Radon-rich waters in Russia

A. N. Voronov

Abstract In radon mineral curative waters, according to Russian mineral water classification, the radon concentration should be greater than 185 Bq/l. There are about 30 mineral waters with high levels of radon in Russia. Radon-rich waters have high therapeutic effects. It is proven that natural background radiation stimulates the human immune system. Radon is a natural radioactive gas that has no taste, smell or color. Radon-222 is one of the heaviest elements in the zero groups of inert gases. It is a gaseous radioactive element. All radon isotopes are α-emitters while the transformation of its decay products is accompanied by the emitting of α-or β-particles. The main products of radon decay are short-lived isotopes Po, Pb, Bi, and TL. Belonging to the uranium and thorium decay chain, radon isotopes form directly during the decay of radium isotopes. Therefore the radon concentration depends upon the concentration of its parent's isotope in water and rocks washed by it as well as upon the amount of radon emanation. Loose rocks or rocks with a great number of cracks are characterized by higher radon concentration (zones of tectonic disturbance, weathering crusts, etc.).Crystalline rocks usually have higher uranium concentrations than the average bedrock. Examples of rock types, which often have enhanced uranium concentration >5% ppm U includes the following: granites, syenites, pegmatite, acid volcanic rocks and acid gneisses. In the earth's crust radon migrates either in a gaseous or dissolved state. It can go to the surface without any chemical reaction. Formation of the radon-rich therapeutics waters of Russia has been analyzed, and most of them are genetically connected to crystalline acid rocks that have exceeded uranium-radium mineralization. The radon content in Russia reaches more than 8,000 Bq/l. Radon-rich waters of this type occur in the Altai, Karelia, St Petersburg and Trans-Baikal regions. Another type is connected to geodynamic activity of regions and secondary radioactivity. A well-known example of radon-rich waters of the second type is Pyatigorsk in the North Caucasus. Mixing confined carbon dioxide-hydrogen sulfide water and unconfined groundwater forms radon waters. The radon concentration is 1,170–2,430 Bq/l. The occurrence of radon-rich water deposits in other regions of Russia is described. Further investigation of the radon content in other geological environments will contribute to the environmental safety as well as to the solution to many genetic, hydrogeochronological, paleoreconstructive and prediction problems in hydrogeology.

Keywords Mineral water · Radon · Groundwater · Russia · St. Petersburg

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Introduction

Radon-rich water has the highest therapeutic effect among curative mineral waters. Mineral groundwater with radon as the main curative agent is widely consumed in the world. Much of it is used in health resorts and sanatoriums. Some aspects of using these waters have not yet been resolved. This is a mysterious type of mineral water deposit. Many hydrogeological and balneological aspects of radon-rich water are still to be studied. (For example, necessary dose, influence on human health, complex influence of several balneological components, generation of high-level radon concentration, preservation of chemical composition, etc.).

In radon mineral curative waters, according to Russian mineral water classification, the radon concentration should be more than 185 Bq/l (Voronov 2000). This border value is less symbolic because different countries have other standards. (Table 1). Besides, therapeutic effects can be regulated by the duration of radiation treatment.

There are two types of curative radon-rich mineral waters—pure radon waters and waters where, besides

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Table 1
Minimum admissible norms of radon concentration (Bq/l) for mineral waters in different countries

Italy	48
Poland	74
Russia	185
France	370
Czech	1,192
Germany	6,885

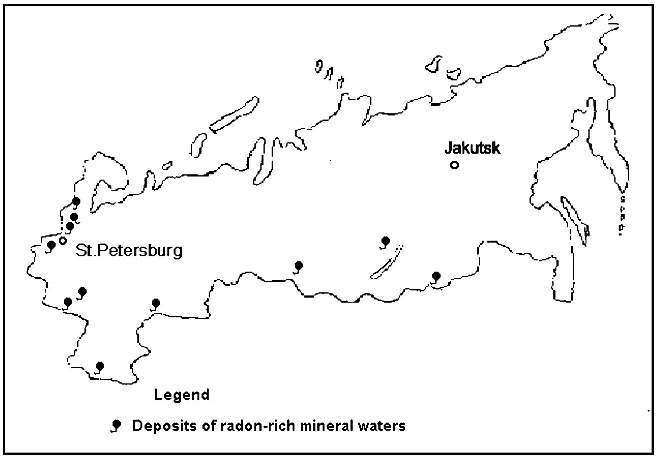

Fig. 1
Radon-rich mineral water deposit of Russia

radon, there are other balneological components, for example, iron, silicon, carbon dioxide.
There are about 30 mineral water deposits with high levels of radon in Russia (Fig. 1). They belong to different geological structures; among them there are sedimentary basins. Many Russian and foreign scientists studied radon in groundwater. A few references are: Ovchinnikov (1963), Ogilvi (1914), Kafri (2001), Przybilski (2000), Gudzenko and Dubinchuk (1987), Yu, and others (1994), Elmanova and Arbuzov (1981), Akerblom and Lindgren (1997).

