radium in the rocks, and the different climatic conditions of the regions under consideration. Significant differences in the radium content in rocks cause a difference of more than 100 times in the EEC of radon progeny, which reach 89,000 Bq m^{-3} at the mouth of adit # 10 of the Beshtaugorskiy mine and 6653 Bq m^{-3} in the Kungur cave. In addition, in the Beshtaugorskiy mine, the airflow velocity on the lower adits ranges from 0.4 to 6.8 m s^{-1} in winter and from 0.4 to 8.4 m s^{-1} in summer, i.e., it does not show significant seasonal fluctuations. At the same time, in the Kungur cave, as mentioned above, the winter values of the flow velocity vary from 0.5 to 3.85 m s^{-1}, while in summer the air velocity is only felt, but cannot be measured by instruments. This sufficient difference in summer and winter air velocity is probably because in the relatively harsh climate of the Middle Urals (T_{an} = +2.1 °C; T_{Jule} = +18.1 °C, T_{Jan} = −15.7 °C), in winter the temperature gradient between the cave and the surrounding atmosphere is much higher than in summer. The higher values of the air velocity in the Beshtaugorskiy mine compare Kungur cave are most likely associated with the large scale of underground cavities system. The elevation difference between the lower and upper levels of the Beshtaugorskiy mine is about 200–250 m, while the elevation difference between the entrance to the Kungur Ice Cave and the top of the Ice Mountain does not exceed 50–60 m.

It is generally considered that high radon concentrations in underground cavities are primarily associated with the accumulation of radon due to a decrease in air exchange. However, results obtained show that in the Beshtaugorskiy uranium mine, the maximum radon concentrations corresponded to the highest air velocities. The correlation coefficient between these parameters was 0.75. Even in the Kungur Ice Cave, weak airflows are present in summer, and long-term microclimatic studies conducted in the cave earlier (Kungur Ice Cave, 2005) showed that the volume of air entering the cave in the summer season is only 4 times less than in winter. Thus, increased concentrations of radon in the underground space can be formed not only due to low air exchange, but by the entry of radon enriched air from cracks and cavities in the rock. So, in summer, the air enters from the karst upland surface to the Kungur Ice Cave via the cracks and cavities across a layer of carbonate-sulfate rocks with a thickness of at least 50 m (see Fig. 5). Seeping through cracks, the air can significantly enrich with radon. Moreover, in the Beshtau mine, air moves up or down in the mining system for up to 200 m, seeping, among other things, through uranium ores.

The data obtained suggest that the convective air circulation (i.e., chimney effect) determine the movement of air and fluctuations in the radon concentration in various types of underground cavities. It is important to note that this effect prevails provided that caves or mines are located above the local base level of erosion and have entrances and other air channels located at different elevations. However, the last condition is almost always satisfied, because caves and tunnels can be dead-end only for humans, but not for air. The air penetrates relatively freely through numerous cracks and karst cavities that are always present in the rock massifs and comes from caves or mines to the outside in a variety of places. The pattern of radon variations in caves depends on the position of the cave relative to the neutral pressure plane. In the caves located below NPP, the highest radon concentrations are observed in summer, and in the caves located below NPP, high concentrations are recorded in winter. Studies conducted in a number of dead-end caves in Europe indicate that they also show significant seasonal fluctuations in radon concentration with maximum in summer most likely associated with air convection (Briestenský et al., 2022). The case of a cave with a winter radon maximum is the Castanar Cave, Spain (Alvarez-Gallego et al., 2015). In large multi-level systems of underground cavities with numerous entrances located at different elevations on slopes with different exposures, a complex mode of air exchange with various patterns in different parts of these systems is formed.

The results of measurements at the mouth of the tunnels of the Beshtaugorsky mine allow us to estimate the equilibrium factor F and its seasonal fluctuations. The annual average value of the equilibrium factor at all measurement points was about 0.4. At the same time, distinct seasonal fluctuations were recorded. In the lower part of the mine, below the altitude of 900 m a.s.l., the average values of the equilibrium factor were 0.2 in summer and 0.6 in winter. In the upper part of the mine, on the contrary, in summer the equilibrium factor averaged 0.6, and in winter it decreased to 0.2. Thus, in the mine air flow released to atmosphere the equilibrium factor is about 0.2, and in the atmospheric air drawn into the mine F is about 0.6, which completely coincides with the estimates given in (ICRP, 2017).