Who are you, mister radon?

Radon was discovered about 100 y ago in 1899. Radon is a natural radioactive gas that has no taste, smell or color. Radon-222 is one of the heaviest elements in the zero groups of inert gases. There are three isotopes of radon—actinon, toron and radon. All radon isotopes are α-emitters while the transformation of its decay products is accompanied by the emission of α- or β-particles. The half-life of radon is only 3.8 days. The main products of radon decay are short-lived isotopes Po, Pb, Bi, Tl.
Belonging to the uranium and thorium decay chain, radon isotopes form directly during the decay of radium isotopes. Uranium is the first element in a long series of decay that produces radium and radon. Uranium is referred to as the parent element, and radon and radium as the daughter elements. Radium and radon also form daughter elements as they decay.
Radon is highly soluble in water: 500 ml of radon dissolves in 1 l of water at the temperature 0 °C. The dissolution of radon in water increases then lowers the temperature and concentration of other salts. Radon is 7.6 times heavier than the air, and for this reason, radon collects in the basements of buildings. The process by which radon escapes from the mineral grain of rocks is called "emanation". Emanation depends on several factors such as temperature, density and mineral composition of rocks. Radioactivity is commonly measured in picocuries (pCi), named after the French physicist Marie Curie, who was a pioneer in the research of radioactive elements and their decay. One pCi is equal to the decay of about two radioactive atoms per minute. Another unit is the Becquerel (system SI). One Becquerel is equal to one disintegration per second.

Radon in groundwater

The main sources of radon in groundwater are rocks. All rocks contain some uranium, although most of them contain very small amounts–between one and three parts per million (ppm). (Akerblom and Lindgren 1997). Some types of rocks have uranium contents higher than average. They are light-colored volcanic rocks, granites, and dark shale, sedimentary rocks that contain phosphate and metamorphic rocks derived from these rocks. Such rocks and their soils may contain as much as 100 ppm of uranium. Other sources of radon in groundwater are secondary deposits of radium salts.
Most of the radon produced within a mineral grain remains embedded in the grain, 10–50% escapes to enter the pore space. This value is called the emanation coefficient. It is a ratio of emitted and original radon. The emanation coefficient depends on the type of rock, its structure and porosity. Because of the presence of water in the pore space, the radon atoms can dissolve in groundwater.
Therefore, first of all, the radon concentration in groundwater depends on the radioactivity of rocks and the emanation coefficient. Another factor is the amount of water in the rocks and the velocity of the water circulation. Radon concentrations in intermontane water have the highest level. Water from uranium rich rock, e.g. uranium-rich granites and pegmatite commonly have radon concentrations in excess of 500 Bq/l, with maximum concentrations of 20,000–60,000 Bq/l. Groundwater from fissures of basic magmatic rocks has considerably lower radon concentrations. Fissure groundwater from limestone, sandstone, and shale has a normal radon concentration of 50–150 Bq/l. Sometimes water has higher radon concentration. Groundwater in the upper hydrodynamic zone and soil layers generally has a radon concentration no higher than 50 Bq/l.
This conclusion can be applied, of course, only to aquifers in which the radon originated. Groundwater flow from one aquifer to another and contact between groundwater and

Table 2
Maximum allowable concentration of radon in water for public water-supplies

Country	Bq/l
Finland	300
Sweden	100
Czech Republic	50
Ireland	100
Russia	60

Table 3
Classification of radon-rich mineral water

Class	Radon concentration, Bq/l
Very low radon water	185–740
Low radon water	740–1,480
Medium radon water	1,480–7,400
High radon water	>7,400

rocks with high concentrations of uranium and radium complicates radon distribution. Another source of radon-rich water is the build up of radium-rich material on the surface of cracks and fissures from which radon emanates directly into water in through there cracks.
The radon concentration in surface water may be as low as 2 Bq/l, mainly because radon has time to decay. Different concentrations of radon in water are due to different geological and hydrogeological conditions.
Some areas in the world have exceptionally high concentrations of radon, such as, Brazil, India, Iran, and Canada. There are many springs with high levels of radon in Iran. In Scandinavia, there are high regional levels of radon. The USA and Russia possess many zones with radon-rich groundwater. Because of different geological conditions, countries have different maximum allowable concentrations of radon in the water for public and domestic water use (Table 2).
In Russia "norms of radioactive safety", (NRS 1999) established a maximum allowable concentration for radon of 60 Bq/l. Previously, the maximum allowable concentration of radon in water for public use was 120 Bq/l.

Radon—enemy or friend?

Nowadays, much attention is paid to the harmful effects of radon and many countries have made special maps showing radon-hazardous areas. (United Nations Environmental Programme 1985).
Most radiation comes from natural sources due to the particular minerals in the area. Radon in water may be a health risk, due to the inhalation of the radon gas released from water and the ingestion of the water. When radon-rich water is used, up to 90% of the radon escapes into the air. The more the water is vaporized, processed or heated, the greater the release. In air, radon gas decays into its progeny, which is inhaled along with it, and whose radiation may cause lung cancer.
When radon-rich water is ingested, a dose of radiation impacts the digestive system from the radon gas and its progeny. The radon decays in the body and the continued decay of radon's short and long-lived progeny, impart radiation to the various organs of the body, which is the greatest risk associated with the ingestion of water containing radon. Radon and radon progeny are considered to cause stomach-colon cancer and other organ cancers. Radon is also suspected as a cause of leukemia.
A large doze of radiation may not always prove fatal. It depends on the individual peculiarity of the human organism and complex genetic and environmental factors. Some consequences appear only after many years, sometimes only in later generations.
At the same time the radon irradiation may be beneficial for human health. This is supported by the long history of the use of radon-rich water in balneology. Short-time radiation treatment may mobilize the protective strength of the human organism.

Mineral radon-rich waters

Mineral groundwater with radon as the main curative agent is widely consumed in the world. The famous resorts of Jachimov, Baden-Baden and Brombach use radon-rich water. Radon water is used in water and mud baths, drinking, and air inhalation. It is also used for the treatment of the nervous system, cardiovascular system, and pulmonary system and for rheumatic conditions. Concentrations of radon in curative mineral waters vary widely. For example, the radon concentration in Barbanstein (Austria) is 2,200 Bq/l, Baden-Baden (Germany) 780 Bq/l, Jachimov (Czech Republic) 6290 Bq/l. In Russia the radon concentration in mineral curative waters can be more than 185 Bq/l. Radon-rich groundwaters are divided into four classes (Table 3).
Radon-rich waters are divided into two groups by their chemical composition. The first group is groundwater, where radon is the only balneological component. The second group includes groundwater with more complex chemical composition. Groundwater of the first group has mineralization less than 1 g/l. Groundwater of the second group includes water with a high concentration of iron, carbon dioxide, and silicon.
The genetic classification of radon-rich water is interesting. The first group is connected with acid crystalline igneous rocks with high levels of radioactive minerals. A high-radon concentration in fossil groundwater is also caused by low-water saturation of rock and its disunity. The last one produces the emanation coefficient.
This is the most widespread group of radon groundwaters. Many of them are connected with areas of crystalline massifs and areas of postpaleozoic folds, such as are Baltic, Voronezh, Aldan and Ukraine massifs and in the Baiykal, Ural and Altay fold areas in Russia.
The second genetic group of radon-rich water is connected with zones of contact of the aquifer with uranium-bearing

rocks. Such deposits are widespread, for example in sedimentary artesian basins in contact with crystalline acid rocks. They may also occur in sedimentary rocks with regionally or locally high levels of radioactive minerals in artesian basins (Russian, East Siberian platforms).
The third group is associated with secondary deposits of radium minerals. Such conditions may produce thermal groundwater with dissolved radium salts. A secondary deposit of radium produces radon, which dissolves in the groundwater. Examples are found in the northern Caucasus.
An optimal combination of generation and emanation of radon and groundwater flow is necessary for the formation of radon-rich groundwater

Examples of radon-rich groundwater deposits

The long period of exploitation and study of radon-rich mineral water allows fa full picture of the formation of deposits of all three types (Elmanova and Arbuzov 1977).