Extremely high values of EEC registered in the studied underground spaces require saying a few words about the effective doses from inhalation of radon, which the technical staff could be exposed. For the received annual average EEC values of 21870 Bq m^{-3} and 1130 Bq m^{-3} in Beshtaugorskiy mine (adit #10) and Kungur Cave, respectively, occupancy of 1296 h a^{-1} (4 h × 6 days × 54 weeks), and the dose conversion factor as given in (ICRP, 2017) for tourist cave, the following annual effective doses are derived (UNSCEAR, 2000):

$$\text{Beshtaugorskiy mine}: 21870\ \text{Bqm}^{-3} \times 1296\ \text{h} \times (1.5 \times 10^{-5})\ \text{mSv}(\text{Bq hm}^{-3})^{-1} = 416\ \text{mSv}$$

$$\text{Kungur Ice Cave}: 1130\ \text{Bqm}^{-3} \times 1296\ \text{h} \times (1.5 \times 10^{-5})\ \text{mSv}(\text{Bq hm}^{-3})^{-1} = 21,5\ \text{mSv}$$

Taking into account the significant seasonal fluctuations in the radon concentration in the studied underground spaces, the effective dose will also fluctuate throughout the year. According to calculations using the dose conversion factor mentioned above, the hourly effective dose rate in the adit # 10 of the Beshtaugorsky mine will be 0,65 mSv h^{-1} in summer and 3,8*10^{-4} mSv h^{-1} in winter. The hourly effective dose rate in the Kungur Ice Cave are 0,034 mSv h^{-1} in summer and 4,8*10^{-4} mSv h^{-1} in winter. The estimated effective doses agrees well with the values derived in the other natural caves (Martín Sánchez et al., 2015; Akbulut Özen et al., 2019; Fijałkowska-Lichwa and Przylibski, 2021) and abandoned uranium mines (Fijałkowska–Lichwa, 2016).

5. Conclusion

Studies conducted at the abandoned Beshtaugorskiy uranium mine (North Caucasus) and in the Kungur Ice Cave (Middle Urals) have shown that both in the mine adits and in the grottoes of the cave, strong seasonal radon variations are observed. The EEC of radon progeny in the air varies from 11 to 6653 kBq m^{-3} and from 10 to 89,020 kBq m^{-3} in the Kungur cave and the Beshtaugorskiy mine, respectively. It has been established that the variations are caused by the same process, namely convective air circulation due to the temperature difference between the interior of the mountain massif and the surrounding atmosphere (chimney effect). Both in the mine and in the cave, the air movement occurs similarly, despite significant differences in conditions and scale of these underground spaces. Due to convective air circulation, underground spaces are periodically intensively ventilated with atmospheric air, and then, on the contrary, they are filled with radon-enriched air that seeps into caves or adits from rocks and ores.

The pattern of radon variations in caves depends on the position of the cave relative to the neutral pressure plane. In the caves located below NPP, the highest radon concentrations are observed in summer, and in the caves located below NPP, high concentrations are recorded in winter. The data obtained suggest that the convective air circulation (chimney effect) determines the movement of air and fluctuations in the radon concentration in various types of underground cavities which are located above the local base level of erosion and have entrances and other air channels located at different heights.

The obtained values of the EEC of radon progeny periodically by several times exceed the International and Russian permissible levels for the work spaces (310 Bq m^{-3} in Russia) both in the Beshtaugorskiy uranium mine and in the Kungur Ice Cave. This requires improving our knowledge of the underground environment and underline the importance of radon monitoring to assess radiation risk to humans and workers. The next step could be the organization of continuous monitoring of radon in the Kungur Ice Cave, as well as in adits of the Beshtaugorskiy mine, where the organization of an underground radon and climate therapy center is planned.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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