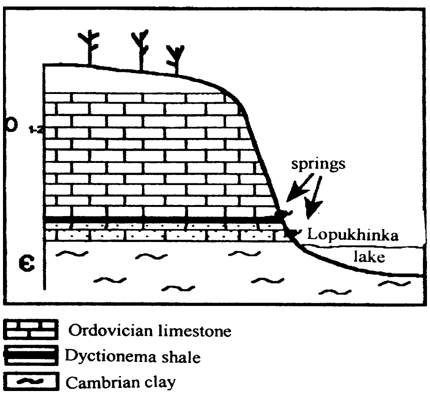

Fig. 2
Schematic cross section of the cliff near Lopukhinka Lake

All fossil deposits of radon mineral water moving in crystalline rocks belong to the first genetic group. There are Baltic crystalline deposits in the northwestern part of Russia, for example, the Pittkiranta deposit. This deposit is on the northern coast of Ladoga Lake, near the small settlement of Pittkiranta. Radon-rich water flow is connected with granite seams. There is an upper hydrodynamic zone. Mineralization of water is 0.1 g/l and pH is 7.2. Concentration of radon in the groundwater of flowing wells is about 4,400 Bq/l. The radon-rich water is caused by circulation of infiltration water in the fissured zone of uranium-rich crystalline rocks (granite). Concentration of radium in groundwater is $8{\times}10^{-12}$ g/l.
Another example of deposits of the first group is Belokurikha, which is in the northern part of the Altay Mountains; it is not far from the town of Biisk. This name means "white vapor" due to clouds of vapor near the springs, because the temperature of the groundwater is 60 °C. Massif "Belokurikcha" is biotite granite, which is very fissured and occupies an area of about 500 km^2. Therefore, the recharge zone is very large.
Thermal waters have very low mineralization (less than 0.3 g/l). Sodium is a predominant cation and sulfate is a predominant anion. Radon concentration is up to 480 Bq/l. Groundwater contains silicon (concentration up to 0.58 g/l). It is an additional curative factor. Belokurikcha is a very popular spa in Siberia.
Lopuhinka is an example of radon-rich deposits of the second type (Voronov 1997). It is in the southern part of the Izore Plateau, not far from St. Petersburg. The main aquifer is an Ordovician limestone above a regional aquiclude, which consists of Cambrian clays (Fig. 2). The thickness of the aquifer is 50 m. The main source of groundwater is precipitation and the water has a bicarbonate chemical composition. Mineralization of groundwater is 0.4 g/l. The aquifer is underlain by dictionema shales, which contain a great concentration of uranium. Concentration of uranium is 0.01–0.17%. The contact of the shales with the aquifer leads to water enrichment by radon. Springs along the glint (southern part of the Izora Plateau) form a small lake with high levels of radon.

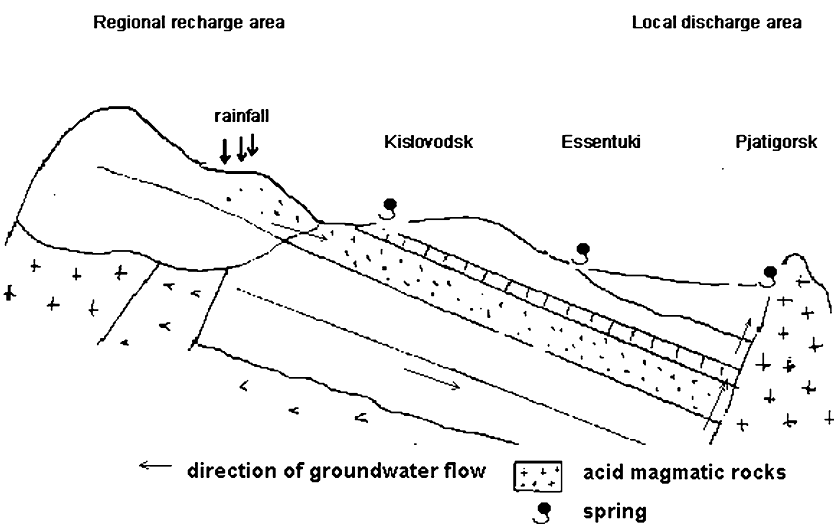

Fig. 3
Schematic section along the groundwater flow path of Caucasian mineral waters

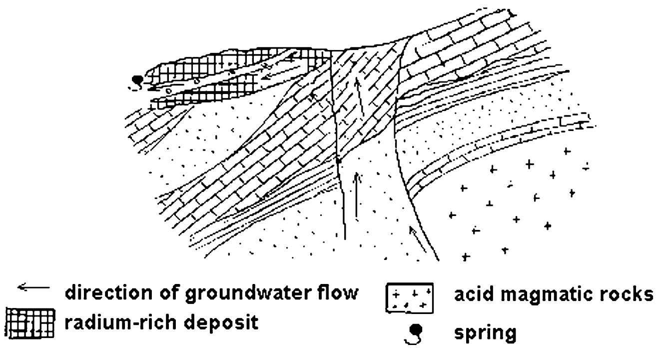

Fig. 4
Scheme of Pjatigorsk deposit of radon-rich mineral water (after Elmanova and Arbusov 1977)

Concentration of radon in the springs is about 200–400 Bq/l and during the middle of the nineteenth century it was a very popular health resort near St Petersburg. It is very curious to note that nobody knew about radon at this time!

One of the most well known and interesting deposits of radon-rich mineral water of the third type is at Pyatigorsk in the northern Caucasus, where there are many other springs of mineral water of different composition, for example Narzan and Essentuki (Fig. 3). These waters are bottled and sold in different parts of Russia.

The geology of this area is well known, as many Russian hydrogeologists studied them (Ovchinnikov 1963; Ogilvi 1914; Slavjanov 1936). The aquifer consists mainly of carbonate rocks, which are found at progressively greater depths away from the recharge area in the mountains and in the direction of the discharge zone in the central part of the Azov-Cuban basin. The aquifer is interrupted by a number of volcanic intrusions, which form picturesque mountain peaks, for example Mashuk, Verblud, and Lysaya. In these zones the CO_2-rich groundwater rises through a series of fractures to emerge at the surface in the discharge zone. The chemical composition of the waters varies greatly, ranging from bicarbonate to chloride-magnesium.

The radon-rich water of Pyatigorsk occurs beside the mountain Mashuk (Fig. 4) at a contact of sedimentary and acid-igneous rocks. The thermal groundwater containing radium rises through fissures and, losing carbon dioxide, pressure and temperature, forms deposits of radium salts (travertine). The deep water contains radium up to 3.5×10^{-11}. Mixed groundwater flow (infiltragenic and deep water) receives radon from travertine. The radon concentration in these waters varies greatly from 600–1,300 Bq/l. It depends on the emanation coefficient, water flow and generation of radon. The mineralization of groundwater varies greatly between 0.4–4.6 g/l. The chemical type of the water is still permanent—chloride-sodium. The temperature of the groundwater is 18–24 °C.

Conclusion

The use of radon-rich waters has an important future, owing to their wide distribution and therapeutic effect. New resorts which can be located at radon-rich water springs and wells are in picturesque landscapes with exclusive cultural and historical value.

There remain questions of origin and use of radon-rich waters that have to be analyzed. For example, is the use of radon-rich waters useful or harmful? What are other balneological effects of water on human health?

Also, in the field of investigation of radon-rich water, there are theoretical problems of origin and distribution of this water, water quality, and its protection from contamination.

References

Akerblom G, Lindgren J (1997) Mapping of groundwater radon potential. European Geologist 5. European Federation of Geologists, pp 13–22

Elmanova NM, Arbuzov BA (1981) Radioactive waters. In: Hydrogeochemia and geochemia of curative mineral waters (in Russian). Nauka, Moscow, pp 62–73

Gudzenko VV, Dubinchek VT (1987) Radium and radon isotopes in natural waters (in Russian). Nauka, Moscow, 158 pp

Kafri U (2001) Radon in groundwater as a tracer to assess flow velocities: two test cases from Israel Environ Geol 40(3):392–398

Norms of Radioactive Safety (NRS) (1999) Goscomepidemnadzor (in Russian). Moscow, 115 pp

Ogilvi AN (1914) The problems of origin of Essentuki mineral water (in Russian). Russian Academy of Sciences, St. Petersburg

Ovchinnikov AM (1963) Mineral water (in Russian). Gosgeoltechizdat, Moscow, 375 pp

Przylibski TA (2000). Radon-222 concentration changes in medicinal groundwaters of Ladek Zdroj (Sudety Mountains, SW Poland). J Environ Radioact 48 (3):327–347

Slavianov NN (1936) Mineral waters springs of North Caucasus (in Russian). In: Guidebook on water recourses of the USSR. Gosgeoltechizdat, Leningrad

United Nations Environment Programme UNEP (1985) Radiation, doses, effects, risks. New York, 80 pp

Voronov AN (1997) Radon in groundwater of Leningrad oblast (in Russian). ProcSt. Petersburg Univ 7(1):22–28

Voronov AN (2000) Some features of mineral waters in Russia. Environ Geol 39(5):477–481

Yu KN, Guan ZJ, Young ECM (1994) A preliminary study on the radon concentrations in water in Hong Kong and the associated health effects. Appl Rad Isotopes 48(7):809–810